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THE UPTAKE AND RETENTION OF P^{32} AND Zn^{65} FROM THE CONSUMPTION OF COLUMBIA RIVER FISH

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THE UPTAKE AND RETENTION OF P^{32} AND Zn^{65} FROM THE CONSUMPTION OF COLUMBIA RIVER FISH (a)

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INTRODUCTION

The Hanford plant in Southeastern Washington uses Columbia River water to cool several large plutonium producing reactors. The cooling water, after passing through the reactors, is discharged to the Columbia River. In its passage through the reactors, the water accumulates small concentrations of radioactive materials, largely the result of neutron activation reactions. In the Columbia River these radioactive materials enter into numerous biological, physical, and chemical reactions which affect their distribution. Some, entering the algae - insect - fish food chain, are reconcentrated and contribute to the radioactivity in some of the food fish removed from the river downstream by fishermen. This pathway of radiation exposure to the public was determined to be the most significant of the several possible pathways which have been examined in detail (1). The radionuclides which prove to be of most interest in this exposure pathway are P^{32} and Zn^{65}, because both zinc and phosphorus enter metabolic processes.

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From the standpoint of exposure dose resulting from eating Columbia River fish, the radionuclide of greatest significance is P^{32}. It represents by far the majority of the radioactivity in Columbia River fish. Detailed analyses of the possible exposure dose to individuals from this source depend on the amount of Columbia River fish considered to be part of their diet. Based on liberal assumptions concerning the amount of fish consumed by individuals, it is possible to estimate that they could obtain up to 40 per cent of the permissible body burden of this radionuclide. The parameters which were used by various agencies to arrive at the permitted body burden of P^{32} were evaluated on the basis of experimental evidence. This experimental study was conducted to contribute to this evidence. The experiment studies the uptake and retention of P^{32} by volunteer subjects, making use of the available Columbia River fish which are already tagged with P^{32}. Similarly, the uptake and retention of Zn^{65} from these fish was also studied because, although Zn^{65} does not significantly contribute to the exposure dose from eating Columbia River fish, it is sometimes used as a convenient measure of the amount of fish in individual diets.

The amount of radioactive material found in fish from the Columbia River is known to be a complex function of fish species, season, river flow level and temperature, reactor operating conditions, and the location of the catch in the river. The concentration of a radionuclide in several samples from a selected species of fish taken from a given sampling location on any given date will display a wide variance about their mean. The variance contributed by these factors determines the confidence with which one can estimate the radiation exposure from fish consumption. It is generally necessary to choose some average value for all the fish

sampled and to depend on the application of conservative assumptions to assure that the resulting estimate is as high or higher than an actual case is likely to be. In Figure 1 are shown the P^{32} and Zn^{65} concentrations averaged for all fish samples taken from the Columbia River during a three-year period. No attempt is made here to distinguish species. The seasonal effect on concentration is clearly evident from the monthly averages shown.

The experiment encompassed an eleven-week period during which Columbia River fish were eaten by seven volunteer subjects. In an effort to make sure that sufficient P^{32} would be present to permit ready analysis of the subjects' body burdens and excreta, a deliberate attempt was made to obtain fish apt to be high in P^{32}. Columbia River whitefish were chosen because they normally contain the most P^{32} and are easy to catch during the fall season. The fish were obtained from the sampling sites which were known to provide some of the highest concentration levels. The experiment began with an intense consumption schedule of one meal of fish per day for five days followed by a period of one meal of fish per week. During the initial week and immediately following the end of the experiment, excreta were collected from the subjects and analyzed for Zn^{65} and P^{32}. Figure 2 displays the course of the experiment. As can be seen from this figure, regular body burden measurements were made for both P^{32} and Zn^{65} during the ingestion schedule and during a decay period following. As a matter of fact, the individuals' Zn^{65} body burdens are still in a process of decay.

To simplify the problem of assuring a uniform ingestion rate, a large enough supply of fish for all subjects was obtained and filleted each week. The fillets were then ground in an ordinary food chopper and

thoroughly mixed. The chopped fish was sampled for laboratory analyses and the remainder weighed in 200-gram portions. These 200-gram portions were frozen until they were eaten by the subjects. Analyses of duplicate samples of the mixed ground fish showed excellent uniformity. The subjects ate the entire portion provided them, preparing the fish in the form of stew or a casserole dish to avoid any losses from frying or broiling.

PHOSPHORUS-32

The P^{32} body burden accumulated as a result of eating Columbia River fish was measured using a special beta counter developed and calibrated for P^{32} in the human body. A description of the counter and its operation has been reported by Palmer [2]. The development and successful demonstration of this counter made possible the measurement of P^{32} *in vivo* for the first time and encouraged the design of this experiment. This detector consists of a 6" x 6" x 1" gas flow proportional counter mounted on a 5" x 1 mm thick NaI crystal and a photomultiplier tube, the counters connected through a coincidence circuit. Beta rays emerging from the surface of the skin produce pulses in the NaI counter and the proportional counter at the same time and are then accepted by a 400 channel pulse height analyzer. Gamma rays and X-rays which do not efficiently produce pulses in the proportional counter are rejected, thereby reducing the background of the counter significantly. The detector was placed at the side of the head of the subject during the counting time of twenty minutes. This particular geometry was used to obtain a minimum background from the influence of K^{40} beta rays. The average counting rate at the side of the head was 1.0 counts per minute per nCi of P^{32} in the body and the

detection level was 40 nCi of P^{32} at the 95 per cent confidence level.

The instrument used to make whole-body measurements of P^{32} actually measures the beta emission from a rather small volume of flesh in the cheek. Thus, the P^{32} in a combination of body tissues including blood, muscle, and skin is detected. The natural K^{40} content of these tissues gives a rather high background, equivalent to a body burden of about 150 nCi of P^{32}. This background is large compared to the P^{32} concentrations that were obtained as a result of eating Columbia River fish, and represents the most severe limitation on the precision of our data. Also, the instrument does not detect deposition of P^{32} in deeply buried organs and tissue such as bone, except as this deposition affects the P^{32} content of blood, muscle, and skin.

The problem of phosphorus metabolism was examined with the analog technique developed by Watson, et. al.(3). It was possible to make only a single test using the best transfer parameters readily available. The resulting hypothetical P^{32} concentrations in various tissues were plotted from the analog program. These results are filled with many uncertainties because of our faulty knowledge of the values of the transfer coefficients. However, the curves probably show the relative positions and shapes of most of these concentration values and give us some idea of how the P^{32} body-burden measurements can be interpreted. The analog data indicate that the concentration of P^{32} in skin is very low and this was borne out by our failure to detect P^{32} in samples of skin scrapings. The shape of the concentration curves for blood and for muscle are shown in Figure 3. In general, it is evident that blood concentration has a prompt acute peak while the muscle concentration curve reaches approximately the same

values at a somewhat later time and decays off much more slowly. Using these curves as a guide, and actual whole-body counting points, a rough composite curve might be shaped as shown in the upper solid line at the left in Figure 3. The actual points obtained tended to indicate the double-peak shape. To the right on a considerably longer time scale are shown the P^{32} decay curves obtained from the whole-body counting instrument for three subjects. The best fit decay curves for these three subjects have slopes representing effective P^{32} half-lives of 10.2, 9.6, and 8.7 days, respectively. The composite points for all three subjects give a best fit slope of 10.2 days.

From the whole-body data obtained very early in the experiment, it was determined that all subjects absorbed P^{32} quite completely. The comparison of body burdens and diet gave P^{32} uptake fractions indistinguishable from 100 per cent. The best interpretation of these early results would indicate that all subjects studied had greater than 95 per cent P^{32} absorption.

In Figure 4 the body burden measured for each of seven subjects eating Columbia River fish is shown during the course of the experiment. The points near time zero indicate the measured background for the individuals, which was subtracted from subsequent whole-body measurements. The solid line is a calculated body burden assuming an effective P^{32} half-life of 9.3 days and a fractional uptake of 1.0. These values were obtained from the first analysis of the experimental data and may be revised by more detailed study. From these data, it is clear that a diet of 200 grams of Columbia River fish per week may be expected to result in a P^{32} body burden of around 200 nCi with the fish concentrations obtained during

this experimental period. The scatter in the various points reflects both the individual variations among subjects and the uncertain precision of our measurements. Note that the maximum body burdens attained were little more than four times background. The scatter in the final measurements at the end of the experiment was too great to permit adequate decay measurements with these data. However, they do not seem to be in marked disagreement with the 9.3 day half-life used in the theoretical calculation.

One of the difficult problems encountered in applying health physics data to estimates of exposure for individuals in the population is the uncertainty concerning individual variations in the pertinent metabolic parameters. It was for this reason that seven subjects were utilized in this experiment. The scatter of the whole-body counting data shown in Figure 4 reflects, at least in part, the individual variations in P^{32} retention by the subjects of this experiment. All of the subjects had the same P^{32} ingestion schedule.

ZINC-65

Zinc-65 has a suitable gamma component to permit whole-body measurements using a conventional whole-body counter. The Hanford whole-body counter has been described elsewhere [4]. For purposes of this experiment a 9 3/8" x 4" NaI(Tl) crystal shielded by 26 cm of iron was used. A counting time of twenty minutes per measurement permitted an accuracy of $\pm$ 5 per cent and a lower detection level of approximately one nCi.

In the absence of a method for measuring P^{32} *in vivo* it has been our custom to estimate what possible P^{32} body burdens might result from a fish consumption reflected by the Zn^{65} body burdens measured. For this

reason, it was of considerable interest to us to measure the Zn^{65} content of the subjects eating Columbia River fish and to compare these data with the P^{32} levels. An earlier fish consumption experiment at Hanford was conducted using a single subject (5). In this case a zinc absorption fraction of 39 per cent and an effective Zn^{65} half-life of 150 days were obtained. These values may be compared with those reported by the I.C.R.P.(6) of 10 per cent and 194 days.

In the experiment reported here the average Zn^{65} absorption from fish consumed during the first two days was 40 per cent, ranging from 31 to 50 per cent among the individuals. There was evidence that the individual fractional uptakes changed during the course of the experiment, varying over a range similar to that between individuals. At the end of the experiment the accumulated body burdens indicated an average fractional absorption of 35 per cent, the individual values ranging from 25 to 45 per cent. The loss of Zn^{65} from each subject following the period of fish consumption indicated an average effective Zn^{65} half-life in the total body of 299 days, considerably higher than the 150 days measured earlier or the I.C.R.P. value of 194 days. The individual values for the seven subjects ranged from 197 to 448 days. The Zn^{65} retention data from this experiment are suspect, however, because the subjects had some opportunity for routinely obtaining small amounts of Zn^{65} from unmonitored portions of their diets. Such ingestion during the decay period would tend to increase the apparent effective half-life of Zn^{65} in the body. Local residents having occupations similar to the subjects of this experiment are routinely found to have Zn^{65} body burdens of 2 - 3 nCi Zn^{65} from all diet sources (7).

The measured body burdens of Zn^{65} of the seven subjects are plotted in Figure 5. The solid line shown in this figure is the theoretical Zn^{65} body burden calculated for 39 per cent absorption and 150 days effective half-life. The first set of points at the left appear high, probably because of unabsorbed Zn^{65} in the subjects' gastrointestinal tract at the time of counting. All of the subjects had the same fish diet, so the scatter shown must result from differences in their absorption and retention of the radionuclide together with any differences in Zn^{65} present in other diet items. The intake of low levels of Zn^{65} from other than fish would not greatly affect the position of points on this plot, but could have a major effect on the calculated effective half-life determined from a least-squares fit of decay data.

DISCUSSION

This experiment provided an opportunity to demonstrate the application of the instrument developed for measuring P^{32} *in vivo*, and to contribute new information on metabolic parameters for P^{32} and Zn^{65}. The experimental application of beta measurements for whole-body P^{32} determinations was, on the whole, successful. The calibration of the instrument made with other subjects and under different conditions gave whole-body values which seemed to agree well with the analyses of the fish consumed assuming a rather complete absorption of P^{32}. The analyses of excreta tended to indicate a somewhat lower absorption, but the interpretation of these data is not complete. The application of these data to exposure dose calculations must be made judiciously because the instrument can measure P^{32} only in blood, muscle, and skin. The deposition of P^{32} in bone is not measurable with

this instrument. However, the experiment demonstrated that an equilibrium distribution among other tissues is attained which can be measured. It was shown that the distribution of P^{32} among various body tissues changes with time, so that measurements made soon after ingestion probably will not represent the equilibrium distribution. For such prompt P^{32} measurements a special calibration of the instrument is necessary.

The fractional uptake of P^{32} determined in this study appeared to be slightly higher than that reported by the I.C.R.P., while the effective total body half-life was found to be slightly lower. The overall effect of these differences on calculated exposure dose from consumption of P^{32} is not large. The resulting calculated exposure will be little different from that determined previously, the differences remaining within the uncertainties in the determination. Phosphorus-32 is excreted from the body in both urine and feces. While more than half appears in the urine, a surprisingly large fraction of the P^{32} was eliminated in the feces, even after the end of the ingestion portion of the experiment. Blood samples were obtained from the subjects at the same time that P^{32} measurements were made. Analyses failed to find significant P^{32} in blood samples, apparently because the samples were taken too long after eating fish. As is apparent from Figure 3, the concentration of P^{32} in blood decreases very rapidly after ingestion. The samples of fish, excreta, and blood were analyzed using a scintillation technique in a well crystal and measuring the P^{32} Bremsstrahlung. Occasional checks of these analyses were made with chemical separation and beta counting. The difficulty of making accurate determinations of P^{32} in excreta samples at the very low levels encountered in this experiment contributes to the uncertainty in the

absorption calculations.

Zinc-65 analyses were all made with scintillation counting in a well crystal. Approximately 90 per cent of the Zn^{65} was eliminated from the body by the feces. The whole-body counting for Zn^{65} is a straight-forward procedure, complicated only by the possible presence of unabsorbed Zn^{65} in the lower digestive tract.

There appeared to be somewhat greater variation of the metabolic parameters of fractional absorption and effective half-life in the case of Zn^{65} than was apparent for P^{32}. It is not always possible to distinguish between variations in the two parameters. The data suggest that considerable difference may occur between individuals and also from time to time for the same individual. A subject for whom an effective half-life of 150 days had previously been measured over a two-year period, was found to exhibit a 197-day half-life during this experiment.

When an individual is found to have a certain Zn^{65} body burden, this could result from either a single ingestion of a sizable amount or an extended continuous ingestion of a relatively small amount of Zn^{65}. While it is impossible to interpret the Zn^{65} body burden to tell the mode of ingestion, the P^{32} body burden level accompanying the Zn^{65} is very sensitive to the schedule with which the fish were eaten. Figure 6 shows this relationship. This figure shows the P^{32}/Zn^{65} ratio calculated for subjects participating in this study. The bars show the P^{32}/Zn^{65} ratio found in fish. Early in the experiment the high P^{32} content of fi[illegible] and high absorption of P^{32} relative to Zn^{65} gives a large ratio. This ratio declines as the Zn^{65} body burden of the subjects accumulates. The ratio rapidly decreases toward zero during the decay interval during the

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end of the ingestion. It is clear that if a Zn^{65} body burden determination were made at some point for an individual for whom the ingestion history was not known, the interpretation of this result in terms of P^{32} radiation exposure could give uncertainties ranging over several orders of magnitude. The better the diet history of the individual is known, the greater is the confidence that can be placed in the radiation exposure calculations. For situations of the kind depicted here, it is clear that methods of obtaining accurate diet information are very important, and the development of techniques in this field could be rewarding if such vicarious determinations are to be attempted.

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ACKNOWLEDGEMENTS

This experiment required the cooperative efforts of numerous scientists and technicians to collect and analyze samples and to operate the whole-body counter. The contribution of the volunteer subjects who manfully ate ground fish on schedule for eleven weeks was of particular importance, and their cooperation is acknowledged with appreciation. In particular, the assistance of the following scientists is acknowledged: Dr. Guy H. Crook, Dr. Thomas D. Mahony, Mr. Claude R. Henle, Mr. Lloyd S. Kellogg, Mr. John J. Jech, and Dr. Richard F. Foster. Technicians whose interested help was invaluable include: Linda Dalton, Jean Warder, Robert Beaver, and Charles McCoy.

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CONCENTRATION OF ZINC-65 AND PHOSPHORUS-32
IN COLUMBIA RIVER FISH

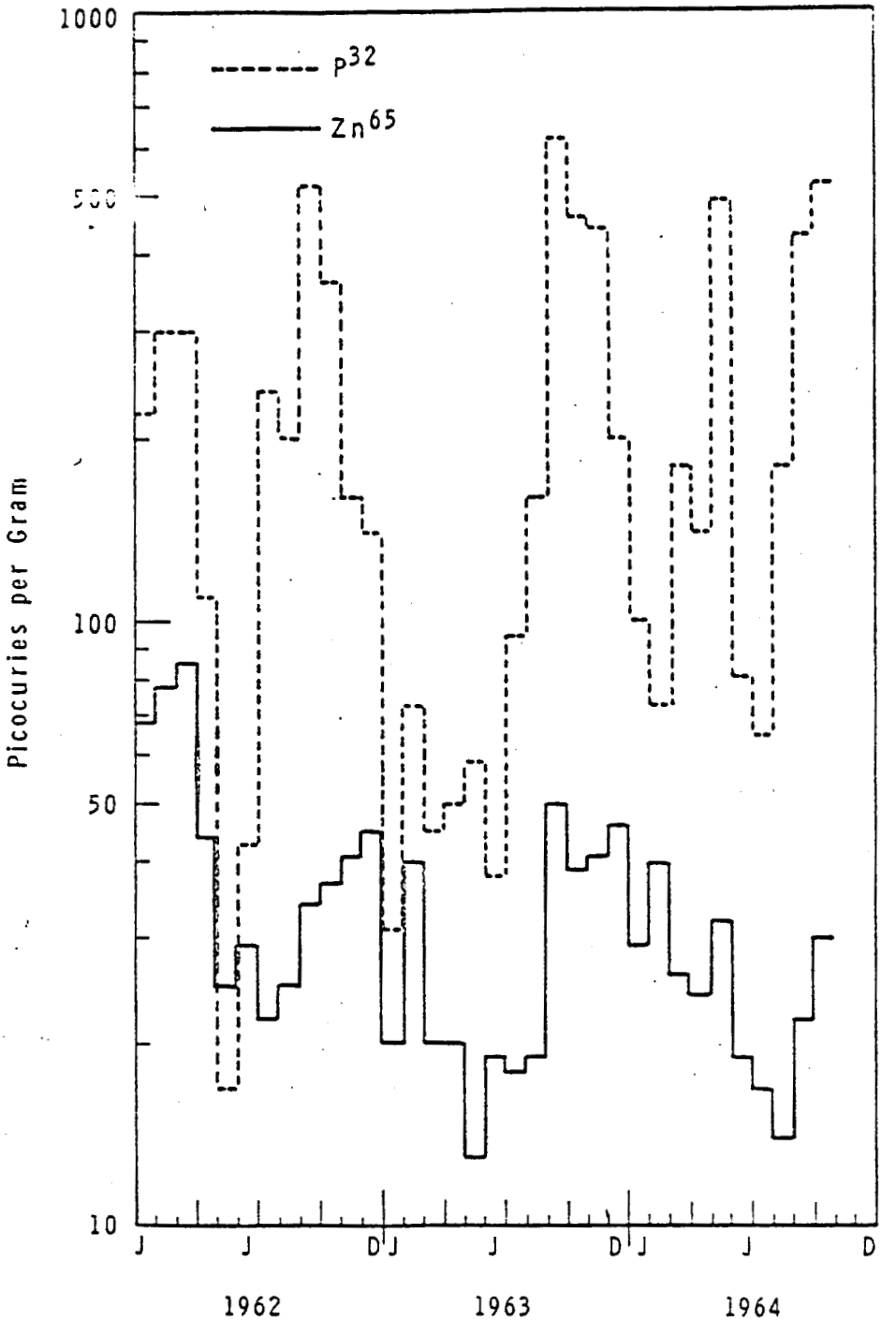

FIG. 1

EXPERIMENTAL DESIGN

"Uptake and Retention of P^{32} and Zn^{65} from the Consumption of Columbia River Fish"

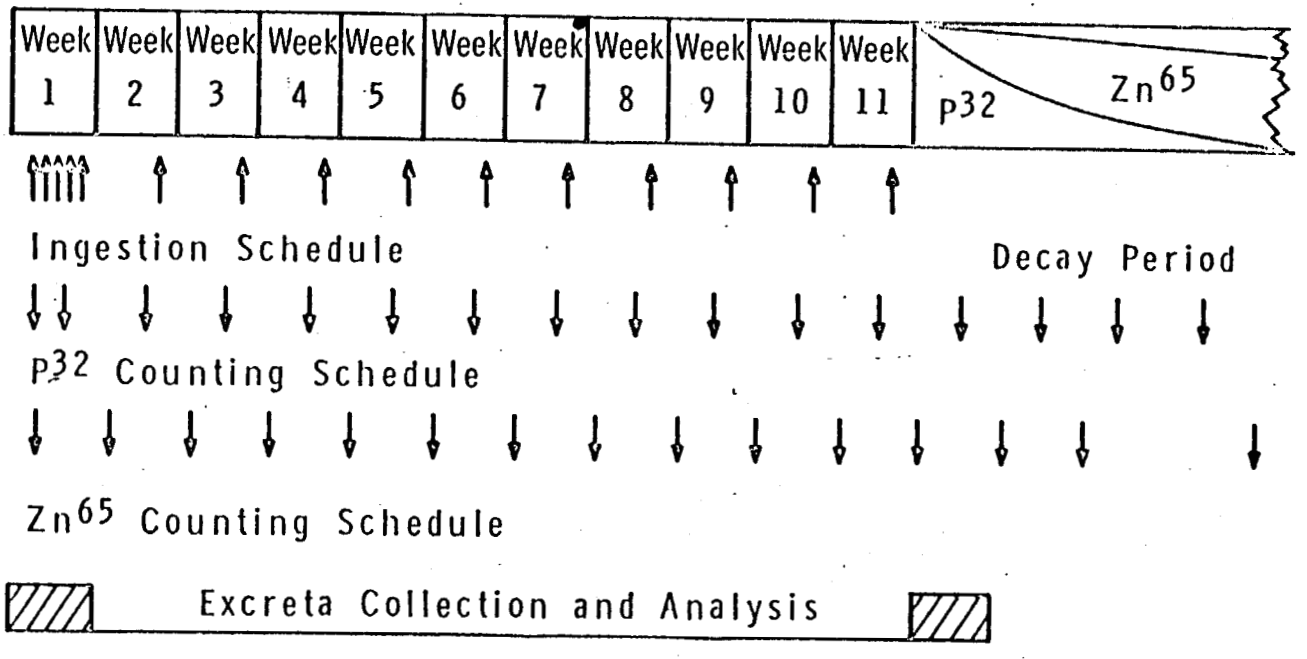

FIG. 2

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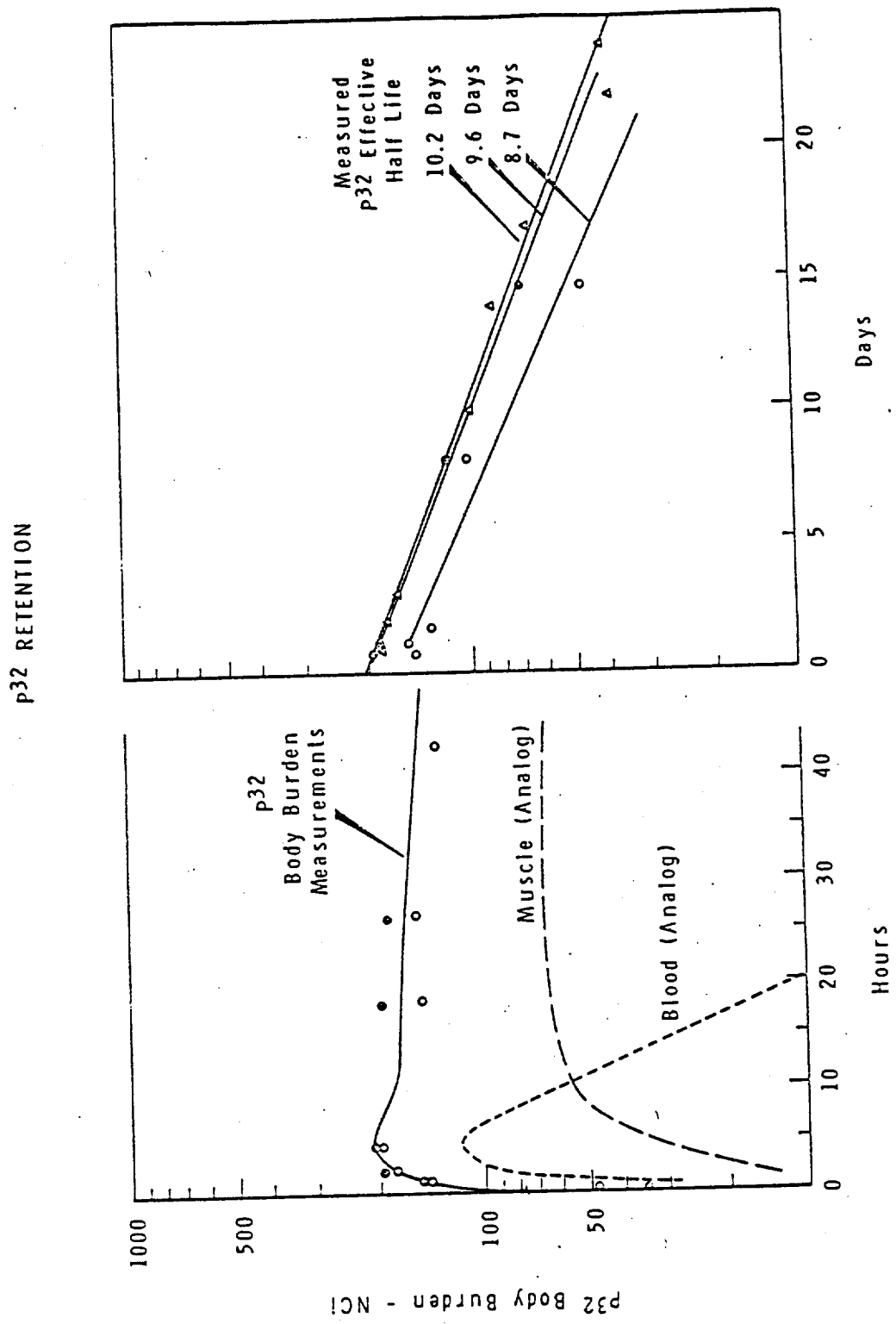
p32 RETENTION
Measured
p32 Effective
Half Life
10.2 Days
9.6 Days
8.7 Days
0
5
10
15
20
Days
p32
Body Burden
Measurements
Muscle (Analog)
Blood (Analog)
1000
500
100
50
0
10
20
30
40
Hours
p32 Body Burden - NCi

FIG. 3

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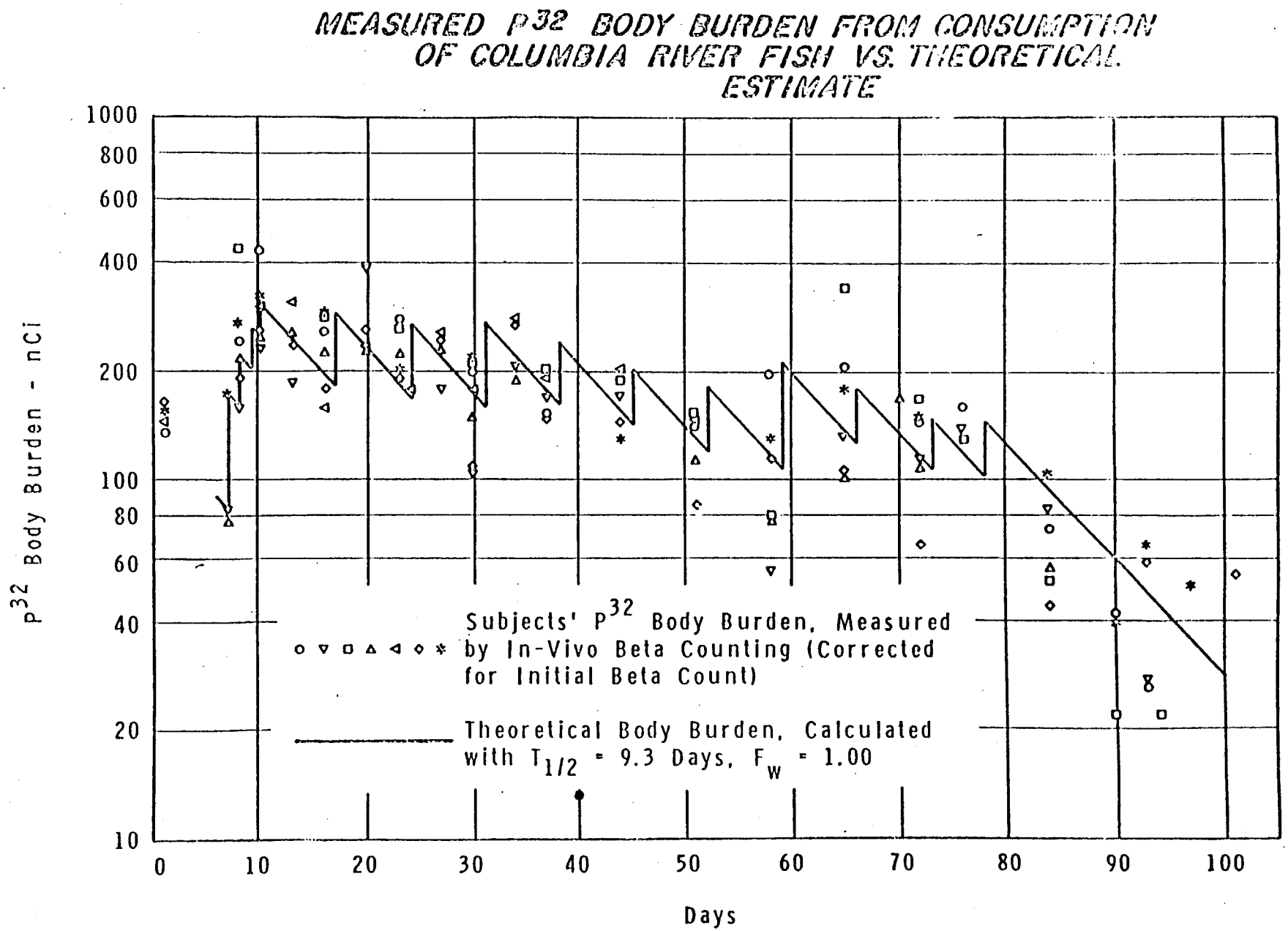

FIG. 4

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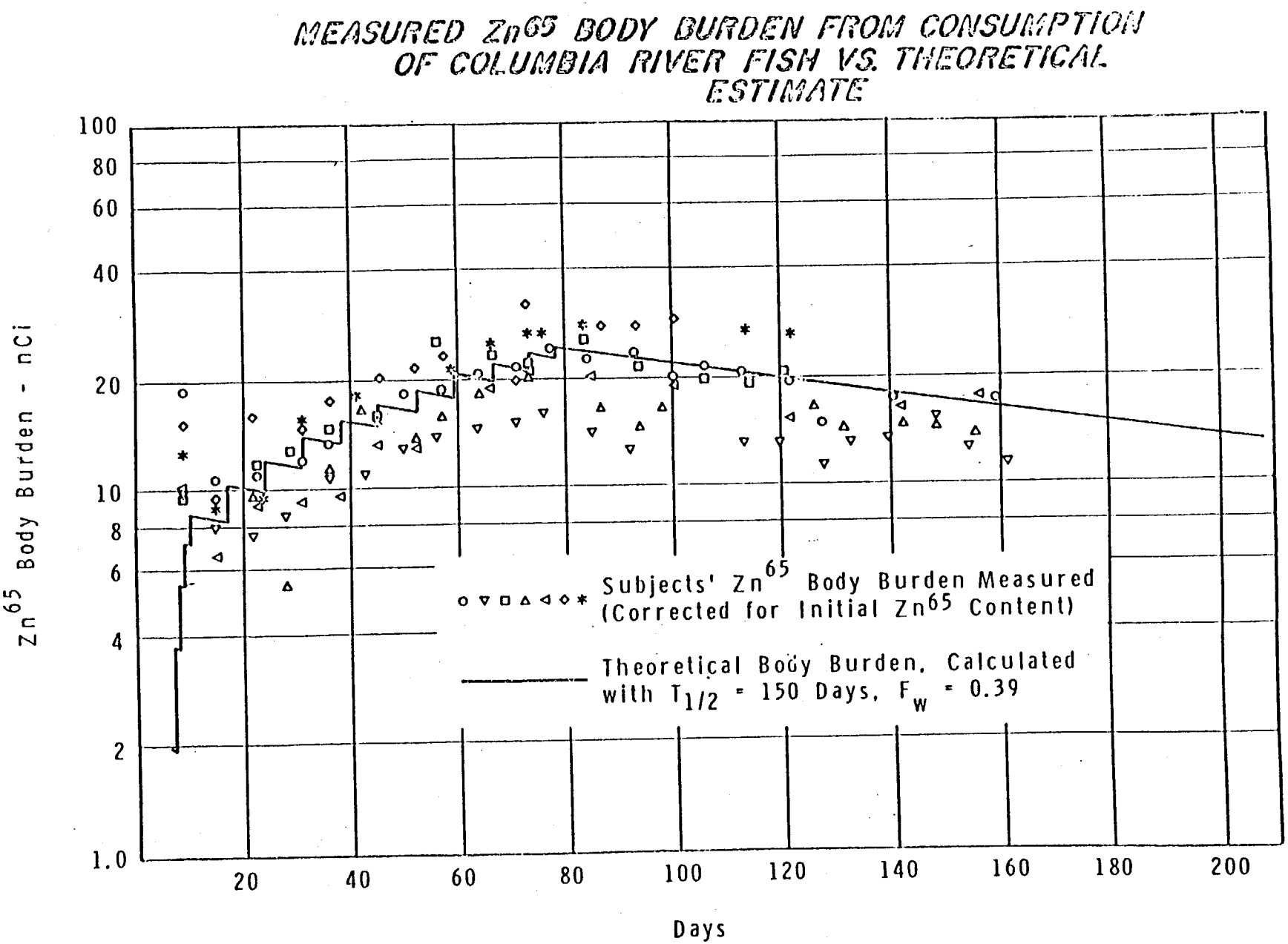

FIG. 5

THE P^{32}/Zn^{65} RATIO IN MAN FOLLOWING PROLONGED CONSUMPTION OF COLUMBIA RIVER FISH
(Calculated)

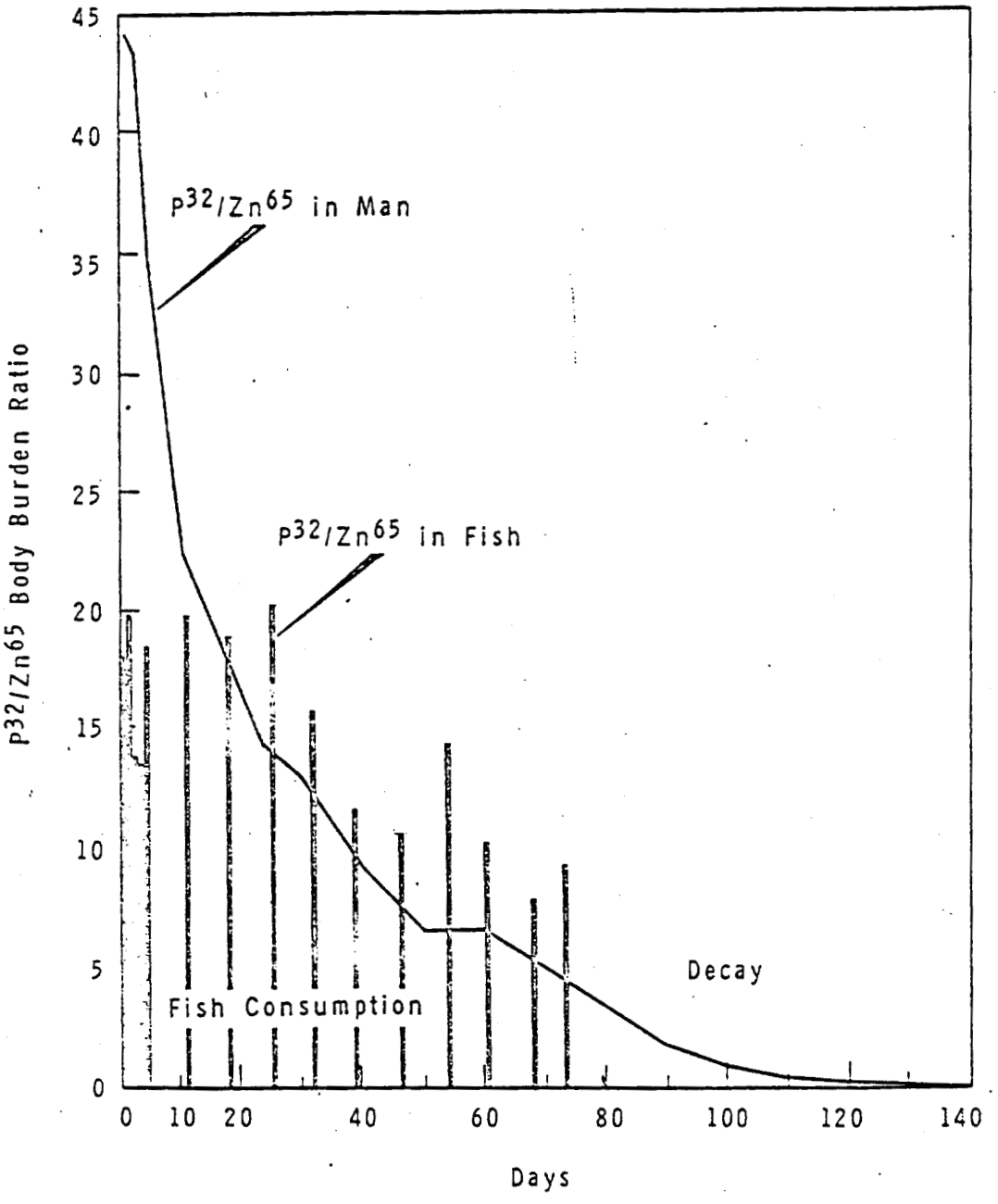

FIG. 6