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ENVIRONMENTAL CONTAMINANTS IN BIRDS: PHOSPHATE-MINE AND NATURAL WETLANDS

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FLORIDA INSTITUTE OF PHOSPHATE RESEARCH

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Florida Institute of Phosphate Research
1855 West Main Street
Bartow, Florida 33830
(863) 534-7160

ENVIRONMENTAL CONTAMINANTS IN BIRDS:

PHOSPHATE-MINE AND NATURAL WETLANDS

FINAL REPORT

Timothy E. O'Meara, Wayne R. Marion, Charles E. Roessler,
Genevieve S. Roessler, Henri A. Van Rinsvelt, and Orrin B. Myers

Department of Wildlife and Range Sciences
University of Florida
Gainesville, FL 32611

Prepared for

FLORIDA INSTITUTE OF PHOSPHATE RESEARCH
1855 West Main Street
Bartow, Florida 33830

Contract Manager: David J. Robertson

Major Research Contributors:

Eduardo Barreto, Daniel Gonzalez, Edward Perry and Michael Stabin

PERSPECTIVE

David J. Robertson, Ph.D.

Florida Institute of Phosphate Research

Uranium is usually found associated with sedimentary phosphate deposits, and the phosphate ore mined extensively in Florida is no exception. The uranium generally is present as a sparingly soluble component of the matrix that is released in quantity only during the manufacture of phosphoric acid. Nonetheless, some uranium is mobilized from phosphate matrix into groundwater. In addition, the uranium associated with phosphate is not in equilibrium with its "progeny" or "daughter" radioisotopes. The uranium daughters that form during the process of radioactive decay may be more soluble than the parent and dissolve in groundwater, they may be physically ejected by recoil when nuclear disintegration occurs, or they may exist in a different state, such as radon gas, which diffuses away. Although the uranium itself is sequestered from the biosphere because of its low solubility, several of the decay products are more problematic.

The isotope of most concern is radium-226. Biogeochemically, radium behaves like calcium, an integral component of vertebrate skeletons. Radium-226 decays through several short-lived isotopes to radon-222. The chemical reactivity of radon is of little inherent concern because it has a short half-life and for all practical purposes is inert. But radon's decay products, particularly polonium-210 and lead-210, are reactive radioisotopes that quickly adsorb onto particulates and can be incorporated into tissue.

Small quantities of radioactive thorium-232 are also present in the phosphate ore body. Generally, thorium occurs in concentrations that are far lower than those of uranium and its progeny.

The radiological quality of reclaimed phosphate-mined land depends a great deal on the type of material that is used to fill the mining excavations. The average radium-226 activity in unaltered surface soil in Polk County, the heart of the central Florida mining district, is 0.6 pCi/g. Where the mining pits have been filled with sand tailings from the beneficiation of the phosphate matrix, the activity averages 3.2 pCi/g. Only slightly higher levels are found in areas reclaimed with overburden, where the average activity is 5.0 pCi/g. Clay settling areas have the highest radium activities, averaging 23.4 pCi/g in central Florida and 14.7 pCi/g in the north Florida mining district.

Some metallic elements were also originally deposited along with phosphate. In general, the concentrations of these elements are extremely low and they are present in forms that are only slightly soluble under most conditions. Nonetheless, a few heavy metals occur

in concentrations that are high enough to have encouraged speculation on their value as a potential by-product of phosphate mineral processing. The metallic elements present in greatest concentrations are titanium, aluminum, magnesium, iron, sodium, antimony, arsenic, and tungsten. Also present, though in much lower concentrations, are barium, cadmium, copper, manganese, molybdenum, potassium, rhenium, and vanadium. Many of these impurities tend to be associated with the smallest fractions of the phosphate minerals. As a consequence, the phosphatic clays tend to have higher concentrations of heavy metals than the ore body in general.

Recognizing the concern over environmental issues associated with reclaimed land, and especially technologically enhanced levels of radiation, several organizations have supported or directly performed research to examine the issue. Among the most active have been the state's Department of Health and Rehabilitative Services, the Florida Phosphate Council, the Department of Environmental Engineering Sciences at the University of Florida, and the Florida Institute of Phosphate Research. To date, the Institute has provided support for 14 projects that directly address radiation. Numerous other Institute projects have radiological components as secondary issues.

The Institute's Environmental Services research program has addressed all aspects of concern over radiation, concentrating on assessment and mitigation of health risks to the public. The program is divided into two areas: (1) public policy and (2) safety and health. The Institute has been most active in supporting research related to health and safety issues that involve direct measurement of the radiological quality of mined land and techniques to reduce public contact with radionuclides. The Institute has funded projects dealing with indoor radon levels, surface and groundwater quality, radionuclide concentrations in natural foodchains, and radioactivity in agricultural products.

Prior to the establishment of the Institute: only one study (by the Florida Game and Fresh Water Fish Commission) had been conducted of radionuclides in wildlife inhabiting areas disturbed by phosphate mining in Florida, Therefore, when the Departments of Wildlife and Range Science and Environmental Engineering at the University of Florida approached the Institute with the concept of performing additional work, the Institute recognized an opportunity to significantly increase information on levels of radionuclides and heavy metals in waterfowl, as well as in their food items and abiotic environment.

The results of this investigation include data on the levels of nine radionuclides and 18 potential heavy metal contaminants in the tissues and skeletons of four species of waterfowl that commonly inhabit wetland areas in the phosphate mineralized regions of central and north Florida. Two of the species, wood ducks and mottled ducks, are often hunted for human sport and consumption, and therefore, represent a potential route for radionuclides to enter into the human food chain. As a result, the investigators prepared a human dose estimate based on consuming these waterfowl.

In addition, levels of the environmental contaminants were measured in the food items of the birds (when they could be identified and collected) and in the water and mud of the birds wetland habitats. Both phosphatic clay settling areas and undisturbed natural wetlands in the mining districts were sampled.

Two other projects with specific goals of developing more data on the radiological quality of reclaimed phosphate mined lands and the ecosystems that have been established on these lands have been funded by the Institute since this project originally was awarded support. In 1981, the Institute began work with Environmental Science and Engineering, Inc. of Tampa on "Ecological Considerations of Reclaimed Lakes in Central Florida's Phosphate Region" (Project #81-03-018). ESE, Inc. compared the radiological quality of reclaimed lakes with that of natural lakes in the mineralized region of central Florida. Radium levels were measured in aquatic vascular plants, zooplankton, phytoplankton, benthic invertebrates, fish, water and lake sediments. Results showed that organisms collected from reclaimed lakes accumulated radium at levels comparable to those in natural lakes in the region.

Four years later, the Institute supported the Florida Audubon Society's efforts to complement the work of earlier investigators. Audubon's project, "Multidisciplinary Study of Radionuclides and Heavy Metal Concentrations in Wildlife on Phosphate Mine and Reclaimed Lands" (Project #85-05-022), included animals that had heretofore not been examined. Audubon selected two aquatic reptiles (alligators and turtles) and one terrestrial mammal (armadillo) based on the criterion that these species have significant proportions of their mass comprised of bony tissue that would likely show elevated radium activities if, in fact, uptake were a problem. Animals from mined, phosphate mineralized, and unmineralized lands in central Florida were targeted for sampling.

The results of Audubon's analyses varied considerably between species. The alligator and armadillo bones contained only low concentrations of radium, and there were no differences between land types. Hardshell turtles did show differences depending on where they were collected. Those sampled from mining-impacted land had, on average, seven times as much radium as those from unmineralized habitats. Softshell turtles also showed location-dependent differences, with lowest activities from unmineralized land, higher levels from mineralized unmined land, and the highest activities from mined wetlands.

Radiation research funded by the Institute has covered at least a portion of all major issues that have been identified by state and environmental organizations. More research is needed in some areas such as groundwater quality and agricultural production. Other studies have indicated the relative insignificance of technologically enhanced levels of radionuclides associated with phosphate mining and processing. However, all research has emphasized the goal of reducing exposure to levels as low as reasonably achievable.

CONTENTS

INTRODUCTION

Phosphate deposits in Florida are located usually from 2-15 m below the surface. As a result of mining these deposits, naturally occurring trace elements, including several trace elements and radionuclides in the uranium and thorium decay series, are redistributed to the earth's surface (Katari et al. 1974, National Research Council Committee on Accessory Elements 1979, Roessler et al. 1979, Wakefield 1980). Processing the ore partitions these elements among marketable rock, sand tailings, and waste clays. Clays typically have an activity of 42-48 pCi/g (Guimond and Windham 1975, Roessler et al. 1979) and retain several trace elements (Katari et al. 1974, Hendry 1978, Wakefield 1980). Many are essential elements, but some can be toxic to animals if present in excessive amounts (Mertz 1981).

Several physical and chemical characteristics of clay minerals make waste clays hydrophilic (Lamont et al. 1975). To conserve and recycle water, a substantial portion of mined-out areas are used as clay settling areas. Mine cuts are diked to accommodate the voluminous waste clays, which exceed available below-ground storage capacity. Settling areas typically are in use for 10-20 years and resemble natural wetlands as succession creates a mosaic of emergent vegetation and open water. These areas serve as attractive breeding and wintering areas for birds (Montalbano et al. 1978, Maehr 1981, Wenner and Marion 1981).

Settling areas produce waterfowl foods (Montalbano et al. 1978, 1979), many of which birds obtain by picking or filtering through shallow water and bottom substrates. Since sediments in wetlands typically are more radioactive than overlying water and may have substantial trace element concentrations (Forstner and Prosi 1979, Ravera 1979), the potential exists

for birds inhabiting settling areas to accumulate toxic concentrations of radionuclides and trace elements from these sediments. Even with contaminant levels in sediments below recognized levels of acute toxicity, synergistic and antagonistic interactions among the elements coupled with added stress factors could be deleterious to wildlife populations (Selby et al. 1970, Truhaut 1975, Dulka and Risby 1976, Ravera 1979, Caren 1981, Fleming 1981, Mertz 1981). Harvest and consumption of contaminated waterfowl by humans may result in threats to human health as well.

Four species in 3 avian families were collected in this study to investigate bioaccumulation of radionuclides and trace elements and to evaluate potential deleterious effects for the birds and humans consuming them. The objectives were:

1. To compare radioactivity and trace element concentrations in tissues of selected avian species collected on phosphate-mine settling areas and unmined (control) wetlands.

2. To determine levels of radioactivity and trace elements in samples of 3 predominant food items for each bird species collected and examine the possibility of, and pathways of, bioaccumulation.

3. To correlate radioactivity and trace element levels in wetland substrates and water with concentrations in bird tissues.

4. To evaluate the potential for deleterious effects to wildlife and man (as a consumptive user of wildlife) resulting from radioactivity and trace elements associated with settling ponds.

The 4 species chosen for study included double-crested cormorants (Phalacrocorax auritis), common moorhens (Gallinula chloropus), wood ducks (Aix sponsa), and mottled ducks (Anas fulvigula). Double-crested cormorants are almost exclusively fish-eaters (Palmer 1976), and herbivory is the dominant feeding mode of common moorhens (Howell 1932). Both are abundant year-round on phosphate mine settling areas and unmined wetlands in Florida, making them suitable subjects for studying contamination in primary and higher level consumers. Wood ducks and mottled ducks commonly breed on phosphate-mined areas, are year-round residents, and also are commonly harvested and consumed by hunters and therefore constitute a potential pathway to humans. Common moorhens are hunted, but harvest and consumption of this species are comparatively low. For purposes of this study, we assumed that ducks constituted the principal potential pathway to humans.

METHODS

Study Areas

Two regions (northern and central) were defined, based on the distribution of subsurface phosphate-bearing strata in Florida (Brooks 1981), for selection of study areas (Fig. 1). Settling areas on Occidental Chemical Company's (OXY) Suwannee River Mine, Hamilton County, were selected in the northern region (Fig. 2). In the central region, settling areas on Noralyn and Clear Springs Mines of International Minerals and Chemicals (IMC) and Agrico's Fort Green and Payne Creek Mines were selected (Fig. 2) .

Control areas having the requisite populations of waterbirds were selected in each region (Fig. 2). Unmined wetlands as close as possible to

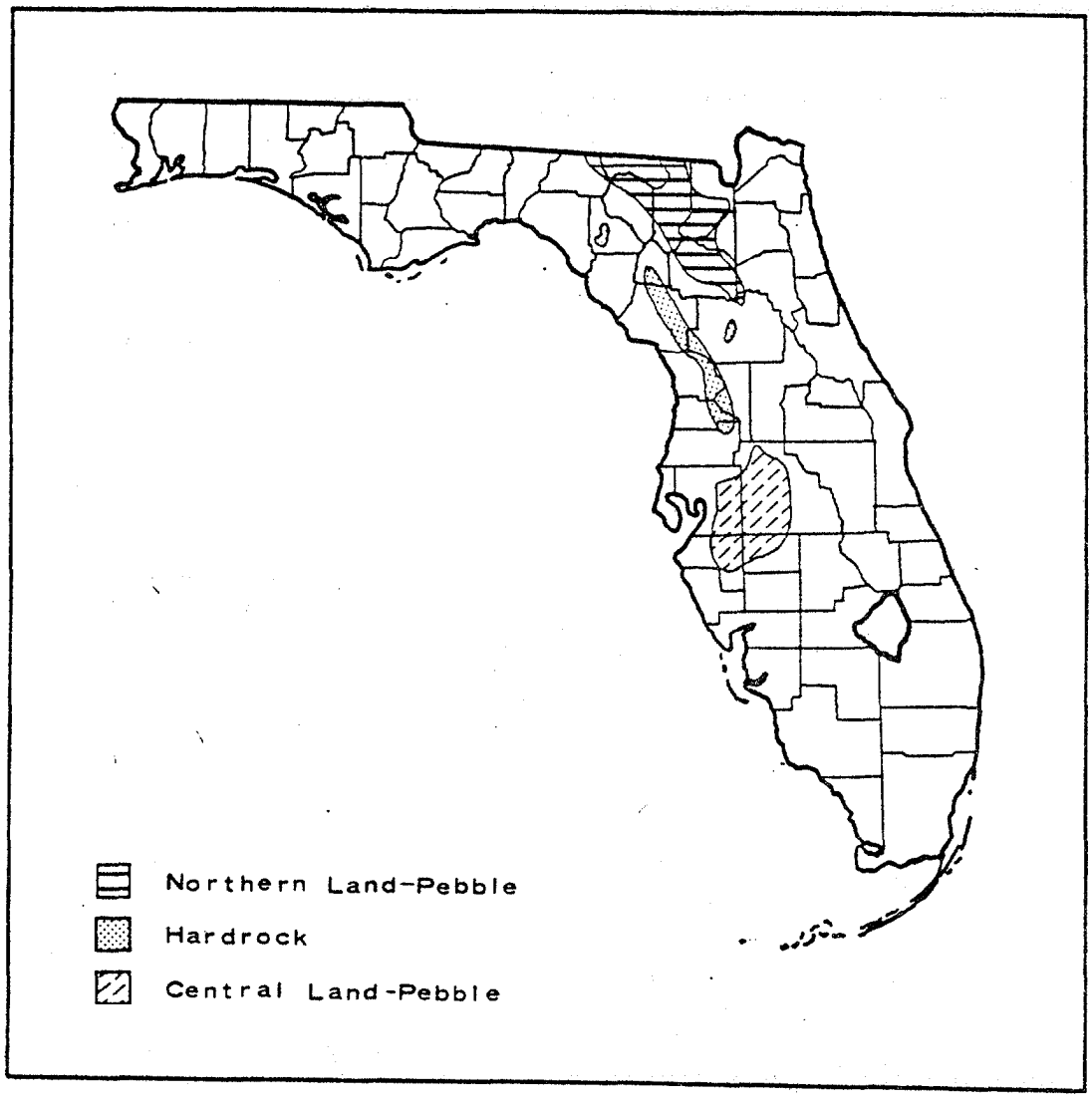

Fig. 1. Distribution of subsurface phosphate-bearing strata in Florida (after Brooks 1981).

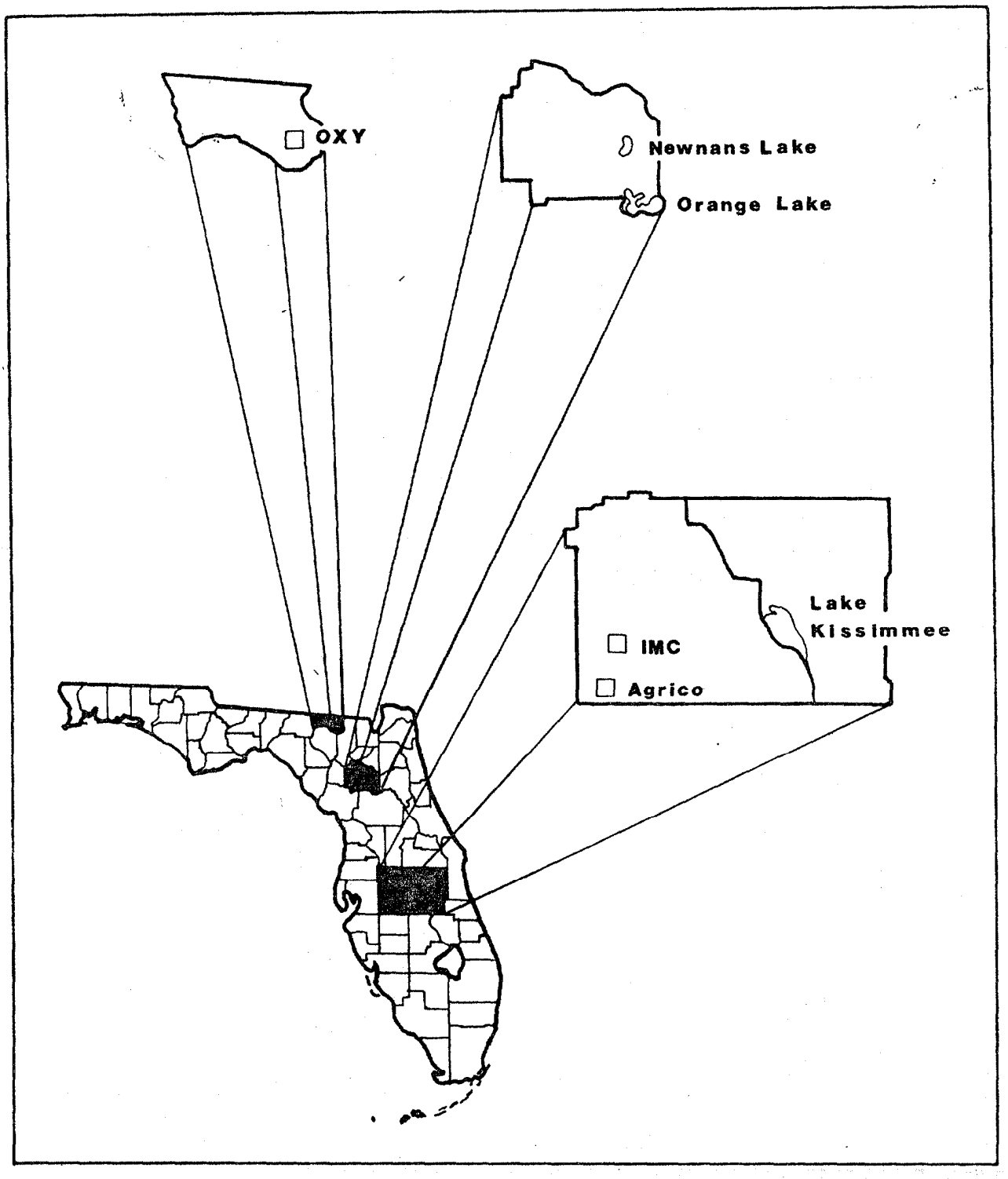

Fig. 2 Locations of study areas in Florida.

areas underlain by phoshpate-bearing strata yet distant enough from settling areas to minimize bird movements between areas were chosen. Newnans Lake and Orange-Lake, Alachua County, were selected in the northern region. The Orange Lake study area included an adjacent freshwater wetland, Right Arm Marsh. Lake Kissimmee, Osceola County, was selected as a control area in central Florida.

Sample Collection

A sample size of 20 birds of each species was sought from each of the 4 study areas (Table 1). Wood ducks and mottled ducks had natural distributions that prevented collecting 20 birds from each area. As a result, mottled ducks and wood ducks were collected in central and northern Florida study areas, respectively. Moorhen collections were limited to adults. Waterfowl and cormorant adults could not be readily and reliably selected in the field; both adults and subadults of these species were collected. All birds were collected with a shotgun using non-toxic steel shot or with a small-bore-rifle to minimize incidental tissue contamination. The major sampling intervals were from June to September, 1981 and 1982. Sixteen mottled ducks also were collected in central Florida (8 control, 8 settling) during March and April 1985 for additional radiochemical analyses.

Soft tissues (muscle, liver, and kidney) and bone tissue, including portions of the femur, tibiotarsus, and humerus, were excised from birds in the laboratory for subsequent trace element and radiological analyses. Sampling of skeletal muscle was restricted to the 2 duck species. To minimize contamination, soft tissues were removed using polystyrene knives and bone was excised using stainless-steel shears. All samples were rinsed with distilled water, placed in ethylene oxide-sterilized polyethylene bags,

Table 1. Numbers of bird, sediment, and water samples collected from 4 study areas in Florida for radiological and trace element analyses.

Area	Double-crested cormorants	Water-fowl[a]	Common moorhens	Sediment	Water
Northern Region					
Settling area					
OXY	20	5	20	12	16
Control Area					
Orange Lake		5	20	10	10
Newnans Lake	20	—	—	2	2
Subtotal	20	5	20	12	12
Central Region					
Settling Area					
IMC	10	10[b]	10	8	9
Agrico	10	18[b]	10	7	8
Subtotal	20	28	20	15	17
Control Area					
Lake Kissimmee	20	25[b]	20	12	12
Total	80	63	80	51	57

[a]Mottled ducks, central region; wood ducks, northern region.

[b]Includes birds obtained in 1985.

and weighed before frozen storage. The upper gastro-intestinal tract, including the proventriculus and esophagus, also was removed from each bird for food habits analyses.

To describe the physical environment, a minimum of 8 water samples and 12 substrate samples were collected from each study area in the vicinity of bird collection sites. Water samples were collected by immersing jars in the upper water column. A plastic scoop or a PVC pipe (3.8 cm diameter) was used to collect substrates. Water and substrate samples were subsequently refrigerated in 3.8 liter screw-top plastic jars.

Detailed analyses of food habits of the collected birds were completed and published (O'Meara et al. 1982). Samples of the 3 predominant food items in each species' diet (Appendix A), based on aggregate volumetric percentages (Swanson et al. 1974), were collected at selected bird-collection sites for evaluation of contaminants in food chains. Vegetative and invertebrate food items were sampled by hand or with dip nets. Fish were sampled using dip nets, gill nets, hoop nets, and by electrofishing.

Sample Preparation

Tissue samples were prepared to ensure representative subsampling for each analysis technique. Soft tissues and food item samples, excluding fish, were weighed, lyophilized for over 24 hours and reweighed to the nearest 0.01 g on a Mettler balance. In most cases, a mechanized shaker was used to convert dried samples to a fine, homogeneous powder. Resistant samples were cooled to below -200°C prior to homogenization. Bone and fish samples did not homogenize well when the above techniques were applied. These samples were weighed, ashed at 400°C and reweighed. Wood duck and cormorant food items were not analyzed for trace element concentrations due to problems with

obtaining adequate samples of wood duck foods and difficulties in homogenizing fish.

Radiological Analyses

Radiological analyses were conducted at the Department of Environmental Engineering Sciences, University of Florida. The primary radionuclide of interest was radium-226. Less extensive information also was obtained for other naturally-occurring radionuclides of the uranium and thorium series.

Substrate samples were oven dried overnight at 100-110°C. Concentrations of radium-226, uranium 238, and thorium-232 were then determined by high resolution gamma-ray spectrometry (Bolch et al. 1977).

Mater and tissue samples were analyzed for radium-226 by the radon-emanation procedure. A portion (up to 1 liter) of each water sample was shaken vigorously to suspend any settled matter. The portion was then filtered with a 0.45 µm pore membrane filter and the resulting filtrate was analyzed for radium-226 by the standard radon-emanation method for drinking water (American Public Health Assoc. 1976). Suspended solids collected on the filter were solubilized for radium-226 analyses according to the procedure used at the EPA's Eastern Environmental Radiation Facility (R. Lieberman 1982, pers. comm.) In this procedure, solubilization was accomplished by repetitive evaporation in hydrofluoric acid, which volatilizes the silica fraction, and hydrochloric acid. The final precipitate was solubilized in hydrochloric acid and diluted to one liter with deionized (D.I.) water. The resulting solution was then analyzed by the same method used for water.

Dried tissues and food items were dissolved using acids either directly or following ashing at 600°C. Resulting solutions were then diluted to one

liter with D.I. water and analyzed by the radon emanation procedure. Inadequate quantities of kidneys to permit radioactivity analyses were available from moorhens and waterfowl from some of the study areas. Minimal amounts of other tissues necessitated compositing tissues from individual birds to achieve adequate volumes for analysis. Some wood duck and cormorant food items were not analyzed for radium-226 concentrations due to problems with sampling and sample preparation, as noted previously.

In the radon-emanation method, counting cells were counted for 900 minutes in a radon cell counting system. Scintillation cells were calibrated and radiochemical recovery was checked using a standard radium-226 solution from the Quality Assurance Division of the Environmental Protection Agency in Las Vegas, Nevada. The laboratory participated in the EPA Quality Assurance Division's quarterly cross-check program for radium-226 in water.

Selected composite samples of water, substrates, foods, and duck tissues from settling and control areas in the central region were submitted to a commercial laboratory* for additional analyses. Isotopes of uranium and thorium, radium-226, and lead-210 were measured to provide information on these radionuclides as well as a cross-check of the in-house procedures for radium-226. The interval between collection and analysis of the initial samples precluded accurate estimation of polonium-210 levels. Mottled duck tissues from the 1985 sampling period were submitted for supplemental analyses of this isotope and the others previously measured. Duplicate bone composites, single muscle composites, and single liver composites from each of the two wetland types were submitted. These composites were submitted

*EAL Corporation, 2030 Wright Avenue,Richmond, CA 94804.

fresh; additional sample preparation was performed by the commercial laboratory.

Lead-210 and polonium-210 concentrations were reported as of the time of collection. This was accomplished for lead-210 by correcting observed concentrations for decay between collection and analysis. This was accomplished for polonium-210 by adjusting for ingrowth from lead-210 and correcting for decay.

Trace Element Analyses

Two techniques were used to quantify trace elements present in a sample: instrumental neutron activation analyses (INAA) and proton induced x-ray emission (PIXE) analyses. Both techniques rely on the interactions between subatomic particles and atoms in the sample to produce measureable radiations characteristic of the elements present. The induced radiations are proportional to the abundance of elements in the sample making quantitative assays possible.

Neutron Activation Analyses. -- Instrumental neutron activation analyses of 700 samples were performed at the Department of Nuclear Engineering Sciences, University of Florida (UF) using the UF Training Reactor at a thermal neutron flux of 1×10^{12} $n.cm^{-2}sec^{-1}$. Substrate samples were irradiated for 10 sec., allowed to decay for 2.3 min., and counted for 500 sec. Tissue, water and food item samples were irradiated for 15 sec., allowed to decay for 2.3 min., and counted for 150 sec. An additional 90 tissue samples were analyzed in a Food and Drug Administration laboratory at the National Bureau of Standards (NBS). These samples were irradiated for 15 sec. at a thermal neutron flux of 6.2×10^{13} $n \cdot cm^{-2}$ sec^{-1}, decayed for 2 min., and counted for 3 min. Blank corrections and NBS standard reference

materials were used to maintain quality assurance for all analyses. Procedures for quantification by INAA were designed specifically for Al, Mg, Na, and V. Other elements which were activated under these irradiation and counting conditions also were quantified and used as a cross-check for the PIXE analyses.

Proton-induced X-ray Emission Analyses. -- Trace elements detectable by proton-induced x-ray emission were analyzed at the Department of Physics, University of Florida, using the 4 MV Van de Graff accelerator. The 2.5 MeV proton beam was diffused by Rutherford scattering and was collimated to a 6 mm diameter homogeneous spot on the target. The beam current was kept below 150 nA to avoid a high counting rate in the detector and eventual evaporation of target material. Samples typically were bombarded for approximately 15 minutes.

X-rays produced during sample bombardment were detected in a 30 mm x 3 mm thick Si(Li) detector in a vacuum at an angle of 135° with respect to beam direction and in the same horizontal plane as the proton beam. A 660 mm thick mylar absorber was inserted in front of the detector to reduce the number of very low energy x-rays. The x-rays were processed by an amplifier/pulse processor and sorted by an LSI-11 based multi-channel analyzer.

Samples were prepared by doping 150 mg of homogenized sample with silver nitrate as an internal standard. After drying at 70° for 6 hours, the powder was again homogenized at liquid nitrogen temperature and fixed to an aluminized mylar backing (thickness 0.7 mg/cm^2; 10/µg of Al/cm^2) with a drop of 1% polystyrene in benzene. Six elements (Ca, Fe, Cu, Zn, Ga, and Sb) normally were present in target backings and were corrected for when quantifying trace element concentrations.

Two types of standards were used to calibrate the PIXE set-up: 1) several multi-element standards specially prepared for this technique in the Department-of Nuclear Chemistry, University of Ghent, Belgium, and 2) standard reference materials from NBS. Normalization between standards and actual samples was achieved with the use of the silver component in spectral data. Elements quantified by PIXE included As,, Br, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Rb, Se, Ti, Y, Zn, and Zr.

Waterfowl Harvest Rate Estimation

Estimates were made of waterfowl harvest rates on settling areas to provide input for determining radiation dose rates to humans consuming waterfowl. A mail survey was used to seek information relevant to harvest parameters and the magnitude and frequency of consumption of waterfowl by hunters. Three mailings of a survey form, cover letter, and self-addressed, postage-paid envelope were accomplished at 4-week intervals to members of the Fin 'n' Feather club having access to hunt waterfowl on IMC-owned land. Average and maximum yearly consumption of duck muscle were extrapolated from harvest figures (see Appendix B).

Data Analyses

Two-way analyses of variance were used to test for differences in trace element levels between settling areas and control areas across regions and to identify interaction effects between treatment and region. T-tests were used to identify differences between settling and control areas within regions in cases where significant ($P < 0.05$) interaction effects were indicated. When sample sizes were adequate, trace element levels in water, substrates, and bird tissues from each species were analyzed using a general linear model for

analysis of variance on unbalanced designs (Freund and Littell 1981). Trace element data were log-transformed prior to analysis to equalize variances between treatment combinations and to improve normality. Samples with visible, physical evidence of contamination by shot or mishandling were excluded from analyses. Trace element results were expressed as parts per million on a dry weight basis. Mean dry-weight/wet-weight ratios were calculated to allow conversion back to a wet weight basis. In cases where an element was not detected in a sample, the level was set at the lowest detectable limit prior to inclusion in analyses.

Log-transformed concentrations of radium-226 were compared between settling and control areas within regions using t-tests. Concentrations in substrate, total water, bone ash, and waterfowl muscle were compared in these analyses.

Trace element and radium-226 levels identified in the 3 major food items of each species were combined by study area to facilitate comparisons. Weighted means were calculated for each element using the volumetric percentage each food item contributed to the total diet as a weighting factor (Appendix A).

Chi-square contingency tests were used to test for homogeneity of bird samples with respect to sex and age composition. Frequencies were tested between study areas within regions using 2x2 tables (moorhens) and 4x2 tables (waterfowl and cormorants).

Tissue weights were analyzed by species using analysis of covariance to identify differences between settling and control areas within regions. Total body weight was introduced into the model as a covariate to minimize allometric biases.

Annual radiation doses to individuals were estimated as the effective committed dose equivalent resulting from an intake at the average concentration of radium-226 found, assuming a conversion factor of 1.1×10^{-3} mrem/pCi ingested (adapted from International Commission on Radiological Protection 1979).

RESULTS AND DISCUSSION

Radioactivity

Radium-226. -- Radium-226 concentrations were greater ($P <: 0.05$) in substrates from settling areas than control areas in both regions (Table 2). Concentrations in substrates from control areas were within the 0.2-3.8 pCi/g range reported for natural soils in Florida (Roessler et al. 1980). Concentrations in individual samples from settling areas ranged from low values (sandy samples) comparable to natural surface soil and natural wetland substrate, to concentrations comparable to those reported for waste clays from phosphate beneficiation plants (Roessler et al. 1980).

Reported concentrations in water samples were higher for settling ponds than for control wetlands but concentrations of dissolved radium-226 were lower in all wetlands than the EPA Water Quality Standard of 5pCi/l for combined radium-226 and radium-228. A major portion of the total activity in water from settling areas was due to suspended matter. The reported concentrations of suspended and total activity may be somewhat arbitrary since the amount of suspended activity would be sensitive to turbulence of the water at the time of sampling and to the amount of disturbance caused by sample collection.

The radium-226 concentrations reported by the commercial laboratory for the submitted substrate and water samples (Table 3) were consistent with the

Table 2. Average radium-226 concentrations in substrates (pCi/kg±SE), water (pCi/l±SE), avian tissues (pCi/kg dry±SE), and diets (pCi/kg dry±SE) from 4 study areas in Florida, June - September, 1981 and 1982.

	Central Region			Northern Region		
	Control	Settling	Observed significance level[a]	Control	Settling	Observed significance level
Substrate	200±100 (13/13)[b]	23,400±3,200 (21/21)	< 0.001	1,400±500 (11/11)	14,700±2,300 (17/17)	< 0.001
Water						
Suspended	0±0.02 (6/6)	2.14±0.96 (7/7)	N.T.	0±0.02 (6/6)	0.41±0.11 (7/7)	N.T.
Dissolved	0.06±0.02 (12/12)	0.60±0.11 (12/12)	< 0.001	0±0.02 (12/12)	0.07±0.02 (12/12)	< 0.001
Total	0.08±0.02 (12/12)	1.98±0.68 (12/12)	N.T.	0±0.02 (12/12)	0.44±0.13 (12/12)	N.T.
Waterfowl[c]						
Bone Ash	322±64 (4/17)	4,440±1,621 (4/16)	< 0.001	685±178 (5/5)	1,935±498 (5/5)	< 0.001
Muscle	13±5 (4/17)	19±5 (5/19)	> 0.1	2±5 (5/5)	9±10 (5/5)	> 0.1
Liver	< 7[d] (3/13)	17±3 (4/17)	N.T.	< 7 (2/5)	23±8 (2/3)	N.T.
Kidney	< 44(4/16)	< 12(3/14)	N.T.	I.S.[e]	155±20 (1/5)[f]	N.T.
Diet	140	365	N.T.	28	1,220	N.T.
Double-crested cormorant						
Bone Ash	263±63 (2/10)	441±44 (3/16)	0.097	165±19 (2/10)	242±14 (2/10)	0.095
Liver	< 6 (3/15)	< 6 (4/20)	N.T.	< 3 (3/15)	2±2 (2/10)	N.T.
Kidney	14±6 (2/21)	3±5 (2/20)	N.T.	11±17 (2/20)	2±6 (1/9)	N.T.
Diet		348	N.T.			N.T.

Table 2. (cont'd)

	Central Region			Northern Region		
	Control	Settling	Observed significance level[a]	Control	Settling	Observed significance level
Common moorhen						
Bone Ash	48±4 (2/20)	455±29 (2/23)	0.002	164±7 (2/20)	352±17 (1/11)	0.06
Liver	< 5 (2/20)	16±6 (2/23)	N.T.	3 (2/20)	1±3 (2/20)	N.T.
Kidney	< 35(1/20)	< 5 (1/12)	N.T.	< 18(1/20)	< 5 (1/9)	N.T.
Diet	144	7,165	N.T.	908	1,014	N.T.

[a] N.T.= not tested.

[b] (Number of composite samples analyzed and averaged/total number of birds included in the average).

[c] Mottled ducks, central region; wood ducks, northern region.

[d] < values indicate that results for all samples in the group were less than the limit of detection. Reported value is 1 standard deviation of the individual analytical determinations.

[e] I.S. = insufficient sample.

[f] For results based on a single composite sample, ± value is the estimated error of the analytical determination.

Table 3. Radionuclide concentrations in substrates (pCi/g±2SD) and water (pCi/l ±2SD, dissolved activity) samples from control and settling areas in central Florida, June-September, 1981 and 1982.

Radionuclide	Substrate		Water	
	Control	Settling	Control	Settling
Uranium Series				
Uranium-238	0.18±0.02	24±2	0±0.1	1.0±0.1
Uranium-234	0.18±0.02	24±2	0±0.1	1.4±0.2
Thorium-230	0.18±0.02	25±1	2±0.2	0.5±0.2
Radium-226	0.09±0.02	24±1	0±0.1	0.3±0.1
Lead-210	0.4±0.1	32±2	0±1	0±1
Polonium-210	0.27±0.06	25±2	0.3±0.4	0.3±0.4
Actinium Series				
Uranium-235	0±0.006	1.3±0.3	0±0.1	0±0.2
Thorium Series				
Thorium-232	0.12±0.02	2.2±0.2	0±0.2	0±0.2
Thorium-228	0.10±0.02	2.2±0.2	0±0.2	0.3±0.2

University of Florida results (Table 2).

Radium-226 levels in avian tissues were consistently high in bone tissue relative to levels in soft tissues (Table 2). Concentrations were below the limit of detection of the method used in many of the soft tissue samples. Radium is an alkali earth with properties parallel to those of calcium and would be expected to be sequestered in bone. As a result, radium-226 levels in bone are probably the best indicators of avian exposure to this element.

Radium-226 levels in bone tissues from all 4 species were greater ($P < 0.05$) on settling areas than control areas in both regions (Table 2). Radium-226 results from the small number of composites submitted to commercial analyses (Table 4) were consistent with the University of Florida results in that concentrations were considerably higher in bone than in soft tissues from both wetland types. Concentrations in bone from settling areas were from 4 (1981-82) to 10 (1985) times those from control areas. The concentration of radium-226 in muscle tissue collected in 1981-82 was apparently greater from settling areas than from control areas. In the 1985 samples, however, there was no difference between the composite samples from the 2 types of areas. The radium-226 concentrations were below the limit of detection in liver composites (1985) from both wetland types.

While birds in settling areas reflected the increased radioactivity of this environment, the tissue increase was not linearly proportional to the substrate increase. Ratios of radium-226 concentrations in bone relative to concentrations in substrates were lower for settling areas than control areas for all species and regions (Table 5). There may be some regulatory mechanism compensating in part for the increased amount of contaminant in settling areas resulting in a nonlinear relationship between substrate and tissue concentrations. Alternatively, settling ponds and natural wetlands

Table 4. Concentrations (pCi/kg dry weight ± 2SD counting error) of isotopes measured by a commercial laboratory in mottled duck tissues collected during 2 sampling periods from phosphate-mine settling ponds and control wetlands.

Tissue	1981-82		1985	
Radionuclide	Control	Settling	Control	Settling
Bone[a]				
Uranium Series:				
Uranium-238	0±10	250±20	9±4;15±5[b]	148±9;80±10
Uranium-234	0±10	240±20	16±6;13±6	160±10;90±10
Thorium-230	0±100	0±40	0±100;0±100	0±100;0±100
Radium-226	270±20	1130±60	140±10;300±20	2400±100;2000±100
Lead-210[c]	930±96	1500±210	490±50;440±50	480±60;430±50
Polonium-210[d]	ND	ND	430±40;360±30	370±40;420±20
Actinium Series:				
Uranium-235	0±5	0±9	0±4;0±2	7±4;0±20
Thorium Series:				
Thorium-232	0±80	0±40	0±100;0±100	0±100;0±100
Thorium-228	0±100	0±100	200±100;300±100	400±100;400±100
Muscle				
Uranium Series:				
Uranium-238	0±0.5	8.6±0.9	0±0.5	0.9±0.5
Uranium-234	0±0.4	10±0.9	0±0.5	0.9±0.5
Thorium-230	8±3	14±3	0±5	0±5
Radium-226	0±0.6	5±0.7	1±0.5	0.8±0.4
Lead-210[c]	0±11	21±11	0±5	0±5
Polonium-210[d]	ND	ND	84±5	89±6
Actinium Series				
Uranium-235	0±0.2	0±0.2	0±0.5	0±0.5
Thorium Series:				
Thorium-232	0±2	0±2	0±5	0±5
Thorium-228	0±2	0±2	13±5	14±5

Table 4 (Cont'd):

Tissue	1981-82		1985	
Radionuclide	Control	Settling	Control	Settling
Liver				
Uranium Series:				
Uranium-238			7±2	13±2
Uranium-234			8±2	14±2
Thorium-230			0±5	0±5
Radium-226			0±3	0±3
Lead-210[c]			50±20	50±20
Polonium-210[d]			1200±60	8700±420
Actinium Series:				
Uranium-235			0±2	0±2
Thorium Series:				
Thorium-232			0±5	0±5
Thorium-228			80±10	130±10

[a]1981-82 bone results reported on an ash weight basis. 1985 bone and all other tissues reported on a dry weight basis.

[b]The double values represent concentrations in duplicate composite samples of bones from birds sampled in 1985.

[c]Lead-210 corrected for radioactive decay between sampling and analysis.

[d]Polonium-210 indicates calculated polonium-210 at time of sampling. Calculated ingrowth from lead-210 has been subtracted out and the remainder corrected for radioactive decay between sampling and analysis.

Table 5. Concentration ratios for levels of radium-226 in substrates, diets, and bone tissues from 4 avian species collected from 4 study areas in Florida, June - September, 1981 and 1982.

	Central Region		Northern Region	
	Control	Settling	Control	Settling
Waterfowl[a]				
diet/substrate	0.70	0.015	0.020	0.083
bone/diet	2.30	12	24	1.6
bone/substrate	1.6	0.19	0.49	0.13
Double-crested cormorant				
diet/substrate		0.015		
bone/diet		1.3		
bone/substrate	1.3	0.019	0.12	0.016
Common moorhen				
diet/substrate	0.72	0.31	0.65	0.069
bone/diet	0.33	0.063	0.18	0.35
bone/substrate	0.24	0.019	0.12	0.024

[a] Mottled ducks, central region; wood ducks, northern region.

may be sufficiently different environments in terms of factors such as ion exchange capacity of the substrate and concentrations of other ions that radium behavior differs in the 2 types of wetlands. For example, uptake of radium by growing plants is dependent on the amount and availability of calcium in the ecosystem (Stabin 1983).

Diet/substrate ratios of radium-226 were less than 1 for all species, indicating that radium was not bioconcentrated in food items relative to levels in substrates. Variation of diet-substrate ratios can be attributed to several factors including differing food items among species and areas and differing weights given to food items in each species' diet. Bone/diet ratios of radium-226 also would be expected to differ among species and areas for these reasons. The fact that only the top 3 food items were averaged to estimate diet concentrations may have contributed to variation in bone/diet and diet/substrate concentration factors as well.

Bone concentrations of radium-226 were appreciably greater in waterfowl than in other species on the same areas (Table 2). Although moorhen diet/substrate concentration ratios were generally greater than or equal to those for waterfowl, bone/diet ratios were consistently greater for waterfowl than moorhens. These characteristics of the data suggest that either waterfowl may bioaccumulate radium from food items at greater ratios than moorhens or items high in radium-226 that were consumed by waterfowl were not included in waterfowl food sampling. Waterfowl, as a result of their mode of feeding, may inadvertantly ingest quantities of radium-bearing sediment not reflected in the diet samples.

Other Radionuclides. -- In addition to reporting radium-226 concentrations, gamma spectrometric measurements of substrates gave an indication of the degree to which uranium-238 and thorium-232 were present.

Table 6. Radioactivity [pCi/g±SE(n)] in substrates from 4 study areas in Florida, June - September, 1981 and 1982, as measured by gamma spectrometry.

Radionuclide	Central Region		Northern Region	
	Control	Settling	Control	Settling
Radium-226	0.2±0.1(13)	23.4±3.2(21)	1.4±0.5(11)	14.7±2.5(17)
Uranium-238	< 0.3[a]±0. 1(13)	30.7±3.5(21)	< 1.4±0.7(11)	17.4±2.9(17)
Thorium-232	0.2±0.1(11)	<0.6±0.1(20)	< 1.0±0.3(11)	<0.7±0.1(17)

[a] < value indicate some samples were below the limit of detection; the limit of detection for these samples was entered into the calculation of the mean standard error.

Average uranium-238 concentrations were comparable to those for radium-226 (Table 6). Thorium-232 concentrations in substrates were low and, in a large fraction of samples, were below the limit of detection for analysis in the presence of the higher uranium series concentrations. Average thorium-232 concentrations were less than 1 pCi/g in the control areas and thus at levels comparable to or less than/radionuclides of the uranium series. Average thorium-232 concentrations also were less than 1 pCi/g in the settling area substrates and hence at least an order of magnitude lower than those of the uranium series radionuclides.

Results of the commercial radiochemical analyses of single substrate and water composites from each wetland type in central Florida are presented in Table 3. The substrate samples clearly illustrated the difference between the control area and the settling area. In the control area, radionuclide concentrations were low. Initial long-lived members of the uranium series, uranium-238, uranium-234, and thorium-230, were in radioactive equilibrium at about 0.2 pCi/g, a result that was consistent with the gamma spectrometric result for uranium-238. The next member of the series, radium-226, was observed by radiochemical analysis of the composite at approximately 0.1 pCi/g, about half the concentration of uranium-238 and about half the average concentrations observed by gamma spectrometry. In the composite sample, the final members of the series, lead-210 and polonium-210, appeared at higher concentrations than earlier members of the series - on the order of 0.3 to 0.4 pCi/g. This enhancement of the latter 2 radionuclides is presumably due to deposition from atmospheric sources.

In the composite substrate sample from the settling area, uranium-238 was present at 24 pCi/g and the series through radium-226 was in radioactive equilibrium. These observations were consistent with the average gamma

spectrometry results. Lead-210 and polonium-210 were in approximate equilibrium with uranium-238 (32 and 25 pCi/kg, respectively). The differences seen in the control area substrates between these nuclides and other members of the series were largely overwhelmed by the higher overall concentrations in the settling area substrates.

Uranium-235 (actinium series) was observed in the composite substrate sample at concentrations much lower than for uranium-238 and 234 (uranium series) and was not detected at all in water or tissue by the method used. The uranium-235/uranium-238 ratio of 0.05 observed in the settling area substrate was comparable to the ratio of 0.0465 expected for natural uranium.

In the composite substrate sample from the control area, concentrations of thorium-232 and thorium-228 (thorium decay series) were about two-thirds the uranium-238 concentration, a result that was consistent with the gamma spectrometric result and that reflected the relative abundances expected for soils. In the settling-area substrate composite, thorium series concentrations were about 10% of the uranium series concentrations. Results from both gamma spectrometric and commercial radiochemical analyses clearly reflect the predominance of the uranium series over the thorium series in phosphate mineral waste clays. Therefore, this supports our decision to place emphasis on radium-226 and other members of the uranium series.

Concentrations of all radionuclides were relatively low in the filtered water samples from both the control area and the settling area. Isotopes of uranium were below the limit of detection in the control-area sample and present at about 1 pCi/l in the settling-area sample. The thorium-230 concentration of 2 pCi/l in the control-area sample appears unusual compared to the 0.5 pCi/l in the settling-area sample and compared to the other nuclides of the series. With only a single composite sample, it was not

possible to determine whether this apparent anomaly was real or an error in sampling or measurement. Radium-226 was below the limit of detection in the control area and 0.3 pCi/l (about one-third the uranium-238 concentration) in the settling area. In water samples from both areas, lead-210 was below the limit of detection (1 pCi/l). The more sensitive polonium-210 analysis indicated a concentration of 0.3 pCi/l from both areas. This latter concentration, about 20% of the uranium-238 concentration in the settling area sample, indicated a low solubility for polonium-210. The fact that control and settling area concentrations were comparable suggested that the major source in both types of water bodies may be from atmospheric deposition rather than dissolution from the substrate.

Thorium isotopes of the thorium series were not detected in water or any of the tissue samples by the method used. These observations indicate that the thorium series is of much less concern than the uranium series in natural wetlands and settling areas in Florida.

In general, concentrations of uranium series radionuclides in duck tissues were higher for settling area samples than for control area samples. However, this pattern was not observed for all radionuclides and all tissues and there were quantitative differences between the 1981-82 and the 1985 sampling episodes (Table 3).

As expected, uranium-238 and uranium-234 concentrations were in approximate equilibrium. Concentrations of these nuclides were higher in settling area samples than in control area samples for all tissues and both sampling episodes.

In control area samples, concentrations of uranium-238 and uranium-234 were below the limit of detection in muscle, were 7 and 8 PCi/kg in liver (1985), respectively, and ranged from below the limit of detection (1981-82)

to 16 pCi/kg (1985) in bone. In the settling area samples, concentrations and enhancement over the control were greatest in bone. The differences between 1981-82 samples and 1985 samples can largely be attributed to differences in reporting on an ash-weight and dry-weight basis. Muscle samples from the initial sampling episode, however, contained about 10 times the uranium concentrations measured in the 1985 samples.

Thorium-230 concentrations were below the limit of detection for bone and liver samples and this radionuclide was not taken up by these tissues in concentrations comparable to uranium. Thorium-230 appeared to be concentrated in muscle to a greater extent than in bone in the 1981-82 samples but not in the 1985 samples. The information provided by these analyses was limited by the fact that the thorium-230 analysis was approximately 1/10 as sensitive as the uranium analysis.

Lead-210 concentrations also were considerably higher in bone than in soft tissues. The concentration in the settling area bone sample was about 60% greater than, that in the control area sample in 1981-82. In 1985, concentrations in bone were comparable in magnitude to the 1981-82 concentrations (accounting for the difference between ashed and dry weights) but there was no apparent difference between the two wetland types. In muscle, concentrations were below the limit of detection in both control samples; the concentrations in settling area samples ranged from 21 pCi/kg (about twice the limit of detection) in 1981-82 to non-detectable in 1985. Concentrations in liver were the same in settling area and control samples in 1985.

Polonium-210 (1985 samples) was observed at about the same concentration as lead-210 in all bone samples from both wetland types. Polonium-210 in bone is probably present as a result of ingrowth from lead-210 rather than

through independent uptake of polonium-210. Concentrations for both wetland types were higher in bone than in muscle and even higher in liver. After correcting for ingrowth and decay, there was no significant difference between settling and control muscle samples but the settling area liver sample had about seven times the polonium-210 as in the control sample. These data suggest that polonium is taken up independently from and preferentially to lead in soft tissue, that the liver is a major organ of polonium uptake, and that a greater concentration of polonium may be expected in settling area livers than in control area livers.

With the exception of one result (7 pCi/g in one 1985 settling area bone composite), uranium-235 concentrations were all below the limit of detection. Similarly, thorium-232 was below the limit of detection in all samples from all sampling episodes.

Thorium-228 was not detectable in any 1981-82 samples but was detected in all 1985 samples. In the 1985 samples, concentrations were on the order of 300 pCi/g in bone, 100 pCi/g in liver, and 14 pCi/g in muscle. There was a suggestion of slightly greater concentrations in settling area samples than in controls. The 1985 thorium-228 concentrations are difficult to explain in that they were inconsistent with thorium-232 concentrations. Unless there was an unusual partitioning, these two thorium isotopes from the same decay series would be expected to be in radioactive equilibrium - as was the case for the 1981-82 substrate samples. Enhanced thorium-228 might result from long-term ingrowth from radium-228 uptake. This, however, would require a radium-228 uptake that is much higher than would be consistent with the measured radium-226 concentrations and the demonstrated thorium series/ uranium series ratio in this environment.

Radiation Dose to Humans. -- The radiation dose to humans consuming duck meat was estimated assuming an average consumption of 1.5 kg/yr and a maximum intake of 10 kg/yr (Appendix B). For the average hunter, effective dose equivalents attributable to radium-226 in duck meat range from 0.001 mrem/year for eating ducks from northern Florida control areas to 0.01 mrem/year for eating ducks from central Florida settling areas.

Evaluating the significance of the observed radium-226 concentration in duck tissue is complicated by the fact that there is no specific standard for radium intake due to specific food items other than drinking water. Stabin (1983) reviewed various standards and concluded that appropriate working values for evaluating radionuclide intake in individual food items would be an annual dose equivalent of 3 mrem or a radium-226 intake of 2700 pCi per year. More recently, the Standards Committee of the Florida Phosphate-related Radiation Task Force (1984) has recommended 500 mrem/yr as the appropriate standard for all exposure of individuals of the general public and has suggested that ingestion of radium-226 be limited to 20 pCi/day. If the intake permitted by the Drinking Water Standard (10 pCi/day, Environmental Protection Agency 1976) is subtracted, this corresponds to 10 pCi/day (3700 pCi/year) or 5 mrem/year from ingestion sources other than drinking water. For consumption of duck muscle from central Florida settling ponds, the area with greatest radium-226 levels, the average and maximum annual intakes estimated for individuals are 0.3% and 2%, respectively, of the non-drinking water portion of the recommended intake limit. Alternatively, at the maximum average concentration of 6 pCi/kg that we found, it would require an intake of over 450 kg/year (980 pounds/year) to achieve an annual effective dose equivalent of 3 mrem. Thus, even at maximum

hypothesized consumption rates, waterfowl meat from settling ponds would not represent a health risk to humans.

Our values for radium-226 in duck muscle were greater than, but the same order of magnitude as, those reported by Montalbano et al. (1983). They found greater ($P < 0.01$) radium-226 concentrations in duck muscle from a central Florida settling pond than from a control wetland, but they also concluded that ingestion rates of radium-226 by humans resulting from these contaminant levels were insignificant.

Radiation Dose to Birds. -- There are no published radiation standards for birds or other animals. The Advisory Committee on Biological Effects of Ionizing Radiation (1972) concluded that existing dose limits to humans can be applied safely across all animal populations.

The Federal Radiation Council (1961) has recommended that radium intake by humans be controlled such that the quantity of radium-226 in the skeleton not exceed 0.001μCi. For the reference man skeleton of 5 kg of bone (International Commission on Radiological Protection 1975) this corresponds to a concentration of 200 pCi/kg wet. By comparison, the average radium-226 concentration in bone from waterfowl collected on the central Florida settling area was approximately 800 pCi/kg (based on an observed 4440 pCi/kg and an assumed 18% ash content).

This comparison provides only a crude evaluation for a number of reasons. Because of small dimensions, the absorbed dose per unit concentration is probably less in bird bone than in human bone due to greater loss of the radon daughter product of radium and the lower absorbed fraction for the gamma radiation. In humans, the concern is for bone cancer which has an induction latent period on the order of 10-40 years. This period would have to be drastically shortened in order for such effects to appear in the

lifetime of the birds collected, which is a maximum of several years. However, birds exhibit greater metabolic rates than humans; it is unknown whether the observed radium concentrations in bird bone would constitute a health hazard to birds. Moreover, the fact that the concentration was 4 times that recommended for human individuals suggests that further dosimetric calculations and research on effects should be performed.

Trace Elements

Substrates. -- Fourteen trace elements were found in greater ($P < 0.05$) concentrations in substrates from at least 1 of the settling areas than in the respective controls (Table 7, Appendix C-1). Of these elements, 7 were found in greater concentrations in settling areas than control areas in northern Florida, and all 14 were found in greater concentrations in settling areas than in control areas in central Florida.

Differences in substrate concentrations between regions may have been due to structural differences in substrates as well as regional differences in element concentrations. One factor affecting trace element concentrations is soil particle size; most trace element pollution is associated with particle sizes between 0.45 and several hundred μm (Literathy and Laszlo 1976). Substrates collected from northern Florida control areas were primarily organic matter and would be expected to have more available surface area for trace element adsorption than the more sandy substrates from the central Florida control area. Also, since substrate sampling locations were based on bird collection sites rather than substrate characteristics, substrate samples differed in the relative amounts of clay and sand they contained. Fewer significant differences in northern Florida may have been due to the fact that nearly one-half of the substrate samples collected from

Table 7. Trace elements found in substrate, water and tissue samples from 4 study areas in Florida, June - September 1981 and 1982. Relative toxicity of these elements and regions within which concentrations were greater ($P < 0.05$) on settling areas than control areas are indicated.

Element	Substrate	Water	Common moorhen			Double-crested cormorant			Waterfowl[a]				Toxicity[b]
			Liver	Kidney	Bone	Liver	Kidney	Bone	Liver	Kidney	Bone	Muscle	
Al	CN[c]		CN					CN				C	S
As													H
Br			CN	CN	CN		N	N					$V(Br_2)$
Co													M
Cr	CN												M-H
Cu	C		CN	CN	N				N	C	N		M
Hg													H
Mn	C					C			CN				M
Mo	CN									CN			M
Ni	CN												M-H
Pb	C									C			M
Rb	C		C	CN		C	CN	C	C	CN	CN	N	N
Se	C		CN	CN		CN	CN		CN	CN		CN	H
Ti	CN												N
V	C		CN		CN	CN			CN	CN	CN		M
Y	CN												S
Zn	C				CN	CN			N				S
Zr	CN												S

[a]Mottled ducks, central region; wood ducks, northern region.

[b]To animals, N=relatively harmless, S=slight, M=moderate, H=high, V=very high (from Bowen 1966).

[c]C=central region, N=northern region.

settling areas in that region were sandy. Although settling areas showed elevated levels of several elements in relation to levels on control areas, trace element concentrations on all areas were within the ranges expected for soils and sedimentary rocks (Bowen 1966).

Water. -- Only 8 trace elements were identified in the combined dissolved and suspended fractions of water samples (Appendix C-2). Most of these were found in low concentrations, and none differed ($P > 0.05$) between the settling area and control areas in either region.

Tissues and Food Items. -- Ten elements were found in greater ($P < 0.05$) mean concentrations in avian tissue samples from settling areas than in the same tissues from the respective controls (Table 7, Appendices C-3 to C-12). Nine of these elements (Al, Br, Cu, Mn, Mo, Pb, Se, V, Zn) are slightly to very toxic to animals (Bowen 1966). With the exception of Al and Se, all were within the ranges of normal concentrations of these elements reported for animal tissues in papers summarized by Bowen (1966) and Underwood (1977). Similarly, all 9 elements, with the exceptions of MO and Al, were found in food items at concentrations below levels identified as potentially harmful in animal diets by studies summarized by the Subcommittee on Nutrient and Toxic Elements in Water (1974), Dvorak (1978), and White and Dieter (1978). Molybdenum was not identified in any of the food item samples.

Fluoride has been identified in phosphate ore (Katari et al. 1974) and water from phosphate mines (Miller and Sutcliffe 1982). Fluorine concentrates in bones and teeth and can cause fluorosis in animals (Underwood 1977). We were unable to measure F in our samples with the assay techniques used and could not assess impacts of this element on birds on phosphate-mine settling ponds. Montalbano et al. (1983), however, found no difference

between concentrations of F in duck muscle from settling ponds compared to duck muscle from a control area.

Aluminum was found at elevated levels in substrates, common moorhen livers, double-crested cormorant bones, and waterfowl muscles on settling areas. Tissue levels for Al apparently have not been reported for birds in the wild. Our results of 11.9 and 6.3 ppm wet (33.0 and 25.1 ppm dry, conversion factors for converting from a dry weight to wet weight basis are in Appendix D) in moorhen livers from settling areas were at least an order of magnitude greater than the 0.4 ppm wet reported for human liver by Hamilton et al. (1972/1973, in Underwood 1977). Aluminum levels in moorhen livers also were substantially greater than the 0.03 ppm dry reported for mammalian liver by Tipton and Cook (1963, in Bowen 1966). Aluminum levels in cormorant bones from settling areas (155 and 160 ppm ash) also were greater than the levels reported by Hamilton et al. (1972/1973, in Underwood 1977) for human bones (60-73 ppm ash). Aluminum in duck muscle from central Florida settling areas averaged 12.0 ppm wet compared to 0.5 ppm in human muscle (Hamilton et al. 1972/1973, in Underwood 1977).

At high levels of intake, Al produces gastrointestinal irritation and can produce rickets by interfering with phosphate absorption. Storer and Nelson (1968, in Subcommittee on Nutrient and Toxic Elements in Water 1974) reported that 500 ppm in poultry feed can reduce growth in immature chickens. Levels of 2200 ppm in feed can cause rickets in chicks (Debold and Elvehjem 1935, in Subcommittee on Nutrient and Toxic Elements in Water 1974). Only moorhen diets on the settling area in the central region exceeded 500 ppm (Appendices E-l to E-4). Items in moorhen diets on this area with highest levels of Al were common duckweed (Lemna minor, 738 ppm Al wet) and green algae (Oedogonium sp., 2621 ppm Al wet). These 2 food items comprised 46% of

moorhen diets on this area, suggesting that Al may be potentially harmful to moorhen growth and reproduction on central Florida settling ponds.

Although Al was found in moorhen food items at levels potentially harmful to these birds, and Al levels in several bird tissues were greater than normal levels reported in the literature, there was no apparent bioaccumulation, or concentration at higher trophic levels, of this element. Ratios of Al concentrations between tissue/food and tissue/substrate were less than 1 for all tissues.

Elevated levels of Al in duck muscle tissues from settling areas apparently posed no threat to humans consuming ducks from phosphate mines. Aluminum levels in mottled duck muscles from central Florida settling areas averaged 12 ppm wet. This amount would be unlikely to cause toxicity problems for humans consuming these tissues, since human diets average 36.4 mg/day of Al in the U.S. (Hamilton and Minski 1972/1973, in Underwood 1977). For example, the intake of a person consuming 227 g (0.5 lb) of duck muscle per day with 12 ppm Al would be only 2.7 mg/day.

Selenium was found at greater levels in all liver, kidney, and muscle samples from settling areas than from control areas (Table 7). Highest mean levels found in each tissue included 22.2 ppm dry (5.8 ppm wet) in cormorant livers and 13.7 ppm dry (3.0 ppm wet) in cormorant kidneys. Levels in waterfowl muscle were greatest on central Florida settling areas (3.6 ppm dry, 1.2 ppm wet). These levels were greater than normal levels of 2.1 ppm dry in mammal muscle as reported by Dye et al. (1963, in Bowen 1966). Our results are within the range of the 5-7 ppm wet in liver and kidney and 1-2 ppm wet in muscles reached by animals at toxic levels of Se intake (Underwood 1977). However, levels of Se in food items of moorhens and mottled ducks that we sampled were well below the minimum dietary levels at which signs of

toxicity will arise (3-4 ppm wet Munsel et al. 1936, in Underwood 1977), and also below the 2 ppm which Arnold et al. (1973, in Underwood 1977) found to have-no adverse effect on growth or mortality of chicks or on the production and hatchability of eggs. Selenium was not detected in water samples from any of our areas, but mean levels in substrates were all equal to or greater than 0.5 ppm. Selenium levels greater than 0.5 ppm in soil are potentially dangerous to animals (J.G. Nagy unpublished manuscript).

Unlike Al, Se concentrations increased with trophic level on the study areas. Liver/substrate ratios of Se ranged from 0.57-22 for the 4 species with only the northern control area exhibiting a value less than 1.0. Kidney/substrate Se ratios were similar to liver/substrate ratios with a range of 1.4-14. Duck muscle/substrate Se ratios also were greater than 1.0, with the exception being the northern Florida control area. Tissue/food Se ratios also indicated bioaccumulation of Se. Liver/food Se ratios ranged from 1.2-26 and kidney/food Se ratios ranged from 1.8-53 for mottled ducks and moorhens. Muscle/food Se ratios were 1.3 and 1.6 for mottled ducks from control and settling areas in central Florida, respectively.

Although Se levels in food items tested were low, tissue levels and concentration factors suggest that Se may be borderline in its toxicity to wildlife. The potential for Se toxicity problems in wildlife is greater on phosphate-mine settling areas than control areas as evidenced by the greater concentrations in soft tissue from these areas. "All degrees of Se poisoning exist, from a mild, chronic condition to an acute form resulting in death of the animal (Underwood 1977: 334)". Seleniferous diets can affect growth and reproduction as well as the health of an animal (Underwood 1977). Since the toxicity of Se varies with amounts and forms of Se ingested, duration and continuity of Se intake, and the nature of the rest of the diet (Underwood

1977), it is difficult to predict effects of Se intake by wildlife on phosphate-mine settling areas.

Cases of Se toxicity in humans apparently have not been reported. It has been proposed, however, that consumption of small amounts of Se by children can increase incidence of dental cavities (Hadjimarkos 1973, in Underwood 1977). Average dietary intake of Se in the U.S. and Canada ranges from 60-220µg Se/day. Consumption of 227 g (0.5 lb) of waterfowl muscle from either of our central Florida study areas would represent an intake of 272 µg of Se. It is not clear whether Se intake at this level would pose a threat to human health. Although dry weight concentrations of Se in duck muscle differed ($P < 0.05$) between settling and control areas in central Florida, wet weight concentrations would contribute Se to human diets at approximately the same rates. If Se is a potential hazard to humans consuming waterfowl, the threat appears to be no greater on phosphate mined lands than on natural wetlands.

Despite elevated concentrations of several trace elements on settling areas, no evidence of unhealthy avian populations was observed. Body weights of each species did not differ between settling and control areas with the exception of mottled ducks, which were heavier on settling areas (Appendix F). Successful reproduction by moorhens on settling areas was observed during the course of the study, and high nest success rates for wood ducks on northern Florida settling ponds have been reported (Wenner and Marion 1981).

One response to trace element contamination may be elevated soft tissue weights. In mammals, hepatic injury induced by organic chemicals can result in focal and zonal necrosis marked by lipid accumulation (Plaa 1980). Renal insult can cause necrosis of the proximal tubules with accumulation of proteinaceous materials in the lumens (Hook 1980). We found elevated ($P <$

0.05) tissue weights for cormorant livers, waterfowl kidneys, and moorhen kidneys from settling ponds. However, it is not clear that these differences in tissue weights were caused by trace element contamination. Other bird studies have shown no meaningful relationships between increased dietary metal concentrations and soft tissue masses (Cain and Pafford 1981, Haseltine and Sileo 1983) or between tissue weights and habitat conditions (Anderson 1969). Factors other than contamination may influence soft tissue weights. Sex and age differences can affect soft tissue weights, but our samples were homogeneous with respect to these demographic parameters ($P > 0.05$). Tissue weights can vary with time of year; Korschgen (1977) found eider livers were lightest after the breeding season and heaviest following a period of hyperphagia and prior to egg-laying. Tissue weights also can vary with parasite loads; Riddle (1947) found doves infested with intestinal parasites (*Ascaridia* sp.) had slightly heavier livers than non-infested birds.

No unhealthy birds were observed during the course of the study, and no evidence of pathological changes in tissues was detected. However, mortality due to chronic or acute toxicity problems is hard to detect in wild populations. Sick and moribund birds are usually eliminated by predators before a terminal condition is reached. Also, sick birds seek concealment in dense cover and bird carcasses quickly decay and vanish in the wild (Bellrose 1976).

Summary

Radiation. -- Greater radium-226 levels in substrates from settling areas than from control areas were reflected in elevated concentrations in bones for all 4 species studied. Diet/substrate ratios of radium-226 were less than 1 for all species on all areas. Bone/substrate ratios differed

between settling areas and control areas. Bone concentrations of radium-226 were appreciably greater in waterfowl than common moorhens, possibly due to direct ingestion of contaminated substrates by waterfowl.

Radium-226 levels in soft tissues were consistently less than those in bone. Average concentrations in duck muscle appeared to be greater in settling than control areas. However, the levels were low, variability was great, and the differences were not significant ($P > 0.05$). Concentrations of radium-226 in waterfowl meat from settling ponds do not represent an increased threat to humans relative to waterfowl meat from natural wetlands in Florida. Concentrations in meat from all areas would pose no threat to humans when evaluated in terms of the radium-226 intake limit recommended by the Standards Committee of the Florida Phosphate-related Radiation Task Force (1984).

Radiation standards for birds have not been published. It does not appear that observed radium-226 levels in bones would constitute a health hazard to birds on settling ponds, given their short life spans. However, the average bone concentration in waterfowl from settling ponds in central Florida was about 4 times the recommended maximum for humans.

Trace Elements. -- Several trace elements were found at elevated levels in substrates and bird tissues from settling areas relative to levels in substrates and tissues from control areas. Only Al and Se appeared in bird tissues from settling areas at concentrations greater than those reported as normal for other animals, and only Al was found in potentially harmful levels in food items from settling areas. Aluminum was found at greater concentrations in 2 items in moorhen diets than concentrations that have been shown to reduce growth in chickens. Selenium, but not Al, exhibited increased concentrations at higher trophic levels. Specific effects on birds

of the Se and Al concentrations found are hard to predict due to the complexity of factors affecting toxicity and the difficulty of detecting toxic effects in the wild. Aluminum and Se levels in duck muscle from settling areas would apparently pose no threats to the health of humans consuming these birds.

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Appendix A. Major food items and percent contribution (volumetric) to total diet of 4 bird species from 4 study areas in Florida, June - September 1981 and 1982.

	Central Region				Northern Region			
	Control		Settling		Control		Settling	
	Item	%	Item	%	Item	%	Item	%
Double-crested cormorant	Gizzard shad (*Dorosoma* spp.)	36	Mosquitofish (*Gambusia affinis*)	33	Gizzard shad (*Dorosoma* spp.)	40	Gizzard shad (*Dorosoma* spp.)	44
	Sunfish (*Lepomis* spp.)	21	Bullhead (*Ictalurus* sp.)	16	Bullhead (*Ictalurus* sp.)	19	Bullhead (*Ictalurus* sp.)	28
	Black crappie (*Pomoxis nigromaculatus*)	7	Sunfish (*Lepomis* spp.)	12	Black crappie (*Pomoxis nigromaculatus*)	10	Mosquitofish (*Gambusia affinis*)	22
	Other	36	Other	39	Other	31	Other	6
Waterfowl[a]	Planorbid snail (*Planorbella scalare*)	28	Smartweed (*Polygonum* sp.)	27	Watershield (*Brasenia schreberi*)	39	Cranefly larvae (*Tipula* sp.)	38
	Wildcelery (*Vallisneria americana*)	13	Water scavenger beetle (Hydrophilidae)	12	Spatterdock (*Nuphar luteum*)	24	Spiders (Aranae)	23
	Apple snail (*Pomacea* sp.)	9	Wax Myrtle (*Myrica cerifera*)	11	Sawgrass (*Cladium jamaicensis*)	16	Soldier fly larvae (Stratiomyiidae)	17
	Other	50	Other	50	Other	21	Other	22
Common moorhen	Planorbid snail (*Planorbella scalare*)	27	Green algae (*Oedogonium* sp.)	20	Hydrilla (*Hydrilla* sp.)	83	Bahia grass (*Paspalum notatum*)	37
	Wildcelery (*Vallisneria americana*)	21	Duckweed (*Lemna* sp.)	12	Planorbid snail (*Planorbella* sp.)	5	Bladderwort (*Utricularia* sp.)	15
	Watergrass (*Hydrochloa* sp.)	15	Bryozoa (*Plumatella* sp.)	11	Coontail (*Ceratophylum demersum*)	4	Maidencane (*Panicum* sp.)	11
	Other	37	Other	57	Other	8	Other	37

[a]Mottled ducks, central region; wood ducks, northern region.

Appendix B. Radiation dose to humans.

Calculation of radiation dose to humans through the consumption of duck meat requires a value for the annual intake of this food item. Such data are not readily available. In reviewing the literature, Stabin (1983) reported estimates ranging from 0.1 to 220 kg/year. He suggested a dose assessment based on a value of 10 kg/year.

In the mail survey conducted during this study, the cumulative response rate was 60.4%, but unexpectedly, many respondents were nonhunters. Fifteen of the 67 responses came from waterfowl hunters, with the remaining coming from club members using IMC land for fishing, upland game hunting, or for unspecified purposes. Based on survey responses, 15 hunters spent 114 hunter-days afield with 94 hunter-days being spent on phosphate-mine hunts. A total of 608 waterfowl were shot during the season with 1 individual claiming to have contributed approximately half of the total harvest. It was assumed that mottled ducks weighed 950 g and that edible tissue was 25% of body weight (Halford et al. 1981). ,By subtracting the proportion given away and adjusting for the proportion shared at meal time, the average hunter was assumed to consume 1-2 kg/year and the maximum intake was estimated as 8-10 kg/year.

The concentrations of radium-226 in duck muscle were converted from a dry weight basis to a fresh weight basis (Appendix B-1). Annual intakes and corresponding effective committed dose equivalents were calculated for the average (1.5 kg/yr) and the maximum (10 kg/yr) intakes. For the average individual, effective dose equivalents attributable to radium-226 in duck meat range from 0.001 mrem/year for eating ducks from north Florida control areas to 0.01 mrem/year for eating ducks from central Florida settling areas.

Appendix B-1. Radiation doses to individual humans attributable to Radium-226 based on average and maximum consumption of waterfowl meat from 4 study areas in Florida.

Location	Radium-226 Concentration pCi/kg		Average (1.5 kg/yr)		Maximum (10 kg/yr)	
	Dry wt Basis	Fresh wt Basis	Intake pCi/yr	Dose[a] mrem	Intake pCi/yr	Dose mrem
Central Region						
Settling	19±5	6.1	9.2	0.010	61	0.067
Control	13±5	4.7	7.1	0.008	47	0.052
Difference		1.4	2.1	0.002	14	0.015
Northern Region						
Settling	9±10	2.2	3.3	0.004	22	0.024
Control	2±5	0.6	0.9	0.001	6	0.007
Difference		1.6	2.4	0.003	16	0.017

[a]"Dose" is the effective committed dose equivalent resulting from a 1-year intake at the average concentration reported. Based on a conversion factor of 1.1×10^{-3} mrem/pCi ingested (International Commission on Radiological Protection 1979).

Appendix C-1. Substrate. Trace element composition ($\bar{X}$ppm ± SE) of substrates from 4 study areas in Florida, June - September, 1981 and 1982.

Element	Central Region		Northern Region	
	Control (N = 12)[a]	Settling (N = 12)[b]	Control (N = 12)[c]	Settling (N = 12)[d]
Al	300±110A[e]	3600±640B	1620±980A	3600±960B
As	1.0±0.1	4.9±2.6	2.5±0.9	1.3±0.5
Br	2.8±1.5	1.4±0.2	39±9A	1.0±0.2B
Cr	2.7±0.1A	97±17B	5.9±1.4A	42±15B
Cu	1.0±0.3A	8.5±1.6B	5.6±1.0	10±4
Hg	3.1±0.7	17±4	3.1±0.7	8.6±3.4
Mn	5.4±1.3A	83±14B	48±6	70±17
Mo	3.2±0.5A	10±2B	4.7±0.5A	7.0±1.3B
Ni	0.7±0.2A	18±4B	5.6±1.0A	11±4B
Pb	8.2±1.9A	31±8B	30±6	30±7
Rb	1.0±0.1A	43±10B	10±3	34±9
Se	0.5±0.1A	1.6±0.5B	1.4±0.2	1.0±0.1
Ti	640±120A	1940±200B	380±70A	1070±270B
V	0.8±0.3A	26±5B	3.0±0.6	19±4
Y	2.5±0.5A	61±10B	20±11A	74±19B
Zn	3.3±1.1A	56±12B	25±5	35±10
Zr	190±70A	300±110B	48±29A	160±50B

[a] N = 11 for Al, Mg, Na, and V.

[b] N = 15 for Al, Mg, Na, and V.

[c] N = 9 for Al, Mg, Na, and V.

[d] N = 10 for Al, Mg, Na, and V.

[e] Different letters indicate significant ($P<0.05$) differences within a region.

Appendix C-2. Mineral composition ($\bar{X}$ mg/l ± SE) of water samples collected from settling areas and control areas in Florida, June - September, 1981 and 1982.

	Central Region		Northern Region	
Element	Control (N=9)	Settling (N=9)	Control (N=8)	Settling (N=12)
Al	1.1 ± 1.1	N.D.[a]	1.6 ± 1.6	1.4 ± 1.4
Br	17.4 ± 6.7	11.5 ± 3.5	10.1 ± 1.7	12.9 ± 2.2
cu	3.0 ± 0.9	2.3 ± 0.9	1.6 ± 0.3	2.3 ± 0.4
Mn	0.13 ± 0.03	0.23 ± 0.14	0.11 ± 0.01	0.18 ± 0.03
V	0.04 ± 0.01	0.04 ± 0.02	0.03 ± 0.01	0.06 ± 0.02

[a]N.D. = not detected,

Appendix C-3. Common moorhen livers. Trace elements ($\bar{X}$ ppm ± SE) in common moorhen livers from 4 study areas in Florida, June-September, 1981 and 1982.

	Central Region		Northern Region	
Element	Control (N = 17)[a]	Settling (N = 17)[b]	Control (N = 19)[c]	Settling (N = 18)[b]
Al	18.2±3.8A[d]	33.0±9.3B	13.1±1.7A	25.1±6.3B
Br	18.6±1.5A	26.4±1.6B	12.5±1.5A	13.7±0.8B
Cu	16.7±2.5A	22.8±2.4B	14.1±2.2A	23.0±4.3B
Mn	8.3±1.0A	4.6±0.8B	5.1±0.7	4.3±0.4
Rb	23.9±2.6A	70.6±6.8B	74.7±6.2A	48.5±4.2B
Se	3.4±0.7A	6.4±0.6B	2.3±0.3A	4.0±0.4B
V	0.13±0.03A	0.43±0.13B	0.10±0.01A	0.61±0.13B
Zn	133±11	146±12	127±7	104±5

[a]N = 15 for Al and V.

[b]N = 13 for Al and V.

[c]N = 16 for Al and V.

[d]Different letters indicate differences ($P < 0.05$) within a region.

Appendix C-4. Common moorhen kidneys. Trace elements ($\bar{X}$ ppm ± SE) in common moorhen kidneys from 4 study areas in Florida, June - September, 1981 and 1982.

	Central Region		Northern Region	
Element	Control (N = 11)[a]	Settling (N = 11)[b]	Control (N = 14)[c]	Settling (N = 11)[b]
Al	15.1±5.3	15.7±3.2	13.5±4.1	20.3±4.0
Br	22.3±2.4A[d]	46.8±21.8B	15.8±1.2A	18.6±1.1B
Cu	15.9±1.5A	26.0±10.2B	14.3±0.8A	18.2±1.3B
Mn	11.2±1.9	11.7±3.6	7.6±0.5	5.8±0.6
Rb	22.3±2.9A	86.5±26.1B	66.0±6.9A	36.6±2.8B
Se	5.7±0.7A	12.4±2.4B	4.8±0.4A	7.1±0.4B
V	0.22±0.06	0.33±0.11	0.20±0.06	0.30±0.12
Zn	89±8	137±43	84±4	92±4

[a] N = 7 for Al and V.

[b] N = 10 for Al and V.

[c] N = 8 for Al and V.

[d] Different letters indicate differences ($P<0.05$) within a region.

Appendix C-5. Common moorhen bones. Trace elements ($\bar{X}$ppm ± SE) in common moorhen bones from 4 study areas in Florida, June - September, 1981 and 1982.

Element	Central Region		Northern Region	
	Control (N = 17)	Settling (N = 18)[a]	Control (N = 20)[b]	Settling (N = 17)
Al	189±41	158±14	142±3	155±27
Br	13.8±1.2A[c]	18.7±2.5B	8.1±0.9A	12.1±1.1B
Cu	8.8±1.0	10.1±1.9	5.7±0.5A	14.1±1.5B
Mn	7.6±0.9	5.9±0.4	5.5±0.4	5.3±0.6
Pb	29.4±24.2	12.6±6.8	17.9±4.3	35.1±22.9
Rb	12.5±1.5	36.0±6.7	31.1±3.6	29.1±3.2
V	0.28±0.03A	0.74±0.27B	0.24±0.02A	0.47±0.08B
Zn	291±12A	302±15B	289±10A	335±15B

[a] N = 17 for Al and V.

[b] N = 18 for Al and V.

[c] Different letters indicate differences ($P<0.05$) within a region.

Appendix C-6. Double-crested cormorant livers. Trace elements ($\bar{X}$ppm ± SE) in double-crested cormorant livers from 4 study areas in Florida, June - September, 1981 and 1982.

Element	Central Region		Northern Region	
	Control (N = 19)	Settling (N = 17)[a]	Control (N = 17)[b]	Settling (N = 20)[c]
Al	11.2±1.2	27.3±17.9	16.7±6.9	33.2±8.5
Br	46.0±3.4	44.3±3.2	38.1±3.3	38.0±3.6
Cu	19.8±1.0	20.5±2.3	14.4±1.2	16.0±0.8
Mn	16.5±0.5A[d]	11.6±1.0B	10.0±0.9	10.1±0.6
Rb	31.1±3.5A	54.1±2.9B	41.4±1.9	44.7±2.8
Se	7.4±1.1A	19.1±5.3B	7.6±2.3A	22.2±4.1B
V	0.12±0.02A	0.10±0.02B	0.25±0.11A	0.09±0.01B
Zn	106±4A	121±6B	96±8A	103±6B

[a] N = 8 for Al and V.

[b] N = 15 for Al and V.

[c] N = 17 for Al and V.

[d] Different letters indicate differences ($P<0.05$) within a region.

Appendix C-7. Double-crested cormorant kidneys. Trace elements ($\bar{X}$ppm ± SE) in double-crested cormorant kidneys collected from 4 study areas in Florida, June-September, 1981 and 1982.

	Central Region		Northern Region	
Element	Control (N = 19)[a]	Settling (N = 18)[b]	Control (N = 17)[c]	Settling (N = 20)[d]
Al	52±38	9±2	10±5	10±2
Br	57±5	57±2	54±4A[e]	40±2B
Cu	13.5±0.4	13.1±0.4	10.8±0.7	11.8±0.6
Mn	8.4±0.4	7.4±0.5	6.5±0.5	6.1±0.5
Rb	27.8±1.2A	62.5±2.9B	47.6±2.4	54.8±2.4B
Se	6.9±0.6A	13.7±2.6B	5.8±0.8A	11.8±1.5B
V	0.35±0.09	0.22±0.06	0.23±0.07	0.15±0.03
Zn	88±2	91±3	78±3	85±3.5

[a] N = 17 for Al and V.

[b] N = 12 for Al and V.

[c] N = 10 for Al and V.

[d] N = 14 for Al and V.

[e] Different letters indicate differences ($P < 0.05$) within a region.

Appendix C-8. Double-crested cormorant bones. Trace elements ($\bar{X}$ppm ± SE) in double-crested cormorant bones from 4 study areas in Florida, June - September, 1981 and 1982.

	Central Region		Northern Region	
Element	Control (N = 19)[a]	Settling (N = 17)[b]	Control (N = 18)[a]	Settling (N = 19)
Al	136±5A[c]	160±15B	130±9A	155±8B
Br	16.9±1.5	21.9±2.5	23.5±2.6A	14.7±1.4B
Cu	4.1±0.5	5.8±0.8	5.0±0.5	4.3±0.4
Mn	5.2±0.5	6.8±1.0	5.7±0.5	5.4±0.4
Pb	2.8±0.7	5.3±2.0	3.3±1.2	1.9±0.5
Rb	12.2±1.7A	22.4±3.5B	21.1±3.0	18.4±2.6
V	0.33±0.06	0.37±0.09	0.36±0.07	0.22±0.04
Zn	204±14	223±26	223±19	224±16

[a]N = 16 for Al and V.

[b]N = 18 for Al and V.

[c]Different letters indicate differences ($P<0.05$) within a region.

Appendix C-9. Waterfowl livers. Trace elements ($\bar{X}$ppm ± SE) in waterfowl livers from 4 study areas in Florida, June-September, 1981 and 1982.

Element	Central Region[a]		Northern Region[b]	
	Control (N = 13)[c]	Settling (N = 17)[d]	Control (N = 5)	Settling (N = 5)
Al	11.1±2.9	25.7±9.6	49.5±40.1	17.1±1.8
As	0.8±0	1.4±0.6	0.8±0	0.8±0
Br	19.2±1.8	24.5±2.2	5.5±0.9	6.3±0.8
Co	5.3±0.9	6.5±1.1	4.3±1.1	6.4±2.5
Cu	81.2±16.1	84.3±25.7	14.1±2.4A	59.0±25.0B
Mn	7.4±0.9A[e]	4.7±0.6B	6.2±0.9A	13.5±2.8B
Mo	3.1±0.5	3.2±0.5	3.1±0.8	3.2±0.7
Ni	0.08±0.04	2.1±1.5	0.4±0.1	0.6±0.1
Pb	20.1±8.0	7.6±2.0	6.4±2.4	4.6±1.8
Rb	16.3±2.2A	30.8±2.1B	44.7±4.1	28.0±5.5
Se	3.2±0.5A	9.9±1.4B	0.8±0.3A	6.3±0.8B
V	0.09±0.02A	0.45±0.11B	0.33±0.23A	0.36±0.08B
Zn	132±8	155±13	89±5A	163±20B

[a] Mottled ducks.

[b] Wood ducks.

[c] N = 8 for Al and V.

[d] N = 14 for Al and V.

[e] Different letters indicate differences ($P<0.05$) within a region.

Appendix C-10. Waterfowl kidneys. Trace elements ($\bar{X}$ppm ± SE) in waterfowl kidneys from 4 study areas in Florida, June-September, 1981 and 1982.

	Central Region[a]		Northern Region[b]	
Element	Control (N = 14)[c]	Settling (N = 17)[d]	Control (N = 5)[e]	Settling (N = 5)[e]
Al	8.4±1.6	39.4±12.1	12.1±2.8	14.5±2.2
Br	32.9±2.6	33±3.0	12.2±1.7	15.1±2.7
Cu	23.5±4.1A[f]	16.4±1.4B	13.8±0.6	35.0±4.0
Mn	8.0±0.9	6.0±0.5	9.4±0.7	11.4±1.9
Mo	2.0±0.3A	3.2±0.4B	1.6±0.8A	3.1±0.5B
Pb	23.0±9.0A	2.8±1.6B	2.3±1.8	0.9±0.1
Rb	17.4±3.0A	34±3.3B	59.9±7.1A	25.4±5.2B
Se	4.6±0.4A	8.2±1.1B	2.0±0.6A	7.5±0.9B
V	0.08±0.002A	0.58±0.08B	0.12±0.03A	1.3±0.6B
Zn	96±4	82.9±5.6	101±2.3	113±5.2

[a]Mottled ducks.

[b]Wood ducks.

[c]N = 8 for Al and V.

[d]N = 9 for Al and V.

[e]N = 3 for Al and V.

[f]Different letters indicate differences ($P<0.05$) within a region.

Appendix C-11. Waterfowl bones. Trace elements ($\bar{X}$ppm ± SE) in waterfowl bones from 4 study areas in Florida, June-September, 1981 and 1982.

Element	Central Region[a]		Northern Region[b]	
	Control (N = 14)[c]	Settling (N = 17)[d]	Control (N = 5)	Settling (N = 5)
Al	152±6	178±9	152±9	147±6
Br	14.8±2.1	14.0±1.3	12.1±2.3	9.3±1.0
Cu	6.0±0.9	6.0±0.9	0.7±0.2A[e]	8.5±0.2B
Mn	8.1±0.8	5.9±0.6	10.9±4.6	26.5±9.5
Pb	220±116	38±15	52±21	30±13
Rb	12.4±2.7A	17.8±1.6B	25±4.8A	8.0±1.3B
V	0.32±0.04A	1.09±0.19B	0.27±0.06A	0.62±0.24B
Zn	347±10	368±16	307±14	322±17

[a]Mottled ducks.

[b]Wood ducks.

[c]N = 13 for Al and V.

[d]N = 16 for Al and V.

[e]Different letters indicate differences ($P<0.05$) within a region.

Appendix C-12. Waterfowl muscles. Trace elements ($\bar{X}$ppm ± SE) in waterfowl muscles from 4 study areas in Florida, June-September, 1981 and 1982.

Element	Central Region[a]		Northern Region[b]	
	Control (N = 14)[c]	Settling (N = 17)[d]	Control (N = 5)[e]	Settling (N = 5)
Al	10.6±1.9A[f]	37.6±8.0B	16.9±5.0	15.9±2.4
As	0.5±0.1	N. D.[g]	N. D.	N. D.
Br	13.9±1.7	13.1±1.3	4.3±0.4	5.9±0.7
Co	0.8±0.1	0.8±0.1	0.7±0.2	0.8±0.1
Cu	14.7±1.0	17.8±2.0	14.4±1.1	16.2±1.7
Mn	1.9±0.3	1.4±0.2	1.1±0.2	1.6±0.3
Ni	0.6±0.1	0.7±0	0.6±0.1	0.6±0.1
Pb	1.1±0.2	0.9±0.1	1.0±0.1	0.9±0.0
Rb	19.7±2.9	28.9±3.3	60.0±7.6A	18.9±2.1B
Se	3.4±1.2A	3.6±0.7B	1.1±0.1A	2.9±0.5B
V	0.07±0.03	0.14±0.03	0.09±0.02	0.09±0.02
Zn	45±2	41±3	31±2	31±1

[a]Mottled ducks.

[b]Wood ducks.

[c]N = 12 for Al and V.

[d]N = 15 for Al and V.

[e]N = 4 for Al and V.

[f]Different letters indicate differences ($P < 0.05$) within a region.

[g]N. D. = not detected.

Appendix D. Average dry weight/wet weight ratios [$\bar{X}$±SE(N)] for soft tissues of birds collected from 4 study areas in Florida, June - September, 1981 and 1982.

	Central Region		Northern Region	
	Control	Settling	Control	Settling
Double-crested cormorant				
Kidney	0.23±0.2 (19)	0.22±.01 (20)	0.22±.00 (20)	0.24±.01 (20)
Liver	0.28±0.00 (17)	0.26±0.01 (20)	0.29±0.01 (20)	0.26±0.01 (19)
Waterfowl[a]				
Kidney	0.21±0.01 (17)	0.24±0.01 (20)	0.37±0.13 (5)	0.32±0.12 (5)
Liver	0.27±0.02 (16)	0.28±0.01 (20)	0.25±0.02 (5)	0.26±0.01 (5)
Muscle	0.36±0.02 (17)	0.32±0.01 (20)	0.28±0.01 (5)	0.25±0.00 (5)
Common moorhen				
Kidney	0.27±0.02 (17)	0.27±0.03 (20)	0.22±0.01 (20)	0.26±0.04 (19)
Liver	0.27±0.03 (20)	0.36±0.05 (20)	0.28±0.01 (19)	0.25±0.01 (20)

[a]Mottled ducks, central region; wood ducks, northern region.

Appendix E-1. Common moorhen diets (wet). Trace elements ($\bar{X}$ ppm ± SE) in diets of common moorhens from 4 study areas in Florida, June - September, 1981 and 1982.

Element	Central Region		Northern Region	
	Control (N=6)[a]	Settling (N=7)[b]	Control (N=6)	Settling (N=4)[c]
Al	14.7±2.8[d]	13.90±340	18.6±4.6	135±37
Br	1.5±0.2	3.8±0.2	1.4±0.1	0.90±0.28
Ca	30,500±10,100	1,080±140	8,650±6,830	247±18
Cu	5.7±0.11	0.88±0.11	0.42±0.16	5.7±1.0
Fe	87.8±18.9	262±30	130±20	23.4±1.3
Mg	3,960±1,590	796±191	113±6	654±66
Mn	43.5±7.7	29.8±3.4	23.0±2.4	17.2±0.7
Ni	0.21±0.05	1.0±0.2	0.10±0.08	0.51±0.12
Pb	12.8±4.3	5.4±1.3	0.02±0.01	31.3±11.0
Rb	0.9±0.2	6.4±0.8	2.1±0.3	8.5±0.2
Se	0.67±0.22	0.03±0.01	0.004±0.005	0.20±0.06
Sr	176±37	66.1±12.2	16.1±8.4	0.21±0.07
V	0.06±0.03	4.2±1.0	0.19±01	0.62±0.18
Zn	3.9±0.4	3.5±0.5	1.8±0.5	19.0±0.4

[a] N = 5 for Al and V.

[b] N = 4 for Al and V.

[c] N = 6 for Al, Br, Cu, Mn, and V.

[d] Means are weighted averages of the top 3 food items in the diet based on aggregate percentages.

Appendix E-2. Common moorhen diets (dry). Trace elements ($\bar{X}$ ppm ± SE) in diets of common moorhens from 4 study areas in Florida, June - September, 1981 and 1982.

Element	Central Region		Northern Region	
	Control (N=6[a])	Settling (N=7[b])	Control (N=6)	Settling (N=4[c])
Al	75.6 ± 28.5[d]	15,800 ± 3,700	255 ± 20	3,350 ± 1,250
Br	11.2 ± 1.8	37.4 ± 2.3	24.0 ± 1.4	1.3 ± 0.4
Ca	52,800 ± 16,700	10,700 ± 1,600	22,000 ± 16,200	342 ± 19
Cu	3.0 ± 0.4	6.8 ± 0.6	4.0 ± 0.6	7.9 ± 1.3
Fe	983 ± 290	2,620 ± 350	2,120 ± 90	32.7 ± 1.8
Mg	7,590 ± 2,560	8,740 ± 2,070	1,560 ± 300	2,550 ± 470
Mn	153 ± 28	246 ± 17	408 ± 54	24.3 ± 1.2
Ni	0.76 ± 0.11	7.9 ± 0.6	0.24 ± 0.20	0.75 ± 0.18
Pb	24.9 ± 7.6	30.1 ± 6.5	1.1 ± 0.1	41.2 ± 14.5
Rb	6.9 ± 1.7	50.5 ± 3.7	30.8 ± 3.5	11.9 ± 0.4
Se	1.2 ± 0.4	0.37 ± 0.11	0.09 ± 0.10	0.28 ± 0.09
Sr	336 ± 61	435 ± 56	115 ± 17	0.3 ± 0.1
V	0.65 ± 0.22	48.1 ± 10.6	2.8 ± 0.1	16.0 ± 5.8
Zn	34.0 ± 5.8	27.2 ± 1.6	24.1 ± 1.8	26.5 ± 0.4

[a] N = 5 for Al and V.

[b] N = 4 for Al and V.

[c] N = 6 for Al, Br, Cu, Mn, and V.

[d] Means are weighted averages of the top 3 food items in the diet based on aggregate percentages.

Appendix E-3. Mottled duck diets (wet). Trace elements ($\bar{X}$ ppm ± SE) in diets of mottled ducks from 2 study areas in central Florida, June - September, 1981.

Element	Control (N=5)	Settling (N=6)[a]
Al	27.2±8.4[b]	28.1±6.1
Br	1.3±0.3	3.7±0.6
Ca	58,700±16,100	1,580±220
Cu	1.17±0.40	5.7±0.9
Fe	47.7±14.4	32.6±1.9
Mg	2,960±1,400	798±81
Mn	31.7±5.2	13.0±0.7
Ni	0.3±0.1	0.75±0.07
Pb	8.9±3.3	0.56±0.05
Rb	0.86±0.24	19.4±2.1
Se	0.50±0.18	0.77±0.27
Sr	189±27	3.4±0.4
V	0.16±0.03	0.15±0.02
Zn	2.2±0.6	25.9±3.9

[a]N = 5 for Al and V.

[b]Means are weighted averages of the top 3 food items in the diet based on aggregate percentages.

Appendix E-4. Mottled duck diets (dry). Trace elements ($\bar{X}$ ppm ± SE) in diets of mottled ducks from 2 study areas in central Florida, June - September, 1981.

Element	Control (N=5)	Settling (N=6)[a]
Al	66.5±16.2[b]	103±17
Br	6.0±0.9	11.0±2.2
Ca	140,000±35,000	4,250±630
Cu	3.6±0.8	17.9±3.4
Fe	150±26	93.1±6.8
Mg	9,110±2,790	2,100±140
Mn	102±15	37.2±2.5
Ni	0.92±0.15	2.1±0.2
Pb	28.7±0.4	1.7±0.2
Rb	2.6±0.4	50.0±5.3
Se	1.4±0.5	2.2±0.7
Sr	534±68	9.5±1.2
V	0.43±0.06	0.40±0.06
Zn	18.7±4.7	81.4±14.8

[a]N = 5 for Al and V.

[b]Means are weighted averages of the top 3 food items in the diet based on aggregate percentages.

Appendix F. Body and soft tissue weights [$\bar{X}$ g ± SE(N)] from birds collected on 4 study areas in Florida, June - September, 1981 and 1982.

Species	Sample	Central Region		Northern Region	
		Control	Settling	Control	Settling
Double-crested cormorant	Body	1752±44(20)	1599±45(20)	1688±39(20)	1688±47(20)
	Kidney	13.0±0.4(19)	12.6±0.5(20)	12.8±0.4(18)	13.1±0.6(20)
	Liver	45.6±1.9(20)A[a]	54.5±2.3(17)B	47.6±2.0(19)A	51.9±2.1(20)B
Wood duck	Body			650±17(5)	594±38(5)
	Kidney			3.1±0.2(5)A	5.5±0.4(5)B
	Liver			14.0±1.8(5)	14.9±1.7(5)
Mottled duck	Body	859±26(17)A	947±26(20)B		
	Kidney	4.8±0.2(16)A	6.8±0.4(19)B		
	Liver	18.6±1.1(16)	23.9±1.4(18)		
Common moorhen	Body	393±11(20)	396±13(20)	413±11(20)	410±10(20)
	Kidney	2.2±0.1(17)A	2.8±0.7(19)B	2.5±0.1(20)A	2.7±0.2(19)B
	Liver	8.5±0.6(19)	8.2±0.8(17)	9.0±0.4(20)	10.4±0.4(20)

[a]Different levels indicate differences ($P < 0.05$) within a region based on analyses of covariance with body weight as a covariable.

Appendix G. Radium-226 in food items.

Sampling of bird food items was designed to represent major dietary components for individual species on the study areas and not to provide a comparison of the various individual food sources. However, these data did provide some information on the radium-226 content of biota other than the 3 bird groups. Because of differing dietary compositions for the various species and locations, few direct comparisons were possible but the data could be examined for patterns. The most striking observation was that highest concentrations in plants were observed in free-floating species such as algae, bladderwort, duckweed, and hydrilla (Appendix G-1). [Although technically rooted aquatics, bladderwort and hydrilla often exist as free-floating mats (Tarver et al. 1978)]. Even in the natural system, hydrilla had nearly 1000 pCi/kg. Also of note was the fact that lowest concentrations were observed in seeds of plants collected on dry land [e.g. bahiagrass (Paspalum notatum), 1 pCi/kg]. However, not all terrestrial plant seeds had concentrations this low.

The radium-226 concentrations observed in various animal species are presented in Appendix G-2. Again, only a limited number of direct comparisons were possible. In the central region, concentrations in whole sunfish (Lepomis spp.) from settling areas were 5 times those from the control areas. It should be noted that the fish were analyzed whole. Since the radium would be expected to concentrate in the bone, concentrations in portions consumed by humans (i.e. muscle) would likely be considerably lower than for whole fish.

Concentration ratios for settling areas are summarized in Appendix G-3. These ratios should be considered as tentative because they are based on so few samples. The highest concentration ratio from water was observed for free-floating plants, 1.7 x 10^4 l/kg (dry weight basis). The concentration ratio for rooted aquatic plants was 10^3 l/kg relative to the water and 0.02 kg/kg relative to the substrate. It was not

possible to separate the relative contributions of root uptake from the substrate and direct uptake from the water. Seeds of smartweed (Polygonum sp.) and wax myrtle (Myrica cerifera) showed a concentration ratio of 0.02 relative to the substrate, This was consistent with the literature which indicated that radium is not concentrated from the soil to plants. Most of the terrestrial plants were actually growing on overburden or sand tailings dikes of lower radium concentration than the clays in pond bottoms. A limited number of insect and fish samples suggested that tissue/water concentration ratios on the order of 100-300 l/kg can be expected in these environments.

Appendix G-1. Concentrations (pCi/kg dry weight) of radium-226 in selected plant groups from 4 study areas in Florida, June - September, 1981 and 1982.

Sample Type	Central Region		Northern Region	
	Control	Settling	Control	Settling
Free-floating				
Algae, green[b]		9078 ± 569[a]		
Bladderwort				4210 ± 359
Coontail			350 ± 23	
Duckweed		9020 ± 418		
Hydrilla		5510 ± 292	986 ± 87	
Rooted aquatic				
Bullrush seed		412 ± 23		241 ± 20
Maidencane seed				64 ± 6
Sawgrass seed			101 ± 10	
Spatterdock			24 ± 9	
Watergrass	226 ± 41			
Watershield			0 ± 7.0	
Wild celery seed pods	106 ± 14			
Terrestrial				
Bahiagrass seed				1 ± 3
Smartweed seed		453 ± 31		
Wax myrtle seed		479 ± 27		

[a] Results are for a single composite of 2 samples.

± values represent the estimated 1 standard deviation uncertainty of the analytical determination.

[b] Scientific names are in Appendix A.

Appendix G-2. Concentrations (pCi/kg ± SD dry weight) of radium-226 in selected animal species from 4 study areas in Florida, June - September, 1981 and 1982.

Sample Type	Central Region		Northern Region	
	Control	Settling	Control	Settling
Bryozoa				
Plumatella sp.		1664±83[a]		
Snails				
Apple snail[b]	224±22			
Planorbid snail	128±4[c]		52±16	
Insects and Larvae				
Cranefly larvae				1590±139
Spiders				610±117
Water scavenger beetle		62±40		
Fish				
Sunfish	28±6	141±9		
Gizzard shad			48±10	
Mosquito fish		425±38		

[a] Samples represented the whole animal. Results are for a single composite of 2 samples unless noted otherwise. ± values represent the estimated 1 standard deviation uncertainty of the analytical determination.

[b] Scientific names are in Appendix A.

[c] Results for planorbid snail are average of individual analyses of 2 samples; ± values represent standard error of mean.

Appendix G-3. Concentrations ratios (dry weight basis) of radium-226 in substrate, water, and selected plant and animal groups from settling areas in north and central Florida, June - September, 1981 and 1982.

Medium	Medium/Substrate	Medium/Water(Dissolved)	Medium/Water(Total)
Water, dissolved activity	2.0×10^{-5} kg/l[a]		
Water, total activity	5.0×10^{-5}		
Plants			
Free-floating	0.3 kg/kg[b]	17,000 l/kg[c]	7,200 l/kg
Rooted aquatic	0.02	1,000	550
Terrestrial, seeds	0.02		
Insects and Larvae	0.003	120	34
Fish	0.006	270	80
Birds			
Duck muscle	0.0007	37	15
Duck bone[d]	0.2	8,200	3,400
Moorhen bone	0.02	1,200	520
Cormorant bone	0.02	920	400

[a] pCi l^{-1} water/pCi kg^{-1} substrate = kg/l
[b] pCi kg^{-1} tissue/pCi kg^{-1} substrate = kg/kg
[c] pCi kg^{-1} tissue/pCi l^{-1} water = l/kg
[d] Bird bone is expressed on an ash weight basis.