HW-3-3442 [redacted]

719097

ACCUMULATION OF RADIOACTIVE ELEMENTS IN FISH IMMERSED IN PILE EFFLUENT WATER

HW--3-3442

DE87 006096

I. Introduction

The discharge into the Columbia River of Pile effluent water containing radioactive elements has led to problems on the effect of the radiations on fish living in the river. One aspect of this concerned the accumulation of radioactive materials in fish. Some work has been done on the metabolism of mixtures of fission products and of several specific elements,(1) but not with the particular mixture of materials discharged in the Pile effluent. The Fish Laboratory provided suitable material for the assay of concentration in fish at this site. This laboratory was under the supervision of R. F. Foster (P Department) who very kindly made special exposures of fish to active water for some tests.

The activity of Pile effluent water arises mainly from Na^{24} and Mn^{56} with about 1% of miscellaneous longer lived elements. The half life of the Mn^{56} is 2.5 hours, requiring rather rapid analysis of fish in order to determine the concentration at the time of removal from the active water. Na^{24} activity, with 14.8 hours half life, could be measured with ease.

The contribution of the H.I. Section as described in the following report was in no way directly connected with the welfare of the fish, but was carried out in an attempt to measure the accumulation of activity so that the dose received by any organ could be calculated.

(1) CH-3233 "Accumulation and Distribution of Radioactive Strontium, Barium-Lanthanum, Fission Mixture and Sodium in Goldfish" by C. Ladd Prosser, Wm. Pervinsek, Jane Arnold, George Svihla and P. C. Tompkins, February 15, 1945

II. Summary

The concentration of active materials in the fish depends to a large extent upon the organ chosen for study. A table of concentration factors (ratio of activity in the fish to the activity in the same weight of water) for steelhead trout is given below.

Table I

	Ratio $\frac{\text{Fish Activity}}{\text{Water Activity}}$	
Organ	Average	Maximum
Liver	30	44
Stomach	16	21
Kidneys	31	48
Muscle Tissue	6	11
Vertebrae	20	33
Gill Arches	32	44

The maximum activity in a fish exposed to this water appears to be reached in 24 hours. After this time, the activity will apparently fluctuate with any long-period changes in the concentration of the water and the concentration of other inactive materials.

The accumulation of long lived activity in such fish as trout and salmon kept in the effluent water is apparently inappreciable. Some material similar in radioactive properties to P^{32} is built up in the suckers feeding from the bottom of the river at the end of the spillways.

III. Procedures

It is to be expected that the skin and scales of any fish in active water would become highly contaminated since these portions are in constant contact with the water. As a result, it was necessary to perform the dissections with extreme care to prevent the spread of activity from one portion to another.

The general procedure was to pin the fish to a board, and by a median ventral incision lay the body cavity open. A new knife was then used to remove the organs. The intestines were clamped off to prevent fluids from flowing into the body cavity, and the organs to be analyzed were then removed. A section of the vertebrae was cut out, along with the surrounding muscle tissue. The tissue was scraped off and the portions treated separately. Each sample was weighed so that the relative concentrations of active materials could be obtained from the total activity present.

[redacted]

Samples were ashed in platinum crucibles in a muffle furnace at 700-800 degrees Centigrade. Residues were dissolved in nitric acid, and transferred to one inch watch glasses.

This ashing method required considerable time for both the ashing and the final evaporation of the nitric acid solution of the residue. A slightly different method was adopted when the primary object became the measurement of short lived material. In this method, the sample was placed on a one inch diameter platinum dish, weighed, and ashed at 700-800 degrees Centigrade by placing the dish directly in the furnace. Several applications of concentrated nitric acid greatly speeded up the ashing process. Samples could be dissected, ashed, and ready for counting within 3-6 hours by this process.

The procedure for analyzing salmon eggs was to boil the egg until the contents were solid, and then carefully peel off the outer shell. Ashing of these pieces was done in solution by the use of hot nitric acid and hydrogen peroxide.

The counting was done on the top shelf of a mica window beta set. Samples were counted for five minutes to one hour, depending upon the total activity present. The results were then calculated to microcuries per kilogram on the basis of 20% geometry.

IV. Results

A. Preliminary Search for Long Lived Activity

1. In Fish Laboratory

The first analyses were performed on 9/14/45. The dissections and further techniques were purely exploratory and the accuracy was consequently rather low. A small amount of activity was detected in the organs, bones, flesh and skin. Decay curves indicated that this activity was mainly Na^{24} with some trace of a longer lived activity.

The second group of fish consisted of five salmon that had been in the water for 30 days and out 3 days before counting. These fish were dissected with extreme care to prevent the spread of contamination. Results were as follows:

Table 2

Activity of Five Salmon from Process Water

Portion	μc/kg (dry)	μc/kg (wet)
Liver	0.5	0.1
Stomach	0.28	0.08
Muscle Tissue	0.21	0.05
Vertebrae	0.32	0.13

Decay curves on these portions i[illegible] the activity after three days [illegible] 70-95% 12-15 hours, and 5-10% of an unknown long lived activity. If the main activity is assumed to be that of Na^{24} and is extrapolated to zero time, the concentration would then be 30-50 times the concentration in the water.

On 10/17/45, six trout were analyzed in two batches of three specimens each. One batch was from contaminated water and the other was from clean river water. The contaminated fish had been in for 48 days and out 12 days. The activity of all portions was less than 0.0[illegible] microcuries per kilogram on both batches, indicating that very little long lived material was present.

2. In River

A sucker was caught in the Columbia River, near a discharge point for the effluent water. The activity of the various portions as counted three days later is given in Table 3.

Table 3

Activity of Sucker from River at 100-F

Portion	μc/kg (wet)
Muscle Tissue	0.08
Liver	0.19
Kidneys	0.29
Vertebrae	0.05
Skin and Scales	0.21

Decay curves on these samples have indicated that the activity had a half-life of 14-15 days. Absorption of the beta radiation in aluminum indicated an energy of about 1.6 MEV. This activity is comparable with that of P^{32}, which has a half-life of 14.3 days, and a beta energy of 1.7 MEV. Mud samples from the bottom of the river at this point gave only 0.02 μc/kg. Energy and decay rate were not measurable on this sample.

B. Measurements of Short Lived Activity

1. Water Measurements

A series of measurements was made on water obtained from the troughs during the period over which the fish were analyzed for the shorter lived activities. These measurements gave values of 0.2 to 0.28 microcuries per liter with an average value of 0.25 microcuries per liter. One water sample taken while the water in the troughs was somewhat murky was allowed to stand for a short time before testing. This sample was then divided into three portions and measured. The two portions from the top of the bottle gave values of 0.23-0.25 μC/L, while the bottom portion gave 0.86 μC/L. This indicates that the flocculent material in the water adsorbs the activity quite strongly.

2. Long Exposures

Three trout that had been in effluent water for five months were dissected and analyzed. Two of the trout (A and B) were removed while the water was clear, and the third (C) was removed while the water was quite dirty from the back-washing of the filters in the water purification area. This last fish had concentrations in the liver, kidneys, and gill arches considerably less than was obtained in the same organs of the other two fish. This was possibly related to the flocculent backwash material. The values obtained on the individual organs of each fish are given in Table 4. All of these activities have been extrapolated from the time of counting to the time of sampling by means of a 15 hour half life.

Table 4

Activity of trout from Effluent Water

Fish	A	B	C
Portion	μc/kg	μc/kg	μc/kg
Liver	10.0	9.5	5.4
Stomach	4.6	5.3	4.9
Kidneys	7.8	12.0	3.9
Muscle Tissue	0.9	1.1	2.8
Vertebrae	3.9	7.4	2.9
Gill Arches	8.8	9.5	0.97

3. Buildup of Na^{24} Activity

In an attempt to explain the changes in activity noted in the three trout analyzed for Na^{24}, a series of fish formerly kept in clean water was exposed to effluent water for times varying from 6 hours to 72 hours. One fish weighing 30-50 grams was used for each exposure time. The times were arranged so that the fish for the 6 hour exposure came out first, the two fish for the 16 and 24 hour exposures came out next, and the two fish for the 48 and 72 hour exposures came out last. The results for the individual organs are given in Figures 1 and 2 attached. In every organ, a maximum is reached in 16-24 hours followed by a significant decrease in the 48 and 72 hour exposures. The overall half-life of the activity in the various organs during the last 48 hours of the exposure is between 30-40 hours. Thus the organs were picking up only about half as much activity as they were losing by radioactive decay during this period.

A comparison of this data with that obtained in the long exposure of the trout shows that the fish removed at 16 and 24 hours correspond approximately in activity to the first two fish removed after a long exposure, while the fish removed at 48 and 72 hours correspond approximately to the last fish removed while the water was dirty. The original buildup of activity [illegible] the three organs which have already been described [illegible] the [illegible] activity [illegible] dirty water.

[redacted]

1140322

The accumulation of activity in non-feeding Chinook salmon fingerlings was also measured. In this case, the exposure times were adjusted so that all the fish came out of the water at the same time. Since the fingerlings were quite small, they were ashed as a unit with no attempt at dissection. The average concentrations as measured by two fish for each exposure time are given in Figure 3. The concentrations are of the order of magnitude expected for the average from the data already obtained for the trout. The slight maximum may well be due to individual fluctuations in the fish, since the individual values for the two fish at 72 hours were 4.4 and 3.6 µc/kg, while the values for the two fish at 24 hours were 4.4 and 4.5 µc/kg. Within the apparent accuracy of both series of tests, these concentrations are in approximate agreement with the results of Prosser, Pervinsek, Arnold, Svihla and Tompkins. The concentration phenomenon is probably a function of the total sodium ion concentration present, which in this case was about 20 ppm.

4. Accumulation of Mn^{56}

No specific measurements were made for Mn^{56} since all analyses were for gross activity. However, a series of half-lives of the activity from the fish gave values of 14-16 hours, while the activity from the water gave values of ~ 12 hours. These measurements were made on the basis of two counts about 20 hours apart, with the first one two hours after sampling. The fish, then, contained little or no Mn^{56} activity, while the water contained about 10-30% of this material.

5. Activity of Salmon Eggs

One group of salmon eggs that had been in cooled process water for 22 hours was examined. The techniques were purely exploratory, and the data was of rather low accuracy. However, the activity inside the shell was low (<0.03 µc/kg) indicating that little sodium or manganese is absorbed through the tough outer shell of the egg.

J. W. Healy
Senior Supervisor
H. I. Special Studies

JWH:gwc

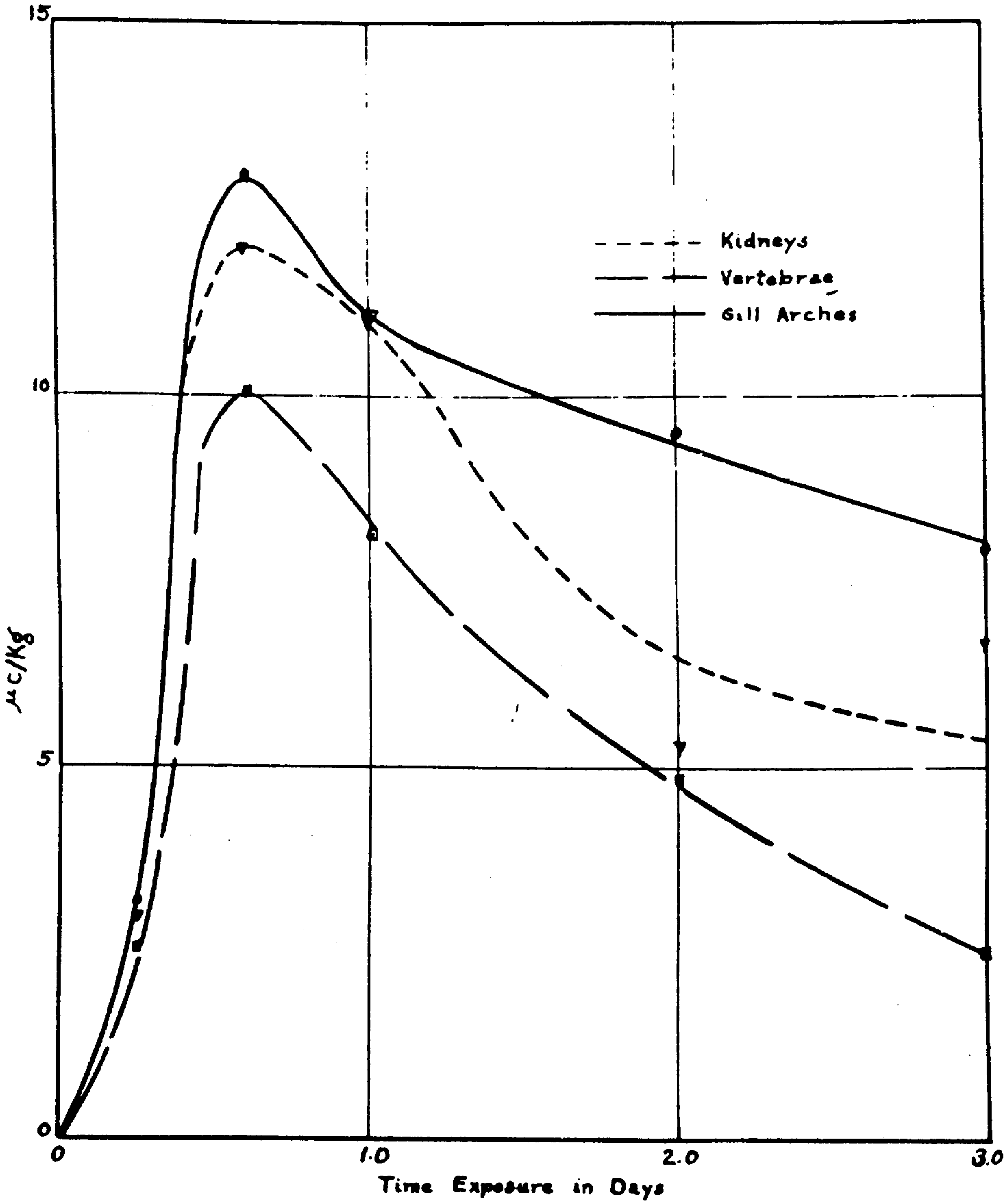
[redacted]

1140323

[redacted]

2-27-46

Fig 1
ACTIVITY INCREASE
In Steelhead Trout in 105 Water
1/8/46 – 1/11/46



[redacted]

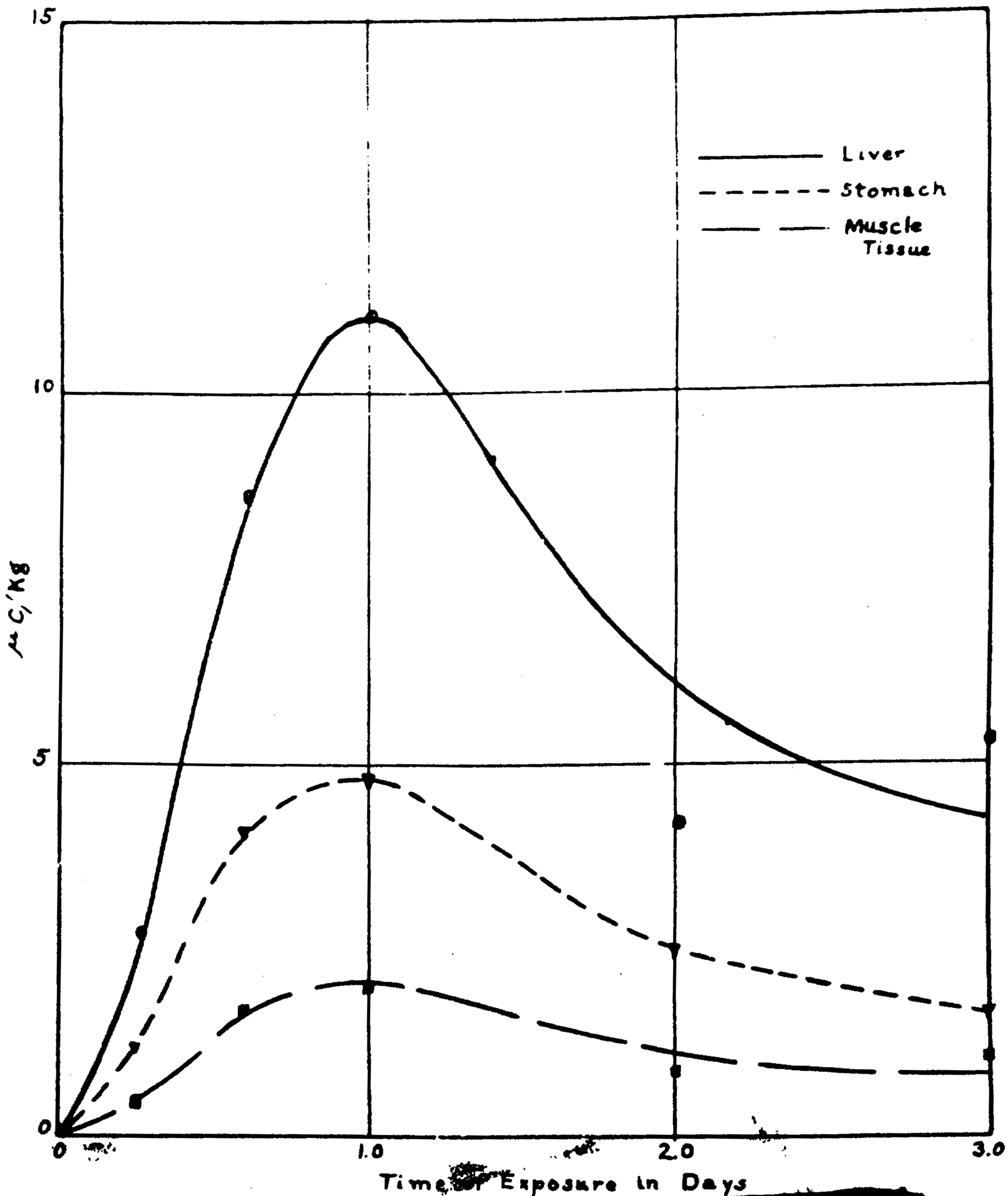
1140324

[redacted]

7-3514
2-27-46

Fig 2
ACTIVITY INCREASE
In Steelhead Trout in 105 Water
1/8/46 - 1/11/46

[redacted]

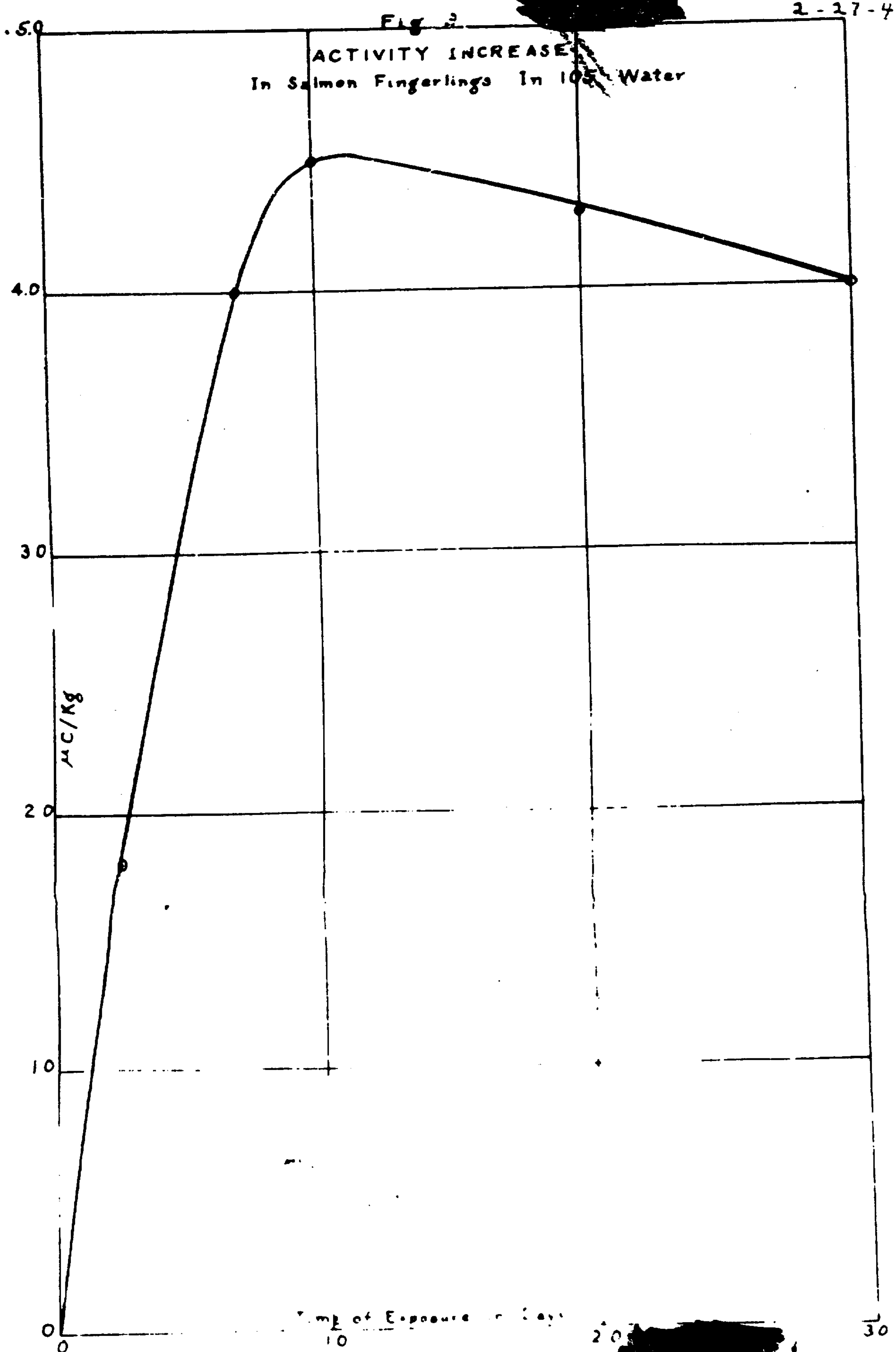


[redacted]

SECRET

1140325

1-35.5
2-27-46



1140326