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ACCUMULATION OF ZINC-65 FROM PROLONGED CONSUMPTION OF COLUMBIA RIVER FISH*

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Abstract—Fish caught downriver from the Hanford reactors were eaten once each week for more than 1 year in order to provide reasonable estimates of the quantity which might be consumed by persons living in the environs and the associated intake of ^{32}P and ^{65}Zn. In a 12-month period this amounted to about 25 lb of fish, 3000 nCi ^{32}P and 800 nCi ^{65}Zn. The accumulation of ^{65}Zn in the consumer was measured each week in a whole-body counter in order to trace the build-up and follow the seasonal trend. A maximum body burden of about 130 nCi ^{65}Zn was reached on day 104 and again on day 310. After the initial build-up toward equilibrium, the body burden approximated seven times the weekly intake—more than anticipated from some published parameters. The reduction of ^{65}Zn body burden following the last meal of Columbia River fish was observed for 511 days, indicating an effective half-life of 162 days.

INTRODUCTION

ZINC-65 is one of several radionuclides released to the Columbia River in cooling water from the Hanford plutonium production reactors.[1] At Pasco, Washington, about 60 Ci of this nuclide are transported by the river each day; based on a concentration in the river water of from 1×10^{-7} μCi/cm^3 to 5×10^{-7} μCi/cm^3. The variation in concentration reflects the quantity of water available for dilution.[2] Because of its biological importance and relatively long half-life (245 days), ^{65}Zn attains measurable concentrations in the flesh of fish and shellfish. It is also found in crops irrigated by water from the river and in milk from cows which have grazed on irrigated pasture.[2]

Members of the population adjacent to the Plant who consume foodstuffs containing ^{65}Zn acquire some of this nuclide and the significance of the intake must be evaluated along with other potential sources of exposure. The γ-emitting ^{65}Zn (1.11 MeV) can be measured directly by whole-body counter techniques. Another radionuclide which is found in some types of local foodstuffs is ^{32}P. Exposure estimates show that for certain individuals this nuclide may provide a significant fraction of the total exposure; however, it has no γ component and therefore cannot be detected by conventional whole-body counting. Phosphorous-32 is of interest primarily in foodstuffs taken directly from the river because its short half-life (14 days) prevents its movement through a long food chain. For this reason ^{32}P is of particular interest in the case of Columbia River fish. Fish will contain both ^{32}P and ^{65}Zn in a predictable ratio when in equilibrium with river water containing Hanford reactor effluent. One of the ways we use ^{65}Zn body burden data, as measured by the whole-body counter, is to estimate the amount of Columbia River fish that might have been consumed by a local resident and from this deduce an upper limit for his ^{32}P intake. For this reason the accumulation of ^{65}Zn from consumption of Columbia River fish is of particular interest.

Calculations of the rates at which ^{65}Zn and ^{32}P are ingested by people in the environs of Hanford (and thus fractions of permissible limits) are now based on rational estimates of

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0028100

the quantities of local fish which might be consumed by the more successful fishermen and their families. Refinement of these estimates has been in progress by a program which includes, (1) determination of the weight of fish flesh consumed at a typical meal, (2) measurement of the body burden of ^{65}Zn associated with a sustained intake of Columbia River fish, and (3) a survey of fishermen to obtain their estimates of the quantities of local fish caught and eaten each year. This paper describes the changes in the ^{65}Zn burden in the body of one of the authors who ate Columbia River fish at approximately weekly intervals for more than one year.

METHODS

The fish used in this study were Rocky Mountain whitefish (*Prosopium williamsoni*), caught from the Columbia River a few miles downstream from the Hanford reactors, usually at a location available to the general public. Although all species of fish in this section contain ^{65}Zn and ^{32}P, the whitefish was chosen because it is among those which have the highest concentrations of these nuclides, because it can be caught throughout the year, and because it is sometimes caught in large numbers by sportsmen. The fish were caught by hook and line each week by members of Hanford's Environmental Monitoring group as a part of the regular Plant surveillance program.

A sample of the flesh of each fish was removed and sent to the laboratory for radiochemical analysis. The fish were then taken home, prepared as a regular meal, and eaten. Several different methods of cooking were used to provide variety. The fish was weighed before and after cooking, and at the end of the meal the remaining bones and scraps were weighed so that the exact quantity consumed could be determined. The quantities of ^{65}Zn and ^{32}P consumed at each meal were calculated later when the laboratory results were available.

The amount of ^{65}Zn assimilated from the fish and retained was determined with Hanford's whole-body counter, which has been described by SWANBERG.[3] This counter uses a $9\frac{3}{8} \times 4$-in. thallium-activated sodium iodide crystal positioned over the subject seated in a reclining chair. The duration of each counting period was 20 min. An attempt was made to obtain a count once each week, but it was not always possible to maintain this schedule. No counts were taken sooner than 20 hr following a fish meal.

RESULTS

Before starting the whitefish diet, a whole-body count was taken for reference. The results showed a burden of 3.2 nCi ^{65}Zn, which is typical of persons living in Richland who do not eat fish or drink water from the Columbia River.[3] During the course of the experiment, the author's weight was in the range of 73–76 kg.

In all, there were fifty-six fish meals in 67 weeks. One hundred and fifteen fish were eaten for the study and the average consumption was 257 g per meal. The smallest meal was 130 g and the largest 489 g. The quantity of fish eaten at most meals was more than ample as the main dish. From this experience an estimate of 25 lb of fish per year as the consumption rate for a standard man who eats fish once each week seems reasonable.

The total intake of ^{65}Zn with the fish was 870 nCi but, because of seasonal and other variability in the ^{65}Zn content of the fish, the amount consumed at individual meals ranged from 0.3 to 50 nCi. The total intake for the first 12 months was 770 nCi, which is about 0.1 per cent of the maximum rate specified for occupational workers by the NCRP.[4] Use of the Radiation Protection Guide, recommended by the Federal Radiation Council for total body exposure for the general population (0.17 rem per year), implies a permissible intake rate of 27,000 nCi of ^{65}Zn per year.

The ^{65}Zn ingested with the first meal of fish was easily detected with the whole-body counter and, of course, the body burden increased with successive meals. Figure 1 shows the γ-spectrum obtained with the whole-body counter just before fish-eating was begun and 1 year later when the ^{65}Zn burden was at maximum.

Changes in the body burden of ^{65}Zn during course of the study are shown in Fig. 2. A maximum of 130 nCi ^{65}Zn was reached on the 104th day but the burden then declined because of low concentrations in the fish and several missed meals. Later there was a gradual increase back to a burden of 120 nCi, but this was not quite maintained, again because of

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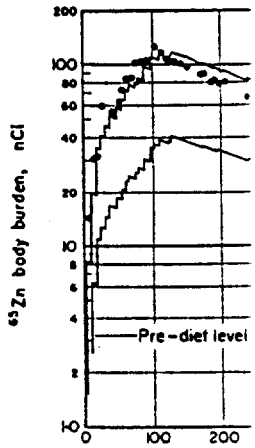

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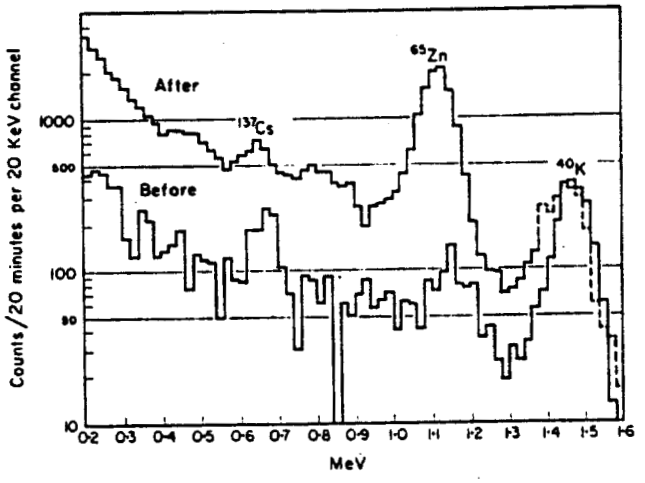

FIG. 1. Whole body spectrum before and after prolonged consumption of fish.

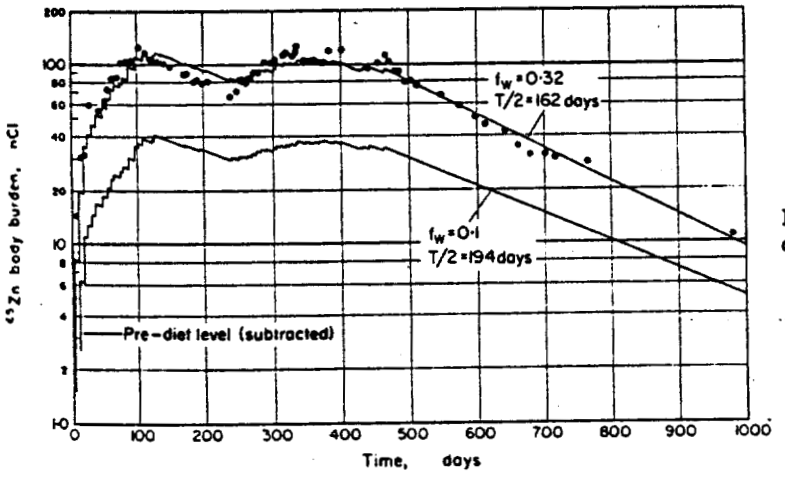

FIG. 2. Observed body burdens of ^{65}Zn vs. theoretical estimates.

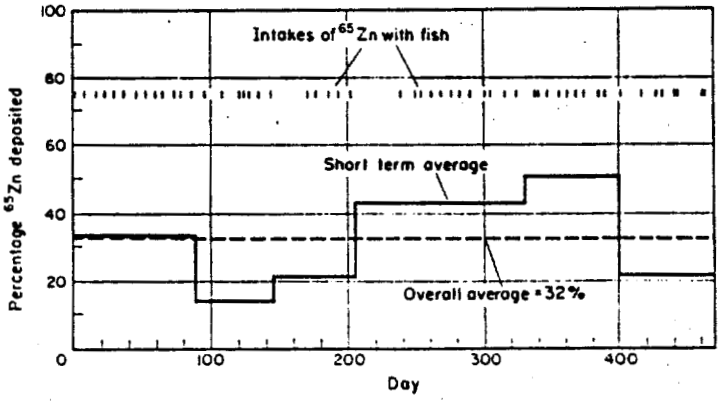

FIG. 3. Percentage of ingested ^{65}Zn deposited

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reduced ingestion. The peak body burden of 130 nCi may be compared with the NCRP recommendation of 60,000 nCi as a maximum for occupational workers.[4] For average members of the general public, a maximum burden of 2000 nCi may be inferred from the recommendations of the FRC.[5,6]

Although this work was not primarily designed to provide data on the human metabolism of ^{65}Zn, a substantial amount of information in this area may be inferred from the results. Loss of ^{65}Zn from the body during periods of no ^{65}Zn ingestion starting on day 150 and again on day 205 provide opportunities to measure an effective half-life. The calculated values for these two periods are 169 and 123 days respectively. The measure of effective half-life of ^{65}Zn which was obtained during the period following the last fish meal is more reliable because the available observation period is much longer and includes more whole-body counts. The effective half-life obtained for this 511-day period was 162 days. The first two decay periods are probably too short (21 and 34 days respectively) for the loss to have approximated a simple exponential. The value of 162 days agrees well with the value of 154 days obtained by RICHMOND *et al.*,[7] as an average for four subjects. Further, empirical curves developed from the intake data to show theoretical body burdens of ^{65}Zn indicate that an effective half-life of 162 days fits the observed body burden data. Such a plot is included in Fig. 2 to show the agreement.

In order to establish the theoretical body-burden curve from the consumption experience of this study it was necessary to determine the fractional adsorption of ^{65}Zn from fish. This parameter, together with the effective half-life, provides the theoretical body burden for individuals when a single exponential elimination model is assumed. In Fig. 2 this amounts to shifting the curve vertically on the log scale to obtain the best fit with the body-burden measurements. The least-squares estimation of the fraction adsorbed from the data from this study, if an effective half-life of 162 days is assumed, was determined to be 32 per cent. The theoretical body-burden curve of Fig. 2 is the result. Also shown in Fig. 2 for comparison is the theoretical curve obtained from using the ICRP "standard man" values of 10 per cent fractional absorption and 194 days effective half-life (from drinking water).

From the positions of whole-body counting points relative to the theoretical curve in Fig. 2, it is clear that the exponential model with effective half-life of 162 days and a fractional absorption constant of 32 per cent does not fit all of the data from the consumption study. There were periods during the experiment when measured body burdens were consistently low and other periods when they were consistently high. These variations were examined by assuming a single exponential elimination model but with the fractional absorption value changing seasonally, possibly in response to seasonal dietary changes. The results of this examination are shown in Fig. 3. Here the fractional absorption is calculated for data points collected during arbitrarily chosen "seasons", which seemed to reflect the variance in the f_w value. For each time period the ^{65}Zn present in each fish meal eaten was adjusted to the end of the period by the appropriate elimination factor, using an effective half-life of 162 days. The ratio of the increase in ^{65}Zn body burden to the sum of the adjusted ^{65}Zn ingestions during the period was identified as the average f_w during the period and are plotted in Fig. 3. The fractions calculated in this way ranged from a low of 14 per cent to a high of 51 per cent. The actual values were found to be sensitive to the specific periods chosen for examination; for example, in one case it was found that by extending the period by 2 days to include one additional point, the f_w value changed from 0.04 to 0.21. The results are too inconclusive to warrant a hypothesis concerning the reason for this variation. In general, the retention of ^{65}Zn appears to have been relatively low during the spring and early summer and relatively high during the fall and winter.

Laboratory investigations of ^{65}Zn metabolism by RICHMOND *et al.*,[7] based on the results of a single administration of radioactive zinc provide retention functions that can be represented by multiple rate equations composed of three exponential terms. The effective half-lives represented by the three elimination constants are 0.55 days, 9.59 days, and 154 days. Within 6 days of ingestion more than 85 per cent of the

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zinc body burden is following an elimination curve described by the third (154-day effective half-life) term of the function. In the experiment reported here zinc is administered by chronic ingestion of fish and the resulting body burden is accumulated over months of regular addition of small increments. In this case only a fraction of the increment supplied by the most recent fish meal is eliminated by the shorter-lived terms in RICHMOND's model and we have neglected these terms in the analysis of our data.

CONCLUSIONS

The fraction of ^{65}Zn ingested with Columbia River fish which was retained was, for any single intake, subject to substantial variation. With prolonged consumption (67 weeks), however, the average fraction retained (f_w) was 32 per cent when an effective half-life of 162 days was used. There was evidence that the fraction retained may have varied within the range from 14 to 51 per cent. The parameters for ^{65}Zn listed by the ICRP[8] (194-day effective half-life, $0.1 f_w$) were not characteristics of the results obtained in this test with a single subject. Application of the ICRP values to the ^{65}Zn intakes in this study result in the theoretical body burdens shown in the lower curve of Fig. 2, which are only about one-third of those observed.

Acknowledgements—This project could not have been carried out without the skillful efforts of many people. We are particularly indebted to the following: Mr. R. W. MEISINGER and members of his Environmental Monitoring team, especially Messrs. J. D. MIXON, J. A. DEARDORFF, R. J. BEAVER, C. A. MCCOY, and M. R. QUARDERS for obtaining and sampling the fish; Mr. F. E. HOLT and members of his Analytical Laboratory team, especially Mr. D. P. ARGYLE, for radiochemical analyses of the fish; Mr. F. SWANBERG of the Internal Dosimetry Operation and especially Mrs. JOYCE BERNARD and Mrs. LINDA DALTON for making the whole-body counts; Mrs. DARLENE MOORE for data reduction and summarization; and the senior author's wife for cooking the fish and having uncommon patience.

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0028104