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DISTRIBUTION OF ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{241}Am and ^{244}Cm IN POND B, SAVANNAH RIVER SITE

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Abstract The distribution of ^{137}Cs, ^{90}Sr, ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm in the biotic and abiotic components of an abandoned reactor cooling impoundment were studied after a twenty year period of equilibration. The impoundment is located at the U.S. Department of Energy's Savannah River Site in South Carolina, USA. It received radioactive contaminants via cooling water discharges from R Reactor from September 1961 to June 1964. Sampling was carried out in 1983 and 1984 to estimate radionuclide concentrations and total ecosystem mass, of water, seston, sediments, zooplankton, aquatic macrophytes, benthic invertebrates, fish, turtles, and waterfowl. Chemical, physical and biological relationships to the radionuclide distribution patterns were investigated. Radionuclide inventories were estimated and compared with reported discharges from R Reactor. Although the biotic components of the system contained comparatively high radionuclide concentrations in reference to filtered water, most of the radioactivity resides in sediments. The principal mechanisms of loss appear to be radioactive decay and periodic outflow of water and suspended materials; biotic export and seepage appear to be inconsequential. Strontium-90 was much more mobile than the other radionuclides. Aquatic macrophytes dominated the biotic component radionuclide inventories and the dynamics of this component appear to exert a strong influence on the spatial distribution of radioactivity in the ecosystem. Pond B and similar impoundments that have been contaminated with long-lived radionuclides are capable of supporting a diverse and productive flora and fauna. The ultimate use of such systems by humans for recreation is feasible, providing adequate monitoring is conducted and established radiological health guidelines are adhered to.

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INTRODUCTION

The gradual senescence of present-day operating nuclear facilities, together with the possibility of nuclear accidents (e.g. the Chernobyl reactor accident; Medvedev, 1986; Mascanzoni, 1987) and resultant contamination of aquatic and terrestrial ecosystems, emphasize the importance of understanding the behavior of radionuclides in the environment. Such understanding is crucial to decisions under emergency conditions, as well as to more deliberate evaluations and management of ecosystems contaminated by routine operations. Systematic study of the distribution of radionuclides in existing ecosystems contributes importantly to such understanding. Furthermore, observations and deductions concerning mechanisms of radionuclide transport can contribute significantly to knowledge of fundamental ecological processes (Odum, 1965; Odum, 1971; Whicker and Schultz, 1982). This study emphasized the ecosystem-level distribution of several long-lived radionuclides in an abandoned reactor cooling impoundment after a twenty year period of chemical and biological equilibration.

Nuclear production reactors on the U.S. Department of Energy's Savannah River Site in Aiken and Barnwell Counties, South Carolina, USA, utilize a system of canals and reservoirs to disperse heat from the reactor cores. This study concerns Pond B, an 87 ha impoundment that received cooling water discharges from R Reactor between September 1961 and June 1964 (Fig. 1). The reservoir also received inputs of the fission products ^{137}Cs (30 y half-life) and ^{90}Sr (28 y half-life) and transuranium elements such as ^{239}Pu (24,000 y half-life). Radionuclide inputs were greatest during 1963 and 1964 and are believed to have originated from faulty fuel elements that leaked during storage in a water-filled disassembly basin in the reactor building. Some of this water was discharged to the R Canal, which carried contamination to Pond B (Fig. 1).

In June, 1964, the R Reactor was shut down for an indefinite period, and Pond B apparently ceased to receive any reactor or disassembly basin discharge. The water

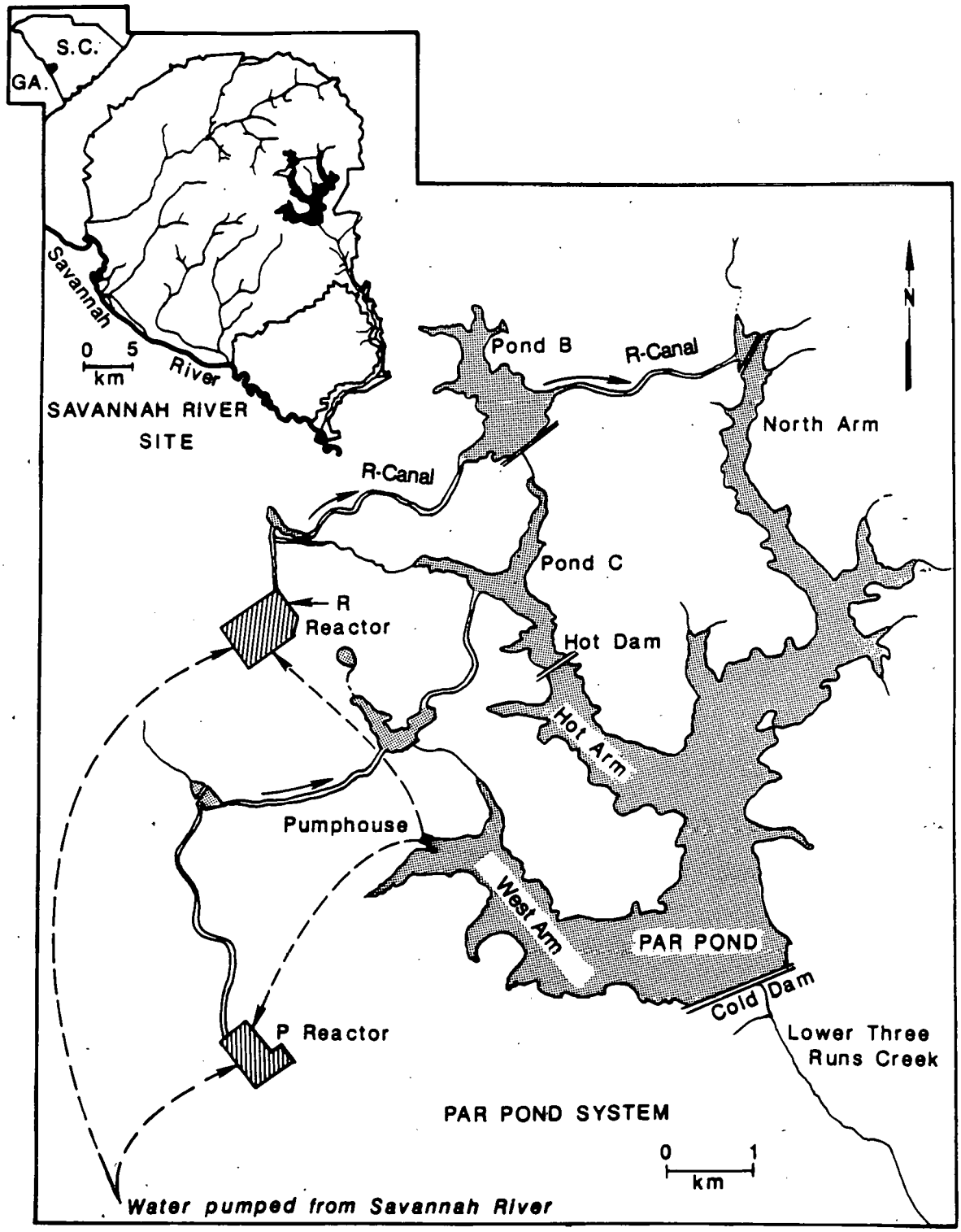

Fig. 1. Map of the cooling water system for R and P reactors, including Ponds B and C, par Pond, and connecting canals.

level has been maintained by precipitation, natural near-surface ground water drainage, and a spillway leading to a continuation of R Canal toward Par Pond (Fig. 1). The system has remained relatively undisturbed and unexploited since June 1964 and a diverse and abundant array of aquatic plants and animals has become established. The present water chemistry reflects the soft, acidic, highly organic subsurface drainage water that feeds the system.

This study was initiated in September 1983. The principal questions asked about Pond B were: (1) What are the total inventories of the more abundant long-lived radionuclides in the system; (2) What fraction of the estimated R Reactor discharge to R Canal is accounted for by these inventories; (3) How are these radionuclides distributed among and within the major components of the ecosystem (i.e. water, seston, sediments, aquatic macrophytes, invertebrates and vertebrates); (4) Can the relationships between radionuclide concentrations in water and other components of the system be predicted from previous literature, and if not, what features of the Pond B ecosystem might explain discrepancies between predicted and observed relationships; and (5) Will Pond B and other similar abandoned reactor cooling reservoirs ever be suitable for normal uses (e.g. recreation, irrigation, etc.) and, if so, what are the time scales and what qualifications should be established for various uses?

STUDY AREA

History

Pond B was filled in July 1961 after completion of a 530 m long, 20 m high earthen dam across Joyce Branch of Lower Three Runs Creek. The dam was faced on the upstream side with asphalt to deter water erosion. A concrete spillway to R Canal (Fig. 1) was constructed at the outlet bay. The basin was cleared of timber and brush prior to filling. The present bottom contours and stratigraphy indicate little earthmoving prior

to filling, except for limited areas near the dam. We found charcoal in numerous dredge samples, indicating that brush piles may have been burned to provide a clean basin.

In September 1961, Pond B started receiving thermal discharge water from R Reactor via R Canal at approximately 11 m^3/s. This water was supplied partially from the Savannah River and partially from the West Arm of Par Pond, which in turn was ultimately supplied from the Savannah River via P and R Reactors (Fig. 1). The initial sources of water to Pond B would have produced a water chemistry similar to that of the Savannah River and Par Pond (Tilly, 1975), but the current chemistry is governed by that of near-surface ground water.

Judging from the appearance of Pond C (Fig. 1), which currently receives thermal discharge from P Reactor, it is very probable that Pond B contained fewer plant and animal taxa during the period it received hot water than at present (Parker et al., 1973; Taylor and Mahoney, 1988). The ends of the more remote bays were probably less impacted by hot water than was the main lake body and may have therefore experienced a more rapid biological succession when thermal inputs ceased.

During 1963 and 1964, periodic radionuclide discharges to R Canal totalled approximately 1.9×10^{15} Bq ^{3}H, 5.7×10^{12} Bq ^{137}Cs, 4.4×10^{11} Bq ^{90}Sr, and lesser quantities of other radionuclides (Ashley and Zeigler, 1980). It is not known what fractions of these quantities were retained by the various segments of the receiving system, nor how the radioactivity might have been distributed between ecosystem components. Discharges from R Reactor could have reached Par Pond either via Pond B or Pond C (Fig. 1). However, the discharge fractions going to either impoundment are not evident from the available records.

Since June 1964, when R Reactor was shut down, Pond B has been maintained by natural, near-surface drainage from its 380 ha watershed, a mean annual precipitation of about 115 cm, and runoff from storm events. Additional drainage from a 280 ha area near the R reactor may also flow into Pond B. Geochemical and biological succession

toward the present state have occurred under a normal temperature regime. With the exception of a radiological monitoring program (e.g. Ashley and Zeigler, 1980; Anonymous, 1984; Zeigler et al., 1986) and several scientific investigations of water chemistry, radionuclide cycling, and biology or ecology, the reservoir has been undisturbed.

Tilly (1975), examined the water chemistry of Pond B while Ruhe and Matney (1980) characterized sediment properties. Further studies of the general water chemistry and interactions of dissolved organic carbon with major and trace elements in both the water and sediments have also been conducted (Alberts and Collins, 1980; Alberts and Dickson, 1985; Alberts et al., 1988a; Alberts et al., 1988b).

Estimates of concentrations of actinide elements in sediments of Pond B were made by Alberts et al. (1986a), and the effects of seasonal physiochemical changes in the pond on the concentrations of ^{232}Th, $^{239,240}Pu$ and ^{241}Am in water have also been reported (Alberts et al., 1986b). Studies were conducted on the release of ^{137}Cs from Pond B and Par Pond sediments during summer thermal stratification and hypolimnetic anoxia (Alberts et al., 1979; Evans et al., 1983), and numerous laboratory studies using Pond B and Par Pond sediments have been undertaken to determine the effects of physiochemical parameters and organics on partitioning of ^{237}Np, $^{239,240}Pu$, ^{241}Am and ^{244}Cm to sediments (Alberts and Orlandini, 1981; Sibley et al., 1984; Sibley and Alberts, 1984; Sibley et al., 1986; Alberts et al., 1986c).

The vascular plants, fish and reptiles of Pond B have been listed by Parker et al. (1973) and compared with the species found in other parts of the Par Pond reservoir system. Domby et al. (1977) measured ^{137}Cs concentrations in wading birds at Pond B, and a number of surveys of the biota of the entire Savannah River Site include mention of species particularly found in or around Pond B (Norris, 1963; Gibbons and Patterson, 1978; Murphy, 1981; Bennett and McFarlane, 1983; Mayer et al., 1986; Brisbin, 1989).

Physical and chemical characteristics

The Pond B surface is at an elevation of 76 m above mean sea level, covers an area of 87 ha, and has a maximum length and width of 1.8 and 0.79 km, respectively. The maximum depth is about 12.5 m. It has four small islands, six distinct bays and 9 km of shoreline (Fig. 2). Bottom slopes, which range from 0 to 13%, average about 5-6%. The mean depth, determined from soundings at 244 intersections of a square grid placed over the impoundment surface (July, 1984), is 4.3 m and the volume is estimated as 3.9×10^6 m^3. The water turnover rate fluctuates with season and rainfall, but has not been measured. Surface outflow through the spillway is intermittent.

The reservoir stratifies from April through October, the thermocline being most pronounced at depths ranging from 4 to 7 m, depending on season. Surface water temperatures in July average about 30 deg C, whereas temperatures below 8 m may be less than 15 deg C. January water temperatures vary little with depth and average about 7 deg C (Alberts et al., 1986b). The reservoir may be classified as warm monomictic (Hakanson and Jansson, 1983).

Dissolved oxygen ranges from < 0.5 g/ml in the summer hypolimnion to 8 g/ml at the surface (Alberts et al., 1986b; Alberts et al., 1988b). In winter, readings of 9-13 g/ml (near saturation) are typical at all depths. The summer anoxia in the hypolimnion limits biological activity to anaerobic processes and appears to create a seasonal remobilization of ^{137}Cs from sediments to the water column (Evans et al., 1983). This process is thought to be caused by ion displacement of ^{137}Cs by cations such as NH_4^+ released under anoxic conditions. Other radionuclides may also be remobilized from sediments during anoxia.

Conductivity of surface water is fairly independent of season and ranges from 20 to 30 S. During hypolimnetic anoxia, the conductivity at 11 m progresses from 40 S in April to 140 S in October (Alberts et al., 1988b). Dissolved and suspended organics are much higher in the hypolimnion than in the epilimnion. SCUBA diving visibility in July

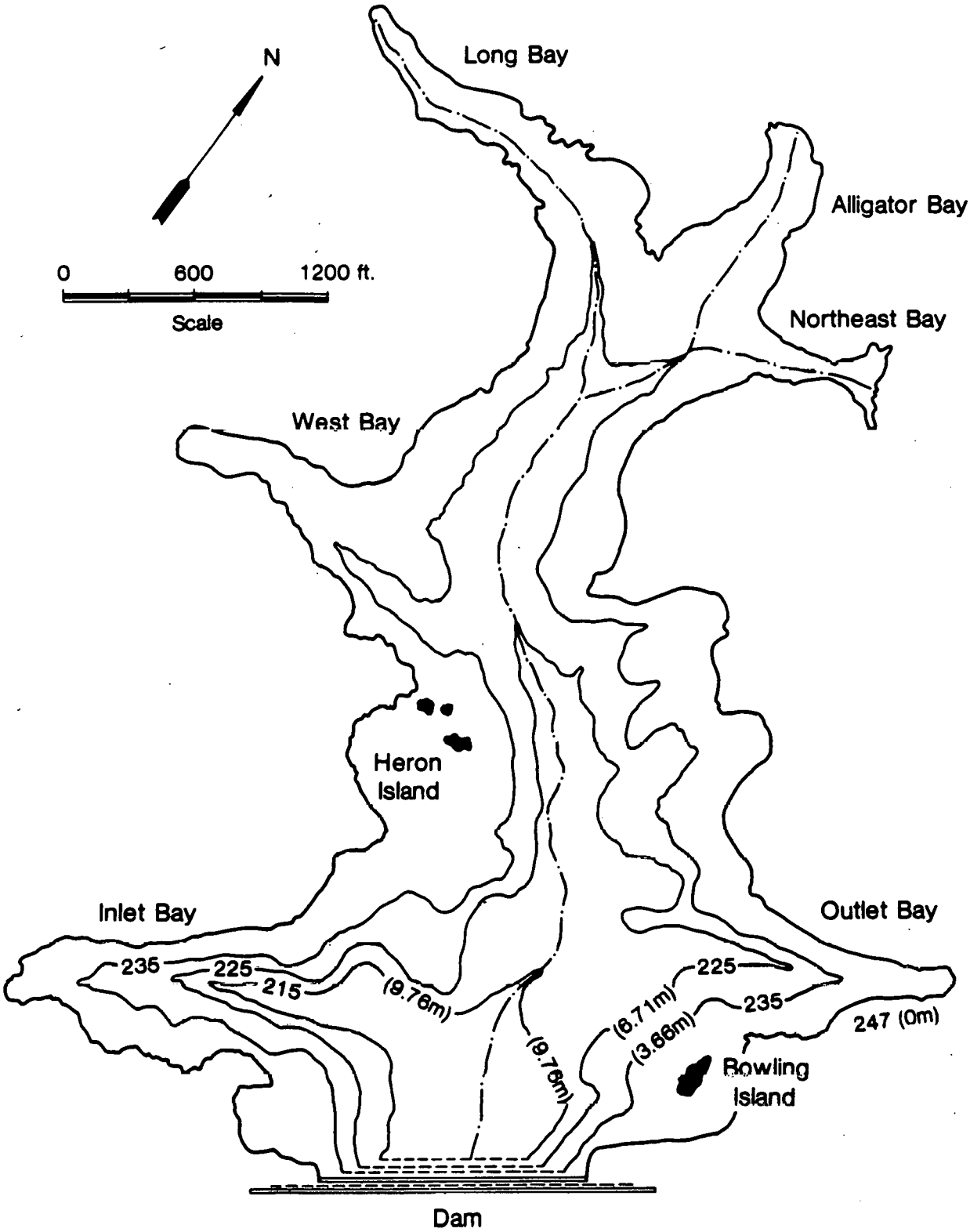

Fig. 2. Map of Pond B, showing bottom contours and names of principal features. Elevations above mean sea level given in feet and depths in meters (m).

drops from 1.0-2.0 m in the epilimnion to < 0.5 m in the hypolimnion. Transparency in October 1983 ranged from 20 to 60%, depending on depth (Alberts et al., 1988b). Secchi disk visibility ranges from 1.7-6.1 m (Chimney et al., 1985). In 1984, mean organic carbon concentrations within the euphotic zones ranged from 3 to 8 mg/L (Chimney et al., 1985).

Pond B water is slightly acidic, with pH values ranging from < 5.3 to 6.8 (Alberts et al., 1988b). Some stratification with depth seems to occur from February through September, with deeper water tending to be more acidic, especially in March and April. A general comparison of the water quality between Pond B and other reservoirs and lakes in the southeastern U.S. is found in Chimney et al. (1985).

Sediments in Pond B vary dramatically by location. They range from wave-washed sand on windy points to 0.5 m thick layers of biogenic (mostly macrophytic) detritus in the ends of the four northernmost bays to kaolinitic clays and silt overlaid by a 2-10 cm layer of organic material (Ruhe and Matney, 1980). Field observations indicate the likelihood of encountering clay-silt sediments in the channel bottoms and sandy material in the areas that would have been described as upland prior to impoundment. Based on analyses of acetate extracts by the Soil Testing and Plant Analysis Laboratory, Athens, Georgia, USA, the mean contents of exchangeable P, K, Ca, and Mg in 20 sediment cores from randomly selected locations (mean length = 26 cm) were 10, 7, 98, and 13 µg/g, respectively.

Biotic composition

Rooted aquatic macrophytes are very abundant in the shallower, wind-protected areas of Pond B. We found vegetation at 98% of the locations sampled in water < 5 m deep. No vegetation was observed at depths > 6 m. The most common species observed include Nymphoides cordata, Brasenia schreberi, Nymphaea odorata, Cabomba caroliniana, and Utricularia floridana (nomenclature from Godfrey and Wooten, 1981).

The major taxonomic groups of phytoplankton that are characteristic of North American freshwaters are found in Pond B, including Bacillariophyta, Chlorophyta, Chrysophyta, Cyanophyta, Euglenophyta and others (Chimney et al., 1985). The average density was about 3.0×10^3 organisms/ml. Mean primary productivity was about 40 g $C \cdot L^{-1} \cdot day^{-1}$ (Chimney et al., 1985). The zooplankton community is composed of four main taxonomic groups: Protozoa, Rotifera, Cladocera and Copepoda. The mean zooplankton density was found by Chimney et al. (1985) to be about 900 organisms/L.

Nine major taxa of benthic macroinvertebrates occur in Pond B and densities of these taxa vary by season and water depth (Whicker, 1988). Some of the more abundant taxons included Annelida, Gastropoda, Amphipoda, Diptera, Coleoptera, Trichoptera, and Odonata.

According to Bennett and McFarlane (1983), Parker et al. (1973) and more recent observations by the authors, the most abundant species of fishes in Pond B include mosquitofish (Gambusia affinis), bluegill (Lepomis macrochirus), largemouth bass (Micropterus salmoides), and yellow bullhead (Ictalurus natalis). Gizzard shad (Dorosoma cepedianum), brook silversides (Labidesthes sicculus), swamp darter (Etheostoma fusiforme), warmouth (Lepomis auritus), black crappie (Pomoxis nigromaculatus) and 5 other less common species have also been observed. It is probable that many of these populations became established from larval stages present in non-thermal water pumped in from the Savannah River and Par Pond (Crawford, 1978). Fish survival during thermal periods likely occurred only in cool water refuges at the ends of bays (Block et al., 1984).

The reptiles and amphibians of the Savannah River Site have been described by Gibbons and Patterson (1978), and those species of reptiles known to occur specifically in Pond B have been listed by Parker et al. (1973). These include the American alligator (Alligator mississippiensis), banded water snake (Nerodia sipedon), yellow-bellied slider turtle (Trachemys scripta), eastern mud turtle (Kinosternon subrubrum), snapping turtle

(Chelydra serpentina) and chicken turtle (Dierochelys reticularia). Additionally, the brown water snake (Nerodia taxispilota) and musk turtle (Sterothaerus odoratus) are now also known to occur in Pond B. Bullfrogs (Rana catesbeina) are the most prominent amphibian of the reservoir although a number of species of smaller frogs, treefrogs, toads and salamanders also use the reservoir, particularly for breeding in the spring and summer months. The slider turtles commonly move overland as much as 5 km and thus are capable of exporting radionuclides (Gibbons, 1986).

Numerous species of aquatic birds are known to occur on the Savannah River Site (Norris, 1963) and any of the waterfowl and/or waders listed in that study could possibly be found at Pond B. Mayer et al. (1986) list 12 waterfowl species whose occurrences have been documented on Pond B. Ring-necked ducks (Aythya collaris) were the most abundant duck species counted on census flights with other abundant species including bufflehead (Bucephala albeola), ruddy duck (Oxyura jamaicensis) and lesser scaup (Aythya affinis). American coots (Fulica americana) are also commonly found on Pond B but were not counted on the aerial censes. These species are present on Pond B almost exclusively as winter visitors. During the summer months, the Pond B waterfowl community consists primarily of a few scattered breeding pairs of wood ducks (Aix sponsa) and common moorhens (Gallinula chloropus) and their broods.

In addition to the waterfowl, several species of waders are found at Pond B, the most important being green-backed herons (Butorides virescens) and little blue herons (Florida caerulea) which may breed in small numbers along the pond margins and on the larger islands (Domby et al., 1977). Numerous common grackles (Quiscalus quiscula) and red-winged blackbirds (Agelaius phoeniceus) also nest in these same areas. Occasional sightings of ospreys (Pandion haliaetus) and bald eagles (Haliaeetus leucocephalus) have been made on Pond B.

METHODS

Sampling and sample preparation

Water, sediments and biota were sampled by designs that would permit estimation of total mass, mean radionuclide concentration and thus total radionuclide inventory for the Pond B ecosystem. Sample numbers were maximized within time and budgetary constraints. Sampling was conducted over a period of 2.5 y. Most samples for radionuclide assay were taken during November and December 1983, thus the radionuclide data represent the state of the system at that particular point in time. Seasonal dynamics and long-term trends were not directly addressed. Mass estimates for most ecosystem components were conducted from February through August, 1984. Additional data were obtained in the summers of 1985 and 1986.

Water. Water samples for ^{137}Cs and ^{90}Sr analyses were taken on November 21, 1983 at 26 locations randomly selected from a square grid consisting of 244 intersections superimposed over a map of Pond B. Each sample consisted of 4 L of water taken midway between the bottom and surface. These midpoints ranged in depth from 0.5 to 5.2 m. On the day of sampling there was no thermal stratification and the water temperature averaged 14 deg C. Samples were taken with a polyethylene 3 L Van Dorn bottle. Samples were transported to the laboratory in 4-L polyethylene bottles and maintained in darkness under refrigeration. Estimates of $^{239,240}Pu$, and ^{241}Am in the water column were available from Alberts et al., 1986b.

Water samples were filtered the day following collection through a 0.45 μm Millipore particulate filter (HAWP-293-25), and the filtrates were acidified with 40 ml concentrated HCl. Stable Cs and Sr carriers were also added after filtration. The filters were folded, air-dried, and placed in labeled vials for storage. The filtrates were evaporated in 1 L Pyrex beakers to a volume of about 50 ml, transferred to a 40 dram polystyrene vial using a 1 M HCl rinse, and brought to 100 ml with 1 M HCl. After

mixing, 20 ml were placed in a separate vial for ^{90}Sr and transuranic radionuclide assay, and the remaining 80 ml were used for gamma ray spectrometry.

Sediment. Sediment cores were taken between November 22, 1983, and March 23, 1984, from 46 randomly selected locations on the same 244 point square grid used for water sampling. Each core was 2.4 cm in diameter. Core lengths ranged from 6-49 cm, depending on the hardness of the substrate; the mean (±1 sd) core length was 23 (±9) cm. These core lengths were determined from counting 1 cm depth segments to be sufficient to capture > 95% of the total ^{137}Cs and transuranic inventory. Water depths above the sediment cores ranged from 0.7 to 12.0 m and averaged (±1 sd) 4.6 (±3.3) m. The distribution of radionuclides and other characteristics were studied in 1 cm thick increments of 15 cores, while the remaining 31 cores were homogenized in total prior to assay.

The cores were taken by driving a 2.7 cm inside diameter aluminum pipe into the bottom. The aluminum pipe contained a plastic tube approximately 70 cm in length to retain the core. Sections of the aluminum pipe were screwed together to form a pole of sufficient length to reach the bottom. A sharpened coupling fitted to the bottom section of pipe retained the plastic tubing and eased insertion into the sediment.

The aluminum pipe was pushed vertically into the sediments by hand until sufficient resistance was met to require driving with a hammer. Final driving formed a plug of compacted sediment in the bottom of the tube, which prevented loss of the core upon withdrawal. The cores were maintained in a vertical position until frozen in the laboratory. Inspection of the clear plastic tubes indicated minimal vertical displacement or smearing and excellent preservation of the stratigraphy. Slight vertical compression of the cores may have occurred (Baxter et al., 1981), but this was not evaluated.

Frozen cores were cut into 1 cm thick sections. The plastic tube and frozen contents were cut as a unit to maintain integrity. Each core section was placed in a

pre-weighed aluminum dish, allowed to thaw, and the plastic ring was removed and rinsed with deionized water over the dish to transfer residual sediment. The samples were dried 48 h at 80 deg C, cooled, weighed, and stored in 20 ml glass vials.

Sediment cores to be used solely for inventory estimates were thawed, quantitatively transferred to pre-weighed 600 ml Pyrex beakers, dried at 80 deg C for 48 h, cooled, and weighed. The sample was transferred to a mortar, crushed and homogenized with a pestle, then transferred to a 40 dram polystyrene vial for storage.

Aquatic macrophytes. Rooted aquatic macrophytes were sampled to determine radionuclide concentrations, percent cover, and biomass. To obtain representative samples for radionuclide assay, 60 sampling locations were chosen at random from 325 equally-spaced points along the Pond B shoreline and marked with aluminum rods. Transects extending perpendicularly from the shoreline at each location to a point in 6 m of water (or the deepest point in shallow bays) were used for sampling. Each species intercepted by a 1.5 m wide transect was recorded and sampled for radionuclide assay. Samples were taken from a boat in the approximate center of the vegetation bed where intercepted by the transect. Water depth was recorded at each sampling location. Some 273 samples representing 25 taxa were collected November 1-6, 1983. Species identifications were confirmed by R. R. Sharitz, Savannah River Ecology Laboratory, Aiken, South Carolina, USA, with nomenclature from Godfrey and Wooten (1979, 1981).

Macrophyte samples for radionuclide assay were taken by hand, garden rake or Ekman dredge, depending on water depth. We attempted to collect all plant material extending more than $\simeq$2 cm over the bottom. When material came up with roots attached, the roots were cut off and discarded. Material was thoroughly rinsed in the surface water to remove visible sediment or loosely attached periphyton. Material was sorted by taxa and placed in sealable plastic bags for transport to the laboratory.

Percent cover and biomass of aquatic macrophytes were estimated using the 244 point square grid. Each grid point was marked in the reservoir with a numbered float

anchored by string to a weight resting on the bottom. When a species touched the taut, vertical string at a given grid location, it was scored as present. Cover, calculated as the frequency of occurrence of a given species across the set of grid points, was determined by SCUBA diving at each location in water shallow enough (< 6 m) to contain rooted vegetation. Ponar grabs and diving searches confirmed the absence of vegetation in water depths > 6 m. Vegetation occurred at 68% of the locations.

A random subset of 84 of the vegetated locations was selected for sampling of plant biomass. Sampling was conducted July 25-August 7, 1984, using a square sampling frame placed on the bottom by SCUBA diving. The frame was centered over the location of the grid point anchor. Sampling frames were constructed of 1.9 cm diameter plastic plumbing pipe. One side of the frame was open so that it could be easily inserted into position on the sediment surface. Two frame sizes were used (0.09 and 0.31 m^2); the smaller being more convenient for very dense vegetation and the larger being more representative for sparse or heterogeneous beds. All vegetation rooted within the frame was clipped 1-2 cm above the sediment surface, placed into a nylon mesh bag, brought to the surface, rinsed to remove adhering sediment and epiphytic periphyton, sorted by species, and placed in plastic bags for transport to the laboratory.

Aquatic macrophyte samples were allowed to drain 1-2 h on clean, absorbent paper, placed in preweighed paper bags, dried to a constant mass at 65 deg C, and weighed. Samples for radionuclide assay were ground in a Wiley mill to < 1 mm and were well-homogenized after oven drying.

Benthic macroinvertebrates. Benthic macroinvertebrates were sampled for biomass estimation in February, May, and July-August, 1984 (Whicker, 1988). The estimates were based on samples from a 15 x 15 cm Ponar dredge at 52 locations in Pond B. The locations were randomly chosen within four water depth strata (0.5-1.5, 2.0-3.0, 3.5-5.5, and > 6 m), along 20 of the 60 shoreline transects used for aquatic macrophyte sampling. Samples were obtained for radionuclide assay by Ponar dredge in

January 1984. These sampling locations were not formally randomized because many samples from various locations had to be pooled to obtain sufficient mass for analysis. Because of the difficulty in obtaining adequate mass for radionuclide analysis, a very limited number of samples was available for radionuclide determination.

After triggering and hauling to the surface, the dredge was emptied into a plastic bucket. The benthos samples were washed in lake surface water using a No. 30 (590 μm) U.S. Standard sieve. This washing procedure removed clay, silt and small detritus particles. Washed material was placed in a sealable polyethylene bag and covered with 80% ethanol for transport and storage. Visible invertebrates were separated from detritus in white porcelain trays under a magnifying lamp (Whicker, 1988). Animals were sorted by taxa and dried 24 h at 60 deg C before weighing. Dried samples were stored in 10 dram polyethylene vials.

Fish. Fish were collected for radionuclide analysis with graduated mesh (2.5 to 7.5 cm, stretched) gill nets and angling from October 19, 1983, to February 23, 1984. The 90 by 1.5 m gill nets were set perpendicularly to shore in water ranging in depth from 1 to 5 m. Smaller mesh ends were placed in the shallower water. Locations ranged from vegetation-sparse areas of the main body of Pond B to heavily vegetated bays. Nets were set evenings and retrieved the following morning. Angling was pursued over most types of habitat. A few small bait fish were also obtained by opportunistic dip-netting. Fish were placed in sealable polyethylene bags over ice and taken to the laboratory where they were identified to species, weighed, measured for length, and frozen. Later, samples were thawed and dissected to separate muscle and bone tissues. Muscle tissues were placed in 40-dram polystyrene vials for gamma ray spectrometry. Bone samples were scraped and briefly boiled in water to facilitate removal of flesh. They were then oven dried, weighed, ashed at 400 deg C for 5-7 d, reweighed, crushed with mortar and pestle, and stored in vials.

Turtles. Turtles were trapped with 6 baited hoop nets in water approximately 1 m deep. Locations were subjectively chosen to maximize trap success. A major trapping effort was made in 1984 although some individuals marked in previous years were still in the population and retrapped. Captured Trachemys scripta were taken to the laboratory, weighed, sexed, measured and marked by drilling holes in specific marginal scutes (Cagle, 1939). Most turtles were then released back into the reservoir at the site of capture. Ten animals ranging in mass from 142 to 1,837 g were bagged and immediately placed in a freezer for radionuclide assay.

Turtles that were frozen for radionuclide assay were thawed, then dissected. Soft tissues (muscle, intestine, blood, viscera, skin, liver, heart and kidney) were separated, weighed, and placed in 40-dram vials for gamma ray spectrometry. After counting, these samples were freeze-dried for storage. Shell and bone samples, after careful removal of adhering soft tissue, were weighed, oven dried at 60 deg C for 7 days and reweighed. The material was then broken into pieces < 3 cm, and the pieces were placed in porcelain crucibles and ashed at 400 deg C until they could be ground to a powder with a mortar and pestle (usually 1-2 weeks). The powder was transferred to 40-dram vials for gamma ray spectrometry. After determining ^{137}Cs, which volatilizes at about 450 deg C, the ash was placed in a 900 deg C furnace for a few days to produce a completely white ash which was stored for further radionuclide assay.

Waterfowl. Seven coots were collected by shooting from Pond B on February 14, 1984, for radionuclide assay. The birds were dissected and tissues processed in a manner comparable to fish and turtles. Soft tissues were placed in 40-dram polystyrene vials for ^{137}Cs assay. Bone samples were treated in the same manner as fish bones.

Analytical procedures

Cesium-137. Analysis of samples for ^{137}Cs was accomplished without any chemical separation because of the penetrating 662 keV gamma ray that accompanies

94% of the nuclear disintegrations (Whicker and Schultz, 1982) and the lack of significant quantities of interfering radionuclides in Pond B samples. Samples were normally homogenized and placed in containers of standard size and geometrical configuration. Containers were positioned in a consistent manner relative to a lead-shielded 10.2 cm thick by 15.2 cm diameter NaI(Tl) crystal or a 4.6 cm thick by 4.3 cm diameter Ge(Li) coaxial detector. Electrical pulses representing total absorption of the 662 keV gamma ray were recorded with a multichannel analyzer (Canberra Model 8100 for the NaI(Tl) and Canberra Series 85 for the Ge(Li)).

When using the NaI detector, corrections were made for counter background and Compton or photoelectric interactions from naturally occurring radionuclides (^{40}K; ^{238}U daughters; and ^{232}Th daughters). The latter types of corrections were generally insignificant, however, due to the comparatively low levels of natural radioactivity in the samples. The high resolution of the Ge(Li) detector (1.9 keV at 1.33 MeV) simplified the problem of dealing with interferences from other gamma rays. Since neither the detector background nor radionuclides other than ^{137}Cs produced elevated count rates above the baseline in the region of 662 keV, it was only necessary to determine the net counts above the spectral continuum between 658 and 666 keV.

Phantoms of identical geometrical configuration and similar density were prepared as standards to estimate counting yields (c/m per Bq). The phantoms contained water, sand or sawdust and these were uniformly spiked with known quantities of ^{137}Cs (Amersham; traceable to standards prepared by the U.S. National Bureau of Standards). Various media volumes were prepared so the best-fitting mathematical relationship of volume to counting yield could be determined. Thus samples of various volumes could be measured with good accuracy. Count times were sufficient to produce counting standard deviations of < 5% of the actual estimate (Currie, 1968).

Strontium-90. With the exception of bone ash, most samples were sent to the EAL Corporation (Now Thermo Analytical, Inc.), Richmond, California, USA, who performed

the analyses for ^{90}Sr. Biological samples were reduced to ash at 500-900 deg C prior to shipment to EAL; water was sent as a concentrated liquid; and sediments were sent dry. Ash was completely dissolved with $HCl-H_2O_2$, while sediments were rigorously leached with a strong acid prior to analysis to remove ^{90}Sr. Solutions were made to a known volume and yttrium carrier was added and allowed to equilibrate. The solution was extracted with HDEHP at a pH of 1, back-extracted with concentrated HCl, chemically purified by fluoride, hydroxide and oxalate steps, then converted to yttrium oxide. The yttrium oxide was weighed and mounted on a low background planchet with copper backing. The planchet was counted for ^{90}Y (the ^{90}Sr daughter) beta particles on a low background beta counting system. The ^{90}Y activity was corrected for ingrowth and decay, chemical yield, and counting yield.

Bone ash samples were crushed to a powder with mortar and pestle and about 3 g were packed into a pre-weighed 2.54 cm by 0.64 cm high steel beta counting planchet. After weighing, the planchets were directly beta counted with a 3 cm diameter end-window Geiger-Mueller tube connected to a Canberra Model 2071 timer-scaler. Count rates were corrected for machine background, contributions from natural radioactivity in bone, and ^{137}Cs in bone. Counting yields were determined by uniformly contaminating several uncontaminated bone ash samples with a standard solution of ^{90}Sr obtained from Amersham. These were then counted under a geometrical configuration identical to that of the samples. The counting yields were independently verified by an EAL radiochemical analysis of 7 bone ash samples that were measured by the direct beta counting technique. Regression of the direct beta counting result on the EAL radiochemical result gave a regression coefficient of 0.99 and a correlation coefficient of 0.98, indicating agreement between the two methods.

Transuranics. Analyses for isotopes of plutonium, americium and curium were all done commercially by the EAL Corporation. Liquid aliquots from the ash dissolutions or acid leaches (see the Strontium-90 section) were equilibrated with ^{236}Pu and ^{243}Am

tracers. The plutonium was absorbed on AG 1x8 resin (Bio-Rad Corp.) in 8 N HNO_3, while the effluent and wash were saved for Am and Cm analysis. The absorbed Pu was eluted with HCl-NH_4I and electrodeposited on a stainless steel counting disk from an ammonium sulfate electrolyte.

A calcium carrier was added to the Am and Cm fraction and calcium oxalate was precipitated at pH 3.5. The oxalate, which carried Am and Cm, was dissolved and reprecipitated several times to purify the precipitate. The Am and Cm were separated from calcium on a $Fe(OH)_3$ precipitate, which was dissolved in HCl for passage through a cation exchange column. After elution, Am and Cm were electrodeposited on a stainless steel disk.

Classical alpha spectrometry measurements were made on the disks to determine the kinds and amounts of radioisotopes present. Tracer recoveries were used to estimate chemical recovery of the entire procedure.

Stable elements. Samples of water, aquatic vegetation, benthic invertebrates, bone ash and muscle tissue were assayed for calcium and potassium, because of their influence on the behavior of Sr and Cs, respectively (Whicker and Schultz, 1982). Samples were submitted to Environmental and Chemical Sciences, Inc., Aiken, South Carolina, USA, for analysis by standard methods.

Compartmental mass estimation

Compartmental mass estimates were required for the calculation of radionuclide inventories in the various ecosystem components. Estimates for the abiotic compartments were obtained from morphometric measurements and calculations. The methods used to estimate the morphometric parameters were adopted primarily from Hakanson (1981). Basic information available for Pond B included a topographic map compiled prior to the construction of the dam, aerial photographs taken October 27, 1978, and a set of soundings taken at 244 square grid intersections in late July, 1984.

Data from each of these forms of information were combined and reconciled to produce a base map with bottom contours (Fig. 2). Using the scaled contour map and a planimeter, the area of each depth zone was determined and a hypsographic curve was constructed, from which the volume (3.9×10^6 m^3) was estimated. Since sediments were sampled by area from a vertical tube, the total sediment area was taken as the area of the lake surface, namely 87 ha.

The total dry mass by depth zone of each of the 7 most abundant aquatic macrophytes was estimated as the product of the frequency of occurrence (cover), the mean biomass (dry g/m^2), and the surface area. All species other than the 7 most abundant ones were pooled into a single additional category. Frequency was estimated as the number of occurrences divided by the number of grid points sampled within a specific depth zone. Mean biomass was taken as the sum of all measurements of dry mass per unit area in a depth zone divided by the number of measurements. The surface area representing a given depth zone was computed from the morphometric data. Total dry mass was summed across all depth zones to yield the estimate for the total system.

Dry mass of the benthic macroinvertebrate component was estimated by taxa and depth zone (Whicker, 1988). For each taxa and depth zone, the total dry mass was taken as the product of the mean density (individuals/m^2), the mean dry mass per individual, and the area within a depth zone. Summation across all depth zones provided a total system estimate by taxa. Total dry mass estimates for the reservoir in 1984 for all taxa were 775, 396, and 184 dry kg in winter, spring and summer, respectively (Whicker, 1988).

An estimate of the largemouth bass population (15 cm in length) was made in the summer of 1986 by a mark-recapture technique. Over 600 individuals were captured by systematic angling, then measured (mass, length), double-marked with numbered dorsal tags, and released. Recaptured fish were recorded through the summer. Data were analyzed by the program CAPTURE (White et al., 1982) to produce an estimate of the

total population, with confidence limits. Mass of the population was taken as the population estimate (5,434; 95% CI = 3589-7279) times the mean mass per individual (255 g, based on 747 individuals examined). Other fish populations were not directly estimated. Crude estimates were nevertheless constructed by scaling to the largemouth bass population according to the ratios of numbers taken in gill nets, and the mean individual masses of the various species captured.

The slider turtle population was estimated in 1984 by a mark-recapture procedure and a modified Lincoln index. The data were broken down by size class. The number in each size class was multiplied by the mean mass per individual in the class, and these products were summed to estimate the total mass for the system. An estimate of 620 individual slider turtles was obtained (J. Green, Savannah River Ecology Laboratory, Aiken, South Carolina, USA, personal communication).

Population and mass estimates of Pond B waterfowl, together with assessments of the seasonality of their occurrence were obtained from previously published studies (Brisbin, 1974; Mayer et al., 1986; Brisbin, 1989). In calculating radionuclide inventories, the Pond B waterfowl population was estimated to be comprised of 72 coots and 102 diving ducks. Body sizes and radionuclide body burdens of diving ducks were typified by those of the ring-necked duck, the most common diving duck species present. Average live body weights of coots (536 g/bird) and ring-necked ducks (682 g/bird) were based on Brisbin (1989).

The population status of the alligator in Pond B has been discussed by Murphy (1981) and Brisbin (1989). The most recent of these two studies made a direct count of 12 alligators in Pond B, ranging in size from small juveniles to large adults. These counts were based on 8 night-time eye-shine census cruises of the reservoir during the summer of 1988 (Brisbin, 1989).

RESULTS AND DISCUSSION

Water column

Cesium-137. Based on 26 filtered water samples taken November 21, 1983, the mean ^{137}Cs concentration (±1 sem) was 0.76 (±0.04) Bq/L. There was no statistically significant correlation with sampling depth or trend over locations, although two samples from 0.5 m of water in the end of Long Bay (Fig. 2) were atypically low (0.13 and 0.21 Bq/L) in ^{137}Cs. These low values could possibly reflect the presence of local springs, as they were occasionally noticed while SCUBA diving in shallow bays. Oxygen and temperature measurements on this date were uniform from the surface to the bottom, indicating the lack of stratification. Measurements on October 2, 1983, when the water was stratified, gave mean concentrations of 0.43 and 1.10 Bq/L in the epilimnion and hypolimnion, respectively. A substantial concentration gradient for ^{137}Cs in water during hypolimnetic anoxia was previously documented by Evans et al. (1983). The mean (±1 sem) content of cesium's nutrient analogue, K, in the filtered water samples was 0.29±0.01 g/ml (n = 30).

The long-term trend of ^{137}Cs in Pond B water appears to be that of a gradual decline, with an annual cycle superimposed on the trend. For example, in November 1978 the mean concentration was 1.6 Bq/L, over twice the November 1983 value (Alberts et al., 1987). In 1978, monthly measurements revealed a cycle with minimum values occurring in April (0.66 Bq/L) and maximum values in November (Alberts et al., 1987). This seasonal cycling phenomenon for ^{137}Cs, originally documented in Par Pond (Alberts et al., 1979), also appears to occur with similar timing in Pond B, presumably due to the mechanism proposed by Evans et al. (1983).

The content of ^{137}Cs in seston was estimated from analysis of the 0.45 μm Millipore filters through which water samples were passed. Four groups of 6 pooled filters were counted. Results indicated a mean ^{137}Cs content of 0.04 Bq±0.01 Bq (sem)

in the material filtered from each liter of water, suggesting that about 5% of the ^{137}Cs in the water column was associated with particles > 0.45 m.

Strontium-90. Aliquots of the same filtered water samples measured for ^{137}Cs had mean ^{90}Sr concentrations (±sem) of 0.14 (±0.005) Bq/L (n = 24). There was no statistically significant correlation with sampling depth and no clear effect of sampling location. However, as was the case for ^{137}Cs, one sample from the back of Long Bay was much lower (0.037 Bq/L) than the remaining samples; the other Long Bay sample that was low in ^{137}Cs was lost prior to ^{90}Sr analysis. Time series data for ^{90}Sr in Pond B water are not available. Recent data suggest higher ^{90}Sr concentrations in the hypolimnion during periods of thermal stratification and hypolimnetic anoxia (K. Orlandini, Argonne National Laboratory, Argonne, Illinois, USA). This phenomenon has been documented for other alkaline earths (Sholkovitz, 1985; Alberts et al., 1988b). Based on analyses of the 4 groups of 6 pooled 0.45 μm Millipore filters used to filter the water samples, 97% of the ^{90}Sr passed through the filters while 3%±0.3% (sem) was retained. Calcium, the nutrient analogue of Sr, had a mean (±1 sem) concentration in 31 filtered water samples of 1.7±0.1 g/ml.

Transuranics. Eight separate water samples (two replicates at 0, 4, 8 and 11 m depths) were also taken in November 1983 for ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm assay. Replicates were averaged, then average values by depth were weighted for the fractional volumes of water in the four depth zones (0.58, 0.28, 0.11, and 0.03 for 0, 4, 8, and 11 m, respectively). The weighted mean concentrations in filtered water samples were $3.4x10^{-6}$, $8.8x10^{-6}$, $2.3x10^{-5}$, and $6.4x10^{-4}$ Bq/L for ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm, respectively. The value for ^{238}Pu was estimated by an analysis of ^{238}Pu and ^{239}Pu in a November 1984 sample (K. Orlandini, Argonne National Laboratory, Argonne, Illinois, USA). The ^{239}Pu values reported by EAL and Argonne differed by only 11% and there is no reason to suspect a change in the $^{238}Pu/^{239,240}Pu$ ratio. A substantial portion of the transuranic radioactivity passed through the 0.45 μm filters (43, 23, 79

and 93% for ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm, respectively). Plutonium-239,240 and ^{244}Cm tended to increase slightly in concentration with water depth, but relatively high counting uncertainties (1 sd counting errors were frequently > 30% and occasionally >100% of the net count rate) and frequently poor agreement between replicates precluded the conclusion of a depth effect. During periods of thermal stratification and hypolimnetic anoxia, however, the deeper waters were clearly elevated in $^{239,240}Pu$ and ^{241}Am. Total inventory calculations, however, did not support the idea of remobilization of actinide elements from sediments (Alberts et al., 1986b). The increase appeared instead to result from downward transport of material from the epilimnion.

Sediments

Cesium-137. Data from 15 individual sectioned cores were averaged for each depth segment of the core to produce a composite profile of ^{137}Cs concentration (dry mass basis) versus depth (Fig. 3). The trend was for maximum concentrations to occur at or near the surface and for levels to decrease sharply with depth in the sediments for 5-10 cm, and more gradually thereafter. The data approximate an exponential decline with a halving depth of about 3.5 cm.

A slightly different profile was observed when we plotted the ^{137}Cs activity per core segment (i.e. $Bq \cdot cm^{-2} \cdot cm^{-1}$) versus depth. In this case, the activity was expressed on a volume rather than mass basis. Due to the generally higher organic matter content and lower compaction, the upper layers of sediment are less dense than deeper layers. A plot of the mean quantity of ^{137}Cs per core segment versus depth revealed an increase from 7.4 $Bq \cdot cm^{-2} \cdot cm^{-1}$ at the surface to a maximum of 10.0 $Bq \cdot cm^{-2} \cdot cm^{-1}$ at 2-3 cm, followed by an exponential decline with a halving depth of about 4 cm. Numerical integration of the data expressed as quantity per core segment yielded 95% of the total inventory through a depth of 17 cm.

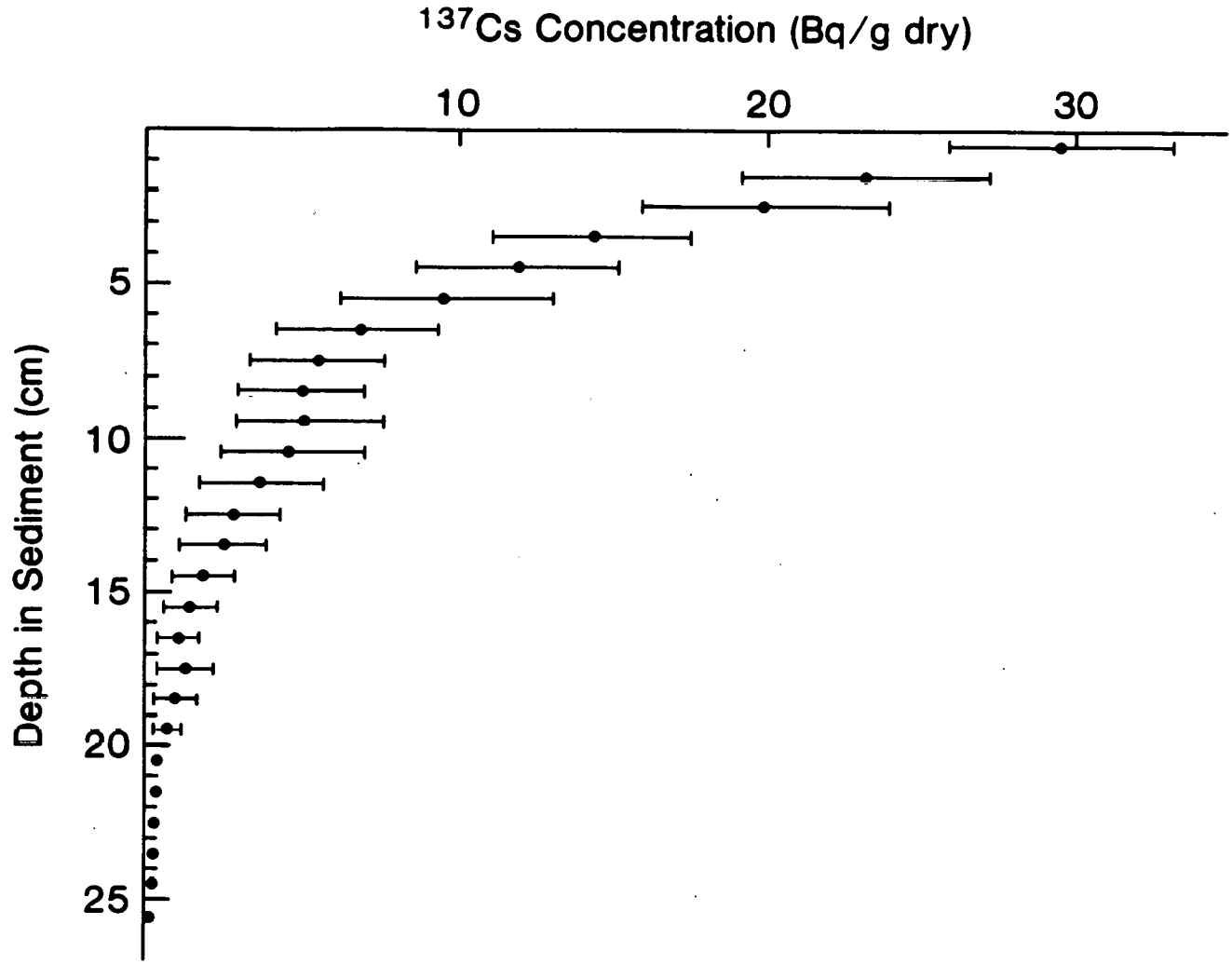

Fig. 3. Mean ($\pm$1 sem) ^{137}Cs concentrations in 1 cm depth increments of 15 Pond B sediment cores.

Individual core profiles were examined with regard to any significant departures from the composite profiles just described. Nine of the 15 individual cores yielded depth profiles that were very similar to the composite profiles. These similarities held with respect to depths of peak concentrations and rates of decline with depth in the sediment, regardless of whether the amounts of ^{137}Cs were expressed as Bq/g or $Bq \cdot cm^{-2} \cdot cm^{-1}$. However, six cores differed from the composite, primarily in the depth of maximum concentrations. Four cores yielded maximal concentrations that were below the 0-1 cm horizon; these maxima ranged from 2-3 cm to 8-9 cm into the profile. Similarly, six cores had maximal amounts per core segment that ranged from 3-4 cm to 10-11 cm in depth.

We believe that the composite core profiles reflect initial adsorption and deposition of radioactivity on the sediment surface, followed by a slow migration into the deeper layers. The composite data also suggest a relatively low sedimentation rate in Pond B since 1964. Actual measurements of sediment accumulation in traps from February 1987 through February 1988 confirm low sedimentation rates in Pond B (<0.02 $g \cdot cm^{-2} \cdot yr^{-1}$ in open water; < 0.2 $g \cdot cm^{-2} \cdot yr^{-1}$ in macrophyte beds). However, in the case of those cores with maximal contents of ^{137}Cs well-below the surface horizon, it seems probable that local sedimentation events occurred after the principal inputs of the radionuclides. All but one of the cores with ^{137}Cs maxima at greater depths were taken in or near the inlet and outlet bays and four of these were taken where bottom slopes were much higher than average. Sediment from erosion of R Canal when operative and/or bottom transport (Hakanson and Jansson, 1983) could have covered the original contaminated sediment surface with several cm of less contaminated material. An alternative hypothesis is that the ^{137}Cs exhibited a more rapid net downward migration in all or some of these atypical locations. We were not able to determine whether the profile differences related to sediment histories or to different ^{137}Cs migration rates. The net vertical directional water flow at the sampling locations was not known.

A total of 46 sediment cores was available to estimate the total ^{137}Cs deposited per unit area. The activity in the 15 sectioned cores was summed to obtain the depth-integrated deposition, while 31 additional cores were physically mixed prior to assay to estimate the total ^{137}Cs content. The total ^{137}Cs deposition in sediment was observed to increase with water depth to a depth of 3 m, then to decline to a depth of 6 m, and then increase again in water > 6 m deep (Fig. 4). Various statistical procedures (non-linear regression and analysis of variance) indicated that the likelihood of observing this particular pattern by random chance was extremely small.

We believe that the low deposition in shallow water may be due to periodic resuspension of fine particles by wave action. The maximum at 2-3 m could possibly be explained by the abundance of macrophytes, which sorb radionuclides from the water column then undergo senescence to form a detrital input to sediments. Furthermore, the macrophytes probably reduce water turbulence, which could enhance particle settling. Sediment traps placed in macrophyte beds in February 1987 accumulated 9 times the mass of material as did traps placed in open water for the following 12 month period. Sedimentation rates in macrophyte beds were about twice the open water rates from February through September, but they were about 30 times the open water rates from October through December. The depth zone from 3-6 m has a reduced macrophyte biomass and the sampling locations had steeper than average bottom slopes. Bottom slopes within the 3-6 m depth zone, estimated from the bottom contour map, ranged from 3.5 to 10.8% and averaged 6.5%. Bottom slopes > 5% are considered unstable and subject to sediment sloughage (Hakanson and Jansson, 1983). Furthermore, the chemocline varies over this depth zone, possibly causing some resuspension. Also, fish may congregate at 3 to 7 m during summer anoxia and create additional resuspension. The profundal zone > 6 m has a relatively flat bottom and would provide a sink for particulates since little resuspension or transport would be expected from this zone.

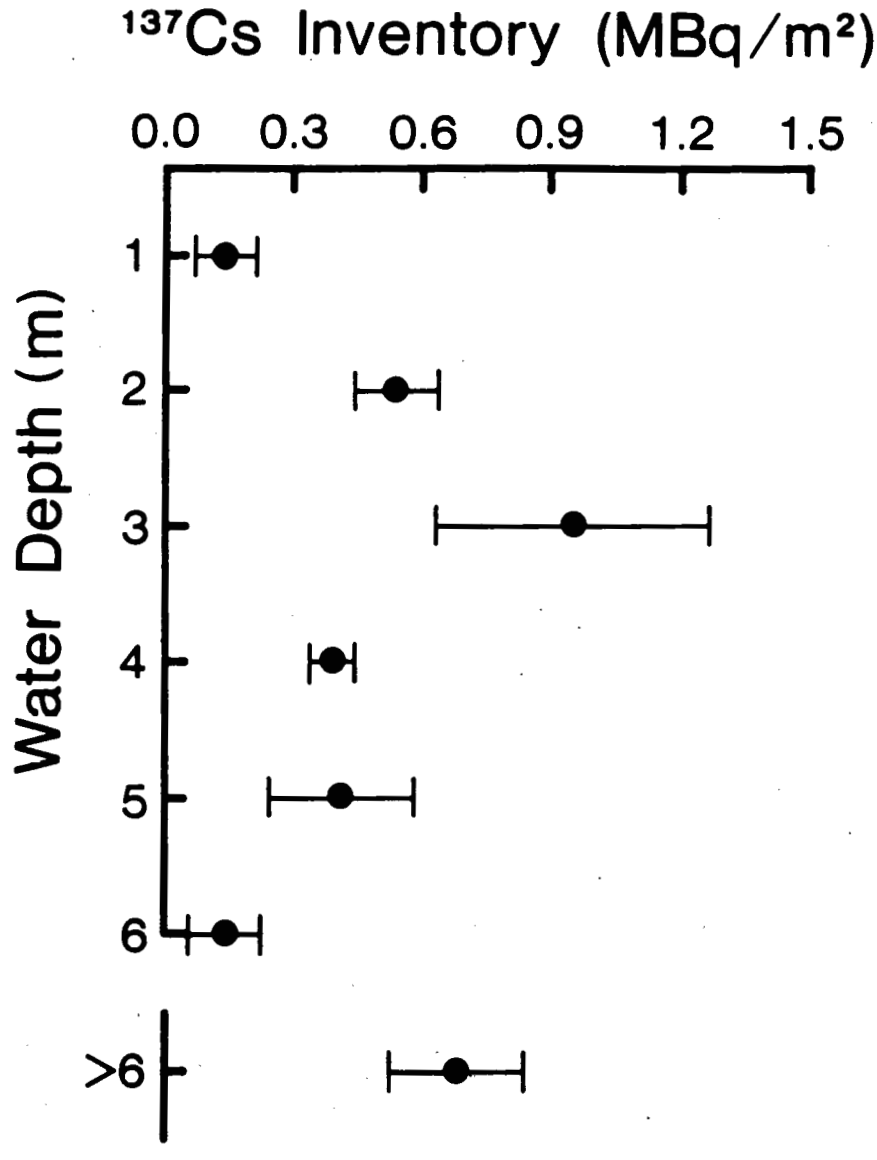

Fig. 4. Total depth-integrated ^{137}Cs per unit area (MBq/m^2) in Pond B sediments as a function of water depth (m). Mean ± 1 sem are shown.

Strontium-90. Concentrations of ^{90}Sr were measured at 1 cm increments in two sediment cores taken in the mouth of the outlet bay in about 7 m of water. These cores, B-10 and B-12, were assayed for all radionuclides included in the study to compare profiles. Strontium-90 concentrations were 1-2 orders of magnitude lower than ^{137}Cs values, depending on depth in the core, and they ranged from 0.03 to 0.35 Bq/g. In core B-10, ^{90}Sr showed no relationship to depth, while in core B-12, it peaked at 5-6 cm and then declined (Fig. 5).

It is evident from Fig. 5 that the ^{90}Sr profiles are quite dissimilar to the ^{137}Cs profiles. In core B-10, ^{137}Cs exhibited a definite peak between 5 and 8 cm, while ^{90}Sr showed no clear pattern. In core B-12, both radionuclides had a defined peak, but they did not coincide. These data suggest a more rapid movement of ^{90}Sr into the deeper sediments than ^{137}Cs, which is consistent with the expectation of a higher K_d for Cs than Sr (Clanton et al., 1964; Huff and Kruger, 1970; Duursma and Gross, 1971). The K_d for Pond B sediment/water, using the mean filtered water concentrations and the 0-3 cm sediment concentrations, is 3.2×10^4 for ^{137}Cs and 1.2×10^3 for ^{90}Sr. These values provide further evidence to indicate that the Pond B system is much less effective in retaining Sr than Cs. In the case of ^{90}Sr, a roughly 10 fold greater fraction of the water+sediment inventory would be expected to reside in water, which carries dissolved material away from the system by mass flow. This concept is also consistent with the relative surface sediment concentrations of the two radionuclides compared to the reported R Reactor release to R Canal. The ratio of Cs/Sr activity in 0-3 cm sediments divided by the Cs/Sr ratio for the reported releases is 11.4, again indicating that Cs has been more efficiently retained than ^{90}Sr, by roughly an order of magnitude.

Twenty-three sediment cores from random locations were assayed in total to estimate the ^{90}Sr inventory. The mean value (±1 sem) was 4.40±0.67 kBq/m^2. The pattern of deposition according to water depth was similar to that of ^{137}Cs, in that minimal values occurred at depths < 1 m (1.15 kBq/m^2) and maximal values occurred at

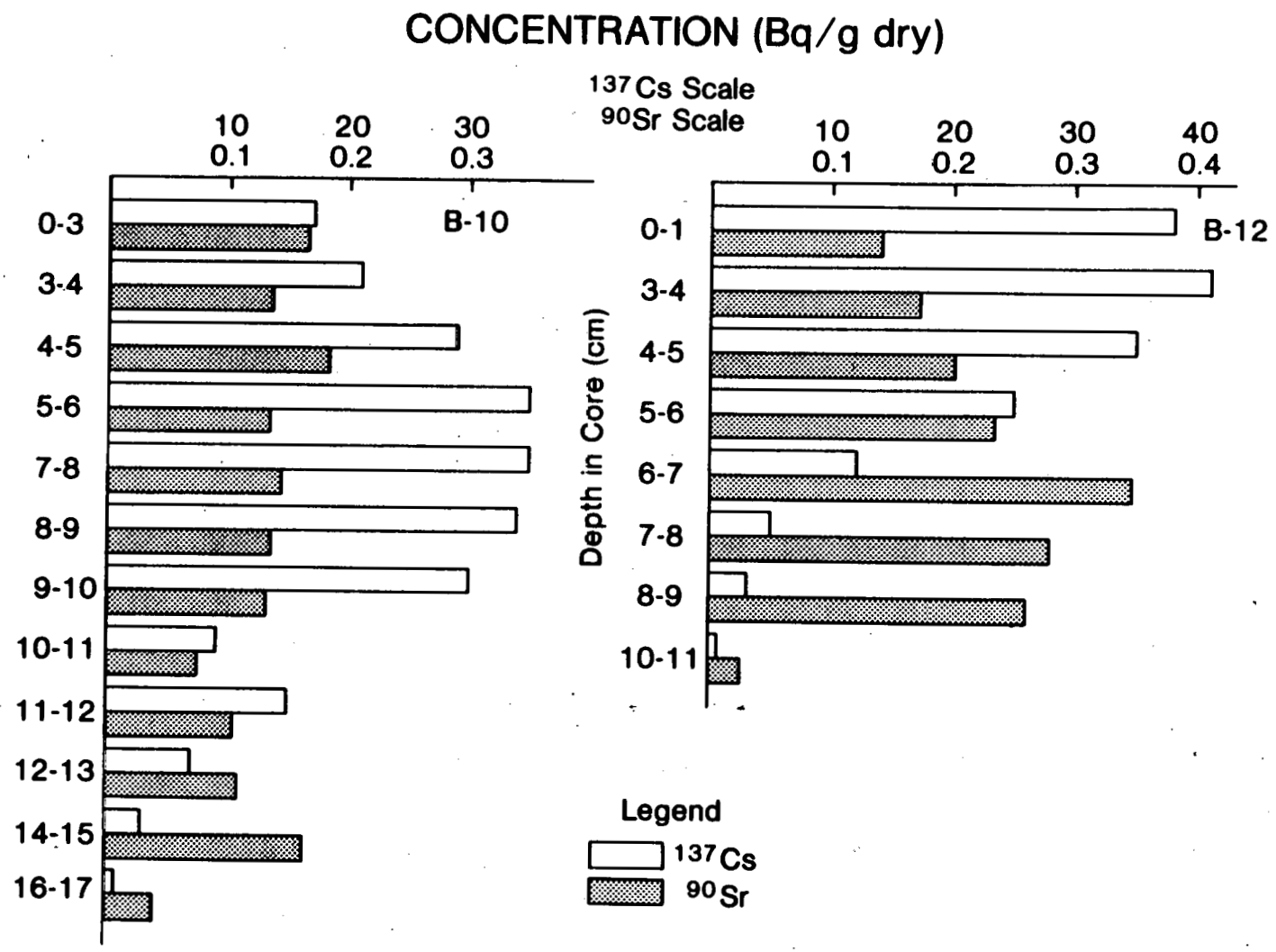

Fig. 5. Comparative depth profiles of ^{90}Sr and ^{137}Cs concentrations in sediment cores B-10 and B-12, Pond B.

depths of 2 to 4 m (4.20 kBq/m^2) and in water > 6 m (7.30 kBq/m^2). In the case of ^{90}Sr, we are less certain than for the other radionuclides that the cores were sufficiently long to account for essentially all the inventory.

Transuranics. Sediment cores B-10 and B-12 were also assayed for ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm. The results reveal well-defined peaks in the two profiles for each transuranic, but there was a difference in the depth of peak concentrations between the two cores (Fig. 6). All four radionuclides exhibited peak concentrations at 8-9 cm in core B-10 and at 2-3 cm in core B-12. These elements are clearly behaving very similarly to each other in sediments and as a group, the transuranic profiles are very similar to those of ^{137}Cs (Fig. 5).

Regressions of $^{239,240}Pu$ and ^{241}Am on ^{137}Cs both yielded correlation coefficients of 0.98 (n = 20), indicating that concentrations of these transuranics could be well-predicted in sediment cores from the much more easily measured ^{137}Cs concentrations. The correlation coefficients of ^{238}Pu and ^{244}Cm on ^{137}Cs were not as strong (r = 0.94 and 0.64, respectively). The slightly weaker correlation of ^{238}Pu to ^{137}Cs, compared to both $^{239,240}Pu$ and ^{241}Am, may be the result of the low concentrations of ^{238}Pu and the greater error associated with its measurement. However, the lower correlation coefficient observed for ^{244}Cm and ^{137}Cs relative to ^{241}Am and ^{137}Cs may be indicative of differences in these chemically similar actinide elements. While it is expected that these elements should behave similarly in aquatic systems, differences in their relative behavior have been noted before (Alberts et al., 1986a; Alberts et al., 1987).

To examine the relative mobilities of these radionuclides through the sediment profile, we examined activity ratios versus depth in the sediment core. The mean ratios of ^{238}Pu, ^{241}Am, and ^{244}Cm to $^{239,240}Pu$ were 0.096, 0.52, and 0.21, respectively, which are similar to previously reported ratios for these elements in Par Pond and Pond

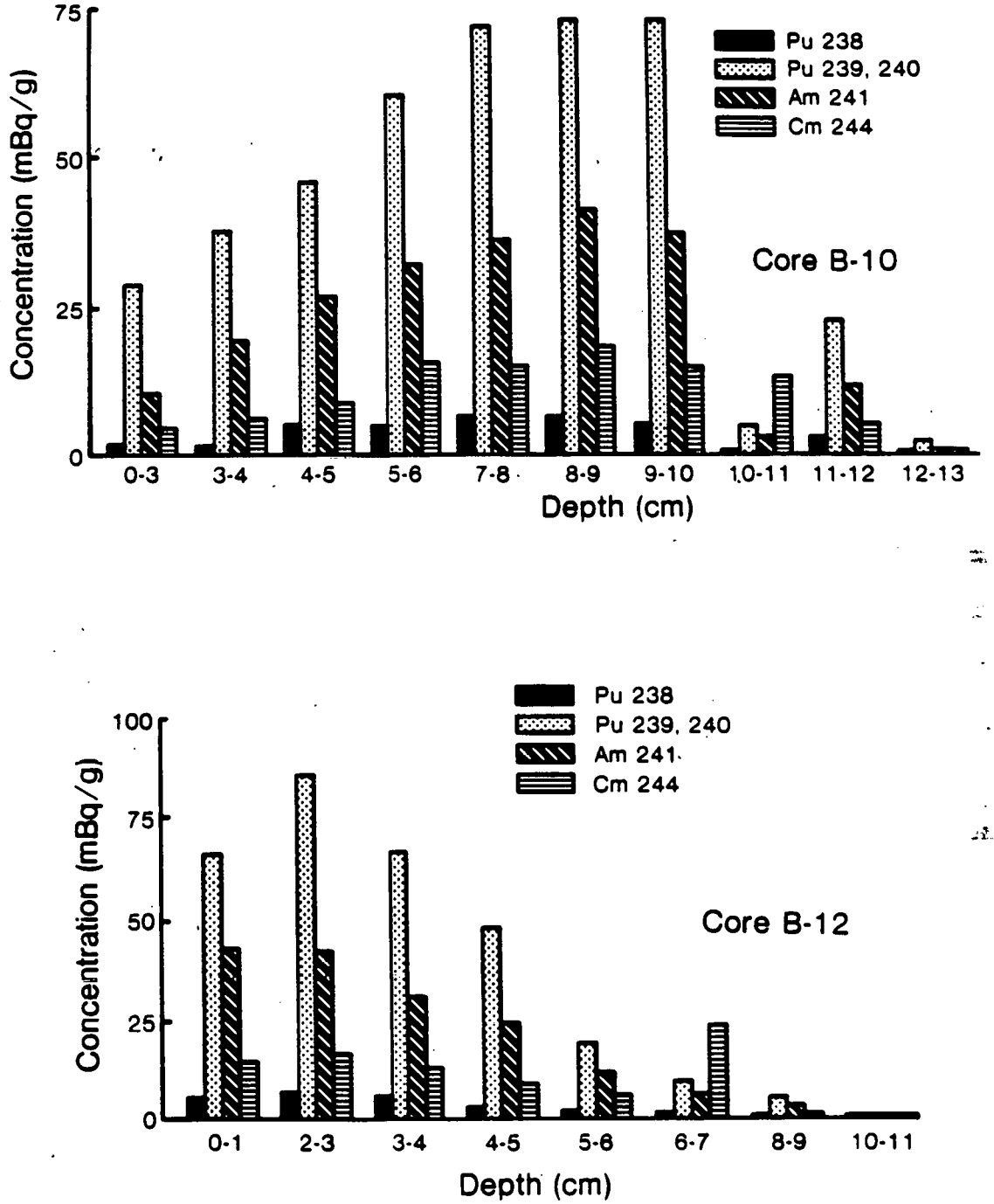

Fig. 6. Comparative depth profiles of ^{238}Pu, $^{239-240}Pu$, ^{241}Am and ^{244}Cm concentrations in sediment cores B-10 and B-12, Pond B.

B sediments (Alberts et al., 1986a). We found no clear trend in these ratios nor in the $^{137}Cs/^{239,240}Pu$ ratio (mean = 562) versus depth in sediment.

The similar behavior of these transuranics in sediments, and their resemblance to ^{137}Cs, was not unexpected from previous published works. For example, K_d values for Pu and Am are commonly in the 10^4 to 10^5 range (Watters et al., 1980) and Cm has been found to exhibit similar K_ds (Sibley and Alberts, 1984; Sibley et al., 1986). This range of K_d values has also been published for Cs (Clanton et al., 1964; Soratheson et al., 1960). The present work for Pond B yields K_d estimates of $1.7x10^5$, $6.9x10^6$, $1.1x10^6$ and $1.5x10^4$ for ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm, respectively. These values are based on depth-weighted mean concentrations in filtered water, and mean concentrations in the 0-3 cm sediment horizon. Even though these K_ds vary by up to two orders of magnitude between radionuclides, all values are so high that the mobilities of these nuclides would be expected to be very low.

Thirty-nine sediment cores were assayed in total to estimate sediment inventories. The depth-stratified means were 38, 492, 276, and 126 Bq/m^2 for ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm. Standard errors were about 15% of the means. The relationship of deposition to water depth observed for ^{137}Cs (Fig. 4) was also noted for the transuranics.

Aquatic macrophytes

Cesium-137. Some 261 samples of aquatic macrophytes, representing 15 taxa, were assayed for ^{137}Cs. For comparison, leaves from a few deciduous shoreline trees were also analyzed. The results (Fig. 7) reveal mean concentrations ranging from 4 to 30 Bq/g dry vegetation for the aquatic species. Individual samples ranged from 1.7 to 49 Bq/g dry. Values ranging from 2.0 to 2.4 Bq/g dry for the individual deciduous tree samples were measured. The mean for all individual samples assayed was 18 Bq/g dry.

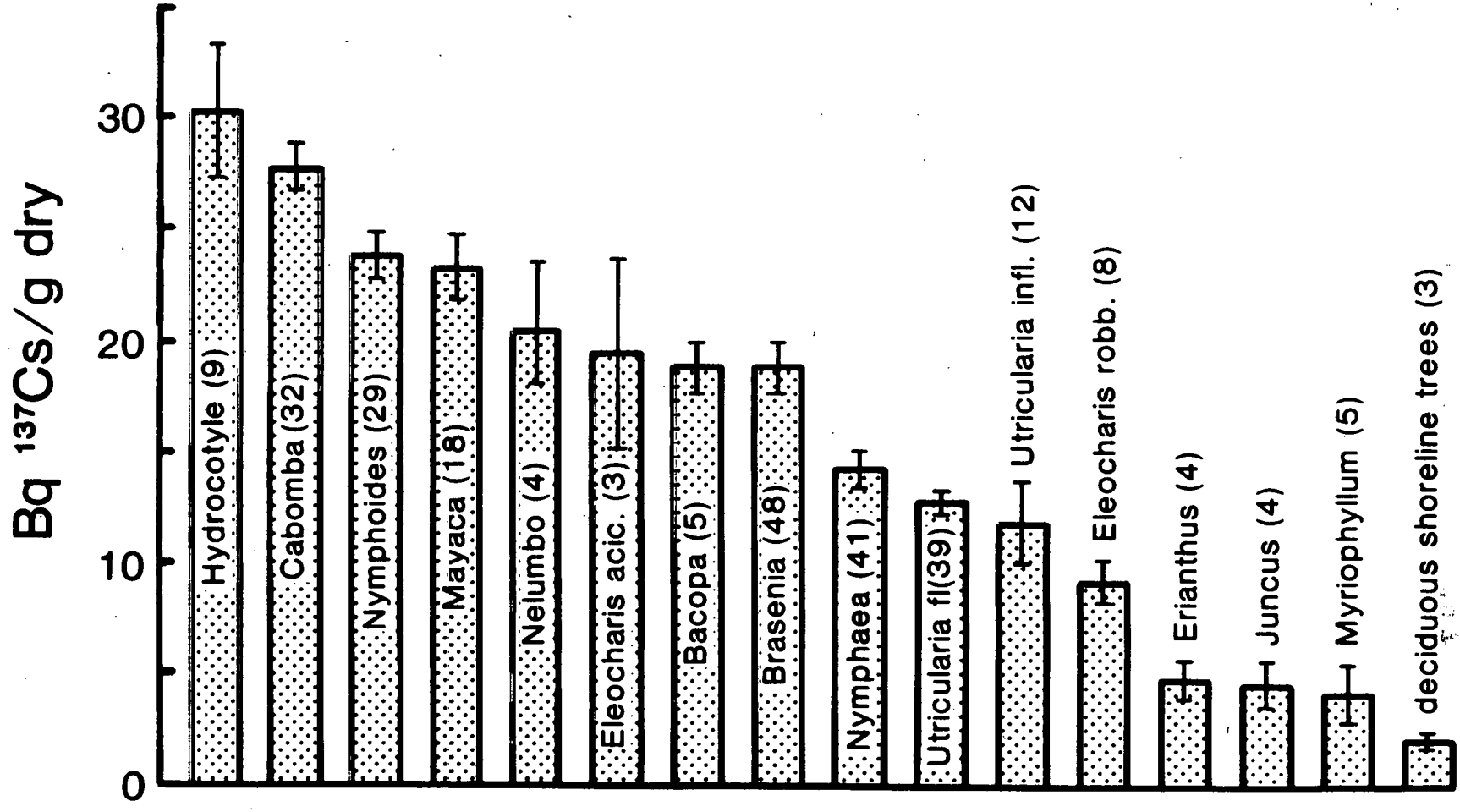

Fig. 7. Mean (±1 sem) concentrations of ^{137}Cs in aquatic macrophytes, Pond B. Sample sizes are indicated (n).

There were clearly species differences in mean ^{137}Cs concentrations, but the reasons for the differences were not specifically investigated nor revealed by analysis of the data. The rankings of five common genera (Hydrocotyle, Eleocharis, Bacopa, Brasenia and Nymphaea) were similar for Pond B and 1978 Par Pond data taken by coauthor J. J. Alberts, although the Par Pond values were all <10% of the respective Pond B values. Mean K contents of the vegetation samples ranged from 3 mg/g dry for Bacopa caroliniana to 19 mg/g dry for Eleocharis acicularis, but the mean ^{137}Cs concentrations were not significantly correlated with the mean K values ($r = 0.44$, $n = 11$). Large coefficients of variation for K values (based on 3-5 samples/taxa), ranging from 15 to 94% and averaging 57%, suggest little internal regulation of K and possibly large effects of available K in the local substrates. A comparison of plants with floating leaves to submerged species also indicated no statistically significant differences.

When ^{137}Cs concentrations of all 261 individual samples of aquatic macrophytes were regressed on the water depth from which each sample was taken, a positive, significant ($p < 0.01$) slope was found. When the same regression was performed for each species, only Nymphaea odorata and Brasenia schreberi exhibited significant effects of sampling depth (Fig. 8). These species occurred over a rather wide range of depths, unlike many other taxa, and a relatively large sample size was available in each case. A regression of mean ^{137}Cs concentration in a taxa on the mean water depth where the taxa was taken indicated a tendency for increase with depth, but the data were too variable to yield a statistically significant regression coefficient.

One possible explanation for the effect of sampling depth is that sediment concentrations tend to increase over the same depth range (Fig. 4), and sediments may be a major source of ^{137}Cs through root uptake. Linear regression of data from all homogenized, depth-integrated sediment cores that were taken in < 3.0 m of water, indicated that the ^{137}Cs content increased by 39 Bq/cm^2 per m of sampling depth

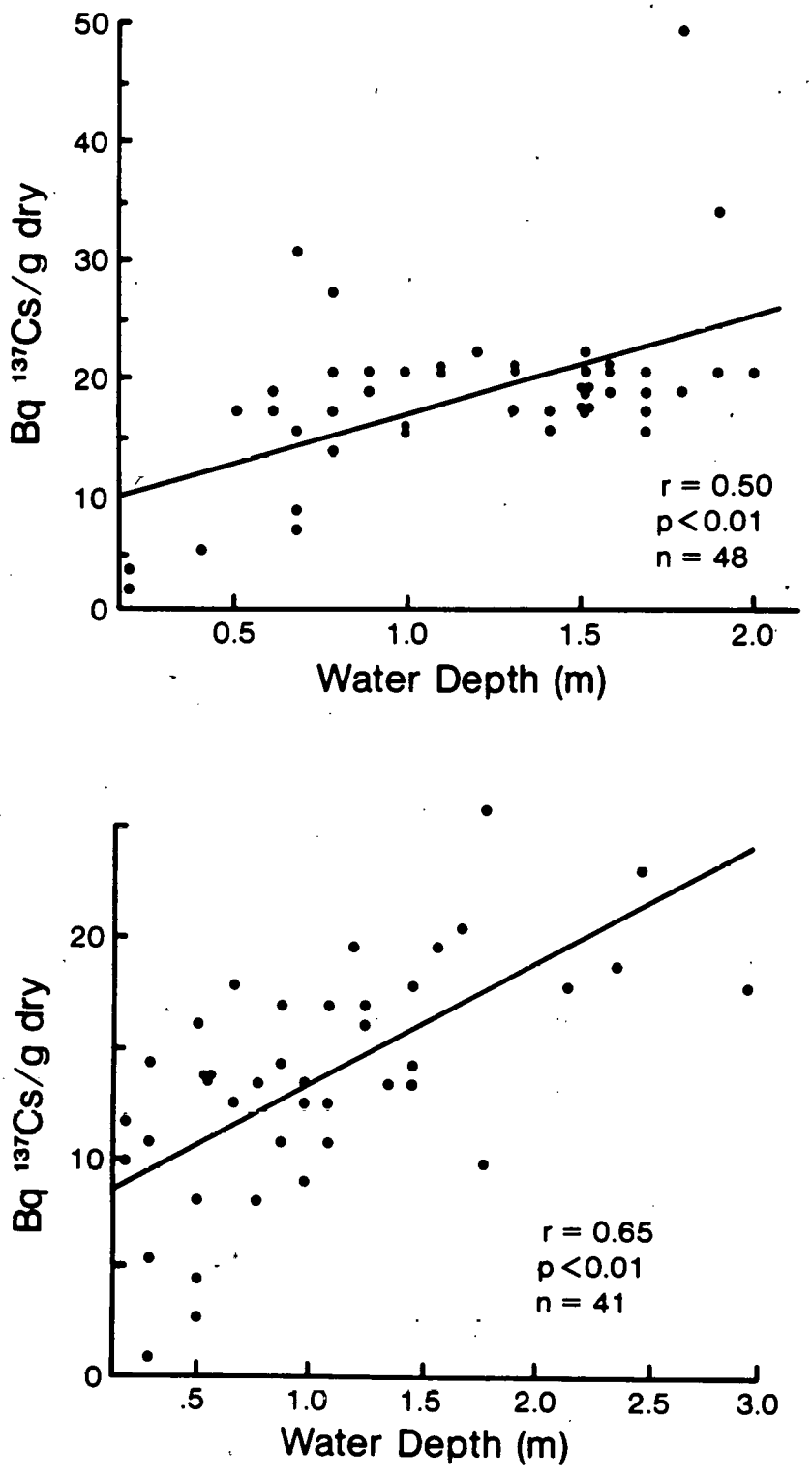

Fig. 8. Concentrations of 137Cs in Brasenia (upper plot) and Nymphaea (lower plot) versus water depth in which each sample was collected.

($r=0.84$; $p < 0.01$). A preliminary transplant study in which Myriophyllum spicatum plants were placed in Pond B and Par Pond sediments, with both systems then being placed in Par Pond, indicated substantial root uptake from the more radioactive Pond B sediments (Evans and Cocke, 1979).

An alternative explanation for the water depth effect in Nymphaea and Brasenia is provided by recent data obtained on Pond B macrophytes by Kelly (1988). These data indicate that root uptake of ^{137}Cs is not a major mechanism of ^{137}Cs accumulation in Nymphaea and Brasenia and that the petioles of these species contain 2 to 3 times the ^{137}Cs concentrations as do the leaves. The deeper the water, the greater is the ratio of petiole to leaf biomass (Wetzel, 1983). In the present study, leaves were not separated from petioles prior to analysis. Therefore, the effect of water depth may simply reflect a greater proportion of petioles relative to leaves in the samples from deeper water. This idea is strengthened by the results for 32 samples of Cabomba caroliniana, which were taken over a range of depths from 0.5 to 4.1 m, but which exhibited no relation of ^{137}Cs concentration to depth. Cabomba is not a floating leaf plant as are Nymphaea and Brasenia. The authors currently favor this alternative explanation over the first.

Strontium-90. Because of the expense of the radiochemical determination of ^{90}Sr, only 53 samples representing 11 species of aquatic macrophytes were analyzed. Mean values for species ranged from 0.2 Bq/g dry for Juncus sp. to 2.3 Bq/g dry for Hydrocotyle umbellata (Fig. 9). The mean of the 11 species means was 0.9 Bq/g dry, which is a factor of 20 lower than the overall mean ^{137}Cs value for the same samples. Hydrocotyle stands out clearly as being a significantly higher accumulator for ^{90}Sr than all other species, while Nymphaea odorata and Juncus were clearly lower than most species.

The reasons for the relative species rankings in ^{90}Sr content remain unclear after several correlation analyses. No significant relationship was found between species mean ^{137}Cs concentrations and species mean ^{90}Sr levels, although Hydrocotyle was the

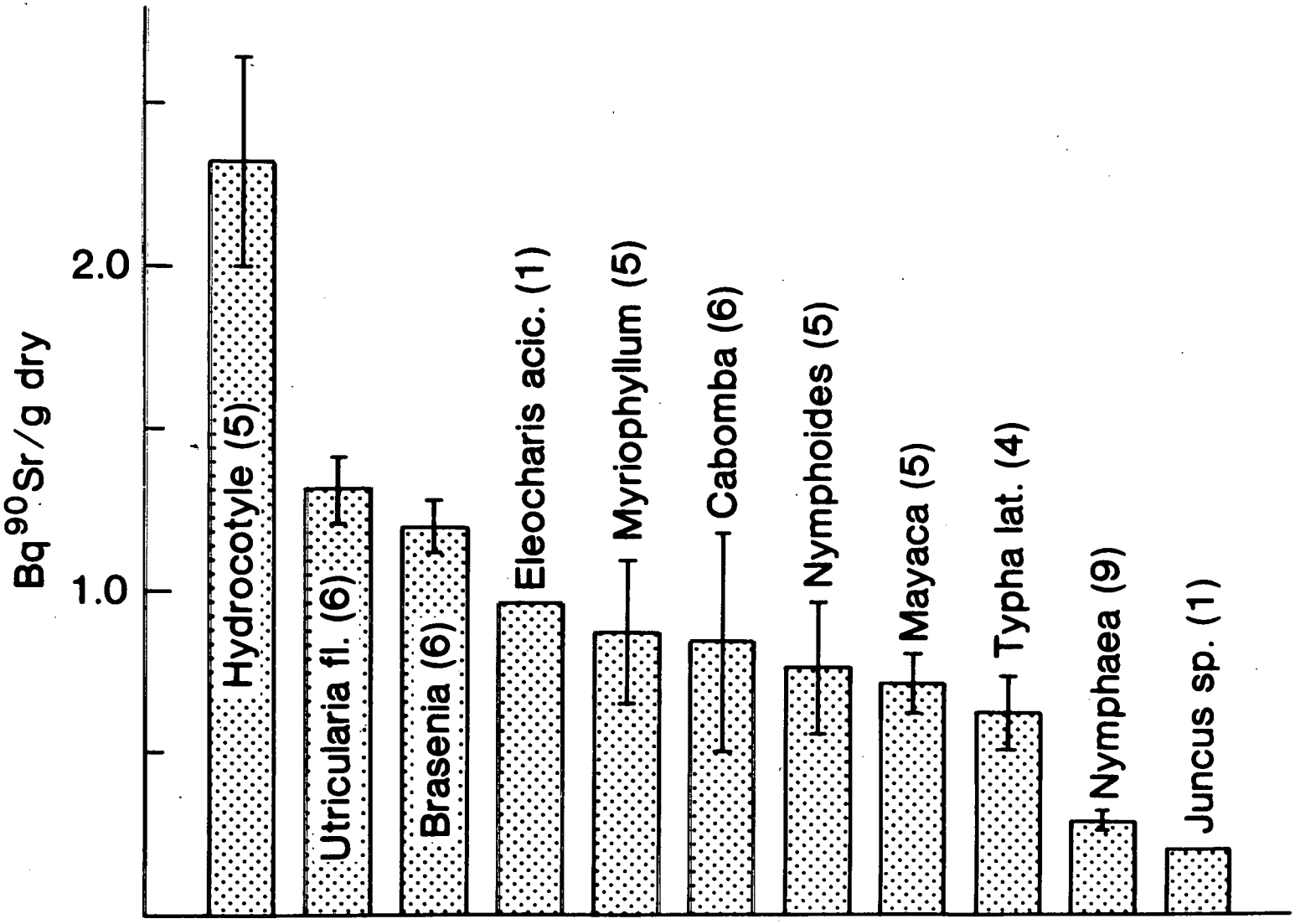

Fig. 9. Mean (±1 sem) concentrations of ^{90}Sr in aquatic macrophytes, Pond B. Sample sizes indicated (n).

highest accumulator for both radionuclides. A regression of species mean ^{90}Sr values on species mean calcium levels also failed to indicate a significant correlation. Again, however, Hydrocotyle stood out as the highest accumulator of Ca, which would be expected to also result in high accumulation of Sr, which is chemically analogous. Calcium concentrations in nine plant species ranged from 1.5 mg/g dry for Eleocharis acicularis to 19 mg/g for Hydrocotyle. As was true for K, calcium concentrations within species were quite variable (coefficients of variation averaged 42% and ranged up to 83%), possibly indicating variable sources and little physiological regulation. Other possible sources of variation included leaf age and proportions of leaves and stems sampled. Floating leaved plants did not differ significantly from submerged species, and sampling locations did not appear to affect ^{90}Sr levels.

The sampling depth relationships observed for ^{137}Cs in macrophytes were not observed for ^{90}Sr. When species mean ^{90}Sr concentrations were regressed on the mean sampling depths for each corresponding set of samples, no significant relationship was found. Furthermore, when individual sample contents within a species for ^{90}Sr were regressed on the corresponding sampling depths, no correlation was revealed.

Transuranics. Isotopes of plutonium were measured in 5 species of aquatic macrophytes, with composite sample sizes ranging from 10-21 per species. The values generally fell within the range of 0.05-0.4 mBq/g dry for ^{238}Pu and 0.05-1.1 mBq/g dry for $^{239,240}Pu$ (Fig. 10). The general species rankings were very similar for the two isotopes. The highest mean concentrations occurred in Utricularia floridana, a highly branched plant with very small diameter stems and leaves, while the lowest concentrations were observed in Nymphaea odorata, a simple floating leaved plant with very large, fleshy parts. The data strongly suggest that plutonium accumulation is enhanced by a high surface area to mass ratio. This would be expected for such insoluble contaminants which accumulate primarily by surface adsorption as opposed to root uptake or direct absorption (Watters et al., 1980).

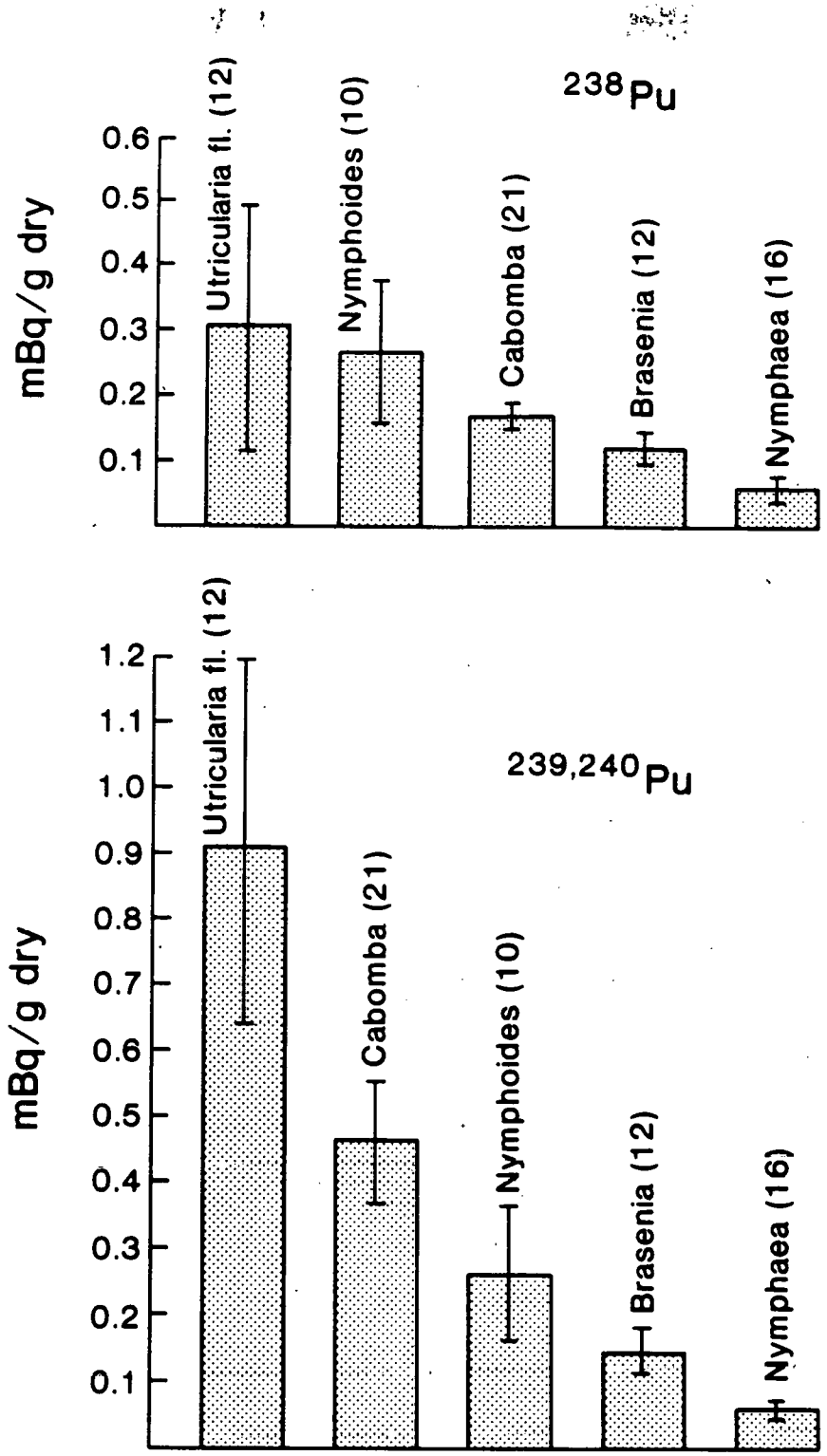

Fig. 10. Mean (±1 sem) concentrations of ^{238}Pu and $^{239,240}Pu$ in five species of aquatic macrophytes, Pond B. Sample sizes are indicated (n).

A few measurements of ^{241}Am and ^{244}Cm were also performed for 4 species of aquatic macrophytes (n = 2 per species). Concentrations of ^{241}Am ranged from 0.03 mBq/g dry in Nymphaea to 1.7 mBq/g dry in Utricularia floridana. For ^{244}Cm, values ranged from 0.05 mBq/g dry in Brasenia to 0.5 mBq/g dry in Utricularia. The species rankings for these transuranics were generally similar to the rankings for plutonium. Again, surface adsorption processes appear important for ^{241}Am and ^{244}Cm.

Regressions between the concentration of the plutonium isotopes and sampling depth and ^{137}Cs concentrations failed to reveal any significant trends. In general, the relative variability among samples for the transuranics was much higher than for ^{137}Cs or ^{90}Sr (Figs. 7, 9, 10). The reasons for this variability are not clear, but most studies on transuranics in the environment have also observed large sampling variations (e.g. Little et al., 1980).

The ratios of ^{238}Pu to $^{239,240}Pu$ in various media can yield insights into the major sources of these materials (Alberts et al., 1986a). For example, the ratio of ^{238}Pu to $^{239,240}Pu$ in Pond B sediments was roughly 0.08. In contrast, the ratio of these isotopes in atmospheric deposition at the distance of Pond B due to atmospheric releases from the Savannah River Site nuclear fuel facilities is approximately 1.0 (Corey et al., 1982). The corresponding isotopic ratios for the floating-leaved plants were 0.85, 1.03 and 1.02 for Brasenia, Nymphaea, and Nymphoides, respectively, providing strong evidence that atmospheric deposition is the primary source of plutonium for these species. In contrast, ratios for submerged species were 0.33 and 0.38 for Utricularia and Cabomba, respectively, suggesting important contributions from the sediments (72 and 67%, respectively) in addition to atmospheric deposition. Resuspension of small, plutonium-bearing sediment particles, followed by adsorption or entrainment of these particles to plant surfaces, is thus probably a dominant accumulation mechanism for submerged macrophytes.

Benthic macroinvertebrates

Pooled, dried samples representing several abundant taxa in Pond B were assayed for radionuclides. Only the aquatic nymphal or larval stages of the insects sampled (orders Odonata, Diptera and Ephemeroptera) were represented. Data were limited primarily by the lack of adequate biomass to provide sufficient radioactivity for reliable assay. The data considered reliable are shown in Table 1, but they were not summarized as means, etc. because of missing data for various taxa and small sample numbers. Concentrations of ^{137}Cs and ^{90}Sr were 1 to 3 orders of magnitude higher than the transuranics, as expected. Concentrations of ^{137}Cs were higher than ^{90}Sr concentrations in the same taxa, except for snails which exhibited higher ^{90}Sr concentrations. The calcareous shells of snails undoubtedly accounted for the elevated concentrations of ^{90}Sr. Samples of damselflies, midges and snails were submitted for assay of transuranic radionuclides. Detectable concentrations of ^{241}Am and ^{244}Cm were found in all samples assayed; ^{239}Pu was measurable in damselflies and midges; and ^{238}Pu was only measurable in midges.

The concentrations of ^{137}Cs measured in macroinvertebrates ranged from 1 to 14 Bq/g dry, whereas macrophyte and surface sediment samples contained roughly 7-30 and 25-35 Bq/g dry, respectively. Thus, no apparent increases with trophic level were evident. The same was true in the case of ^{90}Sr, where insects contained 40-120 mBq/g dry while macrophytes and surface sediments contained 500-1,500 and 200-400 mBq/g dry, respectively. Snails however, contained ^{90}Sr concentrations that were an order of magnitude higher than concentrations in sediments and macrophytes. The invertebrates contained the four transuranics in the range of 1-19 mBq/g dry, compared to roughly 0.05-1.5 mBq/g dry in macrophytes and 3-40 mBq/g dry in surface sediments. The benthic habitat of the invertebrates would encourage detritus ingestion, but we do not know how much sediment may have been in the gastrointestinal tracts or on the surfaces of the animals assayed. However, the particularly strong sorption of $^{239,240}Pu$ to

Table 1. Concentrations of radionuclides in benthic macroinvertebrates collected from Pond B in January 1984.

Classification	Sample No.	Concentrations (mBq/g dry)					
		^{137}Cs	^{90}Sr	^{238}Pu	^{239}Pu	^{241}Am	^{244}Cm
Order Odonata			45[a]				
Dragonflies (Anisoptera)	1	7,600	87				
	2	1,200					
Damselflies (Zygoptera)		3,400	42		2.5	2.2	19.0
Order Diptera							
Midges (Chironomidae)	1		78				
	2	5,500	120	2.9	0.9	8.9	5.9
Biting Midges (Ceratopogonidae)		14,000					
Horsefly (Chrysops)		4,900					
Order Ephemeroptera (Mayfly)			68				
Class Gastropoda (Snails)							
Helisoma	1		7,900			1.8	0.9
	2	930					

[a] A combined sample of dragonflies and damselflies.

sediments, along with the normal biological exclusion of this nuclide, indicates its usefulness as a sediment tracer. Ratios of ^{137}Cs, ^{90}Sr, ^{241}Am and ^{244}Cm to $^{239,240}Pu$ for the benthic insects were 4, 51, 6 and 28 times higher, respectively, than the same ratios for sediments, indicating important accumulation mechanisms other than sediment associations for these radionuclides. The nymphal stages of the dragonflies and damselflies are, like their adult counterparts, carnivorous (Pennak, 1953). The Dyptera larvae analyzed have varied food habits and the snails likely feed mainly on algae and organic detritus (Pennak, 1953).

Nine individual damselfly nymphs were analyzed for stable K and Ca and mean values of 14 and 2.1 mg/g dry were found, respectively. The same analyses for 6 dragonfly nymphs yielded means of 10 and 2.8 mg/g dry for K and Ca, respectively. These values were not particularly useful for interpretation of the ^{137}Cs and ^{90}Sr concentrations.

Fish

Cesium-137. The mean (±1 sem) ^{137}Cs concentrations found in muscle tissue from seven Pond B fish taxa are illustrated in Fig. 11. The greatest confidence can be placed in the data for largemouth bass, bullheads, and gizzard shad because of the sample sizes of 28, 28, and 10, respectively. Largemouth bass were easily obtained by angling, while gill netting proved very effective for bullheads and gizzard shad. Taxon means ranged from 2.1 Bq/g wet muscle for small baitfish (primarily bluegills and brook silversides) to 8.0 Bq/g wet muscle for gizzard shad. A nearly monotonic increase in the mean ^{137}Cs concentration with mean mass per individual within a taxon is evident (Fig. 11). A linear regression of mean ^{137}Cs concentration in Bq/g (Y) on mean fish mass of the taxon in g (X) yielded the predictive equation $Y = 0.011X + 2.1$ with $r = 0.93$ ($n = 7$). When the ^{137}Cs concentrations of all individual fish from all taxa were regressed on fish mass ($n = 82$), a correlation coefficient of 0.31 was obtained ($p < 0.01$). Within species

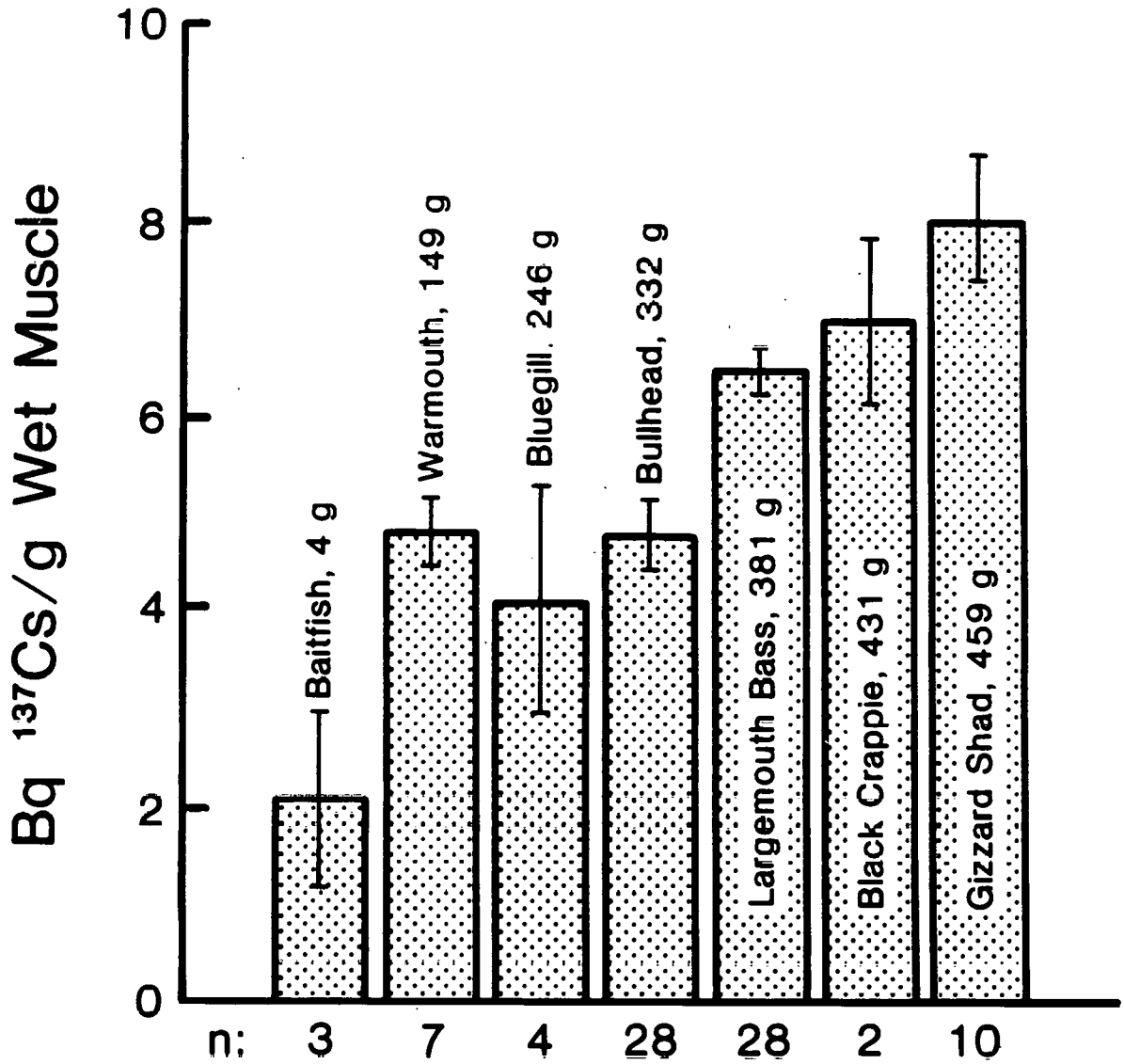

Fig. 11. Mean (±1 sem) concentrations of ^{137}Cs in fish muscle, Pond B. Sample sizes and mean fresh weights of whole animals are indicated.

regressions of ^{137}Cs concentration versus mass were run for largemouth bass, bullhead, gizzard shad and warmouth. These failed to produce statistically significant correlations, even though all the slopes were positive and the regression coefficients approached significance at $p = 0.05$.

The striking tendency for ^{137}Cs concentrations to increase with mean size, especially when several different species are included, may be related to the trophic level increase previously observed for ^{137}Cs. The tendency for predators to contain 2-3 times the concentration of ^{137}Cs in their prey was noted by Pendleton et al. (1965) for both aquatic and terrestrial foodchains. Whicker et al. (1972) found a similar increase from small to large predatory trout in high mountain lakes. It is quite likely that the small baitfish we sampled were consumed by warmouth, bluegill, largemouth bass and black crappie. These species all contained 2-3 times the ^{137}Cs concentrations as the small baitfish. It may also be reasonable to presume that the propensity of the predatory fish to consume other fish increases with size, with invertebrates being relatively more important in the diets of the smaller individuals of the predatory species. Common insect larvae from Pond B generally contained less than 1 Bq/g wet tissue, compared to ^{137}Cs concentrations generally greater than 2 Bq/g for baitfish in the easily preyable range of 1-50 g.

A concerted effort was made in July 1986 to study the effect of size on ^{137}Cs concentrations in largemouth bass by obtaining a much larger range of sizes for inclusion in the sample. In addition to four bass that were in the same size range as those sampled in 1983/84 and discussed above, four much larger individuals, ranging from 1,700 to 3,400 g, were obtained and assayed for ^{137}Cs. In this case, a regression of ^{137}Cs concentrations in muscle versus mass yielded a slope not significantly different from zero ($n = 8$). This result was unexpected because the larger bass certainly must have been capable of feeding on much larger prey than the smaller bass. One difference between the 1986 and the 1983/84 sample was season (summer vs. winter, respectively).

The stomachs of the large bass were essentially empty so we have no information on their actual diet. The mean (±1 sem) for the 1986 sample was 7.0±0.3 Bq/g wet compared to 6.5±0.3 Bq/g wet for the 1983/84 sample, indicating no significant temporal trend.

The highest species mean concentration of ^{137}Cs (8.0 Bq/g wet) was found in gizzard shad muscle (Fig. 11). Gizzard shad are filter feeders that ingest zooplankton, phytoplankton, small insect larvae and suspended detritus (Pierce and Wissing, 1981; Drenner et al., 1982). Cursory examination of the gut contents of gizzard shad from Pond B revealed an abundance of ostracod carapaces, macrophyte fragments and smaller quantities of copepods, desmid algae and sand grains. Using a predictive equation in Drenner et al. (1982), a shad of 35 cm length (the mean length of shad in our sample) can filter 2.7 L/min, or about 1,900 L/day assuming the fish filters at this rate for 12 hours/day. Using our value of 0.04 Bq ^{137}Cs filtered from a liter of Pond B water and an estimated fish filtering efficiency of 0.10, a gizzard shad might ingest on the order of 8 Bq/day, or about 0.4% of the estimated body burden daily. This intake corresponds with, assuming equilibrium conditions and near-complete assimilation of ingested ^{137}Cs, a biological retention half-time of about 170 days in these fish, which is of the same order as other estimates measured experimentally in other species (Hakonson et al., 1975; Vanderploeg et al., 1976; Kolehmainen, 1974). Thus, while we cannot directly explain the relatively high ^{137}Cs concentrations measured in gizzard shad, the values do not seem unreasonable when one considers the large volumes of water they may filter and the estimated ^{137}Cs content of the filtered material.

The ^{137}Cs values presented on a wet muscle mass basis may be converted to a dry mass basis for comparison to other data from the mean (±1 sem) dry to wet mass ratio of 0.22±0.003 (n = 33 samples of primarily largemouth bass and bullhead tissues). Eight muscle tissue samples (two each from largemouth bass, bullhead, bluegill and gizzard

shad) submitted for K analysis had a mean (±1 sem) content of 210±12 mg K/g dry muscle.

Strontium-90. Mean concentrations of ^{90}Sr in fish bone ash ranged from 12 Bq/g in gizzard shad to 23 Bq/g in black crappie (Fig. 12). The elevated concentrations in black crappie relative to the other species may be spurious as the sample size was only 2. The mean across all species was 15 Bq/g with a standard error of 2 Bq/g. Warmouth, bluegill, bullhead and largemouth bass means were all within one standard error of the mean across all species measured. The data may be easily converted from an ash weight to dry weight basis by multiplying by 0.60, the mean bone ash/dry mass ratio which had a standard error of 0.01 (n = 43). Calcium contents of 15 fish bone ash samples averaged 332 mg/g with a standard error of only 4 mg/g. There were no apparent species differences in the Ca content of bone.

The relatively low mean ^{90}Sr values for gizzard shad bone compared to the other species may relate to food habits. Gizzard shad do not consume other fishes which contain elevated concentrations of ^{90}Sr in skeleton and scales. Furthermore, only about 3% of the ^{90}Sr in the water columns is associated with particulate matter in contrast with nearly twice as much ^{137}Cs in particulates. Also, surface sediment material, which is likely to be similar to suspended particulates in terms of radionuclide concentration, was very low in ^{90}Sr (Fig. 5) and about two orders of magnitude lower than bone tissues (Fig. 12) that are consumed to some degree by other species. Thus, the intake of ^{90}Sr by gizzard shad would be expected to be lower than in the other species of fish.

The fish size effect across all species that was observed for ^{137}Cs concentrations in muscle, was not observed for ^{90}Sr in bone ash. However, again in contrast to ^{137}Cs, when ^{90}Sr in bone ash of largemouth bass was regressed against size of the individual, a statistically significant correlation was obtained ($r = 0.40$, $p < 0.05$, $n = 27$). The regression equation was $Y = 0.0053\ X + 12$ where Y was the ^{90}Sr concentration in

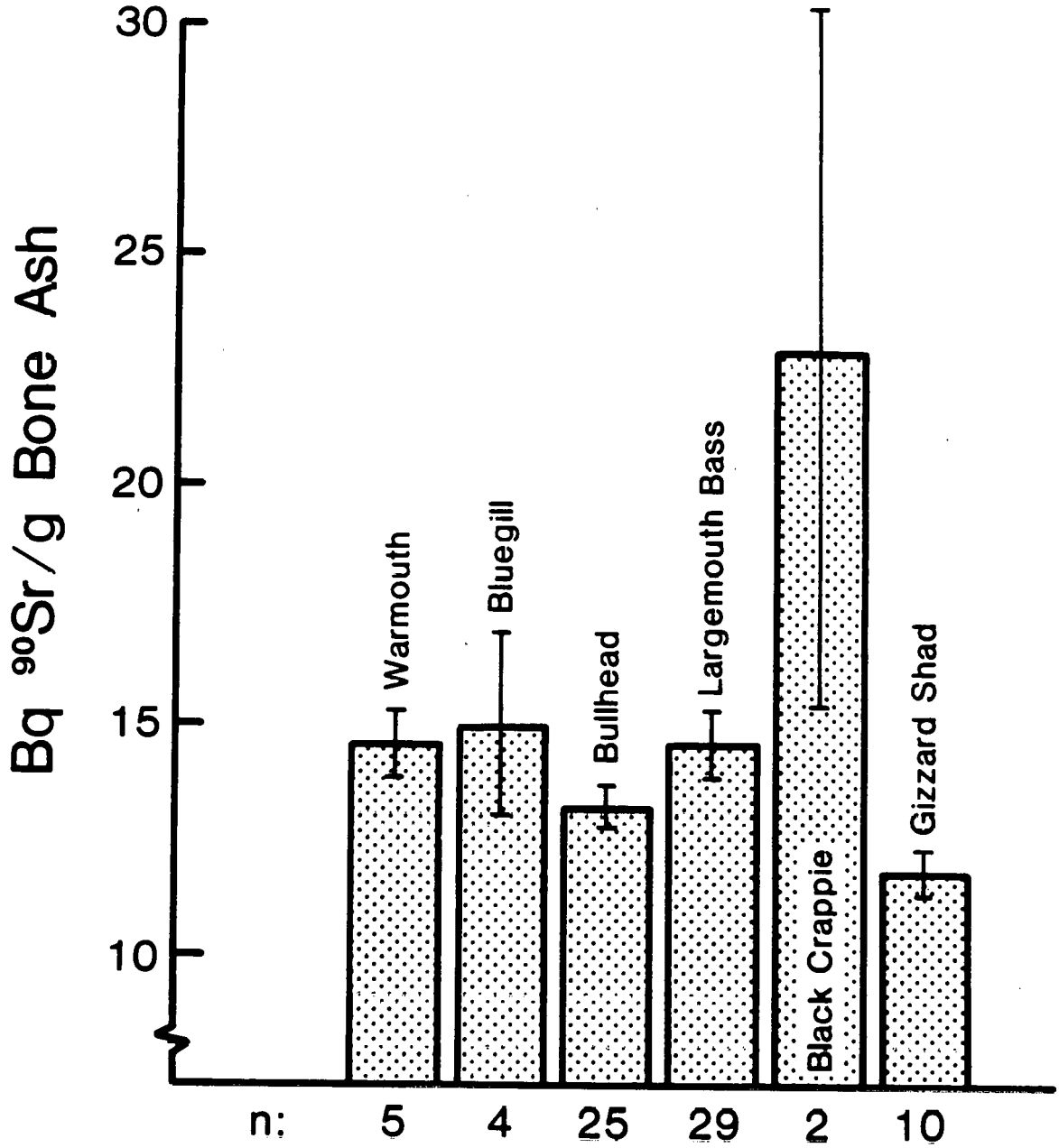

Fig. 12. Mean (±1 sem) concentrations of ^{90}Sr in fish bone ash, Pond B. Sample sizes are indicated for each species. Bars are ordered by mean fish mass.

largemouth bone ash (Bq/g) and X was the fresh whole body mass in g. Similar regressions were run for ^{90}Sr in bone ash versus body mass for other species, but none produced statistically significant ($p < 0.05$) slopes. One would generally expect ^{90}Sr concentrations in bone to increase with mass of the individual because of the predatory tendency of larger fish and because the long retention of the alkaline earths in bone would produce an age effect and size would likely correlate with age. Individual fish diets may vary, of course, which could produce sufficient scatter in the data to obscure a size effect.

Muscle tissue was expected to contain very low concentrations of ^{90}Sr, thus only two samples were submitted for assay. Values of 470 and 86 mBq/g dry muscle were obtained for largemouth bass and bullheads, respectively, confirming the expectation of low concentrations.

Transuranics. The effort to measure transuranic radionuclides in fish was limited by the expense of the analyses as well as the low concentrations in tissues. Muscle, liver and bone ash of largemouth bass and bullheads were assayed (Table 2). Only 17 of 72 separate analyses yielded a statistically detectable quantity of the transuranic sought. The greatest frequency of detection was for ^{238}Pu (6/18); the lowest was for $^{239,240}Pu$ (2/18). A transuranic was most likely to be found in bone ash (3/8 or 37%) and least likely to be found in liver (5/24 or 21%). Analyses of largemouth bass tissues were nearly twice as likely to yield a detectable transuranic concentration as bullhead tissues (11/36 vs. 6/36, respectively).

Concentrations of transuranics, when detectable, were very low, ranging from 0.004 mBq ^{238}Pu/g dry muscle to 11 mBq ^{244}Cm/g dry liver. The counting uncertainties on most samples were relatively large, particularly for small sample masses. Due to the large number of non-significant or zero values, it was not meaningful to compute means and conduct statistical comparisons. One would expect low concentrations of transuranics in fish tissues relative to other radionuclides,

Table 2. Occurrence and concentrations of transuranic radionuclides in largemouth bass and bullhead tissues.

Tissue	Statistic	^{238}Pu	$^{239,240}Pu$	^{241}Am	^{244}Cm
Largemouth Bass					
muscle	frequency[a]	2/5	1/5	2/5	2/5
	range[b]	0.004-0.04	0.25	0.04-0.25	0.2-1.1
liver	frequency	0/3	1/3	0/3	1/3
	range	---	2.7	---	11
bone ash	frequency	1/1	0/1	0/1	1/1
	range	0.1	---	---	1.5
Bullhead					
muscle	frequency	1/5	0/5	0/5	1/5
	range	0.2	---	---	0.29
liver	frequency	2/3	0/3	1/3	0/3
	range	0.56-0.59	---	1.2	---
bone ash	frequency	0/1	0/1	1/1	0/1
	range	---	---	0.16	---

[a] frequency = No. samples with detectable activity/No. samples assayed.

[b] range of values in mBq/g. If only one sample had detectable amounts, the "range" is a single value. Muscle and liver are expressed on a dry mass basis; bone is reported on an ash mass basis.

because of the low concentrations in water, sediments, macrophytes and invertebrates, and because of the generally low foodchain transfer factors of most transuranics (Bair and Thompson, 1974; Eyman and Trabalka, 1980).

Turtles

Cesium-137. Muscle tissues from 10 slider turtles exhibited a mean (±1 sem) ^{137}Cs concentration of 2.1±0.4 Bq/g wet mass. The turtles sampled ranged from 142 to 1,837 g total mass and represented a range of ages from 2 to 6 years, except for the 1837 g female which was too old to age. The considerable variability among individuals in muscle ^{137}Cs concentrations (the coefficient of variation was 0.6) may relate to differences in residence history, size and/or feeding habits of the turtles, the latter of which may be complex and site-specific (Parmenter, 1980). Clark and Gibbons (1969) indicated that slider turtles shifted their diets from animal matter to primarily aquatic vegetation by an age of 1-2 years. Whether or not this dietary shift occurs in Pond B is unknown. In any case, a regression of ^{137}Cs concentrations in turtle muscle on mass failed to indicate any relationship. This finding would argue in favor of variable food habits and/or residence history as possibly being the controlling variables that affect ^{137}Cs concentrations in turtles. Present data are not sufficient to objectively test for the possible effects of these factors.

Strontium-90. The mean (±1 sem) ^{90}Sr content of shell and bone ash samples from the same 10 slider turtles assayed for ^{137}Cs was 12±1 Bq/g. This value was slightly lower than the overall mean (±1 sem) ^{90}Sr concentration in fish bone ash (15±2 Bq/g), but essentially identical to the mean concentration in gizzard shad (12 Bq/g). Like the gizzard shad, slider turtles are not likely to consume fish bone or scales in great quantity, and most likely obtain most of their energy from aquatic vegetation and fish carrion when available (Clark and Gibbons, 1969).

The retention half time of ^{90}Sr in adult turtles is relatively long. Scott et al. (1986) estimated an annual mean retention half-time of 365 days, and more recent data collected by T. G. Hinton (Savannah River Ecology Laboratory, Drawer E, Aiken, South Carolina 29801) suggests a half-time of about 1,000 days. Most turtles were in the range of 2 to 6 years of age, thus an increase in ^{90}Sr content might be expected over this range of ages. When the total body burden of ^{90}Sr in the turtle was regressed on estimated age, a highly significant correlation was observed ($r = 0.97$, $n = 10$, $p < 0.01$). The regression equation was $Y = 0.17\ X - 0.34$, where Y was the total body burden in kBq and X was the estimated age in years. However, this result might simply have resulted from the fact that turtles become larger and accumulate more calcareous tissue with age. Indeed, a regression of ^{90}Sr concentration in whole turtles (Bq/g) on age yielded a slope that was not statistically different from zero. This result may not be inconsistent with a long ^{90}Sr retention half time in turtles. The retention in shell increments may be tenacious, yet new increments are accumulated each year and each might reflect intake during that year. Older shell increments may not accumulate ^{90}Sr readily, once formed. Calcareous tissues in turtles probably cannot be treated as a single compartment (as can soft tissues), and should not be considered conceptually as a tissue in steady state.

A single sample of turtle muscle was submitted for ^{90}Sr analysis. A value of 14 mBq/g dry was obtained, a comparatively low concentration as expected.

Transuranics. Few analyses of turtle tissues were performed for transuranics due to the low concentrations expected and the expense of the analyses. One sample of shell ash contained 0.1 and 0.2 mBq/g of ^{238}Pu and ^{244}Cm, respectively. No ^{239}Pu or ^{241}Am was found in detectable concentrations in shell ash. Five samples of dried muscle tissue were assayed for transuranics. Depending on the radionuclide, 3 or 4 of the five samples did not contain statistically detectable activity, but these data were still used in computing means. The mean concentrations for ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm were 0.05, 0.06, 0.13 and 0.07 mBq/g dry muscle, respectively. Relatively

high counting uncertainties for the transuranic analyses dictate caution in interpretation of these values.

Waterfowl

Since most waterfowl reside only temporarily on the reservoir, length of residence is an important additional variable that affects radionuclide burdens in tissues. However, even though their residence is only temporary, many individuals still have ample time to accumulate measurable concentrations of radioactivity. Departure of the birds causes a very small fraction of the radionuclide inventory to leave the ecosystem and produces the risk that the bird might be shot and consumed by people while its tissues are still contaminated. Therefore, estimates of radionuclide concentrations in waterfowl are particularly useful and relevant.

Analyses were performed on coots because they are very common and they tend to accumulate higher ^{137}Cs concentrations than other waterfowl (Brisbin et al., 1973). The coot is primarily a vegetarian, selecting a variety of macrophytes and algae (Bent, 1963). The mean (±1 sem) ^{137}Cs content of coot muscle, based on analyses of tissues from seven individuals, was 3.1 (±1.1) Bq/g wet tissue, or 15 Bq/g dry tissue. This value approaches the overall mean ^{137}Cs content in fish muscle of about 5-6 Bq/g wet and exceeds the mean value for turtle muscle (2 Bq/g wet). These birds were shot in mid-February 1984 and thus it is possible, but not certain, that they had resided on Pond B for up to several months (Brisbin and Vargo, 1982). A biological half-time of ^{137}Cs in coots was estimated by Potter (1987) to be on the order of 7 days. Thus, a residence time of only 30 days, for example, would, according to a simple exponential model, lead to a fraction of the equilibrium burden of 1-e-((ln2)(30 days)/7days) or 0.95. A bird leaving Pond B with a muscle concentration of 3.1 Bq/g would, after 30 days, contain only 3.1 e-((ln 2)(30 days)/7days) or 0.16 Bq/g, a level well under the safety guidelines

for human consumption, assuming a reasonable consumption rate of 200-300 g/day (International Commission on Radiological Protection 1978).

Only one sample of coot muscle and one of bone was assayed for ^{90}Sr. Values of 14 and 420 mBq/g were obtained for dry muscle and bone ash, respectively. These values are roughly an order of magnitude lower than the respective values in fish. The generally long retention time of ^{90}Sr in tissues, especially bone, and the comparatively short residence time of the coots, could easily explain these relatively low values.

One sample of coot bone and five samples of muscle were submitted for transuranic analysis. Only ^{238}Pu was detected in the bone sample with a reported concentration of 0.1 mBq/g ash. Three or four of the five muscle samples failed to produce a detectable concentration, depending on the transuranic assayed, however, all data were used in constructing means. Mean values for coot muscle were 0.013, 0.008, 0.015 and 0.071 mBq/g dry for ^{238}Pu, $^{239,240}Pu$, ^{241}Am and ^{244}Cm, respectively. Rather large relative counting uncertainties applied to all the transuranic values, thus we urge restraint in their interpretation. Clearly, however, the values are extremely low and of negligible biological consequence.

Concentration ratios

Actual values for radionuclide concentrations in various kinds of environmental materials, are of interest primarily from the standpoint of radiation dose assessment for a specific location and circumstance. Such values are difficult to compare across ecosystems however, because differences may simply reflect source term variations and not differences in ecological or biological processes. The concentration ratio (CR), usually defined as the ratio of the concentration of a substance in a material of interest to that in water, has been used extensively to compare organisms, ecosystems, and radionuclides. Since concentrations are "normalized" to water, the CR should avoid differences due to source term. The CR concept, however, has numerous limitations

and drawbacks (Blaylock, 1982; Whicker, 1984). For example, it is often reported differently (e.g., filtered vs. unfiltered water; wet vs. dry mass of the material), the water and/or material may not be in steady state at the time of measurement, and water may not always be the most appropriate component to normalize to. Furthermore, the CR is subject to numerous variables and for a given medium may change dramatically over time and with circumstance (Polikarpov, 1966). CR values were nevertheless estimated for the Pond B ecosystem because such values are useful for prospective assessments of this and similar ecosystems and for general comparisons.

Concentration ratios for Pond B were calculated from the mean radionuclide concentrations in filtered water and mean radionuclide concentrations in sediments and selected biota, expressed on a dry mass basis (Table 3). The CR values thus estimated, reflect the state of the Pond B ecosystem in the winter season of 1983-84. The few exceptions are noted in Table 3. Conversions of the tabulated CR values from a dry to a wet or ash mass basis can be easily accomplished using factors presented earlier. CR values for less common species not listed in Table 3 can also be calculated from information presented earlier. Data from Table 3 and other information were further condensed into broad taxonomic categories to provide a general impression of mean CR values by selected ecosystem components for ^{137}Cs, ^{90}Sr, and the transuranics (Fig. 13). The transuranic means in Fig. 13 represent the means of the CRs for ^{238}Pu, $^{239,240}Pu$, ^{241}Am, and ^{244}Cm. There may have been significant differences among the transuranics in CR values, but limited sample sizes, low concentrations and frequent non-detection discouraged analysis by individual transuranic nuclide.

Concentration ratios for near-surface sediments, essentially equivalent to field distribution coefficients or K_ds, are clearly highest for the transuranics (CR's ~ 10^6) and lowest for ^{90}Sr (CR ~ 10^3). The transuranics are generally expected to reflect the physical behavior of insoluble particulate matter in ecological systems (Watters et al., 1980) because they attach strongly to particulates and do not readily cross biological

Table 3. Concentration ratios[a] calculated for sediments and selected biota from Pond B.

Medium	Part or Tissue	^{137}Cs	^{90}Sr	^{238}Pu	^{239}Pu	^{241}Am	^{244}Cm
Sediment	0-3 cm	3.2×10^{4}	1.2×10^{3}	1.1×10^{6}	5.9×10^{6}	1.1×10^{6}	1.5×10^{4}
Zooplankton[b]		7.1×10^{4}	3.9×10^{3}	NM[c]	2.3×10^{4}	NM	NM
Macrophytes above-sediment							
Utricularia floridana		1.7×10^{4}	9.4×10^{3}	9.1×10^{4}	1.0×10^{5}	7.5×10^{4}	7.8×10^{2}
Nymphoides cordata		3.1×10^{4}	5.4×10^{3}	7.8×10^{4}	3.0×10^{4}	NM	NM
Cabomba caroliniana		3.7×10^{4}	6.0×10^{3}	5.0×10^{4}	5.2×10^{4}	2.1×10^{4}	3.7×10^{2}
Brasenia schreberi		2.5×10^{4}	8.5×10^{3}	3.6×10^{4}	1.6×10^{4}	2.1×10^{3}	8.4×10^{1}
Nymphaea odorata		1.9×10^{4}	2.1×10^{3}	1.7×10^{4}	6.6×10^{3}	1.4×10^{3}	1.9×10^{2}
Benthic macroinvertebrates							
Insect nymphs, larvae		8.0×10^{3}	5.2×10^{2}	8.4×10^{5}	1.9×10^{5}	2.4×10^{5}	1.9×10^{4}
Gastropods		1.2×10^{3}	5.4×10^{4}	ND[d]	ND	7.8×10^{4}	1.4×10^{3}
Fish							
Micropterus salmoides							
	muscle	3.9×10^{4}	3.4×10^{3}	2.6×10^{3}	5.6×10^{3}	2.5×10^{3}	4.1×10^{2}
	bone	NM	6.3×10^{4}	1.7×10^{4}	ND	ND	1.4×10^{3}
Ictalurus natalis							
	muscle	2.9×10^{4}	6.1×10^{2}	1.2×10^{4}	ND	ND	9.1×10^{1}
	bone	NM	5.7×10^{4}	ND	ND	4.2×10^{3}	ND
Dorosoma sepedianum							
	muscle	4.8×10^{4}	NM	NM	NM	NM	NM
	bone	NM	5.1×10^{4}	NM	NM	NM	NM
Turtles							
Trachemys scripta							
	muscle	1.3×10^{4}	1.0×10^{2}	1.4×10^{4}	6.6×10^{3}	6.6×10^{3}	1.1×10^{2}
	shell & bone	1.1×10^{3}	4.0×10^{4}	1.9×10^{4}	ND	ND	1.9×10^{2}
Waterfowl							
Fulica americana							
	muscle	1.9×10^{4}	1.0×10^{2}	3.8×10^{3}	8.5×10^{2}	6.5×10^{2}	1.1×10^{2}
	bone	2.9×10^{2}	1.8×10^{3}	1.8×10^{4}	ND	ND	ND

[a] Concentration ratio calculated as:

$$\frac{\text{mean concentration in medium (Bq/kg dry mass)}}{\text{mean concentration in filtered water (Bq/L)}}$$

[b] Zooplankton sampled in February 1986.

[c] NM - not measured.

[d] ND - measured but not detected.

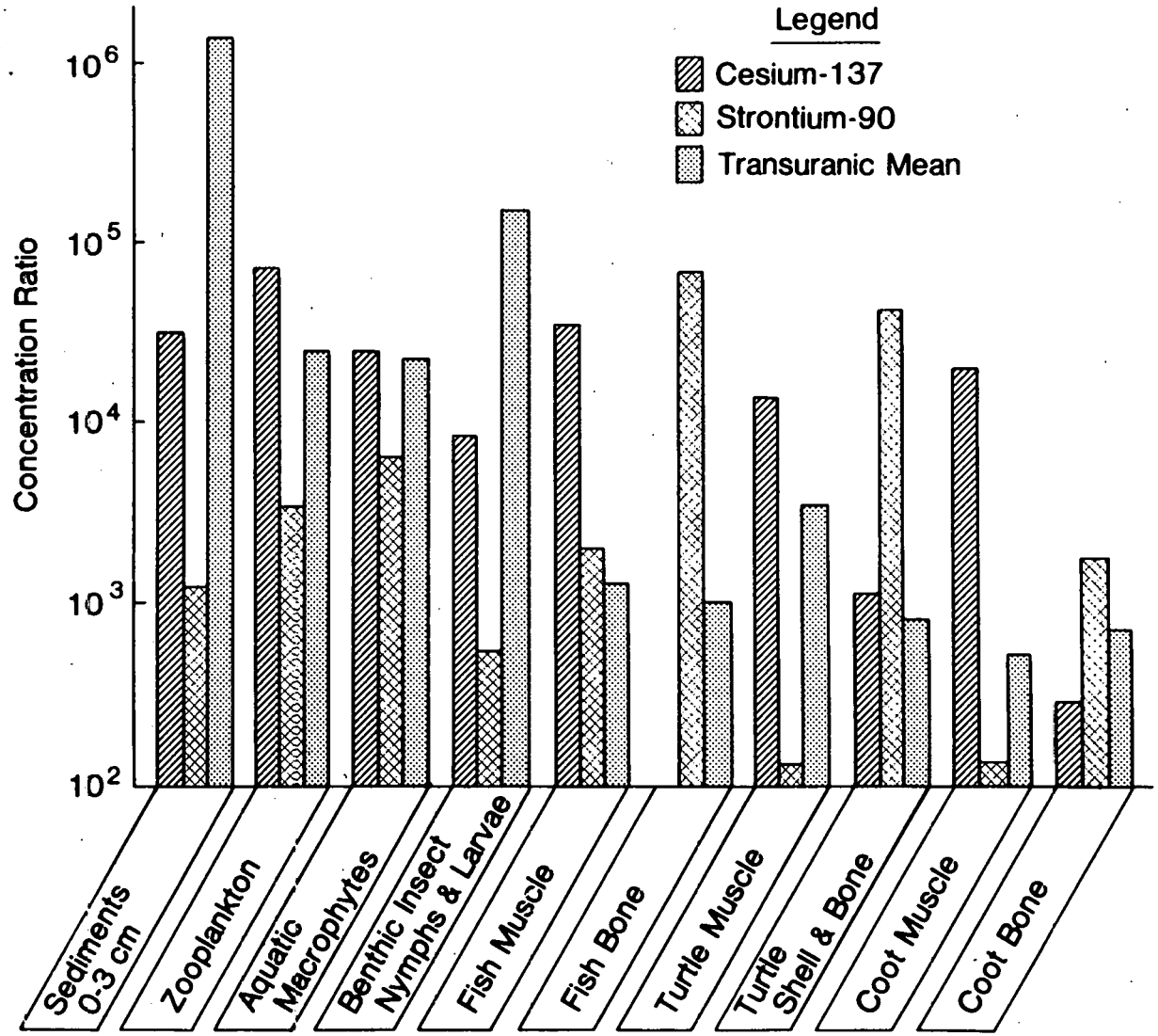

Fig. 13. Mean concentration ratios for ^{137}Cs, ^{90}Sr, and transuranics in selected components of the Pond B ecosystem.

membranes (Bair and Thompson, 1974). Such expected behavior is well-reflected in Fig. 13. Zooplankton and benthic insect larvae have high surface to volume ratios and thus their surfaces, as well as their guts, may carry sediment and sestonic particulates, which in turn could carry measurable ^{137}Cs and transuranic activity. This may explain the relatively high ^{137}Cs and transuranic CR values of these groups (10^4-10^5). Aquatic macrophytes also show a relatively high transuranic CR (~10^4). This could easily be due to adsorption of particulates to the surfaces of macrophytes, which were not cleansed of all periphyton prior to processing. Vertebrate bone and muscle would not be directly affected by such physical transport processes and their transuranic CRs were generally much lower (~10^2-10^3).

Muscle tissues of fish exhibited higher CRs for ^{137}Cs than benthic insects, adding positive evidence to the trophic level increase observed previously for this nuclide (Pendleton et al., 1965; Whicker et al., 1972). However muscle tissues of turtles and coots, both of which are primarily herbivorous, revealed CRs for ^{137}Cs that were slightly lower than the CRs for aquatic macrophytes. Strontium-90 CRs were highest in calcareous tissues (bone and shell) of vertebrates, ranging from 1-2 orders of magnitude higher than macrophytes and invertebrates. On the other hand, ^{90}Sr CRs in vertebrate muscle were generally lower than the values in plants and invertebrates.

Length of residence of vertebrates appears to be generally related to CR values for all three classes of radionuclides. Fish, which are confined exclusively to Pond B throughout their life, usually tend to have higher CR values than turtles and coots, both of which can voluntarily enter or leave the reservoir. In turn, turtle tissues were usually higher in radioactivity than the respective coot tissues. Turtles, though readily capable of moving between separate water bodies (Gibbons, 1970), are certainly less mobile than waterfowl.

In general, CR values for ^{137}Cs, ^{90}Sr and $^{239,240}Pu$ in Pond B organisms appear to be near or exceed the maximum of the range of previously published values (Table 4).

Table 4. Comparisons of concentration ratios:[a] Pond B vs. literature value range.[b]

	^{137}Cs		^{90}Sr		$^{239,240}Pu$	
Biota	Pond B	Literature	Pond B	Literature	Pond B	Literature
Fish Muscle						
Piscivorous	8,500	400-14,000	750	1-130	1,200	0.4-7
Planktivorous	11,000	500-4,100	130	28	NM[c]	NM
Benthic Omnivorous	6,400	270-4,400	NM	NM	920	130-600
Zooplankton	7,100	500-600	350	61	2,200	120-650
Benthic Invertebrates	800	830-3,400	52-5,400	300-720	19,000	590-1,800
Macrophytes	2,400	130-1,500	640	30-220	4,200	1,600-9,000

[a] Fresh weight basis.

[b] Summary in Blaylock (1982).

[c] NM = not measured.

The comparisons in Table 4 are based on the present work and a literature review by Blaylock (1982). The literature values were based on fresh weight, so the Pond B data were adjusted to a wet weight basis. For the various fish categories, the Pond B CR values exceeded the literature range maximum in each case except for ^{137}Cs in piscivorous fish. Even in the latter case, the Pond B value was near the upper end of the published range. Likewise, in nearly all cases, Pond B CRs for zooplankton and macrophytes exceeded the maximum published values. The comparisons for benthic invertebrates, however, are more mixed.

The relatively high CR values for ^{137}Cs and ^{90}Sr in Pond B fish tissues can apparently be largely explained by the low concentrations of potassium and calcium, respectively, in water. Numerous data sets have demonstrated a strong, negative correlation between ^{90}Sr in aquatic biota and Ca in water (Blaylock, 1982; Whicker et al., 1972). A similar but weaker correlation has also been shown for ^{137}Cs and K (Blaylock, 1982). The Pond B CR values, when plotted against their respective nutrient analogue values in water on Figs. 1 and 2 in Blaylock (1982), fits well with expectation from the regressions of the previously published data.

Ecosystem inventories

A major objective of this study was to estimate the total inventories of radionuclides in the Pond B ecosystem and to determine their relative distributions among the major biotic and abiotic components. In most cases, the mean radionuclide concentrations within a given ecosystem component were multiplied by the estimated total mass or volume of the component to obtain the inventory. In the case of sediments, the inventory was estimated from the product of the mean radionuclide quantity per unit area and the area of sediment. Also, there was a distinct effect of water depth on the quantity of radionuclide per unit area of sediment (Fig. 4), so the

inventories were estimated for each of seven depth zones and these were summed to obtain the total inventories.

Detailed inventory data for ^{137}Cs, ^{90}Sr, $^{239,240}Pu$, ^{238}Pu, ^{241}Am and ^{244}Cm are summarized in Table 5 and illustrated in simpler form in Fig. 14. The total inventory of measured radioactivity was dominated by ^{137}Cs (98.9%). The next most abundant radionuclide was ^{90}Sr (0.97%), followed by $^{239,240}Pu$ (0.09%). The vast majority of the radioactivity clearly resides in the sediments. With the exception of ^{90}Sr, over 97% of the radionuclide inventories were found in sediment, thus total inventories of ^{137}Cs and these transuranics in this and similar ecosystems can likely be well-estimated by sampling sediments alone. A much larger fraction of the total ^{90}Sr inventory was associated with the water column and biota than was the case for the other radionuclides (~ 15% versus < 2.5%). This reflects the lower tendency of ^{90}Sr to adsorb to the sediments (see previous discussion on comparative K_d values) and the low calcium concentration in the water, which enhances biological incorporation of ^{90}Sr.

After sediments, the next most important ecosystem components in terms of radionuclide inventory were the water column (filtered water plus seston) and the macrophytes. Water column inventories for ^{90}Sr and the plutonium isotopes exceeded the respective macrophyte inventories by factors of 2.3-4.7, whereas the inventories of ^{137}Cs were equally distributed between these components. The comparative distributions for ^{241}Am and ^{244}Cm were not clear because of incomplete macrophyte measurements. Filtered water contained considerably more activity than seston for all radionuclides except $^{239,240}Pu$, possibly reflecting the extremely high field K_d measured for $^{239,240}Pu$ (5.9×10^6). The ratio of the inventory in filtered water to that in seston was highest for ^{90}Sr (32), reflecting its comparatively low field K_d (1.2×10^3).

Of the total biotic inventory that we were able to estimate (some components were not sampled because of low abundance), macrophytes accounted for by far the largest percentage, ranging from 97% for ^{90}Sr and $^{239,240}Pu$ to > 99% of the total for

Table 5. Pond B radionuclide inventory estimates (I) by major ecosystem component.

Component	Mass, Volume, or Area	^{137}Cs		^{90}Sr		$^{239,240}Pu$		^{238}Pu		^{241}Am		^{244}Cm	
		I (Bq)	% of Total	I (Bq)	% of Total	I (Bq)	% of Total	I (Bq)	% of Total	I (Bq)	% of Total	I (Bq)	% of Total
Filtered Water	3.9E6 m^3	3.0E9	0.66	5.5E8	12.3	3.4E4	7.9E-3	1.3E4	3.9E-2	9.0E4	3.7E-2	2.5E6	2.2
Seston > 45 µm	from 3.9E6 m^3	1.6E8	0.035	1.7E7	0.38	1.1E5	2.6E-2	1.8E4	5.4E-2	2.4E4	1.0E-2	1.9E5	0.17
Sediment	8.7E5 m^2	4.5E11	98.6	3.8E9	84.6	4.3E8	99.9	3.3E7	99.5	2.4E8	99.9	1.1E8	97.6
Zooplankton[a]	4.0E2 dry kg	2.2E7	4.7E-3	2.2E5	0.005	8.0E1	1.9E-5	NM	---	NM	---	NM	---
Macrophytes													
Cabomba	9.9E3 dry kg	3.0E8	0.065	6.7E6	0.15	4.5E3	1.1E-3	1.7E3	5.1E-3	4.8E3	2.0E-3	2.4E3	2.1E-3
Utricularia fl.	9.7E3 " "	1.4E8	0.031	1.3E7	0.29	8.9E3	2.1E-3	2.9E3	8.7E-3	1.6E4	6.7E-3	4.9E3	4.3E-3
Brasenia	2.3E4 " "	5.6E8	0.12	2.8E7	0.62	3.3E3	7.7E-4	2.8E3	8.4E-3	1.1E3	4.6E-4	1.2E3	1.1E-3
Nymphoides	5.5E4 " "	1.3E9	0.29	4.2E7	0.94	1.5E4	3.5E-3	1.5E4	4.5E-2	NM	---	NM	---
Nymphaea	2.9E4 " "	4.4E8	0.096	8.6E6	0.19	1.5E3	3.5E-4	1.7E3	5.1E-3	9.3E2	3.9E-4	3.5E3	3.1E-3
Other species	1.5E4 " "	2.8E8	0.061	1.8E7	0.40	5.6E3	1.3E-3	2.5E4	7.5E-2	NM	---	NM	---
All species	1.4E5 " "	3.0E9	0.66	1.2E8	2.67	3.9E4	9.1E-3	4.9E4	1.5E-1	---	---	---	---
Benthic Invertebrates													
Insects	6.1E2 dry kg	3.9E6	8.5E-4	4.5E4	0.001	1.0E3	2.3E-4	2.5E2	7.5E-4	3.4E3	1.4E-3	7.3E3	6.5E-3
Gastropods	1.6E2 " "	1.5E5	3.3E-5	1.3E6	0.029	ND	---	ND	---	2.9E2	1.2E-4	1.5E2	1.3E-4
All species	7.7E2 " "	4.1E6	9.0E-4	1.3E6	0.030	1.0E3	2.3E-4	2.5E2	7.5E-4	3.7E3	1.5E-3	7.5E3	6.7E-3
Fish													
Largemouth	1.4E3 wet kg	7.7E6	1.7E-3	7.4E5	0.016	7.0E1	1.6E-5	9.9	3.0E-5	15	6.2E-6	1.8E2	1.6E-4
Bullheads	1.2E3 " "	4.9E6	1.1E-3	5.7E5	0.013	ND	---	9.0	2.7E-5	10	4.1E-6	13	1.2E-5
Gizzard shad	6.0E2 " "	4.1E6	9.0E-4	2.6E5	0.006	NM	---	NM	---	NM	---	NM	---
Crappie	1.1E2 " "	6.6E5	1.4E-4	9.1E4	0.002	NM	---	NM	---	NM	---	NM	---
Bluegill	1.3E2 " "	4.6E5	1.0E-4	7.1E4	0.002	NM	---	NM	---	NM	---	NM	---
Warmouth	1.3E2 " "	5.3E5	1.2E-4	6.9E4	0.002	NM	---	NM	---	NM	---	NM	---
All species	3.6E3 " "	1.8E7	3.9E-3	1.8E6	0.040	---	---	---	---	---	---	---	---
Turtles													
Pond Slider	2.4E2 wet kg	1.7E5	3.7E-5	3.4E5	0.008	1.0E1	2.3E-6	5.3	1.6E-5	4.0	1.7E-6	9.4	8.3E-6
Waterfowl	1.1E2 wet kg	2.0E5	4.3E-5	1.3E3	3.0E-5	0.19	4.4E-8	0.59	1.8E-6	0.35	1.5E-7	1.7	1.5E-6
Totals		4.6E11	98.9[b]	4.5E9	0.97[b]	4.3E8	0.09[b]	3.3E7	0.007[b]	2.4E8	0.05[b]	1.1E8	0.02[b]

[a] Based on a Feb. 1986 sample for analysis and a mean biomass estimate for 1984 from data in Chimney et al. (1985).
[b] % of total measured radioactivity in the ecosystem.
ND - measured but not detected.
NM - not measured.
Note: 3.0E9 = 3.0×10^9; 7.5E-4 = 7.5×10^{-4}, etc.

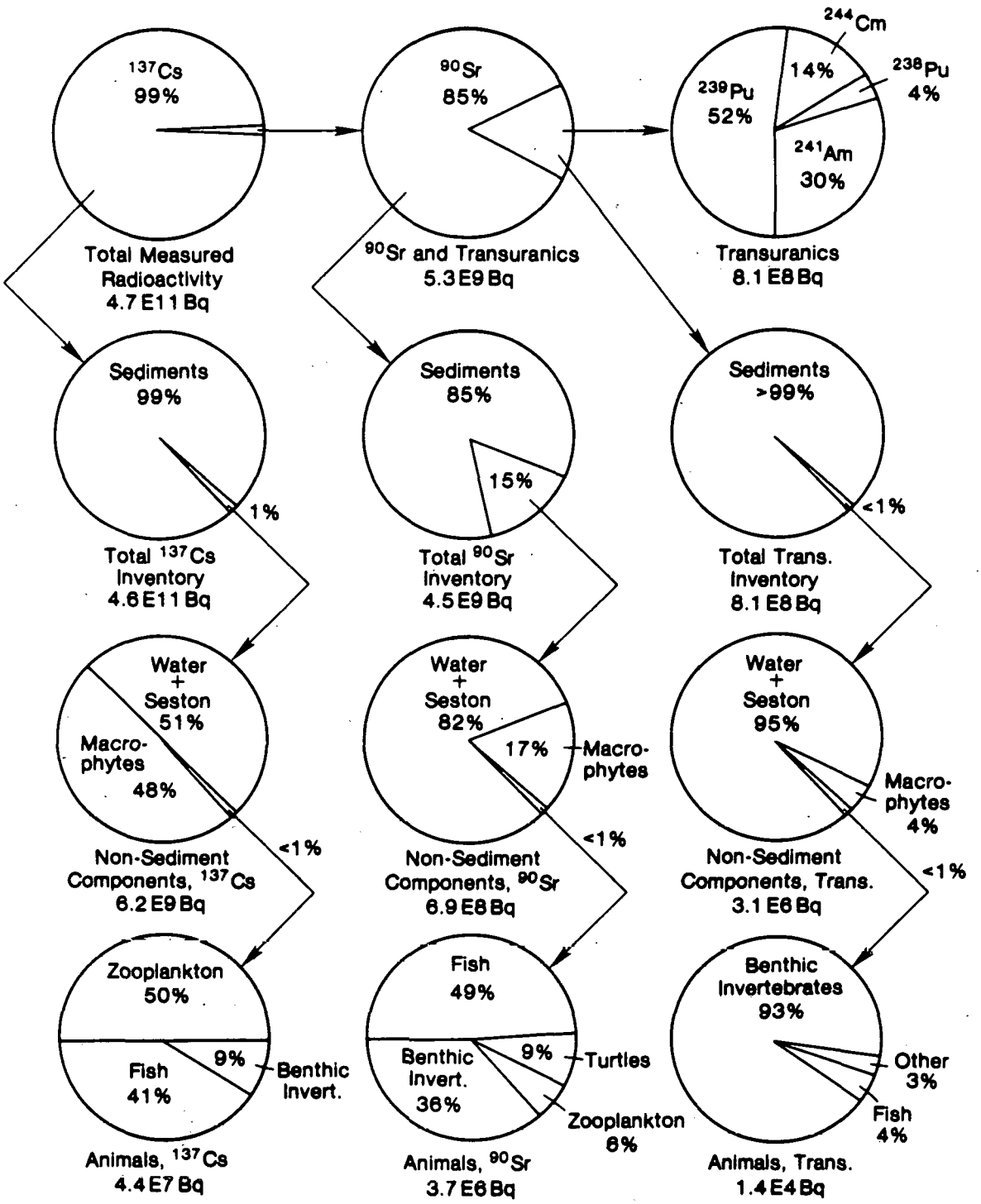

Fig. 14. A simplified illustration of the radionuclide inventories and inventory breakdowns by ecosystem categories for Pond B. Note: 4.7E11 Bq = 4.7×10^{11} Bq, etc.

^{137}Cs and ^{238}Pu. The aquatic macrophytes contained much larger inventories than the animal components, not necessarily because of higher concentrations but rather because of a much larger biomass (~ $1.4x10^5$ dry kg vs. ~ $2.0x10^3$ dry kg). Within the animal biomass, zooplankton contained 50% of the ^{137}Cs and 6% of the ^{90}Sr, while fish contained 41% of the ^{137}Cs and 49% of the ^{90}Sr. Although alligators were not sampled directly in the present study, information presented by Brisbin (1989) yielded estimates of 165 kg and $7.8x10^4$ Bq ^{137}Cs for the total biomass and ^{137}Cs inventories respectively, for this species in Pond B. When compared to values collected in this study for other components (Table 5), these estimates indicated that both the biomass and ^{137}Cs inventories for alligators were similar to those reported here for turtles and waterfowl. About 36% of the ^{90}Sr was found in the benthic invertebrates. Although the data were less complete for the transuranics, it appeared that these radionuclides were primarily contained within the benthic macroinvertebrate biomass. This was due more to higher concentrations in the invertebrates than to a greater biomass. The very low relative inventories in the biotic components indicate that export from the ecosystem through biotic processes such as movements, loss to predators, etc. would likely be extremely low. Loss via surface overflow of water and seston and radioactive decay would appear to represent the principal mechanisms of loss from the ecosystem for the radionuclides studied.

The total inventory of ^{137}Cs measured in the Pond B ecosystem, decay corrected to 1964, represents about 13% of the total reported discharge to R Canal (Ashley and Zeigler, 1980). The respective value for ^{90}Sr is 1.6% of the total reported discharge, indicating the system's greater capacity to retain ^{137}Cs than ^{90}Sr. The fraction of the reported discharge that actually entered Pond B is not known, therefore the system's true efficiency for retaining these radionuclides cannot be calculated. The efficiencies, however, must have been at least 13% and 1.6% for ^{137}Cs and ^{90}Sr, respectively.

The inventory data (Table 5) represent the state of the ecosystem at a single point in time. The dynamics of the system have not been extensively researched and thus one can only speculate about the processes and transport rates that would produce the observed inventories. However, measurements of some of the more critical rate processes, in combination with the observed inventory data, could lead to the ability to construct a dynamic simulation model of radionuclide transport. Such a model could then be employed to estimate past and future states of the ecosystem and have broad applicability for future managment decisions for Pond B and similar systems.

CONCLUSIONS

At the time of sampling in 1983-84, Pond B contained approximately 4.7×10^{11} Bq of long-lived radioactivity, about 99% of which was ^{137}Cs. Nearly 1% was ^{90}Sr, while the transuranic radionuclides accounted for less than 0.2% of the total. The decay-corrected ^{137}Cs inventory is about 13% of the reported R Reactor discharge, while the ^{90}Sr inventory is < 2% of the discharge. Some of the R Reactor discharge may not have entered Pond B, so its true retention efficiency cannot be estimated from the data in hand. Over 97% of the total inventories of ^{137}Cs and transuranics were found in sediments, thus the total inventories of these materials in similar systems can be estimated by sediment sampling alone. The total inventory of ^{90}Sr was primarly in sediments (~ 85%), but this estimate is less certain, since because of the greater rate of transport down through the sediment profile, the cores may not have captured the total sediment inventory.

The inventories of ^{90}Sr and transuranics in the non-sediment components were primarily found in the water column (filtered water + seston). For ^{137}Cs, roughly half of the non-sediment inventory was in the water column and half was in the aquatic macrophytes. Over 97% of the total biotic inventory of all radionuclides studied was contained in the rooted aquatic macrophytes. This was primarily the result of the

comparatively large biomass of macrophytes, rather than higher concentrations. Concerning the radionuclide inventories in the animal components, zooplankton accounted for 50% of the ^{137}Cs and 6% of the ^{90}Sr; fish contained 41% of the ^{137}Cs and 49% of the ^{90}Sr; and benthic macroinvertebrates contained 36% of the ^{90}Sr. The benthic macroinvertebrates accounted for most of the transuranic inventory in animals.

Providing Pond B remains essentially undisturbed, the radionuclide inventories can be expected to decline gradually. Since the system has had 20 years to equilibrate, the radionuclide distributions among ecosystem components are not expected to change appreciably in the near future. The primary mechanisms of loss from the ecosystem, given the current distribution, are radioactive decay and surface outflow of water and seston. Based on limited measurements, roughly 0.3 of the impoundment volume may flow out per year. This would carry $2.1x10^{-3}$, $3.8x10^{-2}$ and $1.1x10^{-3}$ of the total inventory from the system each year for ^{137}Cs, ^{90}Sr, and transuranics, respectively. Loss rates from radioactive decay are higher for ^{137}Cs ($2.3x10^{-2}$/yr), but similar for ^{90}Sr($2.4x10^{-2}$/yr). Sediments, which hold most of the radionuclide inventory, appear quite stable. Animals, such as insects, turtles, waterfowl, and alligators are capable of leaving the system and thus exporting some radioactivity, but since only a trivial fraction of the inventory is contained in animals, biotic export is a very minor loss pathway.

The concentration ratios measured in Pond B are generally as high or higher than measurements reported for other freshwater systems. Concentration ratios for sediments are in the range of 10^6 for the transuranics, 10^4-10^5 for ^{137}Cs, and 10^3 for ^{90}Sr. For biota, ratios range from 10^2 to 10^5, depending on the radionuclide and type of tissue examined. As expected, ^{137}Cs accumulated markedly in macrophytes and muscle tissues of animals, with CRs often exceeding most values in the literature. However, when one considers the low potassium concentrations in Pond B water, the high CRs for ^{137}Cs are not unexpected. Similarly, the Pond B CRs measured for ^{90}Sr are higher than

most literature values, but these can be explained by very low calcium concentrations in water. The comparatively high CR values measured for $^{238,240}Pu$ in Pond B biota were not expected and these remain unexplained. Concentration ratios for fish generally exceeded those for turtles, which in turn exceeded those for waterfowl. This observation supports the expectation that length of residence in the system is an important determinant of the level of contamination.

Cesium-137 and the transuranics behaved very similarly in sediments. These radionuclides all declined rapidly with depth in sediment cores, and all were deposited in a similar pattern according to water depth. Radionuclide profiles in sediment cores, as well as sediment trap measurements, indicated generally low sedimentation rates in Pond B. Wave action in shallow water and sloughage/resuspension in water 4-6 m in depth reduced radionuclide deposition and accumulation in these zones. Extensive macrophyte beds caused enhanced particulate deposition and radionuclide accumulation in water 2-3 m in depth. Enhanced radionuclide accumulation in the relatively flat, quiescent profundal zone was also observed. Strontium-90 had much lower K_ds than the other radionuclides and its behavior in sediments was generally uncorrelated to that of ^{137}Cs or the transuranics.

Radionuclide concentrations in aquatic macrophytes varied across species, but the data failed to reveal the causes for such variations. In two floating-leaved species, ^{137}Cs concentrations increased with water depth, but in other species the depth effect was not significant. Floating-leaved species were shown to obtain most of their plutonium burden from atmospheric deposition, while submerged species accumulated a large fraction from sediment resuspension.

A general increase in ^{137}Cs concentrations with trophic level, when different fish species and benthic macroinvertebrates were compared, was clearly revealed. Strontium-90 concentrations in largemouth bass bone increased as a function of fish size, but no trends across species were observed for this radionuclide. Concentrations

of the transuranics in fish tissues were very low, quite variable, and often not detectable.

Cesium-137 concentrations in turtle muscle were extremely variable, indicating complex residence histories and/or variable food habits. Body burdens of ^{90}Sr increased in proportion to turtle mass, but concentrations in calcareous tissues were independent of mass. As with other vertebrates, the transuranics were difficult to measure in turtle tissues because of the very low concentrations.

An abundant and diverse array of plants and animals have become established in Pond B since its use as a cooling reservoir terminated in 1964. This is apparently the result of a relatively stable hydrologic regime and the lack of exploitation by humans. Radionuclide contaminants, though readily measureable in all components, are not present in sufficient concentrations to cause apparent gross biological perterbations or stress.

Radiation exposure levels on, in, and immediately around the impoundment are trivial for humans, and normal-sized samples of water, sediment and biota can be handled without particular concern. Consumption of fish and the resultant ingestion of ^{137}Cs would be the limiting radiological factor for human use of Pond B, provided the water level was maintained to provide shielding for gamma rays emanating from sediment. The annual intake limit for occupational exposure is $4x10^6$ Bq ^{137}Cs/year (International Commission on Radiological Protection, 1978). For exposure of the general public, this intake limit should be reduced by a factor of 50 (i.e. $8x10^4$ Bq/yr), which produces a dose equivalent of 1 mSv/year. Assuming the maximum observed concentration in fish (8 Bq/g), some 10^4 g/year can be ingested without exceeding the guideline of 1 mSv/year. This would amount to ~ 50 fish/yr, assuming 200 g of edible flesh/fish. The concentrations of ^{137}Cs in fish can be expected to decline with an effective half-time of about 28 yrs, owing primarily to radioactive decay and secondarily to loss from the system by surface water outflow. The ultimate use of Pond

B and similarly-contaminated, limnologically-comparable reservoirs thus appears feasible, provided that adequate monitoring and radiological assessment preceeds and accompanies such use.

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