CESIUM-137 AND STABLE CESIUM IN A HYPEREUTROPHIC LAKE

Thesis for the Degree of Ph. D.
MICHIGAN STATE UNIVERSITY
L. DEAN EYMAN
1972

This is to certify that the

thesis entitled

CESIUM-137 AND STABLE CESIUM
IN A HYPEREUTROPHIC LAKE

presented by

L. DEAN EYMAN

has been accepted towards fulfillment
of the requirements for

Ph.D. degree in Fisheries & Wildlife

Niles R. Kevern
Major professor

Date July 10, 1972

O-7639

ABSTRACT

CESIUM-137 AND STABLE CESIUM IN A HYPEREUTROPHIC LAKE

By

L. Dean Eyman

The inputs of ^{137}Cs and stable Cs and their distribution among the various components of an aquatic ecosystem were studied in a lake exhibiting an advanced stage of eutrophy. Components sampled and analyzed for these two isotopes of cesium included water, sediments, macrophytes, filamentous algae, zooplankton, and several species of fish.

Most of the cesium pool (87%-^{137}Cs; 98%-stable Cs) was associated with the sediments. Stable Cs enters the lake primarily in an organically bound state as migratory waterfowl excreta and is deposited to the sediments. Cesium-137 enters as a soluble inorganic form and is distributed throughout the system. Specific activity (pCi ^{137}Cs/ng stable Cs) of the sediments is lower than other components of the system due to the different modes of entry of the two isotopes of cesium.

L. Dean Eyman

A trend of increased ^{137}Cs concentration at higher trophic levels is demonstrated for those fish that are free-ranging limnetic feeders. No such trend is evident for stable Cs. Forms closely associated with sediments have higher ^{137}Cs concentrations than expected based on their feeding habits.

On the dates samples were collected, specific activity in limnetic fishes was constant but was variable in other forms. The degree of association of biotic forms with sediments is reflected in their specific activity.

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IN A HYPEREUTROPHIC LAKE

By

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A THESIS

Submitted to
Michigan State University
in partial fulfillment of the requirements
for the degree of

DOCTOR OF PHILOSOPHY

Department of Fisheries and Wildlife

1972

ACKNOWLEDGMENTS

I wish to thank those individuals whose help was essential to the completion of my research: Mr. W. J. Johnson of the Kellogg Bird Sanctuary for his cooperation and aid in obtaining samples and providing data on waterfowl usage of the lake; fellow graduate students J. Eckenrode, D. Jude, and A. Szluha for help in the field; J. Seelye for discussions and suggestions on analytical methodology; Dr. B. J. Mathis for his review and helpful suggestions in the preparation of the manuscript.

I extend gratitude to my graduate committee: Dr. R. C. Ball, Dr. F. M. D'Itri, Dr. J. L. Gill, and particularly to Dr. N. R. Kevern for stimulating discussions on radioecology as well as his advice and guidance during the period of my graduate studies.

For her patience and constant encouragement during my graduate study and her complete involvement in providing a secure home for our children in my absences, I affectionately extend my gratitude to my wife, Ginny. To son Jeffrey and daughter Angela, constant sources of strength, thanks for being.

This research was supported through a Predoctoral Fellowship 5-F1-WP-26,354-03 from the Water Quality Office of the Environmental Protection Agency to whom I am grateful.

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INTRODUCTION

The distribution and biogeochemical cycling of cesium in natural ecosystems have received increased attention since the advent of the nuclear era with the concomitant release of isotopes from weapons tests and use. Contributions of cesium isotopes from atmospheric fallout have declined since the nuclear test ban treaty of 1964. However, a new and potentially more significant source is developing in connection with operation of nuclear powered electric generating facilities. Although nuclear power accounts for only 1% of the present generating capacity, this share is expected to reach 30% by 1980 (Arnold, 1970). Nelkin (1971) estimates cooling water needs will equal 20% of the total annual supply of runoff in the United States by 1985. Release of radioisotopes in cooling water due to fuel cell leakage and activation products will be incorporated into aquatic communities exclusively.

Movement of cesium and availability to various components differ significantly between terrestrial and aquatic ecosystems. Terrestrial studies have demonstrated a correlation between nuclide distribution and various parameters, such as type of bedrock, soil types, rainfall and snowfall patterns, and vegetation type (Osburn, 1967). Accumulation and magnification of ^{137}Cs levels in biotic components of terrestrial ecosystems have been related or correlated to moisture content of soil (Stewart, 1961; Stewart and Hungate, 1967); levels of precipitation (Rickard, 1967; Krieger, Kahn, and Cummings, 1967; Löw and Edvarson, 1959); latitude (Stewart, Osmund, Crooks, and Fisher, 1957); season (Wellford and Collins, 1960; Parker and Crookall, 1961) and feeding habits (Pendleton *et al*., 1964).

Freshwater ecosystems appear to offer a greater potential for radiocesium buildup in upper trophic level predators. This is due to increased availability to aquatic plants and algae, in addition to the presence of a greater number of intervening predatory levels that result in increased concentrations. Sorption and transfer of ^{137}Cs by various components of an aquatic community were described by Pendleton (1965). Williams and Pickering (1961) demonstrated the source of ^{137}Cs in bluegills was food rather than direct absorption from water. Uptake of ^{137}Cs by algae and aquatic plants was studied by Cline (1967), Williams (1960) and Rickard (1969). Levels in aquatic insects were determined by

Osburn (1969) and Krumholz (1967). Radiocesium levels in fish have been correlated with season (Pendleton, 1959; Krumholz, 1954, 1956), feeding habits (Kolehmainen et al., 1967; Hannerz, 1968) and sediment activity (Gustafson, 1969).

Accumulation and amplification of ^{137}Cs at higher trophic levels has been demonstrated by Nelson (1967), Pendleton (1965), Gustafson (1967), Häsänen (1963) and Kolehmainen (1966). Nelson (1969) tested the relationship between specific activity of water and fish. He found the relationship to hold constant under conditions of continuous isotope release into the system.

From the above discussion it is apparent that a wealth of information is available on ^{137}Cs in aquatic ecosystems. The present study deals with the distribution of ^{137}Cs and ^{133}Cs in an aquatic ecosystem. The feasability of using specific activity as a tool for predicting levels of ^{137}Cs in aquatic plants of potential commercial value as well as fish important in sports fisheries is investigated.

STUDY SITE

Wintergreen Lake is located in Kalamazoo County, southwestern Michigan. It has been managed as a waterfowl refuge since 1929 and is presently part of the W. K. Kellogg Biological Station, administered by Michigan State University.

The lake may be classified as temperate, partially meromictic. It is typical of the many moranic basin and pit lakes located in that region of Michigan. Surface area is 15.9 hectares. Mean depth is 3.5 meters; maximum depth 6.3 meters and the watershed area is 140.8 hectares. There are no permanent feeder streams and only one permanent dwelling is located adjacent to the lake.

A high degree of eutrophy is quite evident. Approximately three-fourths of the surface is covered by extensive growths of submergent, emergent and floating macrophytes. Volatile residue of sediments range from 15-20%. Inorganic carbon concentration ($CaCO_3$) ranges up to 10%. Productivity is very high (~ 1200 mg C/m^2/day) as evidenced by diel fluctuations in pH. Values of 9.8 were recorded in mid-afternoon on sampling dates. The primary source of nutrient addition to the lake is waterfowl which use it extensively during migration. Each fall an estimated 10,000 Canadian geese use the lake. Approximately 10,000 ducks are also present during migration. A resident population of ducks, geese and swans numbering 300 use the lake year-round.

Wintergreen Lake was chosen as a study site because of its unique features. It is surrounded by a small watershed without permanent feeder streams and is inaccessible to the public. Data on waterfowl usage and various limnological parameters were available.

Temperature, dissolved oxygen, and specific conductance profiles for the lake are shown in Figures 1, 2, 3, and 4.

Figure 1. Hydrographic map of Wintergreen Lake.

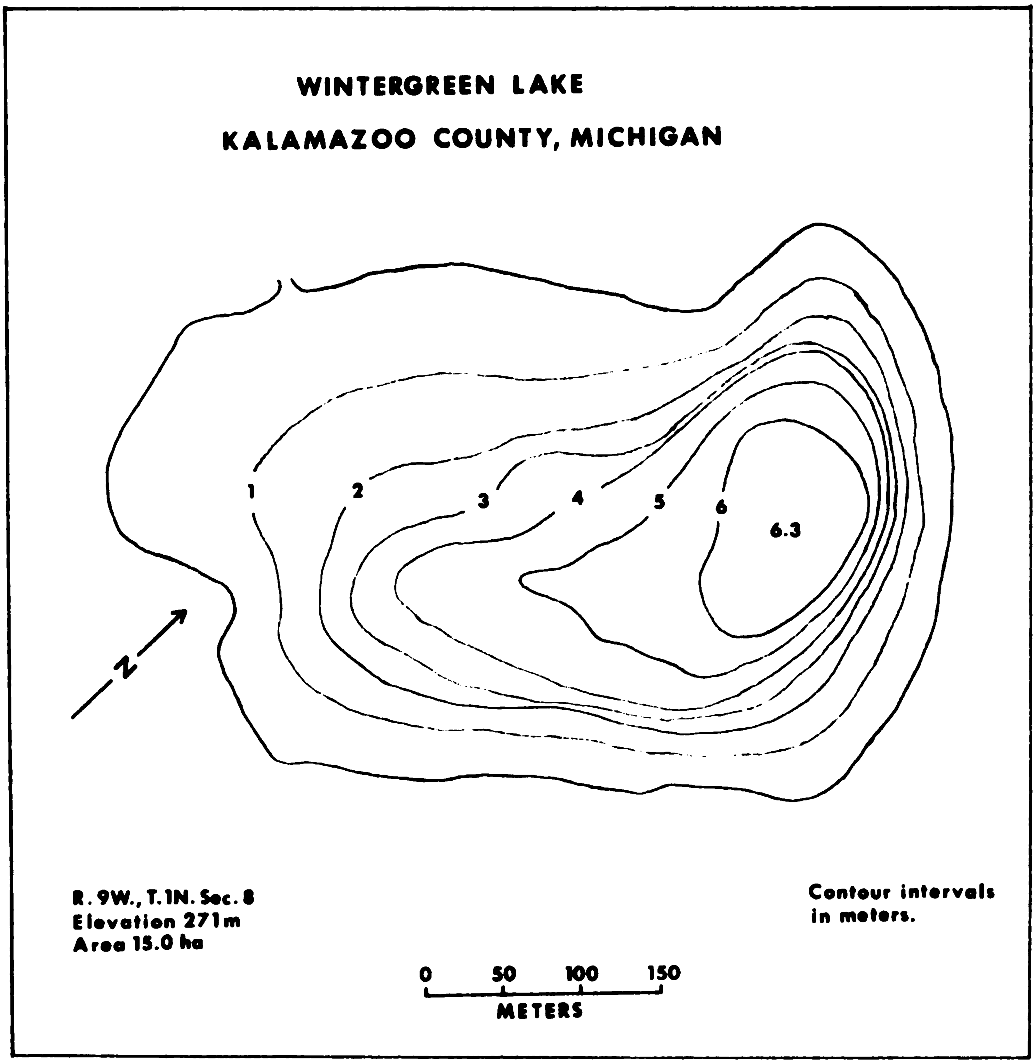
WINTERGREEN LAKE
KALAMAZOO COUNTY, MICHIGAN
1
2
3
4
5
6
6.3
N
R. 9W., T. 1N. Sec. 8
Elevation 271m
Area 15.0 ha
Contour intervals in meters.
0
50
100
150
METERS

Figure 1

Figure 2. Dissolved oxygen (-----) and temperature (———) profiles for September, 1970.

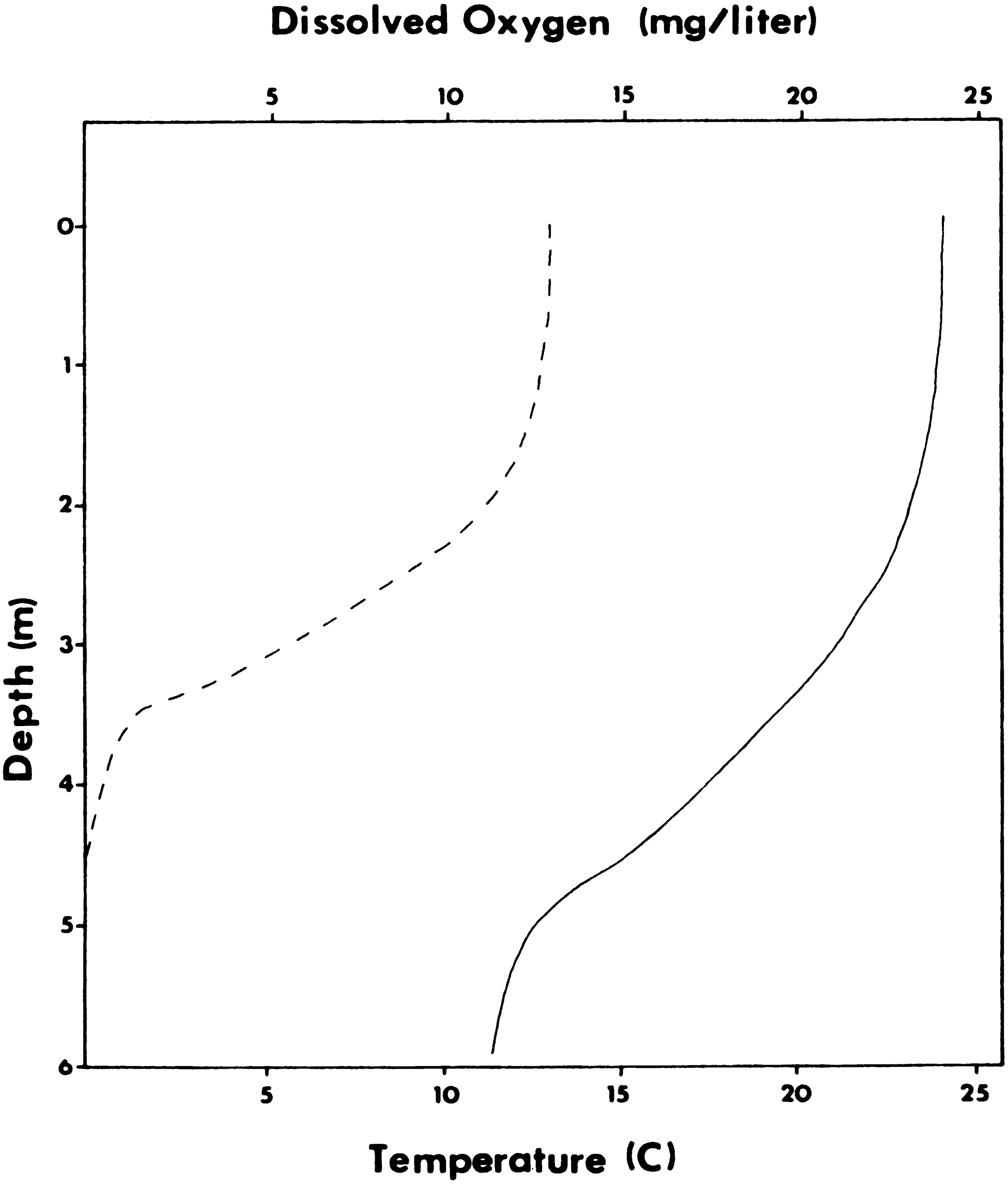
Dissolved Oxygen (mg/liter)
5
10
15
20
25
0
1
2
3
4
5
6
Depth (m)
5
10
15
20
25
Temperature (C)

Figure 2

Figure 3. Dissolved oxygen (-----) and temperature (———) profiles for May, 1971.

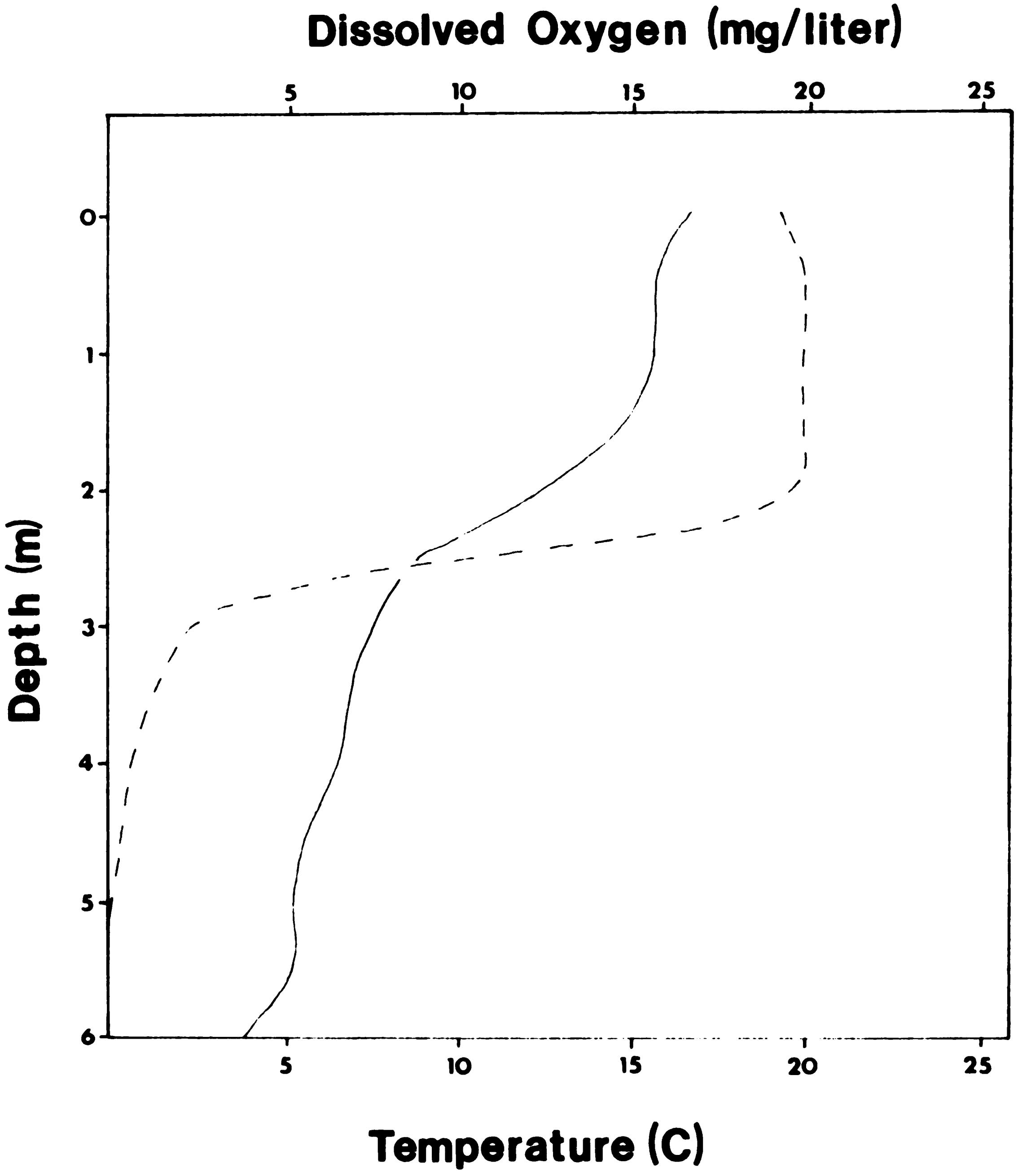
Dissolved Oxygen (mg/liter)
5
10
15
20
25
0
1
2
3
4
5
6
Depth (m)
5
10
15
20
25
Temperature (C)

Figure 3

Figure 4. Specific conductance profiles for September, 1970 (———) and May, 1971 (-----).

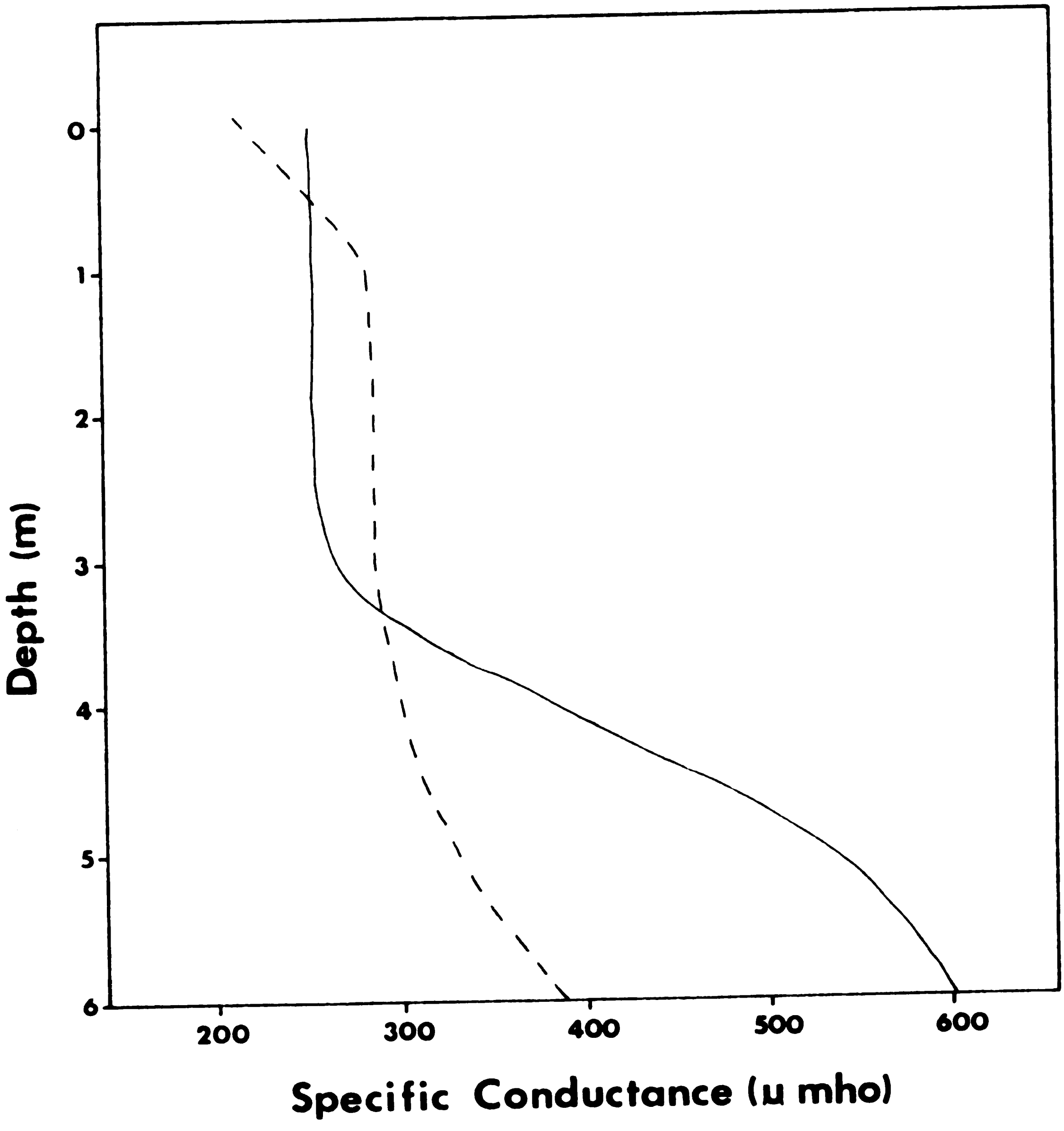
Depth (m)
0
1
2
3
4
5
6
200
300
400
500
600
Specific Conductance (µ mho)

Figure 4

METHODS

Specific conductance, alkalinity, hardness, pH, dissolved oxygen, and temperature profiles of the lake were determined on each collecting date (American Public Health Association, 1971).

Several methods were employed in obtaining fish. Experimental gill nets (32 m in length, 4 mesh sizes ranged from 1.0 to 5.0 cm) were set perpendicular to the shoreline as well as parallel at a depth of 1.5-2.0 meters. Forms not susceptable to capture with gill nets were taken with hook and line. Young of the year were captured by seining.

Macrophytes and algae were collected at random over the lake. Samples of emergent forms included stems and leaves only. Macrophytes analyzed include *Ceratophyllum* sp. and *Nuphar* sp. Dense growths of the filamentous alga *Mougeotia* sp. were entrapped among macrophytic growth which facilitated the collection of large samples.

Zooplankton samples were taken with light traps and plankton net tows. Samples were collected at night.

Sediment samples were collected on a transect along the depth gradient. An Ekman dredge was used, with each sample made up of two grabs.

Droppings from waterfowl were collected from resting areas along the shoreline. Two replicates were obtained for each species tested. All samples were kept frozen until analyzed.

Preconcentration of at least 400 liters of lake water was required to obtain a statistically accurate count of radioactive cesium during a reasonable counting time. This was accomplished with the on-site use of a fast-flow (10 liters/hour) ion exchange apparatus composed of a submersible pump from which water passed through a check valve to a volume recording flow meter. Water then passed through two in-line filters and into a 76 liter reservoir. A float mechanism attached to a microswitch served to maintain a constant pressure head on the ion exchange column. The apparatus was constructed entirely of polyethylene materials to avoid adsorptive losses (Seelye, 1971). Polystyrene vials (2.5 cm by 7.0 cm) packed with 5 g of ammonium hexacyanocobalt (II) ferrate (II) (ACFC) served as columns. Two complete systems were used in the field to obtain duplicate samples.

Preparation of Samples

Biological samples were wet-ashed with concentrated nitric acid. Cesium was removed from the resultant solution with ammonium molybdophosphate (AMP) resin (Feldman and Rains, 1964; Folsom, Young, and Sreekumaran, 1969; Yamagata, 1965). AMP residues were solubilized in sodium hydroxide and extracted with sodium tetraphenylboron (TPB) solution.

Extraction of cesium from sediments was accomplished by leaching with 6 N hydrochloric acid. Samples were then filtered and the filtrate was treated using procedures outlined for digested biological materials (Table A-4, p. 69).

AMP and ACFC resins were counted for ^{137}Cs activity with a single channel gamma scintillation spectrometer coupled with a three inch sodium iodide crystal and photomultiplier. Counting times were calculated to provide minimal counting error ($P < 0.05$) (Overman and Clark, 1960).

Total cesium was determined on TPB extracts by flame emission spectrophotometry. A Jarrell-Ash model 82-800 instrument with a detection limit for cesium of ~ 0.05 mg/l was used.

RESULTS

Inputs of Cesium-137 and Stable Cesium

Cesium enters the lake from two principal sources: atmospheric fallout (rainfall and snowfall), and excreta from migratory and resident waterfowl. Both sources exhibit a seasonal pattern with precipitation at a maximum in April while waterfowl usage peaks in November (Figures 5 and 6).

Waterfowl

Utilization of the lake by waterfowl is extensive. Johnson (1972) estimates a total of 100,000 duck use days and 140,000 goose use days annually. These values are adjusted to time actually spent on the lake or along the shoreline. Kear (1962) estimates excreta produced per day as 3.2% of body weight. Total deposition of ^{137}Cs and stable Cs by waterfowl excreta during 1970 was calculated to be 2.65 mCi and 356 g respectively (Table 1). Most of the ^{137}Cs and stable cesium entering the lake as waterfowl excreta is probably organically bound and is deposited to the sediments.

Figure 5. Seasonal distribution of precipitation at Wintergreen Lake (1926-1960).

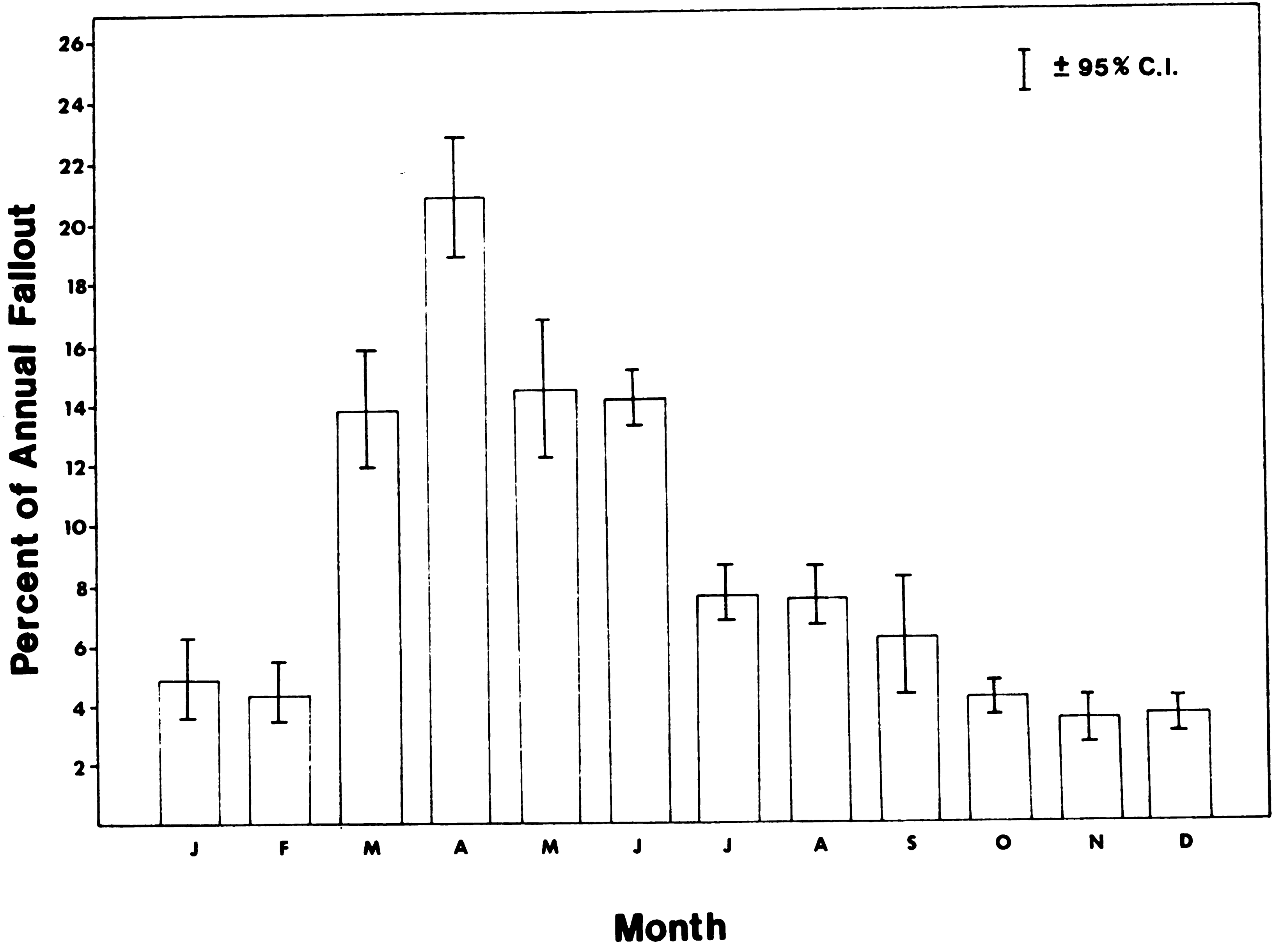
± 95% C.I.
Percent of Annual Fallout
26
24
22
20
18
16
14
12
10
8
6
4
2
J
F
M
A
M
J
J
A
S
O
N
D
Month

Figure 5

Figure 6. Annual pattern of migratory waterfowl usage of Wintergreen Lake.[a]

[a] Data compiled by W. J. Johnson, Kellogg Bird Sanctuary.

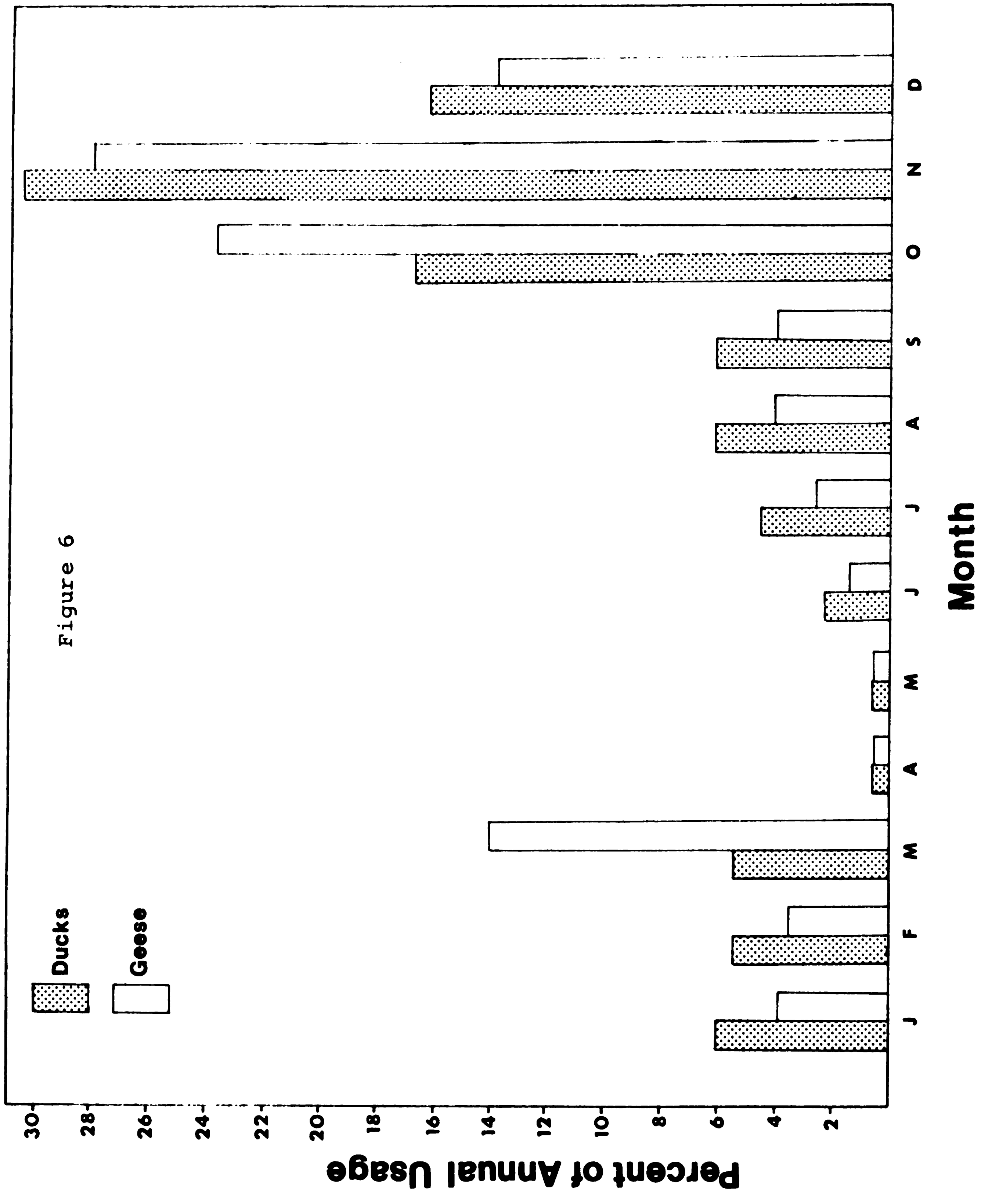
Ducks
Geese
Percent of Annual Usage
30
28
26
24
22
20
18
16
14
12
10
8
6
4
2
J
F
M
A
M
J
J
A
S
O
N
D
Month

Figure 6

Table 1. Estimated input of ^{137}Cs and stable Cs by migratory waterfowl during 1970.

	Geese	Ducks
Bird use days/year	1.43×10^5	1.00×10^5
Excreta/bird/day(g dry wt)	142[a]	32[a]
Excreta/year (metric tons)	20.3	3.2
^{137}Cs (pCi/g)	0.113	0.113[b]
Stable cesium (ng/g)	15.2	15.2[b]
^{137}Cs added to lake in 1970 (μCi)	2.29	0.36
Cs added to lake in 1970 (g)	307	49

[a] From Kear (1962).

[b] Estimated values.

Precipitation

Rates of ^{137}Cs deposition for the period 1954-1969 are presented in Table 2 along with total accumulation (Radiological Health Data and Reports, Environmental Protection Agency). Corrected for radioactive decay, the total accumulation of ^{137}Cs from precipitation on the lake surface is 21.84 mCi. This value assumes no significant export and no addition due to runoff from the watershed. The watershed is predominately sandy loam which has a large exchange capacity for cesium (Squires and Middleton, 1966). Peak deposition rates prior to the atmospheric test ban treaty are quite apparent. Input of ^{137}Cs to Wintergreen Lake from precipitation in 1970 was ~ 300 μCi. The cesium entering the lake as precipitation is in an inorganic soluble state and is generally distributed in the system.

Although data are lacking on addition of ^{137}Cs to the lake from waterfowl for years prior to 1970, it is assumed to follow the general trend exhibited for precipitation (Table 2). Total ^{137}Cs addition in 1970 attributable to waterfowl was approximately 2.5 μCi which accounted for 0.85% of the total. If the assumption of parallel trends is valid, addition of ^{137}Cs to the lake by waterfowl over the period 1953-1970 is insignificant.

Table 2. Deposition rate and accumulation of ^{137}Cs from precipitation.

	Deposition Rate mCi/Km^2/year	Total Deposition[a] mCi/Km^2
1954	1.53	1.04
1955	5.78	5.05
1956	6.12	9.40
1957	5.44	13.36
1958	8.50	19.68
1959	11.90	28.77
1960	2.89	31.01
1961	5.61	35.48
1962	34.51	63.70
1963	52.02	107.14
1964	26.52	129.77
1965	8.50	137.18
1966	3.40	140.22
1967	2.04	142.09
1968	1.70	143.67
1969	2.04	145.63

[a] Corrected to 1970 for decay.

Distribution of Cesium-137 and Stable Cesium in the Lake

The distribution of ^{137}Cs and stable cesium is partitioned into four compartments: fish, plants, water, and sediments. Although this is only an estimate, apparently most ^{137}Cs and stable Cs is associated with the sediments (Table 3). Sediments were sampled to a depth of approximately 10 centimeters. Fish and macrophytes are the only biologic components considered since they account for most of the standing crop. Estimates of the fraction of the total ^{137}Cs and stable Cs pool in fish and plants are high since biomass estimates of one kg/m^2 was used for plants (comparable to sewage lagoon standing crops) and population estimates of fish are based on Fetterolf's (1952) results. Since the carrying capacity of the lake is relatively constant for fish populations and there has been no exploitation, the standing crop is probably comparable to Fetterolf's estimates. Doubling these estimates would not significantly change the percentage of the cesium pool incorporated in fish (Table 3). In spite of the overestimates, approximately 87% of the ^{137}Cs and 98% of the stable Cs pool is incorporated in sediments in the organic fraction or physically associated with the inorganic fraction. No attempt is made to differentiate these two fractions of the sediments.

Table 3. Estimated distribution of ^{137}Cs and stable Cs in Wintergreen Lake.

Compartment	Cesium-137		Stable Cesium	
	mCi	%	grams	%
Fish	1.03×10^{-3}	< 0.01	3.45×10^{-2}	< 0.01
Plants	2.23×10^{-1}	1.8	31.29	0.6
Water	1.39	11.2	47.0	0.8
Sediment	10.82	87.0	5.5×10^{3}	98.6

Cesium Levels in Biotic Components

A pattern of increased ^{137}Cs concentration at higher trophic levels is demonstrated for chubsuckers→ hybrid sunfish→perch→bass (Table 4). No such trophic effect is evident for stable cesium. Previous studies (Nelson, 1969; Kolehmainen and Nelson, 1969) have also failed to demonstrate a trophic effect for stable cesium. Those forms which are more intimately associated with sediments have higher levels of stable cesium relative to radiocesium as reflected in their specific activity (activity/unit mass of element). Since sediments serve as a "sink" for cesium, the lake is, in effect, a two-compartment system.

Those forms which are predatory limnetic feeders, limited to the epilimnion due to hypolimnetic anoxic conditions, would have a specific activity which reflects their food source. A buildup of ^{137}Cs or stable cesium along a trophic scheme is partially explained by several factors: (1) larger animals having a longer biological half-life, (2) differential assimilative efficiencies, and (3) feeding rates.

Forms which feed more exclusively on benthic fauna would all tend to reflect the specific activity of the sediments. Since 87% of the ^{137}Cs and 98% of the stable Cs pool is associated with sediments, forms feeding only

Table 4. Concentrations of ^{137}Cs and stable cesium and specific activities in Wintergreen Lake.

Form	Cesium-137 (pCi/gm) (± 95% CI)		Stable Cesium (ng/gm) (± 95% CI)		Specific Activity (pCi/ng) (10^{-2})		Number of Samples Analyzed	
	Sept., 1970	May, 1971	Sept., 1970	May, 1971	Sept., 1970	May, 1971	Sept., 1970	May, 1971
Largemouth Bass	0.439 ± 0.052	0.415 ± 0.034	15.2 ± 2.4	8.7 ± 2.4	2.89	4.79	19	25
Yellow Perch	0.274 ± 0.034	0.344 ± 0.034	42.2 ± 15.2	25.4 ± 12.6	0.65	1.35	16	18
Hybrid Sunfish	0.138 ± 0.038	0.125 ± 0.012	7.0 ± 2.2	3.8 ± 0.8	1.97	3.29	12	19
Lake Chubsucker	0.230 ± 0.038	0.187 ± 0.044	5.5 ± 1.0	7.3 ± 2.1	4.18	2.56	6	5
Yellow Bullhead	--	0.245 ± 0.040	--	5.3 ± 1.2	--	4.62	--	20
Brown Bullhead	--	0.128 ± 0.054	--	6.7 ± 4.9	--	1.91	--	8
Bowfin	0.222 ± 0.082	--	5.2 ± 2.1	--	4.27	--	3	--
Zooplankton	0.219 ± 0.042	0.044 ± 0.010	33.5 ± 7.6	2.8 ± 1.0	0.65	1.59	4	4
Phytoplankton	--	0.088 ± 0.009	--	10.5 ± 1.6	--	0.84	--	4
Ceratophyllum sp.	--	0.097 ± 0.004	--	10.4 ± 0.6	--	0.93	--	6
Nuphar sp.	--	0.204 ± 0.006	--	79.9 ± 9.2	--	0.26	--	5
Sediments	2.107 ± 0.250	1.990 ± .162	662.1 ± 57.4	697.6 ± 61.5	0.32	0.29	8	8

partially on benthos would have the specific activity of other food sources "masked." Specific activity values in Table 4 demonstrate this point. Fish forms feeding on benthos have a specific activity of 2.0 x 10^{-2} pCi/ng or less. Yellow perch, however, may appear to be an exception. Stomach content analysis revealed they were feeding extensively on larvae of the phantom midge, *Chaoborus* sp. (Table 5).

Aquatic macrophytes also show a difference in specific activity. *Ceratophyllum demersum* had a specific activity of 0.93 x 10^{-2} pCi/ng, while for *Nuphar* sp. it was 0.26 x 10^{-2} pCi/ng or about one-third of the former. This latter value was similar to sediments (0.29-0.32 x 10^{-2} pCi/ng). Since *Nuphar* sp. is a rooted form, one might expect its specific activity to be similar to the sediment in which it is rooted.

Concentration Factors

Concentration Factor (C.F.) is obtained as follows:

$$\text{C.F.} = \frac{\text{concentration of element per unit weight of organism}}{\text{concentration of element per unit weight of water}}.$$

For ^{137}Cs in fish it varied by a factor of three ranging from 1,671 in largemouth bass collected in September, 1970 to 476 in hybrid sunfish collected in the spring of

Table 5. Stomach contents (% volume) of fishes collected in Wintergreen Lake.[a]

	Stomach Contents				
	Fish	Insects	Zooplankton	Vegetation	Miscellaneous
Largemouth Bass	88.0	12.0	0	0	0
Yellow Perch	0	17.5	80.5[b]	0	3.0
Hybrid Sunfish	0	82.0	12.0	3.0	3.0
Brown Bullhead	80.0	16.0	4.0	0	0
Yellow Bullhead	0	0	37.0	0	63.0[c]
Chubsuckers	0	0	100.0	0	0

[a] Stomach contents pooled for each species.

[b] Predominantly *Chaoborus* sp. Although this is an insect, it is placed in the functional category of zooplankton.

[c] Large amounts of unidentified egg masses.

1971 (Table 6). The C.F. for bass is approximately 50% higher than reported by Kolehmainen (1969). The large C.F. present for zooplankton in September is probably due to the presence of large numbers of Chaoborus sp. in the sample which show the influence of their association with the sediments or the sediment-water interface.

Concentration factors for stable Cs in fish are generally lower than for ^{137}Cs. This appears to be due to differential availability of stable Cs. Perch showed a C.F. of 2,009 and 1,209 for stable Cs in the fall and spring respectively. These high C.F.'s reflect their feeding habits (selection for Chaoborus sp.).

Zooplankton samples collected in September had a high C.F. for stable Cs as well as ^{137}Cs. Nuphar sp. exhibited the highest C.F. for stable Cs of any form tested. This, again, is due to the closer interaction with sediments. Concentration factors for stable Cs in perch, fall zooplankton samples, and Nuphar sp. are somewhat misleading since they are based on concentrations in water. They should be compared to available stable Cs levels in sediments. However, since destructive analyses were carried out, that fraction of stable Cs in the sediments available for uptake cannot be estimated.

Table 6. Concentration factors of ^{137}Cs and stable Cs in various components of the biological community, Wintergreen Lake.

Form	September, 1970		May, 1971	
	^{137}Cs	Stable Cs	^{137}Cs	Stable Cs
Largemouth Bass	1,671	723	1,580	413
Yellow Perch	1,043	2,009	1,271	1,209
Hybrid Sunfish	525	333	476	181
Lake Chubsucker	875	262	712	348
Yellow Bullhead	--	--	932	252
Brown Bullhead	--	--	587	319
Bowfin	845	248	--	--
Zooplankton	833	1,595	167	133
Phytoplankton	--	--	334	498
Ceratophyllum sp.	--	--	368	493
Nuphar sp.	--	--	774	3,790

Trophic Level Effect

Data from largemouth bass, yellow perch, hybrid sunfish, and lake chubsuckers, representative of four trophic levels were analyzed for ^{137}Cs, and specific activity using a two-way analysis of variance (ANOVA). Data on stable Cs for those species were not analyzed since a trend for trophic amplification was not in evidence (Table 4). Cesium-137 activities among species (Table 7) were significantly different ($P < 0.005$) while time-species interaction was not significant ($P > 0.05$) (Table 7). Since the data show variance heterogeneity, comparisons of means was completed using Scheffé's (1953) multiple comparison test. All means were significantly different ($P < 0.05$) except those for lake chubsuckers and hybrid sunfish (Table 7). It may be noted that lake chubsuckers were higher in activity than hybrid sunfish. This, again, is attributable to the closer association of the former with the sediments.

Specific Activity

Both specific activity among species tested and time-species interaction were not significant ($P > 0.05$) (Table 8). Although values for the specific activities of these species appears to be different (Table 4) the lack of significant differences among species remains. This is probably due to large variances around the individual

Table 7. Two-way ANOVA with time blocked. Test of hypothesis: no difference in ^{137}Cs levels among species.

Source	SS	df	MS	F	F0.95
Time	0.0005	1	0.0005	0.098	3.93
Species	1.6037	3	0.5346	98.67***	2.69
Interaction	0.0415	3	0.0138	2.55	2.69
Error	0.6481	110	0.0059		
TOTAL	2.2939	117			

$$\begin{array}{cccc} 2^{a} & 1 & 3 & 4 \\ \multicolumn{2}{c}{\underline{0.131^{b} \qquad 0.208}} & 0.309 & 0.427 \end{array}$$

---

[a] Any two means not underlined by the same line are significantly different (P < .05).

[b] 1. Chubsuckers; 2. Hybrid Sunfish; 3. Yellow Perch; 4. Largemouth Bass.

Table 8. Two-way ANOVA with time blocked. Test of hypothesis: no difference in specific activity among species.

Source	SS	df	MS	F	FO.95
Time	0.0108	1	0.0108	2.07	3.93
Species	0.0373	3	0.0125	2.38	2.69
Interaction	0.0218	3	0.0073	1.39	2.69
Error	0.6113	110	0.0056		
TOTAL	0.6813	117			

means. The degrees of freedom were approximated in computing F-critical values for trophic effect and specific activity (Sokal and Rohlf, 1969). This was necessary due to heterogeneous variance. Since this is an approximate test and the calculated and F-critical values are very close for species (Table 4), the null hypothesis of no difference among species in specific activity is acceptable, but questionable.

DISCUSSION

Cesium-137 and stable Cs concentration in aquatic flora and fauna in a lake are influenced by several factors. These include the mode by which isotopes enter the system, degree of eutrophy in the system, behavior and cycling of the isotopes, biological half-life, and assimilative efficiencies.

Two sources contributed ^{137}Cs and stable Cs to Wintergreen Lake. Precipitation, which is the primary source, adds these isotopes to the lake water where they are initially distributed uniformly in the upper strata. The second source, waterfowl excreta, introduces cesium in both a water-soluble and insoluble fraction (Figure 7). The soluble portion, as with precipitation, is generally distributed. The organically bound insoluble fraction is, however, deposited to the sediments, where, due to hypereutrophy of the lake with accompanying anaerobic conditions, it accumulates. This portion is available only to those forms feeding in the sediments and in turn the forms which feed on them. An important route of isotopes from sediments to higher forms is by way of food chains with bacteria as a base (Figure 7). Culver and Brunskill (1969)

Figure 7. Principal pathways of cesium in an aquatic ecosystem.

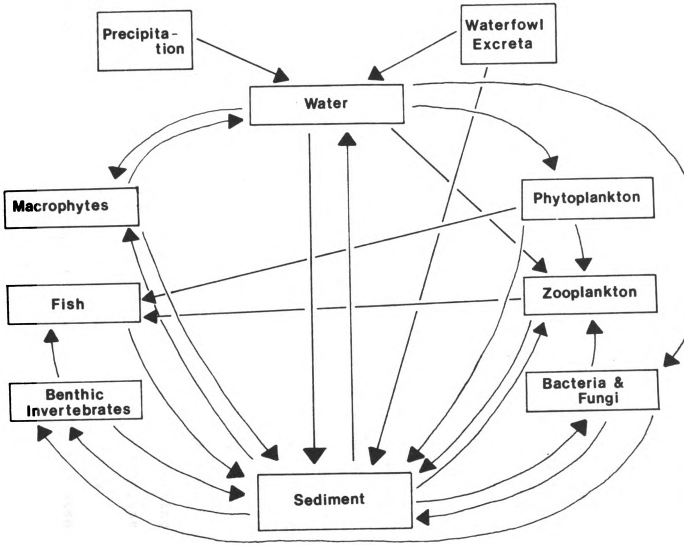
Precipita-
tion
Waterfowl
Excreta
Water
Macrophytes
Phytoplankton
Fish
Zooplankton
Benthic
Invertebrates
Bacteria &
Fungi
Sediment

Figure 7

found zooplankton with their gut tubes filled with sulfur bacteria in a meromictic lake where phytoplankton productivity was low. If bacteria dominate the biomass (under conditions of prolonged anaerobic conditions) Likens (1972) concluded they may provide a very concentrated and important source of food for zooplankton. These observations may explain the high C.F.'s for zooplankton collected in September (Table 6). However, specific data on bacterial populations in the lake are lacking.

The soluble fraction of isotopes from waterfowl and precipitation are ultimately added to both the sediments and the insoluble fraction. Adsorption to suspended particulate matter in the water column with subsequent settling is one mechanism of removal from water. A second mechanism postulated by Manny (1970) for removal of ions from the water column is through marl ($CaCO_3$) formation and deposition. Algal cells, during photosynthesis, remove CO_2 from water in sufficient quantities to exceed the solubility product of $CaCO_3$ in adjacent water resulting in deposition of marl on the cell surface. When sufficient accumulation occurs to exceed a cell's buoyant capacity, it settles to the bottom. As a result, cesium incorporated into algal cells is delivered to the sediments.

Theoretically $CaCO_3$ should resolubilize due to aggressive CO_2 in the lower water strata (Ruttner, 1953).

This apparently does not occur in Wintergreen Lake. Manny (1970) suggests the marl-covered algal cells are coated with a layer of organic nitrogen in the form of large amorphous molecules which prevents attack by CO_2. This prevents dissolution of the $CaCO_3$.

Biological half-life and assimilative efficiency are important factors in determining the concentration of cesium at higher trophic levels. Biological half-life is positively correlated with size of an animal. Since metabolic rates per unit weight necessary for maintenance decrease in larger animals, the turnover rates decrease resulting in a longer biological half-life.

Predatory fish change their feeding habits as they grow. Younger age classes of a population of predatory fish consume zooplankton and bottom organisms. With an increase in size they become predators, consuming small fish. With this change in feeding habits, they are passing from a lower to a higher trophic level. Kolehmainen *et al.* (1968) found large perch to have twice the concentration of ^{137}Cs as small perch in the same lake. The difference was attributed to a change in feeding habits from bottom organisms to small fish. They suggested the assimilative efficiency of perch may be higher for small fish than for bottom organisms. The increase may also be partially attributed to growth. Growing fish would utilize a significant percentage of food intake in the deposition of new tissue in growth. Since the small fish that predators consume have higher concentrations of ^{137}Cs than bottom organisms and zooplankton, a higher concentration of ^{137}Cs is present in the food for the predator. This would result in higher concentrations of ^{137}Cs in the predator. Considering the factors discussed above, one would expect an increase in cesium concentration at higher trophic levels. The expected relative concentrations of ^{137}Cs along a "trophic continuum" is illustrated in Figure 8. Various fish forms analyzed from Wintergreen Lake are ranked (0-10) based on feeding habits presented in the literature (Harlan and Speaker, 1956). A second ranking on the continuum is based on ^{137}Cs concentrations determined for the various species. Largemouth bass were placed at 9.0 on the scale and the mean ^{137}Cs concentrations for bass was divided into mean concentrations of other forms to determine rank on the second scale. Mean ^{137}Cs concentrations for those forms associated with sediments were higher than expected (Figure 8) (i.e., brown and yellow bullhead and lake chubsucker).

Cesium-137 content in bowfin was much lower than expected. No conclusions can be drawn due to small sample size (3 fish) and large variance about the mean concentration.

Figure 8. Ranking of fishes from Wintergreen Lake on a "Trophic Continuum." A. Expected ranking B. Ranking based on ^{137}Cs concentration (pCi/g wet weight).

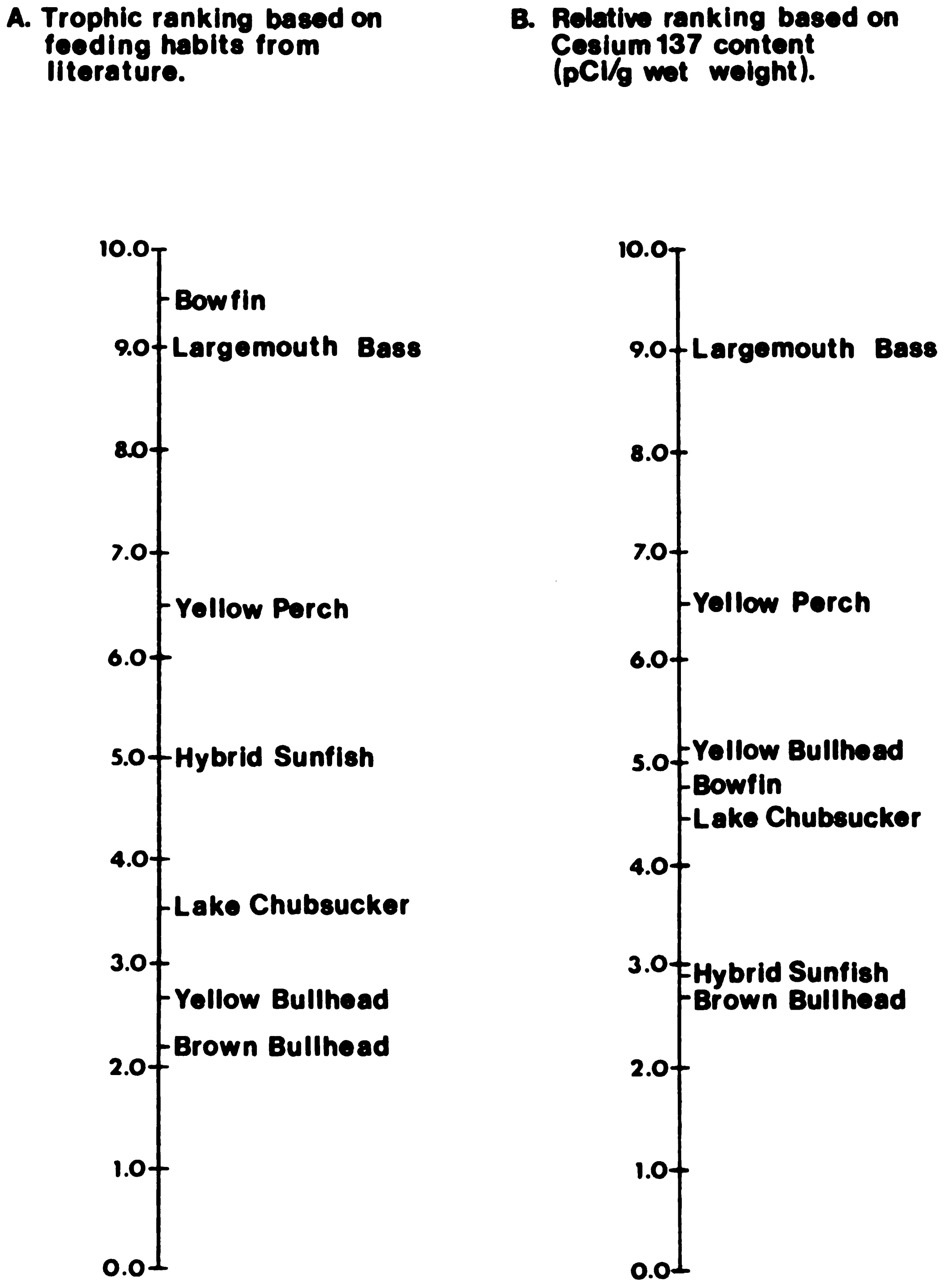
A. Trophic ranking based on feeding habits from literature.
B. Relative ranking based on Cesium 137 content (pCi/g wet weight).
10.0
Bowfin
9.0
Largemouth Bass
8.0
7.0
Yellow Perch
6.0
5.0
Hybrid Sunfish
4.0
Lake Chubsucker
3.0
Yellow Bullhead
Brown Bullhead
2.0
1.0
0.0
10.0
9.0
Largemouth Bass
8.0
7.0
Yellow Perch
6.0
Yellow Bullhead
5.0
Bowfin
Lake Chubsucker
4.0
3.0
Hybrid Sunfish
Brown Bullhead
2.0
1.0
0.0

Figure 8

Trophic level effect on ^{137}Cs concentrations (lake chubsuckers→hybrid sunfish→yellow perch→largemouth bass) is largely explained by common availability of the isotope to these forms. If levels of ^{137}Cs in these forms are compared to levels determined for forms more closely associated with sediments (Table 4) the trophic level effect on ^{137}Cs concentration is not as apparent. Gustafson (1967) found a similar situation in his work with fishes of Red Lake, Minnesota.

The explanation of these data involves several factors. The lake can be thought of as a two-compartment system (water-sediment) with ^{137}Cs exchange between them. Sediments ultimately serve as an autochthanous source of cesium to the water as well as directly to the biological community. Within the trophic structure of a lake community, both compartments are included. Trophic structure, however, does not imply interaction. Several distinct food chains may exist within a given trophic structure. Significant differences in degree of bioamplification between food chains within a trophic scheme may be present. If this situation exists as in Wintergreen Lake where more than 90% of the total cesium pool is in the sediments, it is meaningless to look at trophic effect on cesium concentration without regard to degree of interaction of species tested.

SUMMARY

1. Usage of Wintergreen Lake by waterfowl is extensive. They contribute in excess of 20 metric tons of excreta (dry weight) per year to the lake. The amount of the total annual ^{137}Cs input attributable to them is less than 1%.

2. Of the total cesium pool ~ 87% of the ^{137}Cs and ~ 98% of the stable Cs is incorporated in sediments.

3. It appears that stable Cs enters the system as organically bound (waterfowl excreta) and is deposited to the sediments whereas ^{137}Cs enters in soluble inorganic form and is distributed throughout the system. Consequently specific activity of sediment is lower than any other component of the ecosystem.

4. The intimacy of association of biotic forms with sediments is reflected in lowered specific activity.

5. Concentration factors for ^{137}Cs are higher than for stable Cs except in forms closely associated with sediments.

6. A trend for bioamplification of ^{137}Cs resulting in greater activity at higher trophic levels is

demonstrated. This trend is not in evidence for stable Cs.

7. No significant differences were found in specific activities among forms that are limnetic and predatory.

8. Forms closely associated with sediments have higher ^{137}Cs content than would be expected based on their feeding habits.

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APPENDICES

APPENDIX A

METHODS AND MATERIALS

APPENDIX A

METHODS AND MATERIALS

On-Site Measurements

The following parameters were measured on each sampling date: (1) pH (Beckman Model-N portable pH meter); (2) alkalinity and hardness (American Public Health Association, 1971); (3) temperature and specific conductance (portable conductivity meter and thermistor); and (4) dissolved oxygen (Precision Scientific oxygen analyzer).

Due to low levels of Cs, lake water was passed through ion exchange columns in situ in order to preconcentrate the element in quantities sufficient for analysis. An ACFC (ammonium hexacyanocobalt (II) ferrate (II)) resin was used (Petrow and Levine, 1967). This form of resin was chosen to avoid the presence of ^{40}K since counting of ^{137}Cs was done with a single channel analyzer. ACFC is a stable resin which does not lose the integrity of its crystalline structure producing "fines" which reduce flow rates resulting in variable efficiency (Folsom and Sreekumaran, 1970). A curve of uptake efficiency for ^{137}Cs on the ACFC resin is shown in Figure A-1. Apparatus developed by Seelye (1971) for on-site preconcentration of cesium was used in this study. Four replicates for determination of cesium content of water were obtained on each sampling date.

Polystyrene vials were used as ion exchange columns. The bottoms of the vials were cut out and caps removed when they were in use. Prior to use, vials were stored with caps in place and filled with distilled water in order to have the resin activated thus avoiding efficiency changes associated with resin swelling in the field. The ion exchange column is shown in Figure A-2. Column dimensions were such that they could be placed in the detector and counted directly.

Field Collections

Fish were collected using experimental gill nets (1-5 cm mesh) and hook and line. Young of the year were collected by seining in shallow areas of the lake. Macrophytes and algae were collected randomly over the surface of the lake. Zooplankton were obtained at night using light traps and net tows. Sediment samples were taken along a depth gradient using an Ekman dredge. All samples were kept frozen until analyzed.

Figure A-1. Percent recovery of ^{137}Cs on ACFC (Ammonium Hexacyanocobalt (II) Ferrate (II)) resin verses flow rate in liters per hour.

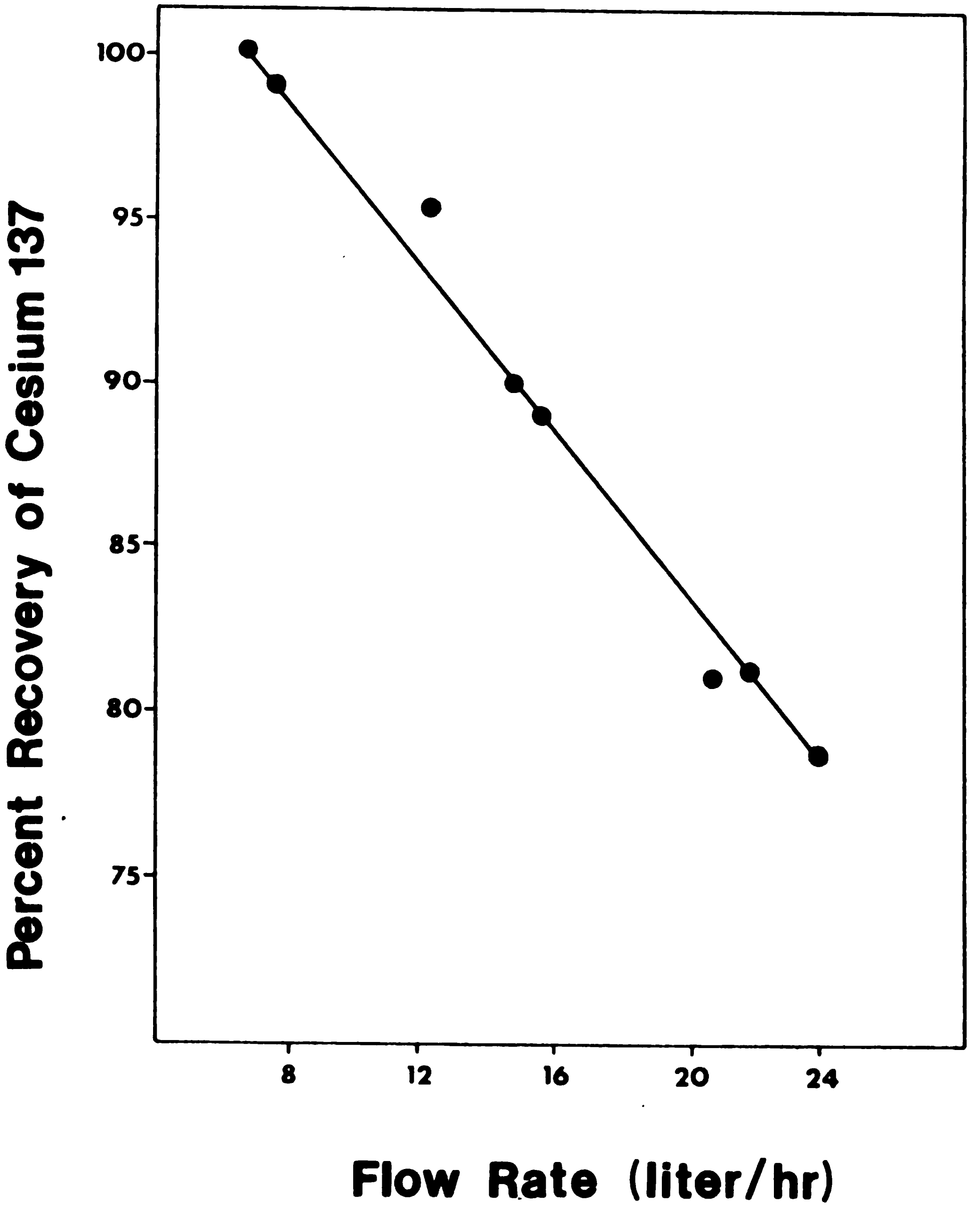
Percent Recovery of Cesium 137
100
95
90
85
80
75
8
12
16
20
24
Flow Rate (liter/hr)

Figure A-1

Figure A-2. Polystyrene ion exchange column.

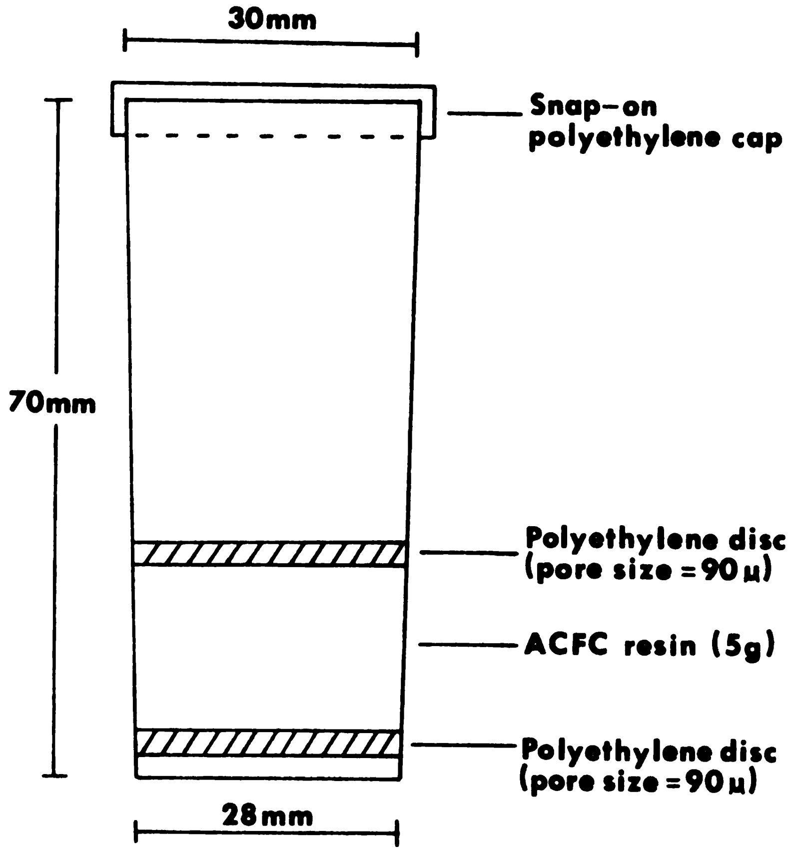
30mm
Snap-on polyethylene cap
70mm
Polyethylene disc (pore size =90 µ)
ACFC resin (5g)
Polyethylene disc (pore size =90 µ)
28mm

Figure A-2

Sample Preparation and Digestion

All biological material was wet oxidized using concentrated HNO_3 at 130 C. Digestion was carried out in 3 or 5 liter round bottom two-necked boiling flasks. The flasks were connected to reflux condensers and fitted with electric heating mantles. Each mantle was connected to a rheostat for temperature control. Methods for sample preparation and digestion are outlined in Table A-1. The digestion apparatus is shown in Figure A-3. When possible, fish were digested and analyzed individually. However in the case of smaller forms (perch and hybrids) it was necessary to pair the fish for analysis in order to have sufficient Cs present to be quantifiable.

Sediment samples were extracted with hot 6 N HCl using methods as outlined in Table A-2. Replicate samples from each depth were analyzed. Filter residue from each sample was counted in order to determine the extraction efficiency. The overall percent efficiency of cesium extraction was 99.2 ± 0.44 (α .05) (Table A-3).

Cesium was removed from the acid solution using a batch method with ammonium molybdophosphate (AMP) as an inorganic exchanger (Smit, 1958; Kahn *et al*., 1957). The batch method was used since the microcrystalline structure of AMP makes its use in a column impractical due to reduced flow rates. The method used was adopted from

Figure A-3. Wet oxidation apparatus (3 or 5 liter flask).

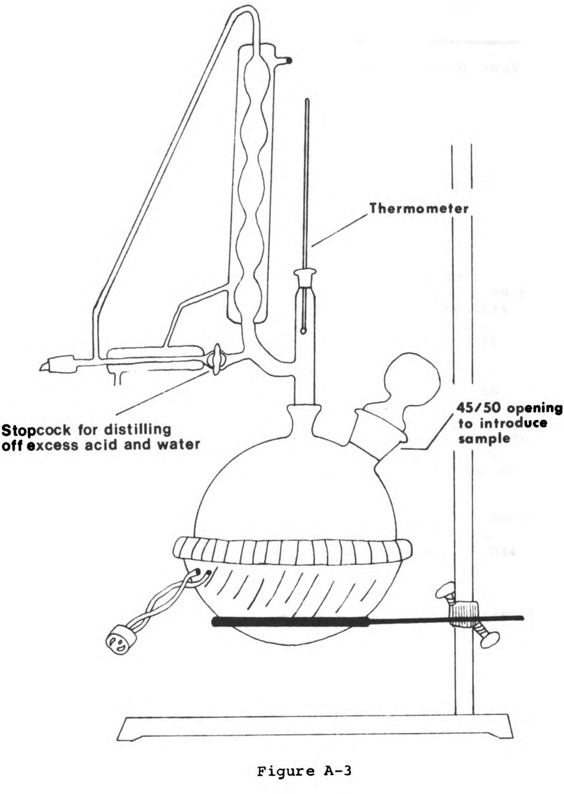
Thermometer
Stopcock for distilling off excess acid and water
45/50 opening to introduce sample

Figure A-3

Table A-1. Nitric acid digestion procedure for biological materials.

1. Divide sample into portions which pass through neck of boiling flask.

2. Determine wet weight of sample.

3. Place sample in boiling flask.

4. Add approximately 5.0 ml of concentrated HNO_3 per gram of sample.

5. Allow digestion to proceed 3-4 hours at room temperature.

6. Reflux until nitrous oxide fumes are no longer apparent. Open stopcock and distill off excess water and acid until approximately 2 ml of acid are left per gram of fish. (Do not distill further since precipitates will form which may result in severe bumping.)

7. Add additional concentrated HNO_3 to bring back to original volume (step 4).

8. Reflux with stopcock closed until no oil can be seen floating on the surface (6-10 hours). Additional acid may be required if oil droplets are present at the end of this time.

9. Allow digestant to cool and remove from flask. Pass sample through glass wool filtering material and rinse flask thoroughly with distilled water. Rinse into polypropylene beaker of at least twice the volume of the sample.

10. Allow to cool to room temperature and proceed with AMP collection of Cs.

Table A-2. Preparation of sediment for analysis of Cs.

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1. Dry sample at 70 C (48-72 hours).
2. Powder sample (mortar and pestle or Wiley mill) and weigh out aliquot (100 g).
3. Place aliquot in Pyrex beaker (1 liter) and add 6 N HCl (5 ml/g). CAUTION: If sediment is high in organic content or contains significant amounts of marl ($CaCO_3$) extensive foaming may result. Addition of an antifoaming agent (silicone solution) may be necessary.
4. Allow reaction to proceed at room temperature for 2-3 hours. Stir sample periodically.
5. Place watch glass over beaker and apply heat. Maintain a temperature just below the boiling point. Allow leaching to continue with heat for 15-20 hours.
6. Allow sample to cool and pass through Whatman no. 41 filter paper.
7. Treat fitrate as outlined in Table A-4.

Table A-3. Efficiency of cesium recovery from sediment samples.

Depth (meters)	Efficiency of Extraction					
	AMP-1		AMP-2		Filter Residue	
	DPM/g	%	DPM/g	%	DPM/g	%
1.5 A	208	88.3	26	11.1	1.6	0.6
1.5 B	202	86.3	31	13.3	1.0	0.4
3.0 A	208	66.1	106	33.6	1.0	0.3
3.0 B	279	90.0	25	8.2	2.9	0.9
4.0 A	151	69.8	62	28.6	3.5	1.6
4.0 B	166	66.9	80	32.2	2.3	0.9
4.5 A	311	87.4	43	12.2	1.7	0.4
4.5 B	325	90.5	33	9.3	1.0	0.2
5.0	148	71.4	56	27.2	3.1	1.4
6.0 A	127	65.4	67	34.6	0	0
6.0 B	120	88.3	13	9.6	2.9	2.1

Percent recovery using:

1. Single addition of AMP = 80.1 ± 7.4[a]
2. Two successive additions of AMP = ... 99.2 ± 0.44

---

[a] Mean ± 95% C.I.

Feldman and Rains (1964) with modifications as suggested by Folsom and Sreekumaran (1970) and as outlined in Table A-4. Two successive additions of AMP to the acid solution were necessary since a single addition produced highly variable recoveries. With two additions the percent recovery of cesium was 97 ± .74 (α .05) (Table A-7). Subsequent cleanup steps eliminated potassium from the sample, thus eliminating interference in counting due to ^{40}K.

Commercially available sources of AMP have been found to contain 0.2 to 1.0 mg Cs/g which renders them unsuitable for use in trace analysis of cesium (Folsom and Sreekumaran, 1970). Therefore AMP was prepared in the laboratory using a procedure which purges it of cesium. The prepared AMP was counted with no detection of ^{137}Cs. Procedures for preparation of cesium-free AMP are outlined in Table A-5. Initially, polyethylene or polypopylene labware was used in all analytical procedures to avoid adsorption losses of Cs. Polypropylene was later used exclusively due to its superior resistance to acid.

The final NaOH solution containing cesium was extracted with 0.1 N sodium tetraphenylboron solution (TPB) (3/1 hexone:cyclohexane). The solution was less concentrated than suggested by Feldman and Rains (1964). However, it gave more favorable resolution (signal-to-background ratio) necessary for analyzing trace amounts

Table A-4. Cs collection and preparation for flame emmission analysis.

1. To acid solution of biological material add 4 mg AMP/g wet wt.
2. Stir solution for 30 minutes and allow sample to settle overnight.
3. Decant supernatant and collect AMP in 50 ml graduated polypropylene centrifuge tube.
4. Repeat steps 1-3.
5. Centrifuge collected AMP (5 min at 1800 rpm) and count ^{137}Cs.
6. Dissolve AMP in 1.0 N NaOH (~ 15 ml per gram).
7. Adjust pH to 3.5 with powdered tartaric acid.
8. Add 0.8 mg AMP/g wet weight and stir for 30 minutes.
9. Repeat step 3 and 5.
10. Solubilize AMP in 1.0 N NaOH (~ 10 ml).
11. Place 10 ml NaOH Cs solution in separatory funnel with 10 ml 0.1 N TPB and shake vigorously for 2 minutes.
12. Allow layers to separate overnight.
13. Retain organic layer for flame emission analysis. (Keep samples refrigerated)

Table A-5. Preparation of low-cesium AMP (Folsom and Sreekumaran, 1970).

Solutions used:

1.--81 g NH_4NO_3 + 81 g citric acid + 102 g $(NH_4)_6Mo_7O_{24}$ + 2140 ml H_2O

2.--391 ml 70% HNO_3 + 455 ml H_2O

3.--Add sol. 1 slowly into sol. 2 stirring without heat

4.--100 g $(NH_4)_2HPO_4$ + 2000 ml H_2O

1. Place solution 3 in a 4 liter Pyrex beaker and add 2 ml of solution 4. Heat to a boil while stirring.

2. Cool solution in water bath and allow yellow ppt. (AMP) to settle.

3. Decant mother liquor (60-80 C) into another 4 liter Pyrex beaker and discard AMP that was made in step 1. The mother liquor has now been purged of cesium.

4. To mother liquor add 100 ml of solution 4 and bring to boil while stirring.

5. Allow to cool and decant supernatant.

6. Wash the ppt. (AMP) with 1 N NH_4NO_3.

7. Collect on Whatman No. 41 filter paper.

8. Dry at 70 C overnight and store in dry place.

The yield will be ~ 71 g AMP.

of Cs. The organic layer was retained for analysis of total Cs by flame emission (8521 Å).

Controls

Reagent blanks were run routinely to check for contamination. Periodically, sample blanks were spiked to check on the efficiency of recovery of cesium (Table A-6). The spiked blank, however, cannot be considered as a true test of methodology since interferences present in acid digests of biological materials would not be present in reagents. In order to test for interferences, successive treatment of the sample with AMP was counted separately. In all cases it was found that all of the activity was removed from the sample with three additions of AMP. The total activity found in the three aliquots was divided into the activity of each to determine the percent Cs recovered in each addition (Table A-7).

Radiocesium Analysis

Cesium-137 activity of water, biological and sediment samples was analyzed gamma-spectrometrically with a single channel solid scintillation counter (0.662 Mev.). A three inch NaI(Tl) crystal with a 1.25 inch by 2 inch well was coupled with a Nuclear Chicago spectrometer and scaler. Counting vials were all one inch in diameter insuring uniform sample geometry.

Table A-6. Reagent blanks and spiked blanks used as controls on methods.

Sample No.	HNO_3 (ml)	^{137}Cs Added (DPM)	Cs Recovered (DPM)			Percent Recovery	Cs (ppb)
			AMP-1	AMP-2	TOTAL		
1	1500	670	621	40	661	98.7	--
5	1500	0	0	0	0	--	0
3	2500	853	791	39	830	97.3	--
4	2500	0	0	0	0	--	0
8	1500	326	297	26	323	99.2	--
2	1500	0	0	0	0	--	0
6	2500	54	49	5	54	100.0	--
7	2500	0	0	0	0	--	tr.

Table A-7. Efficiency of cesium recovery from biological materials.

Sample No.	AMP-1		AMP-2		AMP-3	
	DPM	%	DPM	%	DPM	%
W06-C	178.2	79.5	34.3	15.3	11.5	5.1
W06-E	116.2	88.6	11.0	8.4	4.0	3.0
W06-H	76.1	71.4	26.3	24.7	4.2	3.9
W03-A	107.0	75.8	29.1	20.6	5.0	3.6
W03-C	103.0	75.0	29.7	21.6	4.6	3.4
W03-E	177.5	90.5	15.1	7.7	3.6	1.8
W03-F	111.5	90.0	9.3	7.5	3.2	2.5
W05-A	81.0	80.7	13.3	13.3	6.1	6.0
W05-D	372.0	89.0	40.4	9.7	5.7	1.3
W05-G	896.0	89.8	86.8	8.7	14.5	1.5
W07-A	605.0	82.6	108.8	14.9	18.4	2.5
W07-B	535.0	86.6	71.7	11.6	11.4	1.8
W08-D	578.0	84.2	83.5	12.2	24.8	3.6
W08-F	513.0	80.0	108.0	16.8	20.5	3.2
W09-C	709.0	85.4	104.0	12.5	17.4	2.1

Percent recovery using:

1. Single addition of AMP = 83.3 ± 3.35[a]
2. Two successive additions of AMP = ... 97.0 ± 0.74

---

[a] Mean ± 95% C.I.

Standards were counted daily to determine efficiency which averaged 13.5%.

Counting times were calculated to provide determination of ^{137}Cs activity at 95% level of significance according to the following formulae (Overman and Clark, 1960):

$$T_s = \frac{R_{s+b} + \sqrt{(R_b)\ (R_{s+b})}}{(G^2)\quad (R_s{}^2)}$$

Where:

T_s = Sample time (min)
R_s = Sample rate (cpm)
R_b = Background rate (cpm)
R_{s+b} = Total rate (cpm)
G = percent error (0.05)

$$T_b = Ts/\sqrt{\frac{R_{s+b}}{R_b}}$$

Where:

T_b = Background time (min)

Cesium Analysis

Cesium analysis was accomplished by flame emission spectroscopy. The instrument used was a Jarrell-Ash model 82-800 with an infrared grating and red-sensitive photomultiplier (R446). An air-H_2 flame was used. The detection limit for cesium in hexone-TPB was approximately 0.05 mg/liter. The principle emission line of cesium (8521Å) was used. Cesium standards (CsCl) were prepared in NaOH and extracted with equal volumes of 0.1 N TPB in 3/1 hexone:cyclohexane.

APPENDIX B

LITERATURE REVIEW

APPENDIX B

LITERATURE REVIEW

Cesium, although widely distributed in the earth's crust, is a rare element seldom occurring in significant quantities. Typical of the alkali metals, cesium forms strong bases and its salts are generally water soluble. It is the most electro-positive of all metals and is readily oxidized. The chemistry, radiochemistry and analytical methods for analysis are reviewed by several authors (Finston and Kinsley, 1961; Poluektov and Mishchenko, 1962, Yamagata, 1965).

Stable Cesium in Aquatic Environments

Cesium content in marine forms has been investigated much more extensively than freshwater counterparts. Fish are the only freshwater forms for which data are available on natural cesium levels. Most fresh water studies have dealt with isotopes of cesium occurring in greatest abundance (^{137}Cs and ^{134}Cs). Levels of cesium reported for fresh water and freshwater fish are summarized in Tables B-1 and B-2 respectively.

Table B-1. Cesium content in fresh water reported from various studies.

Location	Cs(ng/l)	Reference
Lakes in Northern Italy	0.043-0.087	Kolehmainen 1969
Clinch River, Tennessee	0.025	Nelson 1969
Lake Mead	0.057	Sreekumaran *et al.* 1968
Colorado River	0.023	Sreekumaran *et al.* 1968

Table B-2. Cesium content of freshwater fish as reported from various studies.

Species	Cs(ng/g)	Location	Reference
White Crappie	12.9	Clinch River	Nelson 1969
Freshwater Drum	8.7	" "	" "
White Bass	16.0	" "	" "
Channel Catfish	4.1	" "	" "
Bluegill	3.4	" "	" "
Mixed Group	9.8-57.0	Northern Italian Lakes	Bortoli as cited by Kolehmainen 1969

Kolehmainen et al. (1968) found that cesium is removed from water to a much greater degree in eutrophic and turbid lakes than in oligotrophic lakes. Removal is probably accomplished by algal bloom uptake in eutrophic lakes (Williams, 1960) and through sorption by suspended inorganic particulate matter in turbid lakes (Tamura and Jacobs, 1960; Kudo and Gloyna, 1971).

^{137}Cs in the Aquatic Environment

Cesium has 21 isotopes ranging in atomic weight from 123-144. The stable isotope is ^{133}Cs. The most abundant radionuclide of cesium is ^{137}Cs which has a physical half-life of 30.23 years. A major component of fallout from atmospheric testing of nuclear weapons, ^{137}Cs is formed late in nuclear detonations by the following reactions:

$$^{137}\text{I} \xrightarrow[\text{sec}]{19} {}^{137}\text{Xe} \xrightarrow[\text{min.}]{3.4} {}^{137}\text{Cs} \xrightarrow[\text{yrs.}]{30.23} {}^{137}\text{Ba (stable)}.$$

Emissions resulting from decay include gamma rays (0.662 mev) and beta particles (0.514, 1.18 mev). A relatively high fission yield (6%), long half-life and energetic emission of ^{137}Cs presents a potential hazard to biotic components of various ecosystems when this isotope is incorporated into them.

Sources

Large-scale atmospheric testing of nuclear devices is the primary source of ^{137}Cs contamination of the environment. Total production of this isotope from detonations prior to the nuclear test-ban treaty of 1962 is estimated at 30 MCi (Machta and Harley, 1969). Due to delayed formation of ^{137}Cs in detonations, it is associated with smaller particles which are injected into the stratosphere (Davis, 1963). Stratopheric residence time is six months to three years. During this period radioactive debris is subject to atmospheric circulation patterns in which air enters the stratosphere primarily at the equator and descends in temperate and polar latitudes during the spring (Davis, Hanson, and Watson, 1963). Spring peaks in fallout have been observed by several investigators (Parker and Crookall, 1961; Stewart _et al_., 1957). Accidental venting from continued underground testing subsequent to the moratorium has resulted in injection of fission and activation products into the atmosphere. Nuclear detonations for the purpose of excavation is an additional potential source of atmospheric ^{137}Cs. The Sedan test (1962) which produced a crater 1,200 ft. wide and 320 ft. deep released an estimated total activity of 2 kCi (Barth, 1967).

Radionuclides are released from reactors both as fission and activation products. Contamination is primarily due to neutron activation of elements in the coolant as well as corrosion products in the reactor cooling system. Small quantities of fission products may enter coolant systems from leakage of fuel elements. Most power reactors recirculate coolant which prevents chronic release of nuclides to the environment. Fission and activation products are removed continuously by purification systems (ion exchange). Nuclides not readily removed by these systems (tritium, xenon, and krypton) are released to the environment. During operation, however, reactor fuel is replaced periodically. Replaced fuel elements are usually stored in cooling ponds for a period before shipping to a processing plant. During this interval activation and fission products (including ^{137}Cs) enter the cooling pond water and are released to surface or ground waters.

A substantial release of nuclides from reactor accidents is a distinct possibility. The Windscale accident of 1957 released 600 Ci of ^{137}Cs (Garner, 1971). Beattie and Bryant (1970) state it is a realistic assumption that in future accidents, escape of "volatiles" (including cesium) is unavoidable.

Garner (1971) concludes there are two sets of circumstances through which the biosphere may be exposed to radionuclides: (1) the "normal" introduction through chronic release from atomic energy operations and (2) introduction of large amounts of activity during a short time period from either atmospheric fallout or accidental release. The former circumstance requires a knowledge of the environmental ramifications of long-lived components of release while in the latter situation, short-lived isotopes assume an ephemeral position of great importance followed by persistent long-lived forms.

Pathways of ^{137}Cs to Aquatic Environments

Since cesium is water soluble the most significant source of ^{137}Cs to ecosystems is via the hydrologic cycle. Fallout levels have been correlated with seasonal patterns of precipitation. In many soils ^{137}Cs is firmly bound to clay minerals. Sandy soils exhibit an apparent complete fixation three years after contamination (Squires and Middleton, 1966). This high affinity of soils for ionic cesium precludes contribution to aquatic environments due to leaching. Fallout deposition in snow is generally more available to aquatic systems since snowmelt with accompanying runoff occurs before soil surfaces thaw. Contributions due to sheet erosion may be significant in localized areas, however, ^{137}Cs remains in a bound form and is incorporated into sediments of lakes or streams.

A second source of ^{137}Cs to water systems is precipitation directly on the water surface. Low level wastes may enter water from power and experimental reactors as well as fuel element processing plants. In localized situations, transport of ^{137}Cs into lakes by migratory waterfowl in the form of excreta may be significant.

Uptake

Upon entering surface waters ^{137}Cs is subject to physical-chemical-biological mechanisms occurring in the system. It is readily rendered unavailable through sorption phenomena (both adsorption and absorption). Particulate suspended matter (organic and inorganic) exhibits a strong affinity for ionic cesium through physical and chemical adsorption. Phytoplankton, zooplankton, and macrophytes also remove ionic cesium from water to a much greater extent than do their terrestrial counterparts (Pendleton, 1962; Rickard, 1967; Williams, 1960). Algal uptake was demonstrated to be almost immediate. Dead algal cells exhibited a high affinity for ^{137}Cs. Williams (1960) determined a concentration factor for dead *Euglena* cells of 16 and *Chlorella* of 418. Apparently, structural components persist in dead algal cells which adsorb cesium from solution. Kolehmainen *et al*. (1968) concluded adsorption of ^{137}Cs to clay particles with subsequent settling out was the main factor removing this isotope in turbid eutrophic waters. This mechanism along with uptake by algal cells with retention upon death and settling results in significant deposition to the sediments. Whether cesium is an essential trace element for metabolism has not been established. A metabolic function, however, may be inferred by the fact of active uptake by plants.

Uptake of ^{137}Cs by other biotic components of aquatic systems is generally thought to be by way of food. Zooplankton absorbs most of its radiocesium from food sources (King, 1964). Assimilation from food is low (~ 21%) which may partially explain the lack of an increased concentration of this isotope over levels present in food sources. Data on benthic forms are lacking. They obtain most of their cesium burden from food since they are predominantly detritus feeders. Although large quantities of clay particles are passed through the gut during feeding, the adsorbed isotope may not be available. In cattle feeding experiments Stewart et al. (1965) concluded 30% of the total ^{137}Cs, that portion bound to clay particles associated with hay, was completely unavailable to the cow. Major contributions of radiocesium to benthic forms is apparently from both autochthanous and allochthanous organic materials.

Uptake by freshwater fish is almost exclusively via food (~ 99%) as opposed to movement across the gills Kolehmainen et al., 1967). Assimilative efficiencies range widely (7-80%) depending on food type (Kevern, 1966). Variable efficiencies are very significant in considering movement of ^{137}Cs through food chains. It may help in explaining the build-up or lack of build-up through trophic levels.

Metabolism

Metabolically, the behavior of cesium is frequently compared to that of its chemical analogue, potassium. Levels of ^{137}Cs in micro- and macro-aquatic plants is regulated by concentrations of the isotope and of potassium in the water surrounding them (Kolehmainen and Nelson, 1969). Stable cesium, at concentrations found in aquatic environments, does not show a carrier effect on ^{137}Cs (King, 1964). At high concentrations (natural levels x 10^3) stable cesium resulted in decreased concentrations of ^{137}Cs in *Euglena intermedia*. This was interpreted, however, as an isotopic dilution effect (Williams and Swanson, 1958). An inverse relationship between potassium content in water and ^{137}Cs content in aquatic plants has been demonstrated (Kolehmainen et al., 1967). Pendleton et al. (1965) described an increased Cs/K ratio at higher trophic levels implying a differential retention time of the two elements. Nelson (1969), utilizing a double tag (^{134}Cs and ^{42}K) on white crappie, observed no ^{42}K excretion in five days while 26% of the ^{137}Cs was excreted in three days. He concluded K was retained more efficiently than Cs under identical diet conditions. Nelson's results imply that increased ^{137}Cs content at higher trophic levels is probably due to differences in assimilative efficiencies and food habits rather than differential excretion rates for Cs and K as indicated by Pendleton *et al*. (1965).

Amplification of ^{137}Cs in Aquatic Food Chains

Increased ^{137}Cs content at higher trophic levels has been demonstrated by numerous investigators (Pendleton, 1962; Gustafson, 1967; Gallegos and Whicker, 1968). Other studies (Kevern, 1966; Kevern and Griffith, 1966; Kolehmainen and Nelson, 1969) are inconsistent with the concept of amplification of ^{137}Cs content at higher trophic levels. Pendleton (1962) has postulated a concentration factor of 3 per trophic level. Gustafson (1967) found pike (*Esox lucius*) contained 4.81 times as much ^{137}Cs as perch (*Perca flavescens*) while perch were 1.85 times higher than small fish they were eating. He also found, however, concentrations of radiocesium in whitefish (*Coregonus* sp.)

and sheepshead (Aplodinotus sp.) to be higher than in perch (P. flavescens) (1.46 x and 2.89 x respectively).

Gustafson's results (1967) point to the influence of feeding habits on ^{137}Cs accumulation in freshwater fishes. Hannerz (1968) found 3-4 fold differences in the radiocesium concentration in several whitefish species in the same lake. Those forms which were planktivorous were consistently higher than bottom feeders. Gallegos and Whicker (1968) found levels of ^{137}Cs in trout to be about twice that found in amphipods they fed on. Kevern (1966) found no significant differences between ^{137}Cs levels in carp and food sources. Kevern and Griffith (1966) observed slightly higher ^{137}Cs activity in bluegill than in gizzard shad or largemouth bass, but the differences were not significant. Kolehmainen and Nelson (1969) found no relationship between ^{137}Cs content and trophic level. Gizzard shad and golden shiners contained similar levels of the isotope (47.03 and 62.61 pCi/g) as largemouth bass (52.75 pCi/g). Their results are partially explained by the time of year samples were collected.

It is apparent from the above studies that trophic level increases of ^{137}Cs in aquatic ecosystems is not a universal principle. The trophic level effect appears to be demonstrable in aquatic environments with relatively low turbidity and those which would be considered oligotrophic.

As stated by Kolehmainen and Nelson (1969), body burden for an organism is influenced by four factors: (1) assimilation, (2) feeding rate, (3) concentration of ^{137}Cs in food and (4) biological half-life. Any of these factors could account for the presence or absence of a trophic level increase in radiocesium content. Levels of ^{137}Cs have been positively correlated with size for various species (Hannerz, 1966; Kolehmainen and Nelson, 1969). This may partially account for apparent trophic level increases since in predatory food chains the size of the organisms is greater at higher trophic levels.

Specific Activity

Specific activity is defined as the ratio of a radionuclide to the total quantity on that element (Nelson and Kaye, 1971). The underlying assumptions and constraints governing the use of specific activity in aquatic systems are: (1) radionuclide and stable nuclides of the element are equally available. This implies they are completely mixed and in the same chemical form, (2) the various nuclides of an element are metabolically indistinguishable by the organisms being considered, (3) organisms are at equilibrium with their environment, and (4) the physical half-life of the radionuclide is significantly longer than the equilibration time for organisms in a given system.

The specific activity concept has been used successfully to predict radionuclide levels in organisms subject to chronic exposure (Preston, 1967; Nelson, 1967, 1969). Other investigators have found specific activity to vary two to four fold in aquatic systems (Bortoli *et al.*, 1967; Merlini, 1967; Kolehmainen and Nelson, 1969). The need for future research in this area is very apparent in order to ameliorate information on specific activity in aquatic systems and develop its potential as a predictive tool.

APPENDIX C

WINTERGREEN LAKE

APPENDIX C

WINTERGREEN LAKE

Wintergreen Lake is located in Kalamazoo County, southwestern Michigan and is one of the many pit lakes in an outwash plain of the Kalamazoo morain (Hough, 1958). The watershed (240.8 ha.) is undulating glacial till covered by well drained sandy loam.

Mean annual percipitation is 86.3 cm. Snowfall averages 129.5 cm per year. High rainfall periods are April-June (28.2 cm) and August-October (24.7 cm) (Climatological data--U.S. Weather Bureau).

Morphometric and chemical data are presented in Tables C-1 and C-2 respectively. Inflow to Wintergreen consists of surface runoff and intermittent flow from a stream originating in dairy farm feed lots. During periods of high lake levels, water flows into adjacent Gull Lake through an outlet located on the west side of Wintergreen (Figure C-1).

Sediments range from pulpy peat to sapropel (Ruttner, 1953). Extensive marl ($CaCO_3$) deposits are found over the entire lake to a depth of 3.5 meters.

Figure C-1. Hydrographic map of Wintergreen Lake with associated ponds utilized by waterfowl.

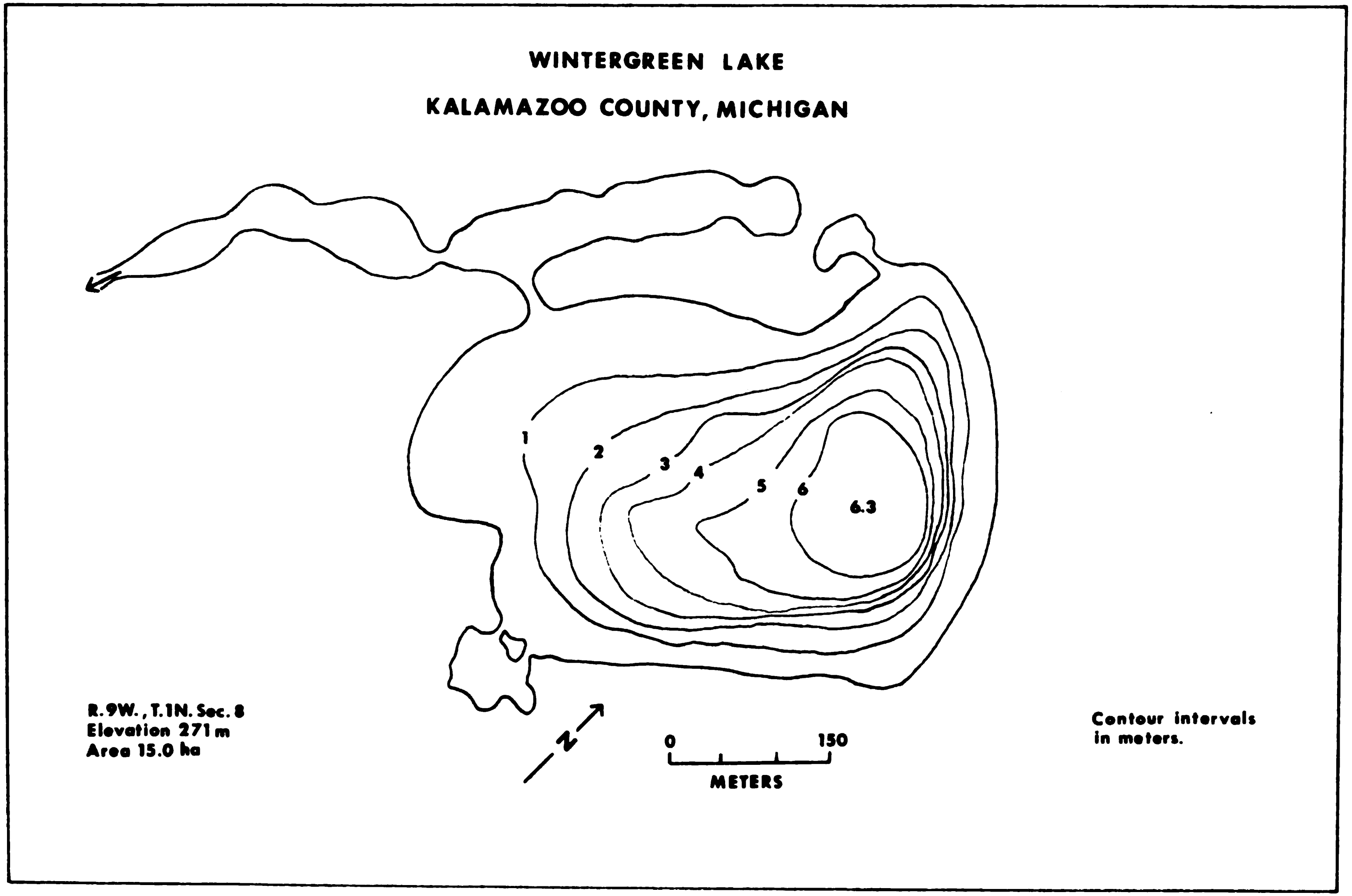
WINTERGREEN LAKE
KALAMAZOO COUNTY, MICHIGAN
1
2
3
4
5
6
6.3
R.9W., T.1N. Sec. 8
Elevation 271 m
Area 15.0 ha
N
0
150
METERS
Contour intervals in meters.

Figure C-1

Table C-1. Morphometric parameters for Wintergreen Lake.

Length (m)	544
Width (m)	375
Area (ha)	14.98
Volume (m^3)	530,584
Mean Depth (m)	3.5
Maximum Depth (m)	6.3
Shoreline Development	1.15

Table C-2. Annual range of chemical parameters at 1.0 m depth.[a]

NO_3 (mg/liter)	0.005-1.340
NO_2 (mg/liter)	0.012-0.040
NH_4 (mg/liter)	0.005-2.320
Total dissolved P (mg/liter)	0.02 -0.13
Total dissolved C (mg/liter)	3.0 -9.1
pH	7.3 -9.5
Alkalinity ($CaCO_3$) (mg/liter)	100-180
Specific conductance (μmhos)	230-280
Annual mean pelagic productivity. (mg C/m^2/day)	> 1200

[a] Data from Manny, 1971.

Percent volatile residue increases with depth to three meters. At depths of three meters or more, volatile residue is relatively uniform (Table C-3).

Submergent, emergent and floating aquatic macrophytes cover 70% of the lake to a depth of approximately three meters. Dominant submergent forms include *Ceratophyllum demersum* L., *Myriophyllum exalbescens* Fernald, *Najas flexilis* (Willk.), *Potamogeton foliosus* Raf., *P. pectinatus* L. *Nuphar advena* Aiton, is the principle emergent form. Floating macrophytes include *Lemna minor* L., *Spirodella polyrhiza*, and *L. wolfia*.

Mean annual pelagic productivity is in excess of 1,200 mg C/m^2/day (Manny, 1971). The productivity, macrophytic standing crop, dissolved oxygen regime (prolonged hypolimnetic anoxia), high organic content of sediments, and pronounced diurnal oscillations of pH in Wintergreen Lake fit the description of other hypereutrophic lakes (Wetzel, 1966; Sloey, 1970). Extensive blooms of the blue-green algal forms, *Aphanizomenon flos-aqae* and *Microcystis aeruginosa* occur regularly each spring.

The contribution of carbon, nitrogen, and phosphorous (particulate and dissolved) by waterfowl as excreta is the most important nutrient source to the lake. Canadian geese (*Branta canadensis interior Linn*.) use

Table C-3. Percent volatile residue of sediments from Wintergreen Lake.[a]

Depth (m)	9/70	5/71
1.5	14.7-15.5	13.0-13.9
3.0	19.4-19.5	--
4.0	10.3-10.5	20.0-21.5
5.0	19.3-19.7	19.0-19.8
6.0	--	19.0-19.7

[a] American Public Health Association, 1971.

Wintergreen Lake as a stopping point on both north and southbound annual migratory flights. The input of excreta per year from Canadian geese is approximately 20 metric tons (dry weight).

Table C-4 presents estimate of annual inputs of nitrogen and phosphorous to the lake from goose droppings. Other migratory waterfowl also use the lake during migration with contributions of nutrients. Based on estimated nutrient input, Wintergreen Lake may be described as "guanotrophic" (Leentvaar, 1967).

Zooplankton are abundant, occasionally reaching a density great enough " . . . that they present the appearance of a false sandy bottom to the eye" (Fetterolf, 1952). Principal forms include <u>Daphnia galeata mendotae</u>, <u>D. pulex</u>, <u>Bosmina longispina</u>, and <u>Chaoborus</u> sp. larvae. The latter exhibit a diurnal pattern in vertical migration resulting in great numbers present in the upper strata at night.

A large, diverse benthic community is present in the lake, especially in the littoral area where macrophytic growth provides a very diverse habitat. Studies of the macroinvertebrate populations of the lake are conducted each year by limnology classes taught at the W. K. Kellogg Biological Station. Schreibner (1958) has also studied the benthic community of the lake.

Table C-4. Estimate of annual nutrient input to Wintergreen Lake from Canadian geese (*Branta canadensis interior* Linn.)

Adjusted goose days	143.000[a]
Excreta/goose/day (g dry wt)	142[b]
% N	1.42[b]
% P	0.37[b]
Input of excreta (kg/ha/yr)	1,269
Nitrogen input (kg/ha/yr)	18
Phosphorous input (kg/ha/yr)	4.7

[a] Adjusted to time actually spent on lake--compiled by W. J. Johnson.

[b] Data from Kear (1962).

Previous studies of the populations were conducted by Fetterolf (1952). A summary of fish species reported for Wintergreen Lake is presented in Table C-5. Although several species of shiners and bluntnose minnows are reported from earlier studies, none were collected during the present study. The more advanced stage of eutrophy exhibited in the lake may explain their absence.

Table C-5. Species of fishes reported from Wintergreen Lake.

Micropterus salmoides (Lacepede).	Largemouth Bass
Lepomis cyanellus Rafinesque.	Green Sunfish
Lepomis macrochirus macrochirus Rafinesque.	Common Bluegill
Lepomis gibbosus (Linnaeus).	Pumpkinseed
L. cyanellus x *L. macrochirus*	Greensunfish x Bluegill
L. gibbosus x *L. macroshirus*	Pumpkinseed x Bluegill
Amia calva Linnaeus.	Bowfin
Erimyzon oblongus (Mitchill).	Creek Chubsucker
Notropis heterodon (Cope).	Blackchin Shiner[a]
Notropis heterolopis heterolopis Eigenmann and Eigenmann.	Northern Blacknose Shiner[a]
Notropis cornutus chrysocephalus (Rafinesque).	Central Common Shiner[a]
Notemigonus crysoleucas auratus (Rafinesque).	Western Golden Shiner[a]
Pimephales notatus (Rafinesque).	Bluntnose Minnow[a]
Ictalurus natalis (LeSueur).	Yellow Bullhead
Ictalurus nebulosus (LeSueur).	Brown Bullhead
Esox americanus vermiculatus LeSueur.	Mud Pickerel[a]
Perca Flavescens (Mitchill).	Yellow Perch
Ambloplites rupestris (Rafinesque).	Rock Bass[b]

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[a] Reported from earlier surveys but not collected by author.

[b] Single specimen collected.