MASTER

THERMOLUMINESCENT DOSIMETRY OF AQUATIC ORGANISMS

by

D. G. Watson and W. L. Templeton

Ecosystems Department

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BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

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ABSTRACT

Lithium fluoride thermoluminescent dosimeters were employed to estimate the radiation exposure to Columbia River organisms downstream from the effluent outfalls of the Hanford plutonium producing reactors. Highest dose measurements were made in the river benthos. Observed dose was inversely related to distance downstream from the effluent outfalls. Radionuclides in suspension or solution in the river water contributed significantly to the total radiation exposure to both fish and benthic organisms.

The dose to fish homogenate and intact fish kept frozen during dosimeter exposure was generally inversely related to fish size. This relationship did not hold for the major gamma emitting nuclides contained in the fish carcass, but appeared to be associated with the body burden of radiophosphorus.

INTRODUCTION

Although the assessment of the radiation dose to aquatic organisms in surface waters receiving radionuclides is at times difficult and uncertain, it is, however, essential to the evaluation of radiation effect. The body burden of radionuclides measured in organisms after their removal from the environment is often used as a basis for these estimates; and it is usually necessary to make assumptions or estimates concerning the distribution of radionuclides within the organisms, its body form, and the external dose obtained from the environment. This approach has been used to calculate the dose to chironomids (Nelson and Blaylock 1963) and to fish eggs (Brown and Templeton 1964, Adams 1968, and Woodhead 1970).

The use of thermoluminescent dosimeters (TLD) to directly measure the radiation dose in an aquatic environment offers some obvious advantages over the indirect method of dose estimation. They can be placed on or within the larger aquatic forms for extended periods of time to obtain an integrated dose measurement during conditions of varying radiation field, or they can be used to directly measure the dose contributions of different parts of the environment, such as the water or bottom sediments. TLD's have been used to measure the differences in radiation dose at the mud-water interface in an experimental pond spiked with ^{137}Cs (Guthrie and Scott 1969), the dose to fish in the sea near the effluent discharge site at Windscale (Ministry of Agriculture, Fisheries and Food 1967), and the dose to periphyton in the Columbia River (Lappenbusch et al., in press).

The purpose of this report is to present the results of TLD measurements in the biota in the Columbia River downstream from the

points of radioactive effluent discharge from the plutonium production reactors at Hanford.

Methods

There are more than 60 radionuclides in the reactor cooling water effluents that are discharged into the Columbia River from the Hanford reactors (Table 1). Both beta and gamma emitters are represented, with energies ranging from less than 0.01 to more than 2 Mev, and physical half-lives of a few hours to thousands of years. Fluctuations in the concentration of these radionuclides in the river result from variations in reactor operation, diurnal river flow variations produced by a power-peaking dam upstream from the reactors, seasonal variations in flow, and incomplete mixing of the effluents in the river near their point of discharge. TLD lithium fluoride-100 (Harshaw Chemical Co.), the phosphor used in the dosimeters, has characteristics that enhance its utility in dose measurements under these conditions. These are, according to Cameron et al. (1968): 1) a nearly linear response over a wide range of energies (0.03 to 2 Mev) and exposures (10^{-2} to 10^3 R), 2) near soft tissue equivalence, 3) dose rate independence, 4) responsive to both gamma and beta radiation and 5) high retention of stored information with time.

Dosimeters were made of approximately 85 mg LiF powder encapsulated in a section of 2.4 mm diameter light-impervious heat-shrinkable tubing, with wall thickness of 0.254 mm and a density of 1.1 g cm^{-3} . The quantity of LiF powder in each dosimeter was sufficient for three separate readings. The exposed powder was read on a Madison Research reader during most of the study; later measurements were made with a Harshaw Model

2000 A-TL detector. All readings were made in a nitrogen gas atmosphere, and calibration of each batch of powder was based on known exposures of dosimeters to ^{226}Ra and ^{60}Co sources. Before exposure the LiF powder was annealed for 1 hour at 400°C , followed by 24 hours at 80°C (Cameron et al. 1968).

Water dose measurements were made with dosimeters suspended on a wire through the center of a meter long section of 3.8 cm diameter pipe attached to the under side of a float anchored in the river. The pipe was used to restrict the light exposure to the dosimeters and thus reduce algal growth. Sediments did adhere to the water dosimeters and occasionally bryozoa would grow on them. Consequently the radiation dose measured with these dosimeters was greater than that from the water and suspended particulates.

Radiation dose to the benthic river organisms was obtained from dosimeters strung on a nylon thread and tied to both upper and lower sides of rocks on the river bottom. After attachment of dosimeters, the rocks were placed in their original position on the river bottom and their location marked with an iron stake. In most cases the same rocks were used throughout the course of the study.

Radiation dose estimates to fish were made in several ways. Initially dosimeters were attached to live fish on their outer surface, just below the dorsal fin; others were implanted in the abdominal cavity through a slit in the abdominal wall just anterior to the pelvic fins. These animals were kept in a fenced enclosure near the river shore where they had access to natural foods and were exposed to external sources of radiation in the river. This approach was abandoned because daily fluctuations in river elevation of nearly two meters made it impossible to keep the enclosure fence intact. Subsequent dose estimates were made on fish that were killed

immediately after collection and kept frozen at -8° C. Dosimeters were attached to the outer surface near the dorsal fin, implanted in the muscle mass, and placed in the abdominal cavity near the mid-gut and anterior kidney. Other measurements were made on whole fish homogenate with dosimeters placed in the center of the tissue mass. On an aliquot of the fish homogenate, radioanalysis were made for ^{32}P , ^{24}Na , ^{51}Cr , ^{54}Mn , ^{60}Co , ^{65}Zn , ^{106}Ru , ^{46}Sc , ^{137}Cs , ^{144}Ce , ^{95}Zr , ^{131}I , ^{59}Fe , ^{140}Ba , ^{140}La , and ^{239}Np .

Results and Discussion

The dominant life forms inhabiting the upper surface of the rocky river substrate include filamentous green algae of the genera Cladophora, Stigeoclonium, and Ulothrix; diatoms of the genera Gomphonema and Melosira; and larval stages of several midge species (Tendipedidae). Invertebrates such as the sponge, Spongilla spp.; caddisfly larvae (Trichoptera), aquatic moth larvae, Argyraetis sp.; and limpets, Fisherola sp. inhabited the under side of the rocks.

The TLD radiation dose rate measurements in water, benthos, and fish are summarized in Table 2. These do not include the low energy beta radiation shielded out by the tubing used to encapsulate the LiF phosphor. Beta particles of energy below 0.15 Mev were excluded, and those slightly above this energy were probably significantly attenuated.

The measured dose in the water and benthos at a distance of 22 km downstream from the nearest reactor outfall remained fairly constant during 1968 and 1969. The mean dose rates to the water during this period ranged from 8.4 to 17 mR/day, and the range to the benthos from 21 to 34 mR/day. During this time there were several changes in the number of operating reactors. Four were operating until February

1968, three to April 1969, two to February 1970, and the remaining reactor was closed in January of 1971. The dose measured on the river bottom did not reflect these reductions in reactor operation until the fall and winter of 1970-71 when there was about a 70% decrease in observed dose. Only a slight decrease was observed from 1968 to 1969 in the concentration of radionuclides in the river water at the city of Richland, Washington, located approximately 70 km downstream from the nearest effluent outfall (Wilson and Essig 1970). Shoreline measurements of gamma radiation exposure showed no change between 1968 and 1969 (Wilson and Essig 1970). Comparison of the water and benthos dose showed that from 30 to 55 percent of the dose to the benthic communities came from the water and its suspended particulates. The burden of radionuclides associated with the sediments and biological growth that accumulated on the outer surface of the water dosimeters undoubtedly contributed to the amount of radiation measured by these TLD's, tending to make the estimates of radiation derived from the water too high.

The radiation dose to the benthos decreased rapidly with distance downstream from the reactor outfall during the period of normal reactor operation (1969). A mean dose rate of 220 mR/day was measured at 7 km below the effluent outfall. This value was nearly the same as the dose rate observed in periphyton communities grown on artificial substrates immediately below the point of effluent discharge in the summer of 1969 (Lappenbusch et al., in press). The radiation exposure at 14 and 22 km downstream from point of discharge was about one-fourth and one-sixth that at 7 km. Differences with distance downstream were much less during the fall and winter of 1970-71, probably as the result of the rather sporadic operation of the single remaining operating reactor.

The radiation exposures to the under sides of the bottom rocks were 10 to 20 percent less than that of the upper surface, probably due to the shielding from the water by rock and the absence of a thick periphyton mat on the under surface.

The only true environmental dose measurement to fish were obtained in the early part of 1968 when live fish were held in the river. Dose rates at the outer surface of the animals were only slightly greater than that in the water, and the dose rate to the gut was about 30 percent greater than that of the water.

The TLD's that were attached or implanted in fish that were killed and frozen during exposure were subjected only to the radiation field provided by the radionuclides contained in tissues of the animal, and this field decreased over the period of exposure due to physical decay of the contained radionuclides. If the dose rate from water is added to that of the whole fish homogenate for the fall period of 1968, an estimated environmental dose rate on the order of 23 mR/day is obtained. This is a conservative figure because no correction was made for loss of radioactivity in the fish through radionuclide physical decay. This is similar to the dose rates measured with TLD's attached to fish in the marine environment near the outfall from the Windscale Atomic Energy installation in Great Britain in 1967 (Woodhead, personal communication, 1967). Dose rates of this magnitude would produce an annual dose of about 8.4 R. Although fish resident in the area immediately downstream from the point of effluent discharge conceivably could be exposed to radiation doses several times this amount, no radiation effect would be expected. Laboratory tests in which fish were exposed to effluent concentrations several times the maximum concentration found in the

Columbia River have not shown any radiation effect. One of the lowest acute lethal exposures, 16 R, has been reported for the embryonic developmental stages of salmon (Donaldson and Foster 1957).

There appears to be a seasonal inverse relationship between total body weight and radiation dose (Figure 1). In the fall of 1968 there was a consistent decrease in measured dose with increase in total body weight in the frozen specimens. This relationship was not apparent, or only slightly so, in fish collected from the river in the spring of 1969. The correlation between body weight of fish and the concentration of the principal gamma emitting nuclides in the whole fish homogenate did not follow this same pattern. There did, however, seem to be a generally inverse relationship between body weight and concentration of ^{32}P . This would seem to indicate that ^{32}P , a beta emitter, contributes significantly to the dose measured with the TLD's.

SUMMARY

The radiation dose to the benthic communities and to fish in the Columbia River downstream from the cooling water effluent outfalls of the Hanford production reactors was measured with LiF TLD's. Dose rates of 220 mR/day and an estimated annual dose of 80 R were measured in the upper surfaces on the rocky river substrate 7 km downstream from the effluent outfall. Radiation dose from the water contributed 30 to 50 percent of the total dose to the benthos. Fish 19 km downstream from the nearest reactor had maximum dose rates of 23 mR/day and an annual dose of 8.3 R. During the fall, when the metabolic rate and the uptake of radionuclides was highest, there was a consistent inverse relationship between the measured dose rate and weight of the fish. This same

relationship was evident to a lesser degree between body weight and concentration of ^{32}P , but did not appear to hold when body weight was compared to the principal gamma emitting nuclides.

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