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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Ecology

January—December 1976

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ARGONNE NATIONAL LABORATORY, ARGONNE, ILLINOIS

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RADIOLOGICAL AND ENVIRONMENTAL RESEARCH DIVISION ANNUAL REPORT

Ecology

January through December 1976

R. E. Rowland, Division Director
D. N. Edgington, Section Head

Preceding Report: ANL-75-60, Part III, January-December 1975

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FOREWORD

During 1976, programs of the Ecological Sciences Section have continued essentially at the level of the previous year. Efforts have been almost equally divided between studies of pollutants, such as sulfur dioxide and trace elements entering the environment from the combustion of fossil fuels, and investigations of radionuclides, which could be discharged from nuclear energy facilities.

The format of this report reflects the changes initiated last year: individual contributions have been shortened, and the emphasis has been shifted to the presentation of current results. More complete accounts of research activities will be presented in the open literature and in occasional topical reports which will be prepared as particular projects are completed.

In the past year, a new field program has been initiated to determine the effects of atmospheric pollutants on the viability of midwestern cereal crops. Stress responses of soybeans exposed to elevated levels of sulfur dioxide are being compared for several varietal types and several soil nitrogen fertility levels.

In the Great Lakes Program, investigations of the effects of coal combustion products on plankton and of the effects of once-through cooling on fish have continued. A comparative laboratory study of cadmium uptake by two important Lake Michigan diatoms has found a factor of two difference in cellular accumulation and variable effects on growth rate between the two species. Short-term field experiments using a new in situ technique and longer-term laboratory experiments have documented zooplankton population and community responses to cadmium stress. Studies of cooling-system impact have focused on both the direct effects on Lake Michigan fishes at water intakes and in thermal discharges, and the less direct relationships between thermal plume exposure of salmonids, rates of food consumption, energetics, and toxic material accumulation.

Results from studies of the biogeochemical behavior of plutonium in the Miami River basin and in the Great Lakes are quite similar. This suggests that the environmental behavior of plutonium is influenced very little by either isotopic composition or source since that found in the Miami River primarily originates from a chemical processing plant and is largely ^{238}Pu, while that found elsewhere is ^{239,240}Pu from weapons fallout. A particularly interesting result is that plutonium has apparently migrated to greater depths in undisturbed soil columns than had been expected.

Preliminary results from studies of sedimentation and trace-element distributions in Lake Michigan indicate that only a small proportion of the elements in the periodic table are enriched in recent sediments (≤ 100 years old), and that the observed annual deposition of many of these elements in the sediment approximately equals the estimates of annual inputs.

04475

A PRELIMINARY STUDY OF VARIETAL SUSCEPTIBILITY TO SULFUR DIOXIDE

J. E. Miller and P. B. Xerikos

The injury response of plants to air pollutants, such as sulfur dioxide, is known to vary in severity and type for different varieties or cultivars of a species. Differences in the susceptibility of soybean varieties to sulfur dioxide have previously been noted,[1] but sufficient information is not available concerning the sulfur dioxide resistance of varieties commonly grown in the Midwest. The following report considers the results of preliminary experiments concerning acute sulfur dioxide effects on 12 soybean varieties.

The soybeans were grown from seed in a commercial greenhouse for eight weeks and then exposed to approximately 4.5 ppm of sulfur dioxide for two hours. The exposure chamber was similar to that previously described.[2] The light intensity in the chamber ranged between 700 and 1000 foot candles at plant height, and the chamber air temperature was 28°C. The plants were graded for foliar injury five days after exposure by estimating the percentage of leaf area injured for leaves less than and greater than 20 mm in length.

The injury symptoms were similar to those previously reported[1] and ranged from cream colored necrotic lesions (generally on younger leaves) to a reddish brown necrotic stipling (on older leaves). Differences in the severity of symptom development for the varieties was evident on both the younger and older leaves (Table 1). No injury was apparent with three of the varieties (Amsoy 71, Corsoy, and Hark). Considerable variation in symptom development was also obtained within varieties, and thus the data were not highly significant, although it is clear that several of the varieties tended to be more susceptible. Overall, the younger leaves (less than 20 mm) elicited more injury, the only exceptions being with the York and Wayne varieties.

Table 1. Foliar injury scores for soybean cultivars exposed to 4.5 ppm sulfur dioxide for two hours

Cultivar	Percentage leaf area injured[a]	
	Younger leaves (< 20 mm length)	Older leaves (≥ 20 mm length)
Harcor	72	8
Beeson	64	0
Hodgson	64	8
Wells	64	32
Woodworth	56	0
York	56	80
Lee	24	16
Peking	16	0
Hark	0	0
Amoy 71	0	0
Wayne	0	32
Corsoy	0	0

[a] Values are the mean scores for fifteen plants.

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04476

DIURNAL PHOTOSYNTHESIS AND STOMATAL RESISTANCE IN FIELD-GROWN SOYBEANS

J. E. Miller, R. N. Muller, and P. Seegers

The process of photosynthesis in green plants is the major determinant of crop yield. Although the effects of air pollutants, such as sulfur dioxide, on photosynthesis has been studied, many unsolved questions remain. This is especially true with regard to reduction of photosynthetic rate under conditions of chronic exposure causing little or no visible injury. It was the purpose of these studies to develop techniques suitable for measuring photosynthetic rates of field-grown plants without dramatically altering the microenvironment of the plants. In addition, attempts were made to define other parameters that modify field photosynthetic rates so that the long-range goal of assessing sulfur dioxide effects might be realized.

Gross photosynthetic rates of soybeans (Glycine max. cv. Wayne) in the field were measured by exposing a small section of representative leaves for 30 seconds to $C^{14}O_2$ in a normal atmospheric mixture by a technique similar to that of Incoll and Wright.[1] A 1-cm^2 section of the area exposed to $C^{14}O_2$ is punched from the leaf and processed for liquid scintillation counting. Since the treatment period is of such short duration, there is little photorespiratory loss of $C^{14}O_2$, and thus, the amount of C^{14} fixed in the leaf can be related to the gross photosynthetic rate. Other parameters measured during the course of these experiments were stomatal resistance, light intensity, leaf water potential, and air temperature.

The course of gross photosynthesis for young soybean leaves at the top of the canopy on a typical day is shown in Figure 1B. A maximum rate of approximately 34 mg $CO_2/dm^2/hr$ occurred at 10:30. This compares favorably to previously reported net photosynthesis values of 18 mg $CO_2/dm^2/hr$ and 35 mg $CO_2/dm^2/hr$ for the same variety.[2,3] The photosynthetic rate was generally a function of light intensity, as illustrated by the morning rise and evening decline. However, a sharp reduction in photosynthesis was often noted by late morning or early afternoon, even though the light intensity had not yet

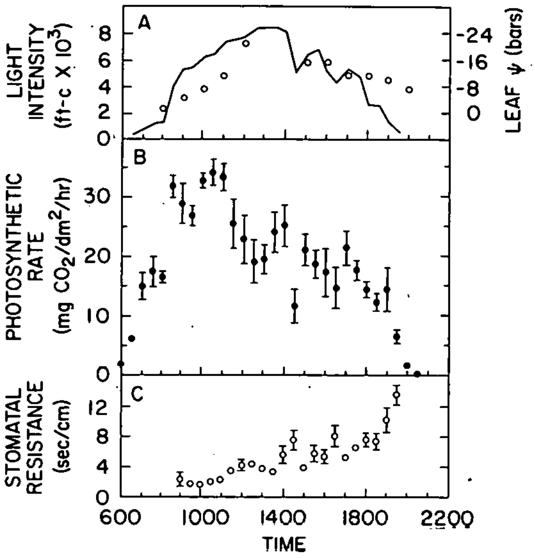

FIG. 1.--The daily course of photosynthesis, stomatal resistance, leaf water potential (ψ) and light intensity in a soybean field.
(ANL neg. 149-77-74)

reached a maximum value. This reduction was attributed to leaf water stress (Fig. 1A) which caused a partial closing of the stomates, as indicated in Fig. 1C, thus reducing gas exchange by the leaves. Boyer[4] found a comparable reduction in net photosynthesis of soybeans at similar levels of leaf water stress. The low photosynthetic value at 14:30 was due to reduced light intensity from a passing cloud and is also reflected in the stomatal resistance value at that time (Fig. 1C). The data indicate that light intensity and leaf water potential account for most of the daily variability in the photosynthetic rate of soybean leaves. Considerable inherent biological variability is also evident, and thus, intensive sampling is necessary.

A study of gross photosynthetic rate as a function of height in the plant canopy indicated a rapid decrease with depth in the canopy. The normalized photosynthetic rates from top to bottom were 1.0, 0.6, 0.3, and 0.1 for four height classes in the canopy. The primary reason for this is shading, although the lower and thus older leaves probably have reduced photosynthetic efficiencies as well. Measurements of leaf area index, vertical leaf area distribution and plant density allowed calculation of the total gross photosynthesis per unit area. A value of 804 kg CO_2/hectare/day was obtained for 7/28, which compares favorably with values obtained previously for maize, sugarcane, and sorghum.[5]

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04477

INFLUENCE OF NITROGEN FERTILITY LEVELS ON SO_2 FUMIGATION RESPONSE IN SOYBEANS

R. N. Muller and J. E. Miller

While species and varieties of crop plants exhibit inherent differences in susceptibility to air pollutant damage, the extent of damage may be significantly influenced by the environmental conditions of growth. In crop plants, where fertility levels of different sites or different farming practices vary considerably, nutrition may strongly influence the degree of damage. This study was designed to assess the interaction of nitrogen fertility levels and sulfur dioxide exposure upon photosynthesis of soybean (*Glycine max* c.v. Amsoy 71).

Plants were grown in pots of washed sand (3 plants per pot, 3 pots per treatment) and watered every three days with 1000 ml of 1/4 strength Hoagland's solution.[1] The four nitrogen levels consisted of 13, 26, 53, and 210 ppm NO_3-N. The plants were grown for 7 weeks and fumigated for 2 hr with 2 ppm SO_2. Stomatal diffusive resistance was measured just prior to fumigation and immediately after. Gross photosynthesis was measured just prior to fumigation and two days after by a procedure similar to that of Incoll and Wright.[2] Significant deviations of postfumigation photosynthetic rates and stomatal resistance from prefumigation values were determined by t-test comparison of the means.

The dry weight of plant material showed no significant difference between treatments and averaged 13.2 g per pot. However, a significant difference was observed between the nitrogen treatments in the proportion of total weight contained in the roots as opposed to leaves and stems. Plants in the low-level treatment contained 61% of the total biomass as roots, while in all other treatments the percentage of biomass as roots averaged 43%. Most of the decreased shoot biomass in the low nitrogen treatment was accounted for by decreases in the photosynthetic tissue rather than the stems of the plants. This reduction of photosynthetic biomass in the low nitrogen treatment suggests that the ultimate yield (both total biomass and seed production) of the mature plants would be considerably reduced.

No visible injury due to any treatment could be observed two days after fumigation. Gross photosynthesis showed no significant differences between treatments prior to fumigation and ranged from 6.0 to 6.9 mg $CO_2/dm^2/hr$ (Table 1). These rates are considerably lower than the maximum photosynthetic rates obtained for field grown plants in full sunlight[3] and would be accounted for, in part, by the lower light levels of the fumigation chamber (~900 ft-c). The lowest nitrogen treatments (13 and 26 ppm) exhibited a significant reduction in photosynthetic capacity two days after fumigation ($p < 0.05$). The greatest reduction was observed in the 13 ppm treatment which fixed CO_2 at 57% of its prefumigation rate, indicating that, while no visible injury could be observed, permanent biochemical damage had occurred.

Stomatal resistance at the three lowest nitrogen levels exhibited no change with fumigation (Table 1). A significant reduction in resistance was observed with the 210 ppm nitrogen treatment after fumigation. This reduction in stomatal resistance has been observed elsewhere[4,5] and appears to be a common physiological reaction to low levels of SO_2. In healthier plants stomatal opening induced by low-level sulfur dioxide fumigation may lead to greater SO_2 uptake over a long exposure term and may ultimately induce greater damage than was observed under the conditions of this experiment.

Table 1. Photosynthetic rates and stomatal resistance for soybean (Amsoy-71) prior to and after fumigation with 2 ppm SO_2 for two hours. Values given are means of 12 observations ± 1 standard deviation. Significant deviations from prefumigation values are indicated.

Nitrogen level, ppm	Photosynthetic rate, mg $CO_2/dm^2/hr$		Stomatal resistance, sec/cm	
	Before	48 hr after	Before	Immediately after
13	6.1 ± 1.2	3.5[a] ± 1.2	15.8 ± 4.1	16.7 ± 7.8
26	6.9 ± 1.1	5.0[a] ± 1.0	14.3 ± 4.6	12.1 ± 3.3
53	6.1 ± 0.9	4.9 ± 0.8	11.3 ± 2.1	10.1 ± 2.1
210	6.0 ± 1.3	4.8 ± 0.5	12.9 ± 2.9	10.6[b] ± 1.1

[a] $p < 0.05$.

[b] $p < 0.025$.

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04478

THERMAL PLUME RESIDENCE AND TEMPERATURE EXPOSURE OF SALMONID FISHES

S. A. Spigarelli, G. P. Romberg, M. M. Thommes and W. Prepejchal

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Typical cooling-system discharges present two potential sources of stress to fish: (1) exposure to elevated temperature and/or temperature shock and (2) exposure to toxic residual chlorine compounds. In order to concentrate our efforts on temperature as a directive factor, we have focussed our studies at the Point Beach Nuclear Plant where no chlorination occurs. In light of our goal to characterize plume residence and temperature exposures of motile fish, we initiated a number of related projects which, in combination, yield a complete description of salmonid responses.

We developed, tested, and employed a nondestructive echo-location technique to estimate the density-distribution patterns of fish and to determine the influence of discharge design and location on fish attraction.[1] Studies were conducted between 1972 and 1975 at the Point Beach, Zion, and Waukegan power plants on Lake Michigan. Preliminary inspection of results indicates seasonal attraction of abundant species, such as alewife, trout, and salmon. In general, fish densities in the plume area tend to be elevated relative to unheated areas during spring and early summer, as can be seen in Figure 1. Power plant location and discharge type apparently affect the magnitude and timing of attraction to discharges. Fish in plume areas generally are observed at elevated temperatures or near temperature interfaces. Data analyses include conventional approaches to detect differences in mean densities over time and space and recent developments in time-series analysis. Predictability of fish responses will depend on the identification of temporal and spatial distribution patterns.

Results of mark and recapture studies show that most salmonid species in Lake Michigan do orient to thermal discharges, but residence times within a particular discharge area seem to be limited to a few days or weeks between April and November (we have no winter tagging data).[2] Limited recaptures of lake trout indicate (1) short-term plume residence during periods of low water

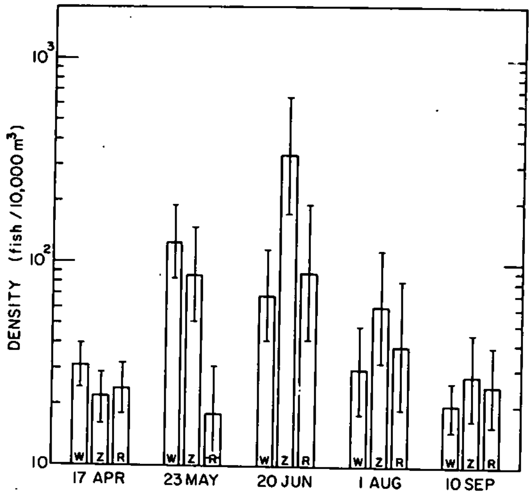

FIG. 1.--Total densities of fish in plume and reference areas at Zion (Z), and Waukegan (W) power plant sites, and reference location (R) on Lake Michigan (acoustic survey technique). Vertical lines represent 95% confidence interval on density.
(ANL neg. 149-77-340)

temperature (cool seasons and upwellings); and (2) inshore-offshore migration patterns that reduce the potential for multiple plume encounters. Other trout species (brown, rainbow, and brook) migrate in shore-parallel directions and encounter numerous thermal discharges (e.g., approximately 50% of total recaptures were made at power plants). Coho and chinook salmon frequent southern basin thermal discharges in the spring, and chinook are commonly found near most discharges in the fall (20% total chinook recaptures at power plants).

Studies employing TLD tags[3,4] show that "plume resident" trout and salmon do not acclimate to maximum discharge temperatures, but maintain integrated acclimation states that approximate optimal metabolic temperatures. Of the species tested, brown trout consistently show higher temperature exposures than do rainbow trout or chinook salmon.

In the course of our sport fishing census at Point Beach,[5] body temperature measurements were made on the majority of fish caught.[6] Body temperatures of fish feeding in this discharge area show considerable variation, ranging from ambient to discharge temperature, and the majority of fish have acclimation states elevated above ambient temperature (i.e., body temperatures exceed avoidance temperatures expected for ambient acclimation).

The results of TLD studies and body temperature measurements are complementary in that both substantiate thermoregulatory behavior by these fishes and provide estimates of integrated acclimation states. A summary of results from these studies during fall seasons of 1972—1974 is given in Table 1. Two important concepts are apparent from these data: (1) trout and salmon that frequent thermal discharge areas attain body temperatures up to maximum avoidance temperatures but tend to "regulate" below final preferred; and (2) excess temperatures (body minus ambient) vary, depending on ambient conditions, i.e., excess body temperatures are highest when ambient temperatures are low.

Following development and testing of a fish tracking system (radiotelemetry)[7] we initiated studies to determine salmonid thermoregulatory responses under variable conditions.[8] Results from telemetry studies indicate diel and seasonal patterns of plume and ambient temperature exposures. An example is shown in Figure 2. This and other tracks demonstrate a number of important

Table 1. Mean body temperatures (T_b) and excess body temperatures (T_b-$T_{ambient}$). Integrated exposure temperautres (T_e) and excess exposure temperatures of salmonid fishes at Point Beach, °C.

	Brown trout	Rainbow trout	Brook trout	Lake trout	Coho salmon	Chinook salmon
1972						
T_b	17.9	14.3	15.2	11.9	17.3	15.7
Excess T_b	5.2	4.5	5.2	3.7	3.8	5.7
n	4	67	16	19	2	1
1973						
T_b	15.7	15.7	16.0	11.6	16.3	17.7
Excess T_b	5.0	3.3	4.2	1.9	5.9	4.6
n	56	370	19	10	28	24
T_e	16.1	14.0	—	—	—	15.5
Excess T_e	1.9	2.1	—	—	—	1.8
n	10	5	—	—	—	4
1974						
T_e	14.7	11.8	—	—	—	—
Excess T_e	6.3	3.4	—	—	—	—
n	11	20	—	—	—	—

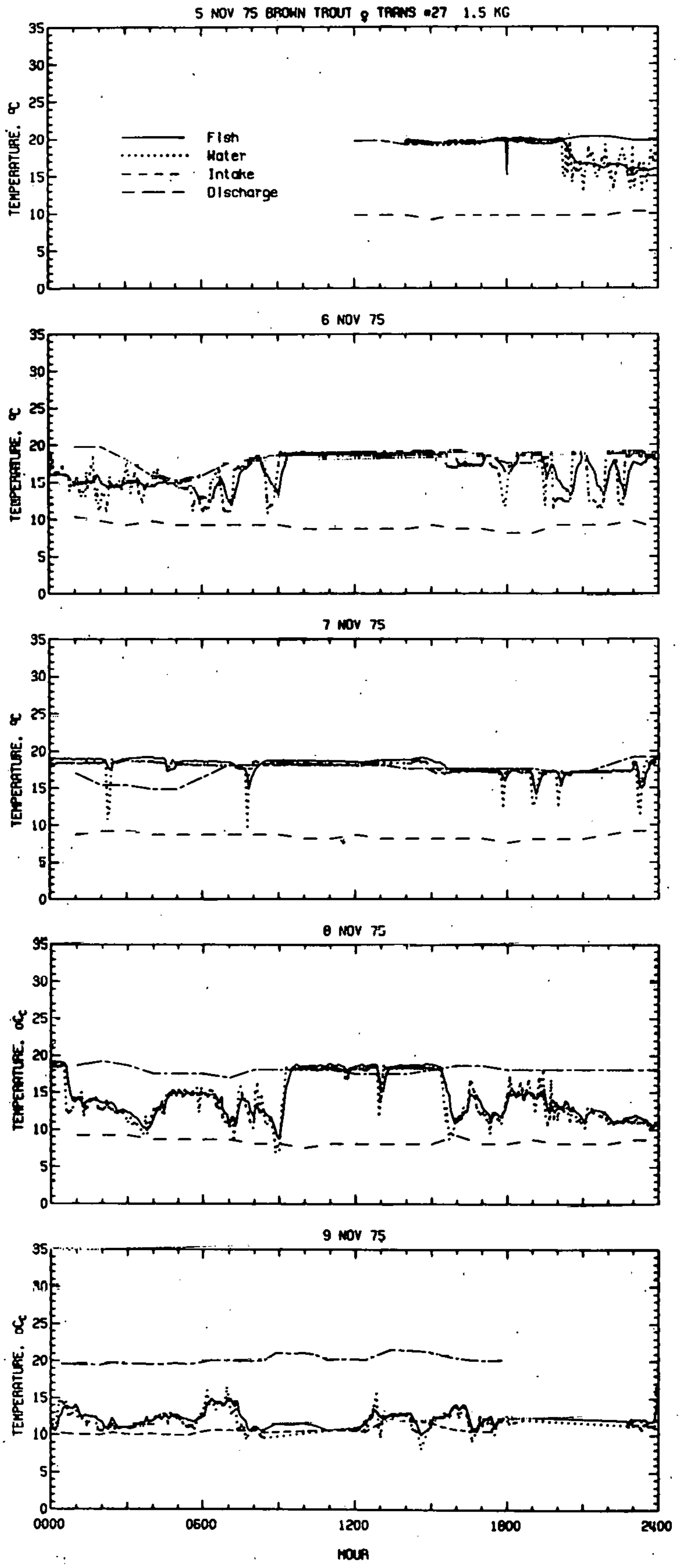

FIG. 2.--Water temperature and body temperature records for plume "resident" brown trout at Point Beach (radiotelemetry technique). (ANL neg. 149-77-341)

concepts about fish thermoregulatory behavior: (1) plume "resident" salmonids do not acclimate to maximum discharge temperature, even when that temperature is below upper avoidance; (2) thermal acclimation is "cyclic" (rather than constant) and results from a partitioning of time between ambient and various discharge temperatures; (3) plume exposure occurs during periods of high activity and by multiple excursions between cool and warm waters; (4) long periods (hours) of residence at discharge temperatures or at variable plume temperatures are common; (5) excursions into heated waters result in rapid external temperature changes as high as 10°C and, in many cases, result in body temperature changes of the same magnitude.

In combination, fish tracks provide detailed time-at-temperature exposure histories for salmonids and will allow controlled duplication of these exposures in critical experiments designed to quantify the consequences of thermal discharge residence.

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04479

FISH IMPINGEMENT AT LAKE MICHIGAN POWER PLANTS

R. K. Sharma,* R. F. Freeman,* and S. A. Spigarelli

A study was initiated in 1974 to survey the magnitude and to evaluate the impact of fish impingement at 20 power plants on the Great Lakes. Data on impingement rates, site characteristics, intake designs and operational features have been collected and analyzed.[1] Interpretive analyses of these data are in progress.

The objectives of this study are: (1) to summarize fish impingement data for Lake Michigan (16/20 plants surveyed are on Lake Michigan); (2) to assess the significance of total and source-related mortalities on populations of forage and predator species; and (3) to expand the assessment of power plant impingement to include all water intakes on Lake Michigan.

Table 1 summarizes the numbers and weights of fish impinged at power plants and other water intakes (i.e., city, municipal, industrial) for one year between 1974 and 1975. Estimates for other power plants and other water intakes were based on mean impingement rates of surveyed plants within statistical districts. An estimated total number of impinged fish of over 50 million (3 million pounds) represents a substantial operational and monitoring problem for the utilities and may constitute a serious reduction in standing crops or recruitment potential of impinged species populations. Also, there is ample evidence to show that modern large power plants which began operation after 1975 will add large incremental mortalities to the estimated totals presented in Table 1. For instance, the Zion plant impinged a total of over 19 million fish (1.5 million pounds) in 1975[2] after initiating full power operation. This total for a 2000 MWe plant is greater than the entire sum of all plants in 1974 and represents an estimated 40% increase in total impingement on Lake Michigan. When operated at full power, the D. C. Cook Plant (similar to Zion in size and design) could account for similarly large impingements and

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Table 1. Annual numbers and weights of fish impinged at surveyed power plants and estimates of impingement at all Lake Michigan water intakes (mid-1974 to mid-1975).

Source	Alewife		Smelt		Yellow Perch		Other Species		All Species	
	no.	wt, lb	no.	wt, lb	no.	wt, lb	no.	wt, lb	no.	wt, lb
Surveyed plants (16)	9,300,649 (86)	532,503 (87)	658,571 (6)	22,456 (4)	132,094 (1)	5,649 (1)	739,026 (7)	50,020 (8)	10,830,340 (-)	610,449 (-)
Other power plants (8)	2,067,455 (82)	114,897 (88)	77,763 (3)	2,195 (2)	8,066 (<1)	375 (<1)	359,495 (14)	13,233 (10)	2,512,779 (-)	130,699 (-)
Total at power plants	11,368,104 (85)	647,400 (87)	736,334 (6)	24,651 (3)	140,160 (1)	6.024 (1)	1,098,521 (8)	63,253 (9)	13,343,119 (-)	741,148 (-)
Other water intakes	31,054,859 (64)	2,025,349 (75)	4,157.014 (2)	71,758 (1)	35,010 (3)	7,067 (4)	2,276,457 (31)	132,027 (20)	37,523,340 (-)	2,236,380 (-)
Total impingement	42,422,963 (65)	2,672,749 (76)	4,893 348 (2)	96,409 (1)	175,170 (3)	13,091 (4)	3,374,978 (30)	195,280 (19)	50,866,456 (-)	2,977,529 (-)

() Percent of total.

increases in total fish losses on Lake Michigan.

Although the monitoring data provided by Lake Michigan utilities were not adequate to evaluate the significance of most operational or environmental factors on impingement rates, it is apparent that intake design has a strong influence on impingement rates. Our results indicate that plants with canal intakes show the highest impingement rates (per volume of water screened), followed by offshore open bay, and porous dike intake designs (order of decreasing rates). The most recently constructed plants on Lake Michigan (Zion and D. C. Cook) employ modifications of the offshore open bay intake design.

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04480

IMPACT OF COOLING SYSTEMS ON LAKE MICHIGAN FISHES

S. A. Spigarelli and G. P. Romberg

The preceding paper[1] summarized fish mortalities due to impingement at water intakes on Lake Michigan. A comparison of these data with available estimates of standing crop biomass, commercial and sport fishery catches, and estimated predation mortality, appears in Table 1. The striking features of these data are the proportions of total mortality due to predation and the lack of accurate basic population statistics such as standing crop biomass and natural mortality for important forage and human food fishes in Lake Michigan.

Although this preliminary assessment would indicate that power plant and total impingement losses constitute an insignificant fraction of total forage biomass, the potentially unstable forage-predator ratios and the apparent high degree of annual fluctuations (year-classes) in alewife, smelt, and perch indicate the need for a more detailed assessment of cooling-system related impact on selected populations. This assessment should include mathematical simulations of the effects of impingement and entrainment on production and recruitment of fish populations.

Table 1. Summary of available information on standing crop biomass and mortalities of three species of Lake Michigan fish (lbs).

	Alewife	Smelt	Yellow perch
Biomass estimates (1973–1974)[a]	$1-2 \times 10^9$	$>31 \times 10^6$	n.a.
Mortality estimates			
Impingement (all intakes)[b]	2.7×10^6	0.1×10^6	0.01×10^6
Commercial fishing (1975)[c]	35×10^6	1.2×10^6	0.8×10^6
Sport fishing (1976)[c]	0	2.8×10^6	1×10^6
Predation by salmonids[d]	325×10^6	13×10^6	0
Total estimated mortality	363×10^6	17.1×10^6	1.8×10^6

[a] Personal communication, E. Brown, Great Lakes Fishery Lab, Ann Arbor, Michigan.

[b] Ref. 1.

[c] Personal communication, L. Wells, Great Lakes Fishery Lab, Ann Arbor, Michigan.

[d] ANL estimates.

n.a. = not available.

Results of studies designed to characterize elevated thermal exposure of salmonids could have significant bearing on the assessment of cooling-system impact on forage species. Our analysis indicates[3] that a significant fraction of Lake Michigan salmonids experience elevated temperature exposures for variable periods during an annual cycle. By employing a set of rather conservative assumptions about (1) total salmonid standing crop (40×10^6 lbs), (2) forage-to-predator conversion ratios (6/1), (3) mean annual growth increments of salmonids (1.5 lbs/yr); and (4) thermal exposure factors (based on adjusted Q_{10} and measured exposures), we estimate that total annual forage consumption by salmonids in Lake Michigan ($\sim 430 \times 10^6$ lbs) is increased by 3% (13×10^6 lbs) owing to increased feeding by thermal plume resident salmonids (assuming equal growth at elevated temperatures). This incremental predation (mortality) of forage fishes is four times higher than losses due to all water intakes.

It is certain that free-swimming salmonid fishes regulate their metabolic state by partitioning time between heated and ambient waters. Active regulation of this type must confer physiological advantage to individual fish in terms of energy expenditure. But the fact that integrated acclimation states of salmonids are elevated over ambient by as much as 9°C (average 3–5°C) indicates the possibility of serious effects on plume resident fish and the entire fishery resource. Despite inherent sampling problems and assumptions related to thermal histories of sampled fish, pilot studies indicate that intermittent plume residence can affect growth,[2] proximate body composition[3] and bioaccumulation of toxic materials[3,4] in Lake Michigan salmonids. Quantification of these effects is extremely difficult by the previous methods of tagging (TLD) and recollecting fish for comparative analyses. Instead, we are studying these effects by exposing captive fish to temperature regimes which duplicate those experienced by fish in Lake Michigan.

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04481

SORPTION OF CADMIUM BY ASTERIONELLA FORMOSA AND FRAGILARIA CROTONENSIS

H. L. Conway and S. C. Williams*

Trace elements such as cadmium, which is known to be toxic at elevated concentrations, are incorporated by phytoplankton and then transferred through zooplankton to higher trophic levels and eventually to man. A subtle response by a lower trophic level organism to a particular trace pollutant may eventually reveal itself as a significant change at the higher level of fish or man. Accumulation of toxic pollutants in the biota can lead to decreased rates of growth[1] and reproduction and changes in the community structure.

The objective of the present study was to determine the sorption characteristics of cadmium by two of the major diatom species found in Lake Michigan.

Short-term (< 48 hr) sorption experiments were carried out using unialgal populations of Asterionella formosa and Fragilaria crotonensis that had been previously grown in steady-state chemostats.[2] Subsamples of the steady-state populations were placed in 2-l Purex bottles. Cells in one bottle were killed by the addition of $HgCl_2$ (35 μg/l). Cadmium (stable plus radioactive, ^{115m}Cd) was added to the live cultures to yield concentrations of 2, 5, and 10 μg Cd/l; the concentration of cadmium in the dead culture bottle was 5 μg/l. The populations were incubated under batch culture conditions,[3] and samples were taken at discrete time intervals for radioactive counting and population cell volumes.

Cellular cadmium content appeared to be a hyperbolic function of time for both species (Figure 1). Populations of A. formosa accumulated approximately three times as much cadmium as F. crotonensis populations after 24 hr of cadmium exposure (Table 1). The cellular cadmium content of the dead A. formosa cells, after 24 hours, was an order of magnitude lower than that of the live cells exposed to the same ambient cadmium concentration; for F. crotonensis

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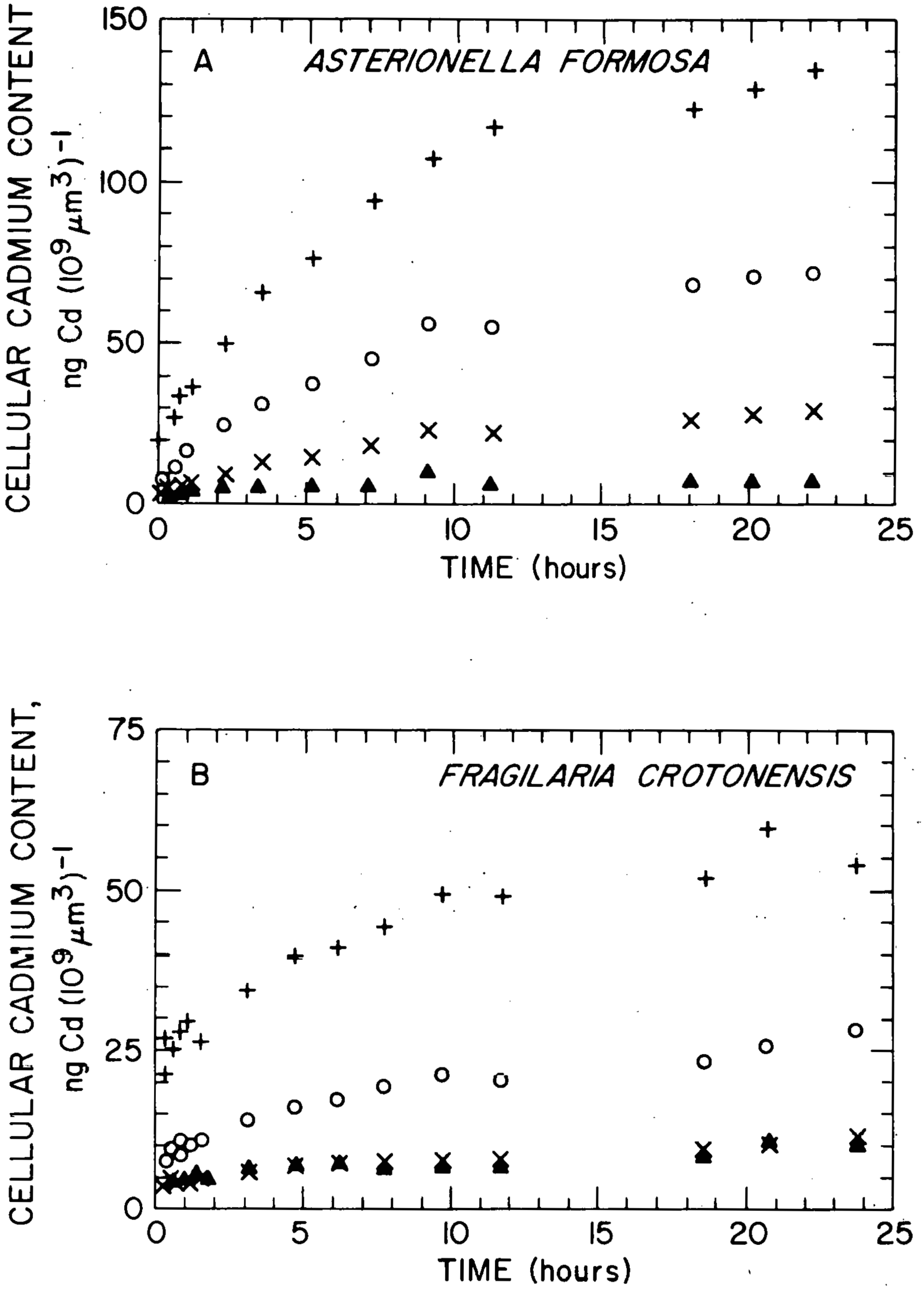

FIG. 1.--The cellular cadmium content versus time at ambient cadmium concentrations of ~ 10 μg liter^{-1} (+); ~ 5 μg liter^{-1}, live cells (O); ~ 5 μg liter^{-1}, Hg-killed cells (▲); and ~ 2 μg liter^{-1} (x). (A) *Asterionella formosa*; (B) *Fragilaria crotonensis*.

Table 1. Cellular cadmium content after 24 hours of exposure

Species	Ambient concentration, μg/l	Concentration in cells, ng/10^9 μm^3
Asterionella formosa	2	31.9
	5	86.9
	10	148
(dead)	5	7.35
Fragilaria crotonensis	2	11.2
	5	28.0
	10	56.7
(dead)	5	10.0

the live cells contained only ~ 3 times the amount of cadmium contained in the dead cells.

The cellular cadmium content of *Asterionella formosa* and *Fragilaria crotonensis* is a function of ambient cadmium concentration, the duration of cadmium exposure, and the response of the individual species. Short-term cadmium sorption appears to be an active process by the cells since the cellular cadmium content and the sorption rate of both live populations greatly exceeded those of the dead populations at the same ambient cadmium concentration. In addition, the two dead populations showed nearly the same sorption rates and cadmium content.

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04482

EFFECTS OF CADMIUM ON THE GROWTH AND ASSIMILATION OF INORGANIC CARBON BY ASTERIONELLA FORMOSA AND FRAGILARIA CROTONENSIS

H. L. Conway and S. C. Williams*

The objective of this study was to investigate the impact of cadmium exposure on the growth and utilization of inorganic carbon by Asterionella formosa and Fragilaria crotonensis.

Short-term (< 48 hr) experiments were conducted using populations that had been previously grown in steady-state chemostats.[1] Subsamples of the steady state populations were placed in 2-l Pyrex bottles, and a cadmium solution was added to the test cultures to yield 5 and 10 µg Cd/l. Cadmium was not added to the control cultures. Following a 12-hr preincubation period with the cadmium, ^{14}C was added to one of the test bottles and one of the controls. The populations were incubated under batch culture conditions,[2] and samples were taken at discrete time intervals for radioactive counting and determination of population cell volumes.

No effects were observed for F. crotonensis at either of the concentrations tested (Table 1). A. formosa showed inhibition of ^{14}C incorporation and a decline in growth rate at 5 and 10 µg Cd/l, the most pronounced effects being observed at the higher cadmium concentration (Figure 1 and Table 1).

The higher cellular cadmium concentration of A. formosa[3] is reflected in the greater inhibition of metabolic activity in this species. The reason for the difference in response of the two organisms is unknown; it may be that F. crotonensis has a mechanism to excrete this toxic element, or A. formosa may have more active sorption sites. Based on the A. formosa data at the two concentrations (5 and 10 µg Cd/l) there appears to be a relationship between the degree of suppression of normal cell function and the cellular cadmium content. Given the slower sorption of cadmium by F. crotonensis, it may be that the cellular cadmium content of these organisms never reached a high

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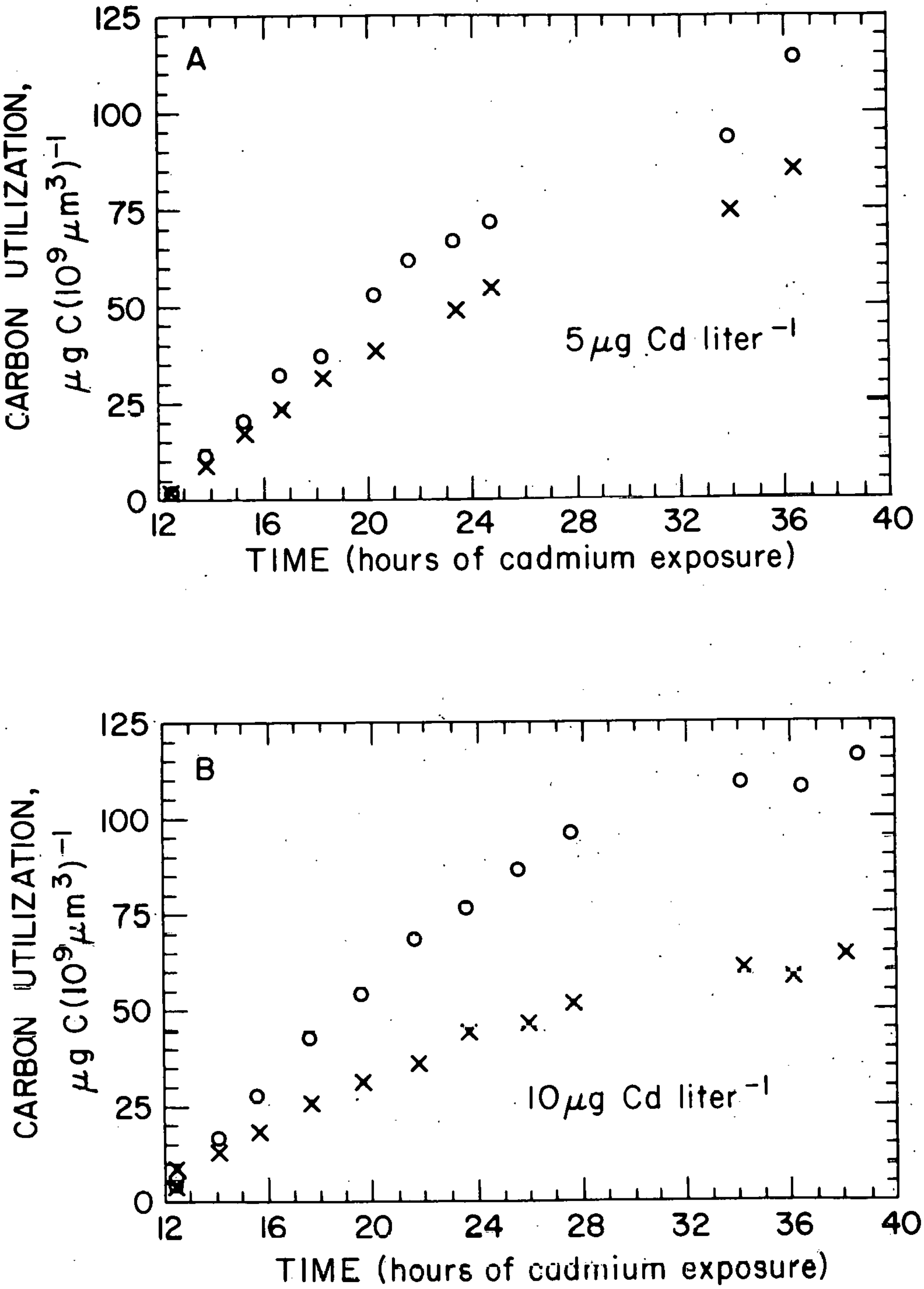

FIG. 1.--The utilization of carbon versus cadmium exposure time for the test population (x) and the control population (O) of Asterionella formosa.

Table 1. Percent reduction in growth rate and ^{14}C assimilation

Species	Ambient concentration, μg/l	Growth rate, %	^{14}C assimilation, %
Asterionella formosa	5	25	25
	10	46	45
Fragilaria crotonensis	5	n.e.	n.e.
	10	n.e.	n.e.

n.e. = no effect (no measurable difference between test and control populations).

enough level during the span of the short-term experiments to cause measurable changes in metabolic activity.

Based on the evidence presented, it is difficult to judge with certainty the metabolic process most vulnerable to cadmium exposure. Carbon-14 assimilation was affected most consistently by the cadmium in the medium and in the cells. Impairment of the vital photosynthetic process may account for the reductions in growth rate observed for A. formosa.

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04483

A METHOD FOR THE IN SITU STUDY OF POLLUTANT EFFECTS ON NATURAL ZOOPLANKTON COMMUNITIES

J. S. Marshall and R. Van Reken

The need for experimental studies of pollutant effects on whole communities and ecosystems, as well as isolated components, has been stressed recently by several ecologists.[1–3] The usefulness of in situ controlled environmental pollution experiments (CEPEX) in assessing the effects of copper on a marine zooplankton community was recently discussed by Reeve et al.,[4] who concluded that, at least in the short and intermediate term (up to 1 month) the zooplankton were probably the most affected component of the system. Although it has been possible to maintain apparently normal zooplankton communities for up to 22 months in enormous (18,000 m^2) enclosures in a small lake,[5] the use of large enclosures for experimental studies of long-term pollutant effects on plankton communities in the Great Lakes does not seem feasible. Some of the factors that would limit the use of large, transparent bags or enclosed water columns for studies of long-term pollutant effects on Great Lakes zooplankton are: (1) high unit cost; (2) susceptibility to storm damage; (3) growth of periphyton community; (4) toxicity of high light intensities to zooplankton enclosed in transparent bags.

During the summer of 1976, we developed and tested a new method for the in situ study of pollutant effects on zooplankton communities in large lakes which does not suffer the limitations of large enclosures. In each of five experiments conducted in northern Green Bay, Lake Michigan, two to four samples of lower epilimnion water for each of four levels of added pollutant (Cd) plus controls were incubated in situ in opaque, polyethylene carboys (2 to 5 gallon size) for 4 to 15 days. In each experiment 2 to 4 additional samples, which were not incubated, were also taken. Special care was taken to ensure that all of the carboys were filled with a homogeneous composite of the lower epilimnion water samples. When all the carboys were nearly filled, the numbered carboys were selected at random for addition of different concentrations of cadmium. After the cadmium was added, each carboy was thoroughly shaken and finally filled completely to exclude air bubbles. The carboys were then suspended in the epilimnion between a spar buoy and a mooring anchor, using 600 ft of 3/8-in nylon line at a bottom depth of 100 ft. At the termination of each experiment, the zooplankton in each carboy were removed by filtering through a nylon net having either 153 μm (experiments 1–3) or 85 μm (experiments 4–5) apertures, narcotized with club soda, and preserved with 5% formalin. In the laboratory, the populations of each category in each entire sample were enumerated in an open-top chambered counting cell, using a low power microscope.

The results, summarized in Table 1, indicated that the effects of enclosure, as measured by changes in species populations in the incubated control samples were mostly insignificant, even after 15 days. This was shown by the relatively high frequency of insignificant differences ($P < 0.05$) in the results of t-tests by two methods of comparison: (1) 80 tests of the differences between the mean populations of each species or category in the incubated control vs. the unincubated initial samples taken at the beginning of each experiment; and (2) 48 tests of the differences between the means in the incubated controls of experiments 1, 3, and 4 vs. those in the samples taken from the lake on the same (or next) day. Both methods gave similar results and showed that 58 to 67% of the differences in the species populations of cladocerans and 50 to 68% of those of the copepod categories were not significant. Most of the significant differences, on the other hand, were accounted for by relatively few species. Holopedium gibberum, Eubosmina coregoni, and immature copepods accounted for 54 to 68% of the significant decreases, while Chydorus sphaericus and Cyclops bicuspidatus thomasi accounted for 57% of the significant increases.

In contrast to the apparently small effects of enclosure, the effects of added cadmium, as indicated by the rates of increase (r) for each species in the incubated samples, were pronounced. The mean linear regression coefficients (slopes) for the relationship between average rate of increase, $r = \ln(N_t/\overline{N}_i)/\Delta t$, (where N_t is the terminal number of individuals of a given species population in an incubated sample, $\overline{N}_i$ is the average initial number,

Table 1. Average numbers of individuals (mean ± standard error) of different categories of crustacean zooplankton per 10 liters in variously-treated samples in five cadmium-enrichment experiments using in situ incubation in Green Bay, Lake Michigan, 1976. Inclusive dates shown for each experiment indicate duration of incubation

Category	Experiment 1 22—26 July			Experiment 2 27 July—1 August			Experiment 3 20—27 August		
	Unincubated initial samples (n = 2)[a]	Incubated samples, added Cd, μg/1		Unincubated (n =2)	Incubated samples, added Cd, μg/1		Unincubated initial samples (n = 3)	Incubated samples, added Cd, μg/1	
		0 (n = 2)	50 (n = 2)		0 (n = 2)	50 (n = 2)		0 (n = 4)	50 (n = 4)
Cladocera									
Leptodora kindtii	0	0	0	0	0	0	1 ± 1	0	0
Polyphemus pediculus	1 ± 1	0	0	1 ± 1	0	0.3 ± 0.3	0.4 ± 0.4	0	0
Holopedium gibberum	2 ± 1	1 ± 1	0	1 ± 1	1 ± 1	0	65 ± 2	4 ± 1	0.3 ± 0.3
Sida crystallina	0.3 ± 0.3	0	0	0	0	0	0.4 ± 0.4	0	0
Bosmina longirostris	127 ± 2	70 ± 10	0.3 ± 0.3	295 ± 25	109 ± 27	1 ± 1	59 ± 6	68 ± 5	2 ± 1
Eubosmina coregoni	44 ± 4	16 ± 1	0	105 ± 11	46 ± 2	0	181 ± 39	156 ± 51	2 ± 1
Daphnia retrocurva	14 ± 4	6 ± 0	0	52 ± 14	50 ± 0	0	135 ± 20	60 ± 13	0.3 ± 0.3
D. galeata mendotae	6 ± 2	3 ± 1	0.3 ± 0.3	7 ± 2	10 ± 0	0.3 ± 0.3	22 ± 4	24 ± 6	0.3 ± 0.3
D. longiremis	31 ± 5	18 ± 1	0.3 ± 0.3	38 ± 20	44 ± 20	0	7 ± 2	6 ± 2	0
Ceriodaphnia lacustris	1 ± 1	0	0	5 ± 1	10 ± 0	0.3 ± 0.3	74 ± 5	56 ± 16	0
Chydorus sphaericus	6 ± 1	6 ± 1	1 ± 1	6 ± 2	6 ± 1	1 ± 1	159 ± 19	233 ± 15	9 ± 1
Copepoda: Cyclopoida									
Cyclops vernalis	4 ± 0	18 ± 2	13 ± 2	1 ± 1	13 ± 1	7 ± 1	4 ± 1	6 ± 3	5 ± 1
C. bicuspidatus thomasi	17 ± 4	39 ± 6	31 ± 2	14 ± 1	40 ± 7	30 ± 6	51 ± 5	54 ± 8	38 ± 5
Mesocyclops edax	3 ± 1	5 ± 1	4 ± 1	1 ± 1	10 ± 1	6 ± 1	9 ± 2	4 ± 1	4 ± 1
Tropocyclops prasinus	5 ± 2	6 ± 1	7 ± 2	20 ± 1	37 ± 9	28 ± 4	30 ± 4	34 ± 5	26 ± 6
Cyclopoid copepodites	84 ± 1	75 ± 2	75 ± 5	67 ± 7	103 ± 5	110 ± 15	228 ± 20	95 ± 14	88 ± 11
Copepoda: Calanoida									
Eurytemora affinis	2 ± 0	5 ± 1	2 ± 1	2 ± 1	6 ± 2	3 ± 1	2 ± 1	6 ± 3	1 ± 1
Diaptomus spp.	6 ± 1	10 ± 1	3 ± 1	3 ± 1	5 ± 0	1 ± 1	6 ± 1	10 ± 1	1 ± 1
Calanoid copepodites	21 ± 1	3 ± 2	1 ± 1	15 ± 3	14 ± 2	3 ± 1	24 ± 3	13 ± 3	0

Table 1. Continued

Category	Experiment 4 27 August—11 September Unincubated initial samples (n = 4)[a]	Incubated samples, added Cd, μg/1 0 (n = 3)	25 (n = 4)	50 (n = 4)	Experiment 5 11—20 September Unincubated initial samples (n = 3)	Incubated samples, added Cd, μg/1 0 (n = 3)	5 (n = 3)	10 (n = 2)	20 (n = 4)	40 (n = 4)
Cladocera										
Leptodora kindtii	0.3 ± 0.3	1 ± 1	0	0	0.4 ± 0.4	0.4 ± 0.4	0	0	0	1 ± 1
Polyphemus pediculus	2 ± 1	0	0	0	0	0	0	0	0	0
Holopedium gibberum	55 ± 1	0.4 ± 0.4	0	0	49 ± 6	1 ± 1	0.4 ± 0.4	0	0	0
Sida crystallina	0	1 ± 1	0.3 ± 0.3	1 ± 1	0	0	0	0	0	0
Bosmina longirostris	63 ± 13	81 ± 33	0	0.3 ± 0.3	37 ± 3	39 ± 3	22 ± 8	18 ± 1	5 ± 1	5 ± 1
Eubosmina coregoni	123 ± 21	20 ± 13	2 ± 1	1 ± 1	144 ± 13	125 ± 9	3 ± 1	4 ± 1	2 ± 1	1 ± 1
Daphnia retrocurva	89 ± 5	26 ± 13	0.3 ± 0.3	0.3 ± 0.3	87 ± 10	42 ± 4	22 ± 6	1 ± 1	1 ± 1	4 ± 2
D. galeata mendotae	15 ± 4	19 ± 5	11 ± 5	0	13 ± 1	10 ± 5	7 ± 2	5 ± 2	3 ± 1	0.3 ± 0.3
D. longiremis	8 ± 5	8 ± 5	1 ± 1	0	2 ± 2	3 ± 3	0.4 ± 0.4	0	0	0
Ceriodaphnia lacustris	25 ± 5	15 ± 4	0	0	15 ± 1	11 ± 4	3 ± 1	1 ± 1	0	0.3 ± 0.3
Chydorus sphaericus	126 ± 6	439 ± 128	13 ± 10	4 ± 2	50 ± 1	94 ± 5	35 ± 7	26 ± 10	4 ± 2	8 ± 2
Copepoda: Cyclopoida										
Cyclops vernalis	3 ± 1	2 ± 1	1 ± 1	1 ± 1	2 ± 1	3 ± 1	5 ± 2	5 ± 3	3 ± 1	7 ± 1
C. bicuspidatus thomasi	16 ± 2	40 ± 10	10 ± 3	10 ± 2	37 ± 9	41 ± 5	47 ± 11	45 ± 6	36 ± 4	32 ± 4
Mesocyclops edax	9 ± 1	3 ± 1	3 ± 1	2 ± 1	6 ± 1	15 ± 2	14 ± 2	9 ± 1	9 ± 2	11 ± 2
Tropocyclops prasinus	58 ± 4	70 ± 4	33 ± 5	8 ± 3	47 ± 1	40 ± 5	38 ± 5	43 ± 2	31 ± 2	17 ± 1
Cyclopoid copepodites	189 ± 24	98 ± 11	46 ± 11	34 ± 9	231 ± 14	169 ± 2	146 ± 14	144 ± 8	128 ± 6	95 ± 13
Copepoda: Calanoida										
Eurytemora affinis	1 ± 1	0.4 ± 0.4	0.3 ± 0.3	0	2 ± 1	2 ± 1	3 ± 2	1 ± 0	1 ± 1	1 ± 1
Diaptomus spp.	6 ± 3	5 ± 2	2 ± 2	0.3 ± 0.3	9 ± 1	12 ± 3	6 ± 1	9 ± 3	5 ± 2	0.4 ± 0.4
Calanoid copepodites	8 ± 3	3 ± 2	1 ± 1	0	14 ± 1	15 ± 3	8 ± 3	9 ± 4	5 ± 1	1 ± 1
Copepoda										
Copepod nauplii	191 ± 19	128 ± 38	31 ± 10	15 ± 4	244 ± 10	115 ± 21	138 ± 34	150 ± 26	120 ± 4	80 ± 10

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[a] Number of samples.

and Δt is the duration of incubation in days) and Cd (ppb) for the three major groups of zooplankton crustaceans are: Cladocera, -0.0132 ± 0.0032; Calanoida, -0.0058 ± 0.0009; Cyclopoida, -0.0010 ± 0.0003. Friedman's two-way analysis of variance by ranks indicates that the differences between the major groups are highly significant ($P < 0.001$). Within each major group, however, there are no significant differences between species.

We conclude that this new method appears to overcome some of the limitations imposed by the use of large, transparent bags or open-top tubes in large lakes for studies of pollutant effects on zooplankton—at least for short- to intermediate-term effects. The duration of in situ experiments employing opaque enclosures will be limited by decreasing food (phytoplankton) and oxygen concentrations in constant darkness; these limitations will be site-specific and will have to be determined for any system in which the method is used.

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04484

LONG-TERM EFFECTS OF CADMIUM IN LAKE MICHIGAN WATER ON AVERAGE NUMBERS AND BIOMASS IN LABORATORY DAPHNIA POPULATIONS

J. S. Marshall

Cadmium has long been recognized as a highly toxic element, but it was not until comparatively recently that concern has been expressed over the impact of anthropogenic emissions of cadmium into the environment. Very little is known about the long-term effects of cadmium on aquatic organisms, although zooplankton now appear to be more sensitive than either phytoplankton or fish and thus may be critical in determining the overall impact of cadmium on plankton-dominated ecosystems, such as the oceans or the Great Lakes. The purpose of this study was to determine the long-term effect of elevated concentrations of cadmium in Lake Michigan water on laboratory populations of Daphnia galeata mendotae, a species of some importance in all of the Great Lakes.

In each of two consecutive experiments, four populations were maintained for 23 weeks (or until they become extinct) at each of five levels of added cadmium: 0, 1, 2, 4, and 8 ppb in the first experiment, and 0, 5, 10, 15, and 20 ppb in the other (40 populations altogether). The medium consisting of filtered Lake Michigan water to which 2×10^4 cells/ml of Chlamydomonas reinhardi as food and prescribed amounts of Cd were added 24 hr before use, was changed daily (Monday through Friday, with a triple ration of food on Friday). Weekly determinations, of estimates based on permanently removed samples, were made on total numbers and average individual dry weight.

The means of the average numbers of individuals, coefficients of variability, and average biomasses (dry wt) of the populations at each added cadmium concentration are shown in Table 1. Mean numbers and biomass both tended to decrease with increasing cadmium, while the mean coefficients of variability tended to increase. Although mean values of each parameter changed progressively with increasing Cd, the mean values for average numbers and biomass at 1 to 2 ppb Cd and for coefficients of variability at 1 to 4 ppb Cd are not significantly different from those for the controls. The

Table 1. Mean average number of individuals, coefficient of variability, and average dry weight (means ± standard errors) in populations maintained at different concentrations of added cadmium. Values smaller than those underlined are significantly different ($P < 0.05$) from those of controls.

Added Cd, µg/l	Number of populations	Average number of individuals	Coefficient of variability	Dry weight, mg
0	8	149 ± 3	29 ± 1	1.09 ± 0.02[a]
1	4	142 ± 2	30 ± 1	1.07 ± 0.03
2	4	142 ± 5	28 ± 2	1.08 ± 0.01
4	4	114 ± 6	32 ± 3	0.99 ± 0.03
5	4	90 ± 3	50 ± 4	b
8	4	62 ± 9	51 ± 12	0.77 ± 0.07
10	4	48 ± 6	84 ± 11	b
15	4	23 ± 3	79 ± 14	b
20	4	9 ± 1	131 ± 10	b

[a] Based on 4 populations in Experiment 1 only.

[b] Not determined.

effect of cadmium on mean biomass is less pronounced than that on average numbers because of an increase in average individual dry weight. At 8 ppb Cd, for example, where average numbers are reduced to 42% of those in the controls, average biomass is reduced only to 71%.

As can be seen from Figure 1, there is a linear relationship between the average number of individuals in each population and the added cadmium concentration at which the population was maintained. The open symbols represent populations that established stable long-term averages and were, therefore, assumed to provide valid estimates of a quasi-equilibrium about which each population was fluctuating. The average numbers for the populations that became extinct are not considered valid in this sense and are represented by filled symbols to emphasize this difference. The downward-pointing arrows indicate that these values were decreasing rapidly at the end of the experiment, whereas the others were not.

It appears that an increase of 0.1 ppd in Lake Michigan would probably not have any readily detectable effect on the population of *D. galeata mendotae*, although there is no evidence for a true threshold for the effects of chronic

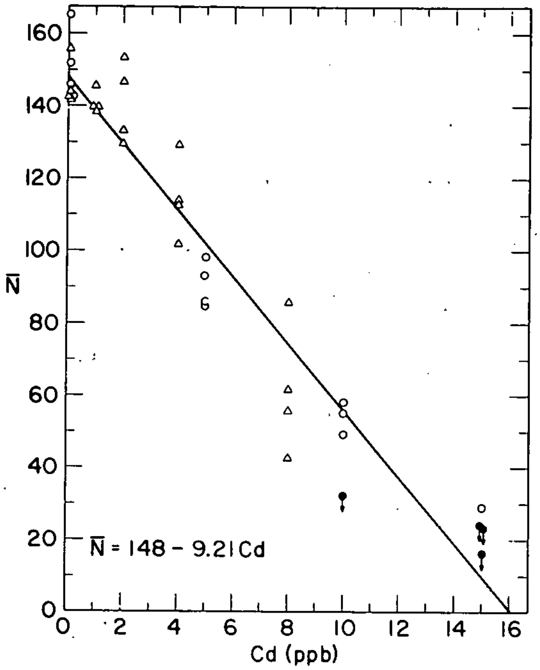

FIG. 1.--Effects of cadmium on average numbers of *Daphnia galeata* mendotae in laboratory populations, based on 23 weekly censuses. Experiment 1, Δ; experiment 2, o●.
(ANL neg. 149-76-332)

Cd stress. However, this conclusion is not conservative because it assumes that the results are directly applicable to the natural environment. More research is needed to assess the significance of several possible interactions of cadmium stress with other variables influencing zooplankton populations in the lake. Fish predation, for example, is likely to interact synergistically with cadmium stress.

04485

PARTICLE SIZE AND CHEMICAL PHASE DISTRIBUTION OF PLUTONIUM IN AN ESTUARINE SEDIMENT

J. J. Alberts, R. N. Muller, and K. A. Orlandini

Plutonium in Lake Michigan sediments has been shown to be associated predominantly with hydrous oxides[1] and exhibits an activity maximum in particles less than 4 μm in diameter.[2] However, since plutonium exhibits variable size and charge characteristics in waters of differing ionic strength and chemical composition,[3] an investigation was undertaken to determine if the particle size and chemical fraction distribution of plutonium in other sediments might be different.

A sample of marine sediment was collected from 60 ft of water in Buzzard's Bay, Massachusetts with sealed Smith-MacIntyre grab having a surface area of approximately 1 ft^2.* The loose floc layer overlying the consolidated sediment was removed by suction and was lyophylized. Three subsamples of the dry material were taken for (1) determination of the ash/dry weight ratios and total $^{239,240}Pu$ concentration,[4] (2) sequential extraction to determine the distribution of plutonium among various chemical fractions of the sediment,[1] and (3) separation into five size classes (> 45 μm, 45–20 μm, 20–4 μm, 4–2 μm, and < 2 μm) by wet sieving and elutriation in a carbonate-oxalate dispersant,[5] followed by the determination of the $^{239,240}Pu$ concentration in each size class.

The results of the sequential extraction (Table 1) clearly indicate that most of the $^{239,240}Pu$ is found in the citrate-dithionite extract as is the case for Lake Michigan sediments. The close agreement of the analyses of the estuarine floc material with those of Lake Michigan indicate that fallout $^{239,240}Pu$, whether it is being brought to the estuary by erosion of soil or through direct input into the overlying water, becomes associated with the same sedimentary phase as in fresh water. In addition, if most of the plutonium material comes from erosion of soils via a freshwater system, the

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*We wish to thank Robert Chase for collecting the sediment sample.

Table 1. Sedimentary phase distribution of ^{239,240}Pu (fCi/g dry wt) of Buzzard's Bay floc

Total	$MgCl_2$ extract	Citrate-Dithionite extract	NaOH extract	Fusion fraction
56.6 ± 3.2	<0.05	56.0 ± 0.6[a]	0.6 ± 0.2	5.5 ± 0.6

[a] The ± value is 1σ counting error.

increased ionic strength of the estuarine water is apparently not sufficient to remove ^{239,240}Pu from the particles. Approximately 10% of the ^{239,240}Pu is found in the fusion fraction and may either represent particles of plutonium formed during detonation, which have settled into the system, or be an indication of a small but finite diffusion of plutonium atoms into the crystalline lattice of the sedimentary minerals.

The distribution of ^{239,240}Pu concentration relative to particle size in the floc (Table 2) is similar to that observed in Lake Michigan sediment with a minimum in concentration occurring in the larger size classes and a maximum in the smaller size classes. As in the case of the freshwater sediments, most of the plutonium (in this case 75%) is found associated with particles less than 20 μm in diameter. This distribution would be expected since the source of the particulate material would be erosion and the smaller particles are more likely to be transported to the estuary than the larger particles. The pronounced maximum in the 4 to 2 μm size class with little apparent overlap in the two adjacent size classes may be an indication of the magnitude of force being exerted on the sediment in this location with the currents being too weak to transport larger material to this point and too strong to allow smaller material to deposit.

Table 2. Sedimentary particle size distribution of ^{239,240}Pu (fCi/g dry wt) of Buzzard's Bay floc

Particle size range, μm				
> 45	45–20	20–4	4–2	< 2
36.7 ± 0.6[a]	33.8 ± 0.9	55.9 ± 0.7	116 ± 2.0	40.6 ± 0.6

[a] The ± value is 1σ counting error.

The total ^{239,240}Pu concentration of the floc material is approximately one-third that observed in Lake Michigan floc.[6] Since this represents only one sample from the estuary, more data must be collected before a definite conclusion can be reached. However, it appears that the estuarine floc reflects both the chemical and particle size relations of freshwater sediments. These findings imply that the material in this sample is of continental origin and that the particles which have been brought in have not been appreciably affected by the change in ionic strength experienced during the transition from fresh to salt water, or that fallout ^{239,240}Pu behaves similarly in these chemically different systems.

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04486

EFFECT OF ANAEROBIOSIS ON THE GEOCHEMICAL CYCLING OF PLUTONIUM IN PAR POND

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Most of the fallout plutonium in Lake Michigan sediments is found associated with hydrous oxides which are significantly solubilized only under reducing conditions.[1] As most of the sediments of Lake Michigan remain aerobic all year long, the possibility of resolubilization of these oxides and reintroduction of the associated plutonium into the overlying water through this mechanism is small. However, in some water bodies, such as PAR Pond, South Carolina, anaerobic sedimentary conditions do prevail for a portion of the year. PAR Pond was studied to determine if the seasonal anaerobic conditions in the sediments lead to a measurable increase in the plutonium concentration of the overlying water.

The PAR Pond located on the Savannah River Project site in South Carolina was chosen as the study site for several reasons. Although the pond is located in the southeastern portion of the U.S., which is dominated by acidic black-water, chloride-sulfate systems, PAR Pond is a carbonate-bicarbonate system with a pH of approximately 8. Thus the pond has similar chemical properties to those existing in Lake Michigan, and, in spite of the high organic content of the water, should permit a reasonable comparison to the behavior of plutonium in the Great Lakes. In addition, PAR Pond sediments become intensively reducing every summer with hydrogen sulfide and dissolved iron present in the hypolimnion of the pond.[2] Finally, the ^{239,240}Pu found in the sediments of the pond is fallout-derived, as is the case in Lake Michigan, and exists predominantly in the hydrous oxide fraction of the surface sediment at approximately 3 times the concentration of Lake Michigan surface sediments.[3] Hence, the pond should offer an ideal system in which to determine if fallout ^{239,240}Pu can be remobilized into the water column by the process of

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anaerobiosis in high pH, carbonate dominated lakes.

Two sampling sites were chosen in the pond. The first is designated as the Hot Arm since this embayment receives heated water from a reactor on site and exhibits elevated surface water temperatures above those in other areas of the pond. Water depth at this location is 9 meters. The second site is designated as the Cold Dam and is located in 16 meters of water (the deepest part of the pond) near the point where water leaves the pond through a spillway. Water samples were taken at various depths from both sites on four occasions between July 1975 and August 1976 with a 10 liter Van Dorn water sampling bottle suspended horizontally, rather than vertically, to allow collection from a narrow lens of water. Fifty liter water samples were taken from each depth. Samples were sequentially filtered through 3.0 and 0.45 µm membrane filters, acidified, and spiked with ^{236}Pu as an isotopic tracer. The samples were analyzed for ^{239,240}Pu after the technique of Nelson et al.[4]

The concentrations of ^{239,240}Pu in PAR Pond water samples are given in Table 1. The concentration of ^{239,240}Pu is low in all cases, compared to values determined in Lake Michigan.[5] In addition, the values are fairly constant

Table 1. Seasonal variations in ^{239,240}Pu concentrations (fCi/1) in water from PAR Pond[a]

Sampling depth, m	July 1975[b]	February 1976[b]	June 1976[b]	August 1976[c]
Hot Arm				
3	0.01	0.10 ± 0.02	0.01 ± 0.03	0.02 ± 0.04
7—9	0.07	0.12 ± 0.02	0.02 ± 0.03	0.03 ± 0.04
Cold Dam				
3	0.05	0.12 ± 0.02	0.04 ± 0.03	0.03 ± 0.04
6—9	0.02		0.05 ± 0.03	0.01 ± 0.04
12	0.02	0.09 ± 0.02	0.03 ± 0.03	0.07 ± 0.04
15	0.38	0.11 ± 0.02	0.03 ± 0.03	0.20 ± 0.04

[a] The stated errors are one standard deviation of the counting statistics.

[b] 0.45 µm membrane filtered water.

[c] Total ^{239,240}Pu concentrations since these water samples were not filtered.

with depth in the water column even though the thermocline was established at approximately 6 m at the Cold Dam site during the three summer samples. The intensity of the thermocline at these times is evidenced by the fact that oxygen was undetectable and the pH dropped from 8 at the surface to approximately 6.5 in the deep waters. This depth distribution differs from that observed in Lake Michigan, where the waters above the thermocline become depleted in ^{239,240}Pu, while the waters below the thermocline retain approximately the same ^{239,240}Pu concentration (~ 0.65 fCi/l).

The data (Table 1) indicate that there is a slight increase in the ^{239,240}Pu concentration near the bottom as compared to the rest of the water column in the July 1975 and August 1976 samples. However, no significant increase in ^{239,240}Pu concentration is observed in the hypolimnetic waters of the pond which could be related to the anaerobic conditions of the sediment and deep waters during the summer months. It appears that even for this pond, the conditions of low redox potential, pH values of approximately 6.5 and a high loading of natural organic acids are not sufficient to solubilize plutonium from the sediments where it is predominantly associated with hydrous oxides.

Acknowledgement

We wish to thank Dr. Lawrence Tilly of the Savannah River Laboratory for his help and cooperation.

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04487

PLUTONIUM DISTRIBUTION IN VARIOUS TISSUES OF FISH FROM A CONTAMINATED POND-CANAL SYSTEM IN THE MIDWESTERN UNITED STATES

J. J. Alberts, G. E. Bartelt, and C. W. Wayman

A system of canals and ponds exists near the Mound Laboratory in Miamisburg, Ohio, which contains high concentrations of ^{238}Pu in the sediments (up to ~0.5 nCi/g)[1] and overlying waters as a result of a spill in 1969. The system contains several species of fish which represent a possible food source for man. Hence, a study of the tissue distribution of ^{238}Pu within these species was undertaken.

Specimens of gizzard shad, bluegill, crappie, smallmouth bass, largemouth bass, black bullhead, and carp were taken from the ponds and one canal by trapping, gill netting, and electroshocking during several field trips in 1975. The fish were rinsed in the water from which they were taken to remove as much of the attached material as possible and then frozen. Later, the fish were thawed and dissected into seven tissue categories: gastrointestinal tract (GI), liver, gills, gonads (when available), skin, muscle (fillet), and bone (including residual tissue). The tissues were dired, ashed, and then analyzed for plutonium by the method of Nelson et al.[2] Depending on the size of the fish, it was not always possible to separate the tissues from a single fish and treat it as a discreet sample. Hence, in the case of gizzard shad, bluegill, crappie, and black bullhead, several individuals of the same species were composited from the same sampling period to constitute one sample.

A compilation of ^{238}Pu concentrations in individual tissues and of all the species studied is beyond the scope of this report. However, a consistent trend in the data is apparent as demonstrated in Table 1. Clearly there is a relatively consistent relationship in the uptake of plutonium by various tissues for the species of fish studied. For all species except the largemouth bass, the GI tract has either the highest ^{238}Pu concentration or equals the gill or liver in plutonium concentration. These tissues always have a higher ^{238}Pu concentration than any of the others analyzed. Also, in all cases, all the species exhibit the same ranking of bone, skin, and muscle with the gonads

Table 1. Ranking of ^{238}Pu activity from highest to lowest in tissues of several species of freshwater fish

Shad (5)	GI > gills ≅ liver > bone > skin > gonad > muscle
Bluegill (2)	GI > gill > liver > bone > skin > gonad > muscle
Crappie (2)	GI > gill ≅ liver > bone > skin > muscle
Smallmouth bass (1)	GI ≅ gill ≅ liver ≅ bone > skin > gonad > muscle
Largemouth bass (1)	gill > liver > GI > bone ≅ skin > muscle
Bullhead (1)	GI ≅ liver ≥ gill > bone > skin > muscle
Carp (6)	GI ≅ gill > liver > bone > skin > muscle

( ) = number of samples examined.

appearing between skin and muscle in those cases in which they were analyzed.

The cause of the elevated plutonium concentrations of these fish is probably adsoption of high ^{238}Pu activity particles in the GI tract and on the gills. Livingston and Bowen[3] and Bartelt et al.[4] have reported elevated plutonium concentrations in the GI tract and Emergy and Klopfer[5] have found elevated levels in the GI tract and gills. In addition, the high plutonium levels reported in livers may indicate some active metabolic uptake of plutonium.

The low values in bone, skin, gonad, and muscle are also consistent with the values reported by the same authors for freshwater organisms[3,5] and are consistent with values found in bones from marine fish.[6] It is difficult to understand why the skin does not contain a higher ^{238}Pu concentration in these organisms if surface adsorption is the important factor in uptake by the GI tract and gills, unless the natural sloughing action of the mucus layer is adequate to remove sorbed particles and keep the plutonium concentration low.

The results of this study indicate that there is little transfer of plutonium to the muscle and skin of freshwater fish even though the fish have lived in a relatively contaminated environment. As these are the tissues most often consumed, it would appear that there is little transfer of plutonium through this pathway to man.

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04488

DISTRIBUTION OF FALLOUT AND STACK-DERIVED PLUTONIUM IN SOME MIDWESTERN SOILS

J. J. Alberts, R. N. Muller, D. G. Sprugel, and C. M. Bobula

It is generally believed that plutonium is immobilized from further migration by adsorption on soil particles. However, recent data indicating that plutonium is being mobilized from a burial site, perhaps via subsurface water movement,[1] has led to a reevaluation of this concept. Since little is known about the mobilization of plutonium in Midwestern soils, a study was begun in the Great Miami River watershed, Ohio, to determine if atmospheric and stack-derived plutonium are mobilized through the soil column.

Soil samples were taken at three locations: Mound State Park located directly east and at a distance of 100 yards from the boundary of the Mound Laboratory in Miamisburg; the Library Park in Miamisburg, located one-half mile north of the Mound Laboratory; and a wooded area near Hueston Woods, approximately 25 miles west of Miamisburg. The Miamisburg sites were chosen because of the high concentration of ^{238}Pu that exists in soils near the Mound Laboratory.[3] The Hueston Woods sample was chosen as representative of background concentrations derived from atmospheric levels of both ^{238}Pu and ^{239,240}Pu. Samples were taken by exposing a soil profile to a given depth and then removing a 10 cm × 20 cm section of soil at 2.5 cm intervals. The material was placed in plastic bags. Prior to analysis for plutonium the samples were dried and ashed.[3]

The plutonium concentration profiles are given in Table 1. The ^{238,239,240}Pu concentrations are higher in the surface soil for both the Miamisburg locations than in surface soil from Hueston Woods, indicating that ^{239,240}Pu, as well as ^{238}Pu, has been added to the surrounding area from the Mound Laboratory stacks. The difference in concentrations of ^{238}Pu and ^{239,240}Pu at the two sites in Miamisburg reflects the greater distance of Library Park from the stack, which is consistent with previous data on areal distribution of these isotopes.[3]

Table 1. Depth distribution of ^{238}Pu and ^{239,240}Pu (pCi/kg dry wt) from three Midwestern soils

Depth of sample, cm	Mound Park			Library Park			Hueston Woods Pasture		
	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
0—2.5	1950 ± 17[a]	175 ± 5	11 ± 0.3	147 ± 3	41 ± 2	3.6 ± 0.1	0.7 ± 0.07	17.5 ± 0.3	0.040
2.5—5.0	2840 ± 11	27 ± 1	105 ± 4	76 ± 2	34 ± 2	2.2 ± 0.1	0.6 ± 0.06	14.6 ± 0.3	0.041
5.0—7.5	4580 ± 15	26 ± 1	176 ± 8	13 ± 1	9.2 ± 0.3	1.4 ± 0.2	0.4 ± 0.04	9.4 ± 0.2	0.043
7.5—10.0	890 ± 6	15 ± 1	59 ± 3	4.5 ± 0.3	3.7 ± 0.3	1.2 ± 0.1	0.15 ± 0.05	4.7 ± 0.1	0.032
10.0—12.5	435 ± 5	7.7 ± 0.6	56 ± 4	3.0 ± 0.2	1.8 ± 0.2	1.7 ± 0.1	0.16 ± 0.03	3.3 ± 0.1	0.048
12.5—15.0	102 ± 2	9.7 ± 0.7	11 ± 1	1.0 ± 0.1	1.1 ± 0.1	0.9 ± 0.1	0.09 ± 0.03	2.4 ± 0.1	0.038
15.0—17.5	65 ± 2	2.9 ± 0.4	23 ± 3	1.0 ± 0.1	1.1 ± 0.1	0.8 ± 0.1			
17.5—20.0	40 ± 1	0.8 ± 0.2	49 ± 12	0.2 ± 0.03	0.7 ± 0.04	0.3 ± 0.05			
20.0—22.5	22 ± 1	0.7 ± 0.2	31 ± 8	0.7 ± 0.05	0.3 ± 0.03	2.2 ± 0.3			
22.5—25.0	13 ± 0.2	0.4 ± 0.04	34 ± 4	0.6 ± 0.05	0.4 ± 0.04	1.6 ± 0.2			
25.0—27.5	2.5 ± 0.1	0.2 ± 0.04	12 ± 3	0.2 ± 0.05	0.4 ± 0.04	0.6 ± 0.2			
27.5—30.0	2.5 ± 0.1	0.1 ± 0.04	25 ± 9	0.2 ± 0.05	0.3 ± 0.04	0.8 ± 0.2			

[a] The ± value is 1σ counting error.

Plutonium is observed to depths up to 30 cm in the park samples and to 15 cm (greatest depth samples) in the Hueston Woods soil. In addition, the Mound Park sample has a maximum concentration at the 5 to 7.5 cm level for ^{238}Pu, while for all other isotopes in all other samples there is a steady decline in concentration with depth. The ^{238}Pu/^{239,240}Pu ratio in both of the park profiles decrease with increasing depth in the soil, but never reaches the value expected for fallout plutonium. For the Hueston Woods soil, on the other hand, the ^{238}Pu/^{239,240}Pu ratio is uniform with depth.

The data clearly show that plutonium, whether from the stack or fallout, is being mobilized downward in the soil column, but at present the mechanism for this mobilization is not known. One possibility is that particles with attached plutonium may be transported downward by burrowing organisms such as earthworms. Such a process would require that the plutonium be mixed uniformly and that the ratio of ^{238}Pu/^{239,240}Pu not change with depth. This may be a suitable explanation for the observed profile in the Hueston Woods sample but is inadequate in the Mound Park sample, where the ratio clearly changes.

Meyer proposed that the medium for chemical leaching of plutonium in soils from Kentucky is water.[1] However, the actual mechanism for the transport, whether through an organic complex, flow along fracture lines or differences in oxidation state caused by the physical characteristics of the soil, is not known at this time. The change in values of ^{238}Pu/^{239,240}Pu ratio suggests that in the Mound Park soil column there is a preferential mobilization of the ^{239,240}Pu since the ratio decreases with depth and there is a peak in ^{238}Pu concentrations at 5 to 7.5 cm without a corresponding increase in ^{239,240}Pu. Such a preferential mobilization is difficult to understand unless the oxidation states or chemical forms of the ^{238}Pu and the ^{239,240}Pu were different when the material was deposited at the soil surface.

The data indicate that for both sources of plutonium there is mobilization downward in the soil column. However, more experiments will be required with other soils to ascertain the mechanism for this transport.

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04489

PRELIMINARY REPORT ON THE DISPERSION AND TRANSPORT OF PLUTONIUM FROM A PULSED EFFLUENT IN A RIVER SYSTEM

J. J. Alberts

The Great Miami River watershed program has been charged, in part, to define the fate of the small quantities of plutonium discharged into a flowing system via the effluent from the Mound Laboratory. Preliminary studies[1] with Rhodamine WT dye have shown that this effluent maintains its integrity as a pulse of water downriver for distances up to 30 miles. Analyses of river water for plutonium have further demonstrated that the plutonium is partitioned rapidly between the "particulate" and "dissolved" components of the river water and approximately the same ratio between these components is retained during transport downriver.[1] There is also an indication that sedimentary particulate material is continually being resuspended in the water and this process is in equilibrium with the removal of new particulate matter by settling. This recycling continues, whether an effluent pulse is passing or not, leading to a situation in which the amount of plutonium in an effluent pulse is small relative to the total amount of plutonium being transported by the river.

In order to define better the processes controlling the fate of plutonium in this river system, a comprehensive field study was undertaken during October 1976. With the assistance of the Mound Laboratory, Rhodamine WT dye was again added as a readily detectable marker to the low level waste storage tanks at the plant as had been done in the previous dye studies.[1] This pulse was followed downstream over a period of days, and comprehensive water sampling stations were occupied at five locations: 3 (Chautauqua), 4 (Franklin), 5 (Trenton), 6 (Bonham Farm), and 7 (Mile 28) (Figure 1). At each station the fluorescence of the dye was continuously monitored while the pulse passed. In addition, 100 ml water samples were collected at specified time intervals, stored in glass bottles and returned to the laboratory, where their dye concentration was again determined. Water samples were taken in triplicate 20 times during the passage of the dye. These samples (ranging in size from 3.8 ± 7.6 liters) were filtered with 0.45 μm membrane filters and both the water and

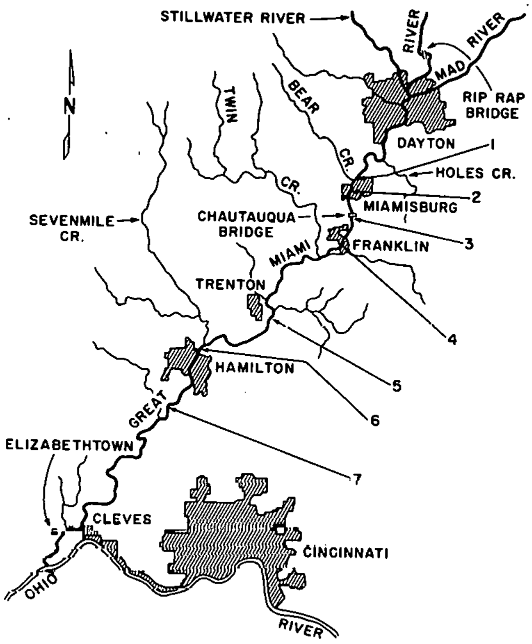

FIG. 1.--The Great Miami River watershed. (ANL neg. 149-6725 Rev. 1)

filters were retained for plutonium analysis. When these samples have been analyzed, a mass balance can be calculated for plutonium within the pulse and any changes can be defined in the "particulate"-to-"dissolved" plutonium distributions as the material passes downriver.

In addition to the studies of the mass balance and compartment distribution, experiments were undertaken to determine if plutonium freshly taken up on particulate material equilibrates with water of low plutonium concentration and conversely if water with relatively high plutonium concentrations equilibrates with particulate matter of low relative plutonium content. This experiment was carried out by filtering several 3.8-liter water samples taken at Rip Rap Bridge, a station located $\sim$20 miles upriver of the effluent discharge (Station 2) and known to have background plutonium concentrations[2] in both water and particulates. A corresponding number of water and filter samples was also taken in this manner at the peak of the pulse at Chautauqua. Filters with "clean" particulates were placed in filtered "high plutonium" water, and "high plutonium" particulate filters were placed in "clean" water. The experiment was run in duplicate and the filters and water allowed to equilibrate for 30 min, 4 hr, and 24 hr. At the end of the equilibration period the samples were again filtered, and the water and filters will be analyzed for plutonium concentration.

Experiments were also undertaken to attempt to define the chemical speciation of the plutonium in the effluent and how this speciation changes with time in the river. To this end, studies were conducted using ultrafiltration to define colloidal plutonium, and anion and cation resins to determine the charge characteristics of the plutonium. These experiments were conducted at sites 3 through 7 and on the effluent just prior to entering the river at the discharge point, site 2. In addition, attempts were made to determine the distribution coefficients for the plutonium on anion and cation resins with filtered water at sites 3 through 7. Finally, an attempt to determine the oxidation state of the plutonium in the river water was made by taking a volume of filtered river water, acidifying it with concentrated nitric acid to a final concentration of 8 M and then passing the solution through large glass columns of Dowex-1 anion resins in the nitrate form. The effluent, which should contain the plutonium in the +6 oxidation state, and the resin, which should contain plutonium in the +4 oxidation state, were retained for analysis.

Measurements of pH, temperature, dissolved and particulate nutrients and carbonate alkalinity were made during the passage of the pulse. Blank samples were run for all experiments using deionized water. Background river samples were collected immediately before the effluent entered the river (site 1, approximately 1 mile upstream of the effluent pipe) and twice a day during the entire field study at site 4. Attempts to collect net plankton samples at the sites failed.

The large number of samples obtained during this study are currently being analyzed. At this time, the data are too sparse to formulate any conclusions regarding the objectives of the study.

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04490

DISTRIBUTION OF FALLOUT PLUTONIUM IN THE WATERS OF THE LOWER GREAT LAKES

J. J. Alberts, M. A. Wahlgren, and D. M. Nelson

Wahlgren and Nelson[1] determined the total concentrations of ^{239,240}Pu in surface water samples collected from the Laurentian Great Lakes in the spring of 1973. The data showed that the surface water concentrations of these nuclides varied between 0.16 and 0.69 fCi/l and was slightly higher in the upper lakes (Superior, Michigan, and Huron) than in the lower lakes (Erie and Ontario). Further studies have shown that in Lake Michigan the concentration of plutonium varies seasonally in the epilimnion but remains relatively constant below the thermocline.[2] In July 1976, samples were collected during a four-day period from all the Great Lakes except Superior to determine if the same spatial distribution of ^{239,240}Pu exists now that was observed in 1973.

Water samples were taken in the four Great Lakes from the Coast Guard Cutter Westwind in the first week of July 1976. Surface water samples were obtained using a submersible pump at a depth of 3 m, while deep water samples were taken with a 30-1 Niskin water sampling bottle. Although the water samples were filtered aboard the vessel following collection and the water and filters were analyzed separately,[3] the ^{239,240}Pu water concentrations reported here are for total samples (determined by addition of the filtered water and filter concentrations) to facilitate comparison with the 1973 values, which were reported for unfiltered water.

The results (Table 1) reveal that the concentrations of ^{239,240}Pu in the surface waters from all the lakes are slightly lower in the 1976 samples than in those from 1973. The same trend of higher concentrations in the surface waters of the upper lakes as in the surface waters of the lower lakes is observed for both years. In addition, the ^{239,240}Pu concentration in samples of deep water collected during the summer of 1976 is higher than in the surface waters but is similar to the surface water values of the 1973 spring samples. This observation is significant in that it suggests that the surface waters of all the

Table 1. Concentrations of ^{239,240}Pu (fCi/l) in Great Lakes water

Station	Location	Depth, m	Summer 1976	Spring 1973
Lake Michigan				
Station 1	44° 48' N	3	0.24 ± 0.01[a]	0.69 ± 0.07
	86° 32' W	220	0.54 ± 0.04	
Lake Huron				
Station 2	45° 20' N	3	0.47 ± 0.05	0.60 ± 0.05
	83° 05' W	125	0.52 ± 0.06	
Station 3	44° 52' N	3	0.41 ± 0.05	0.63 ± 0.05
	82° 28' W	55	0.74 ± 0.06	
Lake Erie				
Station 4	42° 02' N	3	0.13 ± 0.06	0.24 ± 0.06
	81° 36' W	15	0.13 ± 0.04	
Station 5	42° 28.5' N	3	0.02 ± 0.04	0.16 ± 0.03
	79° 55' W	35	0.17 ± 0.04	
Lake Ontario				
Station 6	43° 25' N	3	0.04 ± 0.04	0.24 ± 0.05
	78° 42' W	110	0.35 ± 0.04	
Station 7	43° 38.5' N	3	0.04 ± 0.04	0.24 ± 0.05
	77° 44' W	150	0.35 ± 0.04	

[a] The ± value is 1σ counting error.

Great Lakes undergo a seasonal decrease in plutonium concentration similar to that already observed in Lake Michigan. The results also suggest that there have been little or no concentration changes in the hypolimnetic water. The decrease in surface ^{239,240}Pu concentrations may also explain the apparent lower concentrations observed for all the lakes in the 1976 sampling. However, flushing and dilution with surface waters from their watersheds cannot be ruled out as a possible mechanism of removal of plutonium from the lower lakes leading to the lower concentrations.

Acknowledgements

We wish to thank the U.S. Coast Guard for allowing us to use the cutter Westwind for this study and the officers and crew of that vessel who aided us in carrying out the sample collection.

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04491

SEASONAL CYCLING OF PLUTONIUM IN LAKE MICHIGAN

M. A. Wahlgren and D. M. Nelson

During the summer of 1973, we observed that the concentration of plutonium in the surface water became markedly depleted, coincident with the formation of a stable thermocline. We have followed this seasonal cycling of plutonium from 1973 through 1976; significant inputs of new fallout plutonium occurred during two of these years, 1974 and 1975. Although the new inputs of fallout plutonium are almost negligible compared to the total already in the system, this is not true for the water column which at present contains only 3% of the integrated deposition on the lake. The epilimnion is especially sensitive to new atmospheric deposition because it is physically isolated from dilution with the much larger hypolimnetic volume for a period of several months.

The data for total (dissolved and suspended) plutonium concentrations in Lake Michigan surface water are presented in Figure 1. The concentration of plutonium present in surface water at the end of the fall-to-spring convective mixing period is the same (within analytical error) each year and is independent of the magnitude of the previous year's fallout input. Further, a minimum concentration is reached each summer, although new fallout inputs into the epilimnion may delay the attainment of minimum concentration by 1 or 2 months.

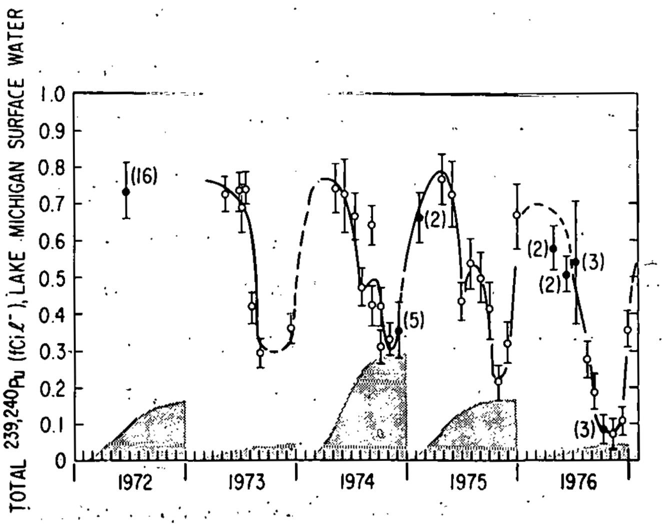

FIG. 1.--Seasonal variations of the concentration of fallout plutonium in the surface water of Lake Michigan.

The properties of the new input of plutonium apparently are similar, or become similar within a month or less, to those of the plutonium previously in the water column, since no significant differences occur in the distribution of plutonium between the seston and surface water through the season.[1]

From samples taken during the spring of 1976 it appears that in the spring of 1976 the concentration of plutonium in surface water at ANL-5 (10 km SW of Grand Haven, Michigan) may have been somewhat lower than in previous years, as indicated by the dashed line in Figure 1. However, analyses of samples of deeper waters from stations across the southern basin, taken in early June, indicate that the plutonium content of the water column was essentially the same as in previous years. This implies either less than complete spring convective mixing at station 5 in 1976, or an earlier than normal removal of plutonium from the surface waters.

There are at least two sequential sources of authigenic particles which form during the period of stratification, either of which can account for the removal of the major part of dissolved plutonium from surface water. From our knowledge of the concurrent decreases in silica and calcium contents of the epilimnion, the plankton concentration factor, and the seston distribution coefficient (K_d), we can make semiquantitative estimates of transport of plutonium out of the epilimnion due to diatom settling (including grazing), and to calcite precipitation (and sinking) which has occurred in August–September each year of the study. Analyses of amorphous silica indicate that diatoms comprise up to 50% of the dry weight of seston during late spring; if these are settling as senescent or grazed organisms, the appropriate K_d is $0.7 \pm 0.4 \times 10^5$ (the ash weight concentration factor from plankton net tows); if incorporated into settling detrital aggregates, the seston K_d of $2 \pm 0.5 \times 10^5$ will apply. The initial silica concentration in surface water is ~ 1 mg l^{-1} or 1 mg cm^{-2} in a 10 m deep epilimnion. Removal of particulate silica as senescent diatoms (which occurs before July) could transport only ~ 0.05 fCi cm^2, but up to ~ 0.2 fCi cm^2 as the diatom-derived portion of detrital aggregates. Calcium concentrations in surface waters decrease from July to September by 1 to 2 mg l^{-1}, or 2.5 to 5 mg l^{-1} as $CaCO_3$, which as sinking seston particles can remove 0.4 to 0.5 fCi cm^{-2}. Since in 1976 the concentration of plutonium in the epilimnion decreased to zero, the total decrease was equivalent to 0.6 fCi cm^{-2}.

Samples taken from four Great Lakes (July 1976)[2] show that the decrease in the plutonium concentration in surface water during the summer is a general occurrence throughout these lakes and that this process probably accounts for the concentration-depth gradients reported from August or September sampling trips on Lake Ontario by Bowen and co-workers.[3]

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04492

A COMPARISON OF THE DISTRIBUTION COEFFICIENTS OF PLUTONIUM AND OTHER RADIONUCLIDES IN LAKE MICHIGAN TO THOSE IN OTHER SYSTEMS

M. A. Wahlgren and D. M. Nelson

The concept of an equilibrium distribution of heavy metals between suspended particulates and the dissolved phase as a mechanism for trace element control in seawater was postulated by Goldberg.[1] Models which utilize this concept to describe the longer-term effects of a steady-state input[2] and of a pulse input[3] have recently been proposed. The distribution coefficient (K_d) for benthic floc and for near-bottom tripton in Lake Michigan is uniformly 3 to 4 × 10^5.[3] These sedimentary materials, in contrast to seston in the water column, are believed to have had equilibration times of the order of years and to have attained relatively uniform compositions through processes of decomposition and aggregation. For plutonium (and for many trace metal inputs as well), the dilution volume of Lake Michigan is so large (5×10^{15} l) that long-term, lake-wide contamination of the lake above levels permitted in drinking water is not physically possible under present conditions. Therefore, in the event of a sudden, localized plutonium input to the lake, the time scale of major interest would be that during which basin-wide dilution occurred, i.e., several months. Although the physical dilution of the contaminated water can be described by dispersion and lake-wide circulation models currently under development, the effective rate of movement of the contaminant (and the availability in drinking water) would be dependent on interactions with both the benthic and suspended particulates. Knowledge of the month-to-month uptake of plutonium by seston is required in order to describe these processes.

Filtration of Lake Michigan water samples has been carried out routinely since 1973, and some plutonium concentrations in the seston have been reported.[4] During 1975 and 1976 a sufficient number of filter samples from various depths was obtained throughout the field seasons to establish whether or not a distribution coefficient also controls the uptake of plutonium by the formation of particulates and their settling from the surface waters. Samples from ANL station 5 (10 km SW of Grand Haven, Michigan, water depth 67 m), of the southern basin, and from the lower Great Lakes have been analyzed for dry weight, ash weight, total organic[3] (loss of weight on ignition), amorphous silica, calcite, and residual minerals. Distribution coefficients were calculated on the basis of each of these solid components, and self-consistent values were observed for depth, season, or lake only, on the basis of dry weight of seston. The results from Station 5 (1975) are given in Table 1. The mean value of the K_d at 3 m depth is $2.0 \pm 0.4 \times 10^5$, at mid-depth it is $2.1 \pm 0.3 \times 10^5$, and 7 m off bottom it is $2.1 \pm 0.5 \times 10^5$. The data for tripton samples from sediment traps at 37 and 60 m depth yield the same K_d on a dry weight basis, i.e., $2.2 \pm 0.5 \times 10^5$. These findings strongly suggest that the uptake of fallout plutonium (including inputs of new fallout during the summer of 1975) is dominated by a surface coating process common to all seston particle types. Alberts et al.[5] have previously reported an association of plutonium in tripton and surficial sediments with the hydrous oxides. An insufficient number of ^{137}Cs analyses are on hand to correlate its uptake to a specific component of the seston, but its behavior is clearly different from that of plutonium.

If the seston-water and sediment-water K_d's for plutonium are controlled by a limited group of chemical variables such as pH, ionic strength, and ligands such as carbonate ion, and by the presence of ubiquitous surface coatings on particles, then generally similar K_d's in otherwise dissimilar aquatic environments are to be expected. A comparison of distribution

Table 1. Seston-water and tripton-water K_d's ($\times 10^5$), ANL station 5, 1975[a]

Depth	Date of Sampling														
(m)	11 Apr		9 May		3 Jun		8 Jul		5 Aug		4 Sept		7 Oct	3 Nov	3 Dec
3	1.4		2.4		1.7		1.7		1.9		2.3		1.9	2.5	2.5
30			1.8								2.0				
37		1.8		1.6		2.3		2.1		2.3		2.8			
60	2.7		2.2		2.0		2.4		2.0		1.1		2.2	1.7	2.9
60				2.1		2.2		1.9		1.7		1.6			

[a] Tripton values are designated by underlined numerals. The tripton collection interval is approximately one month as indicated by sampling dates.

coefficients obtained in different study areas with a variety of sources of plutonium is given in Table 2 and tends to support this hypothesis for plutonium. Also included are data for other actinide elements (Th, U, and Np) and two other nuclides of radioecological concern, ^{137}Cs and ^{90}Sr. The limited data set for ^{237}Np suggests that the Argonne releases are in two different chemical forms, presumably Np(V) and Np(VI).

The remarkable similarity in removal rates of fallout plutonium and cesium from the water column to the sediments of the Great Lakes has been noted previously,[6] as has the covariance of deposition rates in sediments in Lake Michigan.[7] This apparently results from the nearly identical sediment-water K_d's, which these two radioelements happen to exhibit under the carbonate-dominated, pH 8 conditions of the Great Lakes. The low K_d for ^{90}Sr reflects

Table 2. Range of K_d's observed in several natural waters.

Nuclide	Source	Subsystem	No. of samples	K_d ($\times 10^{-5}$)[a]	Reference
^{239}Pu	Fallout	Surface sediment, Par Pond, South Carolina	1	10	8
^{239}Pu	Fallout	Surface sediment, Lake Michigan	8	3.3 ± 0.3	this study
^{239}Pu	Fallout	Seston (4 lower Great Lakes)	31	2.5 ± 0.5	this study
^{239}Pu	Fallout	Seston, Miami River, Ohio	7	1.2 ± 1.0	9
^{239}Pu	Fallout	Water treatment, filter media	3	0.06—0.4	10
^{239}Pu	Plant wastes	Sediments, Irish Sea	10	0.5	11
^{239}Pu	Plant wastes	Sediments, Trombay	8	0.9	12
^{238}Pu	Plant wastes	Seston, Miami River	9	7.6 ± 8	13
^{238}Pu	Plant wastes	Seston, Sawmill Creek, ANL	5	2.0 ± 1.3	14
^{237}Np	Plant wastes	Seston, Sawmill Creek, ANL	2 3	0.21 ± 0.05 0.01 ± 0.001	14 14
^{238}U	Natural	Tripton, Lake Michigan	2	0.01	this study
^{232}Th	Natural	Tripton, Lake Michigan	2	50	this study
^{137}Cs	Fallout	Surface sediment, Lake Michigan	8	~3	this study
^{90}Sr	Fallout	Surface sediment, Lake Michigan		1.2×10^{-3}	15
^{90}Sr	Plant wastes	Surface sediment, Perch Lake, Ontario		1.0×10^{-3}	16

[a] K_d's for seston are calculated on dry weight basis (for sediments and tripton, dry weight ≈ ash weight).

its chemical similarity to calcium and competition for relatively nonspecific ion-exchange sites in the sediments; therefore, its control in Great Lakes waters is primarily hydrodynamic rather than geochemical. Apparently U and Np represent intermediate cases in the competition between flushing and sedimentary deposition.

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04493

THE SORPTION OF ^{237}Pu BY THE DIATOM ASTERIONELLA FORMOSA

H. L. Conway, M. A. Wahlgren, N. Peterson,* and D. M. Nelson

Knowledge of the behavior of the man-made radionuclide plutonium within aquatic environments is of fundamental importance in assessing its potential hazards and ecological impact. The sorption of plutonium by phytoplankton and other algae is the dominant factor in the biological transport of plutonium in the aquatic environment. Wahlgren and Nelson[1] have suggested that sorption by photoplankton may be responsible for the seasonal loss of plutonium from the epilimnion of Lake Michigan. Their findings support the importance of biological sedimentation in plutonium cycling suggested by Bowen et al.[2] and supported by Noshkin.[3]

A unialgal diatom culture was spiked with ^{237}Pu tracer solution[4] in an attempt to simulate the behavior of fallout plutonium observed in field studies.[5] Although the uptake data obtained were limited by collapse of the algal culture, the results were encouraging in that the ^{237}Pu in the filtered lake water medium exhibited strongly anionic properties similar to fallout plutonium in Lake Michigan,[6] with limited sorption on container walls. The purpose of the present study was to extend the investigations of the sorption of plutonium by phytoplankton in a controlled environment using continuous culture techniques.[7]

In Experiment I, the ^{237}Pu tracer in dilute nitric acid was mixed with the influent medium via metering pumps just prior to its addition to the chemostat. The greater portion of the ^{237}Pu in the chemostat was associated with the particulates (Figure 1). Initially ~ 90% of the total activity was associated with the diatom cells; however, by the fourth day of the experiment this percentage was 60 to 70 and remained in this range for the duration of the experiment. The results of short-term sorption studies (< 24 hr), using both live and killed A. formosa cells, were similar to those of the longer term studies: Given a ^{237}Pu spike from the stock solution used in Experiment I, the live

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*Student Associate, January–April 1976, from Luther College, Iowa.

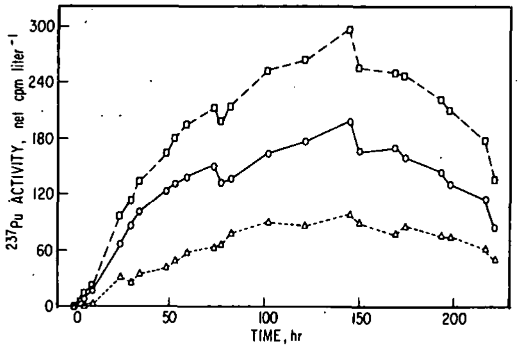

FIG. 1.--The ^{237}Pu activity in the chemostat in Experiment LT-1 versus time. Total activity, ☐ ; particulate fraction (*A. formosa* cells), ○; and soluble fraction, Δ.

cells incorporated between 70 and 95%, and the killed cells between 63 and 87% of the ^{237}Pu activity.

In Experiment II, the plutonium tracer was further treated by heating in carbonate solution; it was then added to 18 l of culture medium. Before the experiment was begun the culture medium containing the plutonium was allowed to equilibrate for a period of ~ 4 days and then filtered. The greater portion of the ^{237}Pu in this experiment was found to be associated with the medium; only 4 to 8% of the ^{237}Pu was taken up by the diatom cells (Figure 2). The bioaccumulation ratio, defined as the ratio of ^{237}Pu in or on the particulate (on a wet weight basis) to that in the medium, was ~ 1700 in Experiment II compared to ~ 100,000 in Experiment I. Short-term studies using the ^{237}Pu stock solution from Experiment II resulted in 8 to 13% of the total ^{237}Pu activity associated with the live cells and 8 to 23% associated with the dead cells.

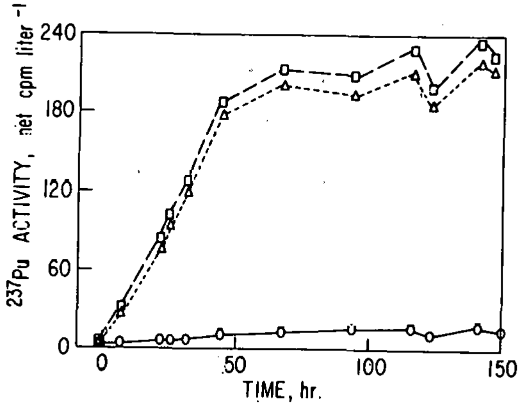

FIG. 2.--The ^{237}Pu activity in the chemostat in Experiment LT-2 versus time. Total activity, ☐ ; particulate fraction (*A. formosa* cells), ○; and soluble fraction,

The contrasting results obtained in the two experiments are associated with marked differences in the properties of the ^{237}Pu solutions used (Table 1). When samples from the ^{237}Pu solution used in Experiment I were run through anion and cation exchange resins, 75 and 98%, respectively, of the initial radioactivity of the solution was retained by the columns. However, when the culture solution used in Experiment II, containing the ^{237}Pu in carbonate solution, was passed through the resins, 94% of the activity was retained by the anion exchange column and only 15% was retained by the cation exchange column.

These experiments indicate the importance of the preparation of the stock solution of ^{237}Pu for use in simulating lake conditions. The ^{237}Pu solution used in Experiment I did not appear to be similar to the form of plutonium found in natural lake water. Both long- and short-term uptake studies with this stock solution resulted in > 60% of the ^{237}Pu activity being associated with particulates. The dead cells in the short-term study had an equivalent amount of ^{237}Pu activity associated with them. These results lead to the conclusion that the ^{237}Pu was taken up by any available surface. Therefore, the ^{237}Pu in this stock solution was not in the form found in the lake (where it is ionic and not colloidal).[1]

Since no significant increase in plankton or particulate uptake was observed following the input of new fallout in 1975,[8] the results from Experiment II approximate far more closely the behavior of both new and old inputs of fallout plutonium in the lake than do those from Experiment LT-1. The form of ^{237}Pu in the carbonate-treated solution was anionic, and a greater proportion of

Table 1. Testing of ^{237}Pu stock solutions through ion exchange resins (net cpm/l)

Stock solution sampled	Initial ^{237}Pu activity	^{237}Pu activity after anion exchange	Initial ^{237}Pu activity	^{237}Pu activity after cation exchange
Experiment I	250	63	240	30
Experiment II	750	41	720	620

the ^{237}Pu activity was associated with the medium; only 4 to 8% of the radioactivity in the long-term studies were incorporated in the particulate portion. This compares favorably to the approximately 5% associated with phytoplankton in Lake Michigan, and the bioaccumulation ratio of 1700 is comparable to that (~ 4600) observed in phytoplankton samples taken from Lake Michigan when A. *formosa* is dominant.

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04494

THE DISTRIBUTION OF SELECTED CHEMICAL ELEMENTS IN THE SEDIMENTS OF SOUTHERN LAKE MICHIGAN

John A. Robbins* and David N. Edgington

Recently Edgington and Robbins have shown that it is possible to reconstruct the history of inputs of nonconservative elements into the Great Lakes by measuring their concentration in the sediment column.[1] They have shown, for example, that the concentration profiles for lead accurately reflect the history of regional lead emissions as a result of burning coal and leaded gasoline. On the basis of concentration and sedimentation rate data, they estimated the net flux of lead in a few cores as a function of time and inferred the present annual accumulation in the sediments of the southern basin.

This study has now been extended to include 30 additional elements measured in cores from forty stations in southern Lake Michigan. The samples were taken from the top of each core (1–2 cm) and from 20 to 24 cm below the surface at which depth the sediment is associated with the presettlement era. Sedimentation rates at each station were estimated from the profile of either ^{137}Cs or ^{210}Pb in the core.[2,3] Trace element analyses were performed using neutron activation and gamma-ray spectrometric methods on whole, dry samples or atomic absorption methods on acid leachates.

A summary of the analytical results are presented in Table 1 as the average values for all cores analyzed. Cultural enrichment factors, defined as the ratio of the concentration near the surface to that at depth, are also given in Table 1. It is clear that most elements shown here are not entering the lake in significantly different amounts than they were about 150 years ago. In fact, there are only 10 or 12 elements which are significantly enriched in surface sediments.

It should be emphasized that these average enrichment factors cannot be directly related to a well-defined time of deposition, partly because sedimentation rates are highly variable over the lake basin. Thus, a given interval

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Table 1. Summary of analytical data, enrichment factors and approximate net annual deposition rates

Element	Mean concentration, ppm Surface sediments (1—2 cm)	 Underlying sediments (22—24 cm)	Enrichment factor[a]	Net annual deposition, metric tons/yr[b]
As	17	9.3	2.2	40
Ba	395	404	1.0	0
Br	57	30	2.4	80
Ca[c]	48000	50000	1.3	-
Cd[c]	3.0	1.5	2.3	6
Ce	42	51	0.88	0
Co	10	12	0.92	0
Cr[c]	64	44	1.6	100
Cs	3.9	4.6	0.93	0
Cu[c]	34	24	1.7	50
Eu	0.88	1.0	0.93	0
Fe[c]	24000	25000	1.1	0
Hf	3.8	3.9	0.95	0
K[c]	5200	7300	0.82	0
La	26	28	0.94	0
Lu	0.31	0.57	0.92	0
Mg[c]	25000	28000	0.94	0
Mn[c]	770	480	1.8	-
Na	5300	5000	1.6	-
Ni[c]	30	29	3.8	0
PO_4[c]	1700	1300	1.7	2000
Pb[c]	89	26	3.8	240
Rb	76	88	0.92	0
Sb	1.4	1.0	3.6	4
Sc	10	11	0.97	0
Sm	4.4	4.9	0.96	0
Tb	0.50	0.60	0.91	0
Th	6.5	7.3	0.91	0
U	1.2	1.5	0.91	0
Zn[c]	170	58	3.3	500
INORGC (%)[d]	2.9	-	-	-
ORGC (%)[d]	2.6	-	-	-

[a] Mean ratio of concentration near the surface (1—2 cm) to that in underlying sediments (22—24 cm).

[b] Approximate values based on uncorrected net fluxes normalized to corrected net lead fluxes; southern basin only.

[c] Using atomic absorption spectrometry

[d] Using carbon analyzer; others using neutron activation analysis

of sediment corresponds to different time periods depending on local sedimentation rates. In addition to such sedimentation rate effects, the homogenization of surface sediments by benthos[4] and other processes, and the further averaging of finite sectioning, result in apparent enrichment factors which underestimate the degree of enrichment of certain elements in sediments deposited in the year the cores were collected (1972). However, the enrichment factors shown in Table 1, while not corrected to a specific time of deposition, illustrate important trends. The elements Pb, Sb, and Zn have the highest and similar degrees of enrichment (about 3.5). The concentration of Sb, however, is low in comparison to many other elements, and little is known of its biogeochemistry. The elements Br, Cd, and As also are enriched in surface sediments to a similar extent (about 2.3) but significantly less so than the first group (Pb, Sb, Ze). Among the next group of elements (Mn, P, Cu, Cr, and Na enrichment factor ca. 1.6) are several which may concentrate significantly by diagenetic processes. Robbins and Callender[5] and others have shown that enrichment of manganese in surface sediments can be accounted for in terms of a recycling process involving burial, dissolution, upward migration and reprecipitation. Similarly, P (along with Fe) may undergo diagenetic remobilization in near-surface sediments although Williams et al.[6] consider that enrichments of phosphorous in Lake Erie sediments are mainly the result of increased anthropogenic loadings. Cline and Upchurch[7] have presented evidence for diagenetic enrichment of copper in surface sediments. Others (see Kemp et al.[8]) have suggested anthropogenic inputs as the principal cause. The observed enrichment of calcium in near-surface sediments is probably significant statistically and may reflect additions of autochthonous calcium carbonate (calcite) which dissolve on burial. However, Kemp et al.[8] have observed both cases of enrichment and depletion of calcium in cores from Lake Erie. The only element which may be significantly depleted in surface sediments is acid soluble potassium. Only 25% of the total (NAA) potassium in near-surface sediments is extracted by acid treatments. Our data, as well as those of Kemp et al.[8] for Lake Erie sediments, indicate no significant enrichment of total in surface sediments. Thus, the effect, if real, may be the result of postdepositional changes in the structure of clay minerals, resulting in increased extractability of potassium in strong acid.

The distribution of organic and carbonate (inorganic) carbon in near-surface sediments, illustrates the complementarity of these variables seen elsewhere in the lakes (e.g., Thomas et al.[9]), as well as their very systematic variation over this lake basin. The elements calcium and magnesium, as well as the fraction of sediment by dry weight soluble in acid, are all strongly correlated with inorganic carbon, which is not unexpected since they originate primarily from allochthonous calcite and dolomite minerals. Because of their mean grain size, these two minerals tend to be deposited preferentially toward the margins of depositional basins of fine-grained sediments. Many elements, including the nonconservative "pollutant" elements, positively correlate with organic carbon, as do elements such as potassium and the rare earths associated with clay minerals. Such elements tend to increase in concentration with increasing distance offshore out to about 20 to 30 km from the eastern side of the lake. This behavior is illustrated also in the distribution of "excess" or net element concentrations in surface sediments. The distribution of excess lead (surface-background concentrations) is shown in Figure 1. As the sedimentation rates are known for many cores either from measurements of ^{210}Pb or ^{137}Cs profiles,[2,3] or by inference from the thickness of postglacial sedimentary member[10] (Waukegan member) where radioactivity data are either unavailable or unreliable, the net flux of each element may be estimated as the product of the net concentration and sedimentation rate in g/cm^2/yr. A contour map for the net flux of lead deposited in the southern basin of the lake is shown in Figure 2. Because mass sedimentation rates tend to increase toward the eastern margin of the basin, highest net fluxes, in contrast to net concentration, are skewed toward the basin margin. Other "pollutant" elements have similar flux distributions. On the basis of such contours, it is possible to estimate net loadings to the southern basin of the lake with greater accuracy than has been possible in other studies. Values for the approximate recent anthropogenic loadings to the sediments are given in Table 1. It should be emphasized that the data here have not been corrected for sedimentation mixing

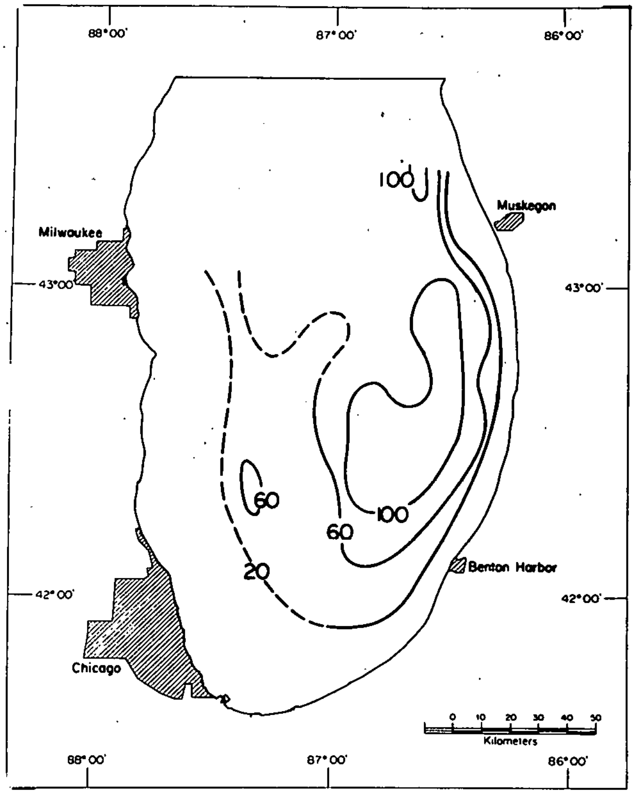

FIG. 1.--The distribution of excess lead in surface sediments of southern Lake Michigan expressed in $\mu g\ g^{-1}$.

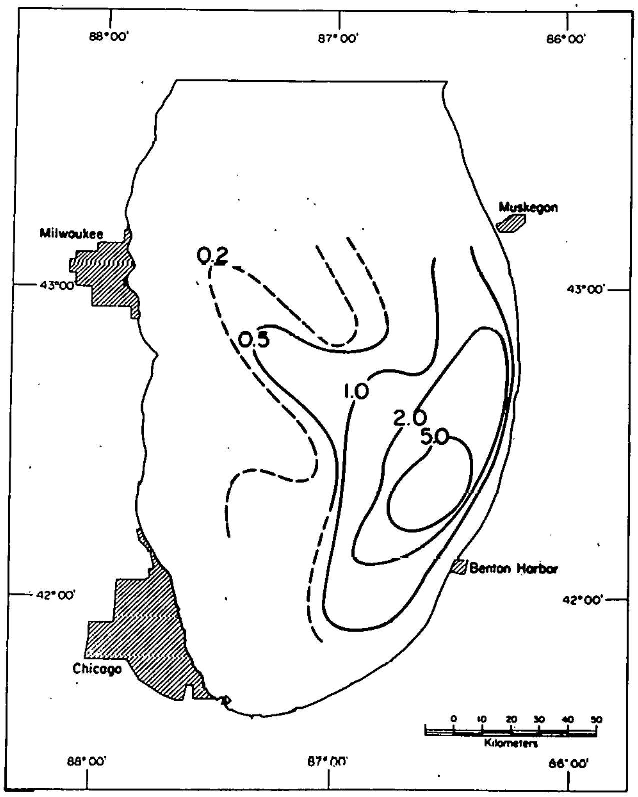

FIG. 2.--The distribution of excess lead in surface sediments of southern Lake Michigan expressed in terms of the annual flux, $\mu g \cdot cm^{-2}\ yr^{-1}$.

and finite interval effects. Since excess lead-210 comes almost entirely from atmospheric inputs, while excess stable lead can originate from both atmospheric and nonatmospheric (river and direct runoff) sources, then if ^{210}Pb and stable lead of atmospheric origin have the same chemical properties, excess ^{210}Pb should be a tracer for stable lead originating from the atmosphere; and variations in the ^{210}Pb/Pb ratio should be indicative of different sources of lead. Previously we have found that the flux of Pb to the sediments near the center of the lake was comparable to the flux from the atmosphere.[2] The value of the ratio is set equal to 1.0 for cores from the center of the basin, and resulting contour map is shown in Figure 3. There is a very systematic trend toward diminishing values of the ratio toward the shore, and therefore, toward river sources of lead, as we had expected on the basis of a few cores previously studied. If the assumptions of our treatment are correct, it appears that excess lead-bearing particles from rivers have a greater range parallel to shore than along a direction perpendicular to shore. Contributions from individual rivers appear to be undiscernible, being averaged out by long-shore transport,

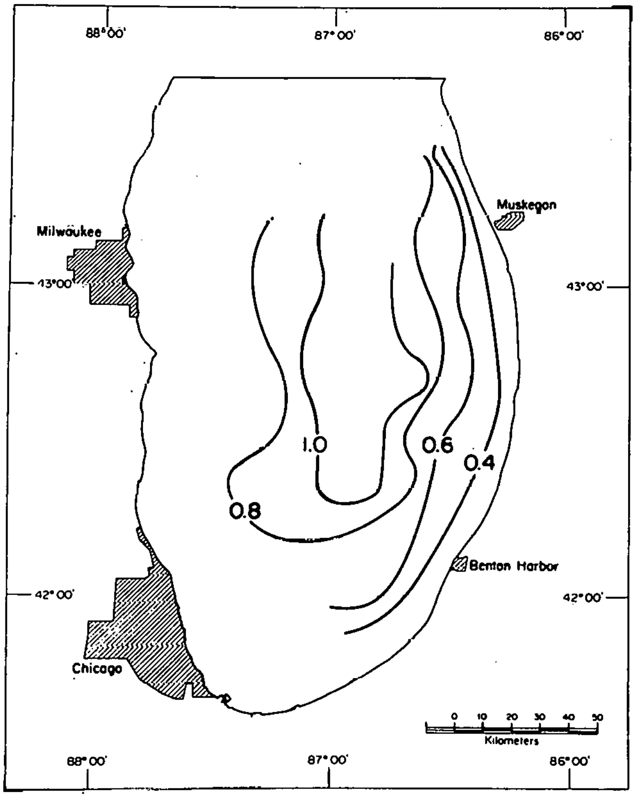

FIG. 3.--The variation in the ratio of the annual fluxes of excess ^{210}Pb and lead in the surface sediments of southern Lake Michigan expressed in $\text{pCi} \cdot \mu\text{g}^{-1}$.

and are apparently constrained to the eastern margin of the basin. Much further work is required to determine the validity and significance of this approach.

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04495

A METHOD FOR DETERMINING FLY ASH CONCENTRATIONS IN LAKE MICHIGAN WATER

J. A. Burger,* C. Seils, and G. T. Tisue

Trace elements mobilized during coal combustion are emitted to the environment largely in association with that fraction of the fly ash which escapes stack gas cleaning devices. As exceptions, mercury, selenium, chlorine, and bromine occur in gaseous form.[1,2] Some elements are both enriched in the very fine fly ash fraction and concentrated near the particles' outer surfaces.[3] Thus, any study of the environmental behavior of trace elements derived from coal combustion must begin with an assessment of the transport and physicochemical behavior of fly ash.

For natural waters, fly ash concentrations, residence times, and leaching rates are of particular interest. Fly ash particles are sufficiently distinctive morphologically and chemically to be distinguishable in common environmental particulate assemblages (Figure 1).[4] This distinctiveness has led to their recognition in air and water samples,[5] but little is known of the concentrations of fly ash particles in natural waters. The problem is similar to the determination of asbestos fibers in water.†

Our initial efforts focussed on the preparation of standards. We found that very dilute slurries, prepared by suspending fly ash (NBS SRM 1633) of > 1 μm diameter in water, could be counted reproducibly by two independent methods. An electronic particle analyzer (Coulter Counter) yielded concentration values that stood in reasonable agreement with those obtained by counting individual fly ash particles in scanning electron photomicrographs (SEM) of 0.45 μm Nuclepore membrane filters through which known volumes of sample had passed. These results are shown in Table 1 for two fly ash suspensions

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†In this regard, we gratefully acknowledge the guidance provided by Prof. W. Hallenbeck (School of Public Health, University of Illinois Medical Center, Chicago), whose work with asbestos served as a model for these investigations.

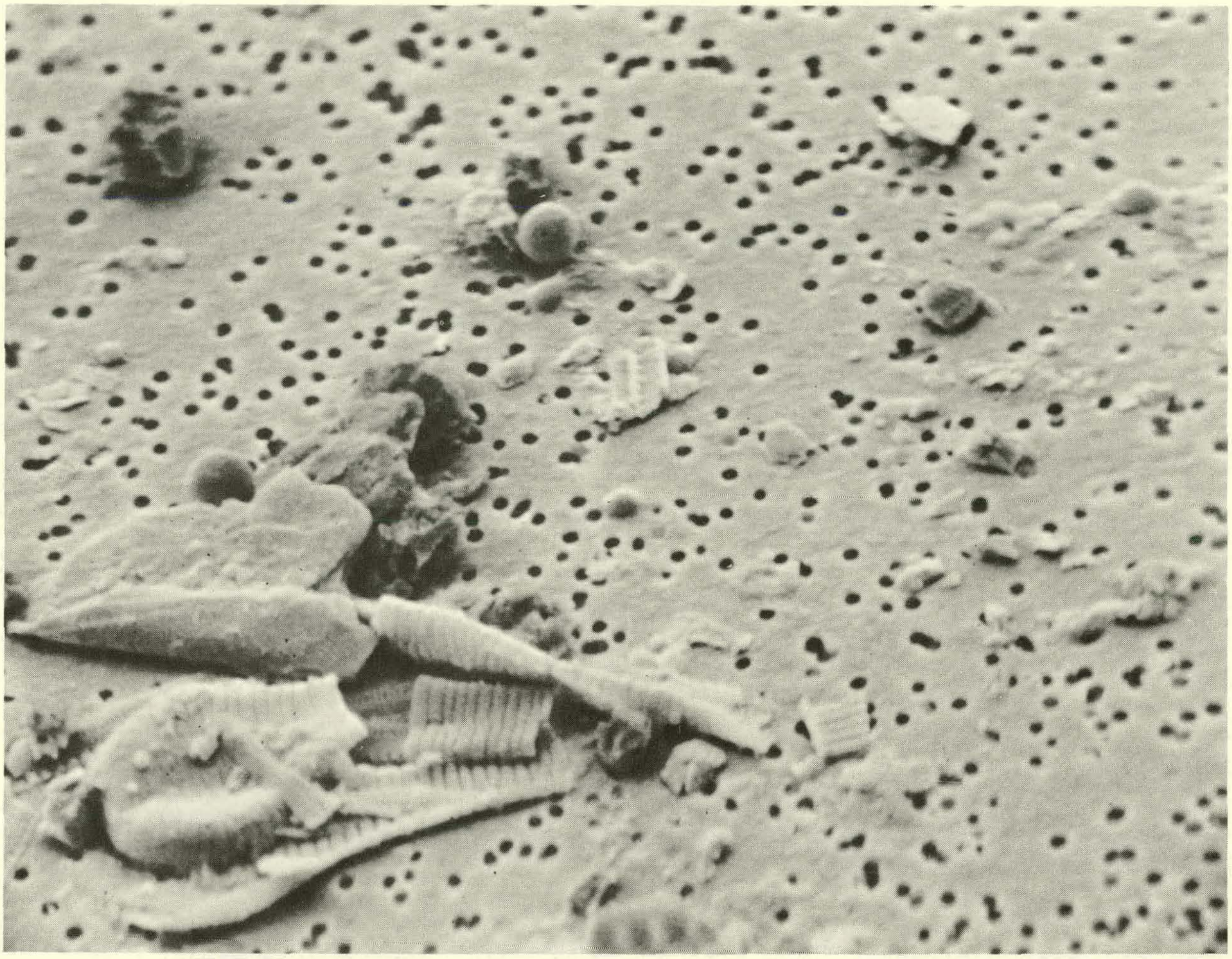

FIG. 1.--Fly ash spheres amidst diatom fragments and other detritus from Lake Michigan surface water collected December 1976, 10 km SW of Grand Haven, Michigan (× 3000 before photographic enlargement; diameter of the membrane apertures is 0.45 μm).

Table 1. Enumeration of fly ash particles in standards

Sample	Enumeration method	Concentration, particles/liter
A (8—20 μm)	Coulter Counter	1.9×10^9 ($N = 3$, $\sigma = 0.1 \times 10^9$)
	SEM	5.39×10^9 ($N = 4$, $\sigma = 0.1 \times 10^9$)
B (<8 μm)	Coulter Counter	1.4×10^8 ($N = 6$, $\sigma = 0.6 \times 10^8$)
	SEM	5.0×10^8 ($N = 2$)

following size classification by elutriation.[6]

We then used the standardized slurries to make known additions of fly ash particles to unfiltered Lake Michigan water. Table 2 shows the agreement between observed and expected concentrations in the spiked lake water. Water collected at a depth of 3 m, 10 km SW of Grand Haven, Michigan, during December 1976, was also analyzed. It contained 5.9×10^5 recognizable fly ash particles per liter.

Enumerating low concentrations of small particles by microscopy at high magnifications involves unavoidable sampling problems. In order to obtain an estimate of precision, and to develop guidelines for determining optimum water volumes and numbers of fields to be examined by SEM, it is desirable to have a mathematical description of the frequency distribution of fly ash counts in scanning electron photomicrographs. We applied the approach suggested by Hallenbeck et al.[7] to the data for the three samples reported in Table 2. These workers found that water-borne asbestos fibers retained on membrane filters exhibited a Poisson distribution. Using the data shown in Table 3, we tested the likelihood that fly ash particles will also be found to have this distribution. Table 4 presents the diagnostic statistics for these data. The frequency distribution of counts of fly ash particles clearly does <u>not</u> follow a

Table 2. Addition of fly ash standard to lake water

Sample	Fly ash concentration, particles/liter
T-40-T[a]	0.7×10^5
8—20 µm standard	2.9×10^5
T-40-T + standard	3.6×10^5 (predicted)
	3.3×10^5 (observed)

[a] After destruction of organic matter with H_2O_2. This sample was collected during August 1975 near the mouth of Grand Traverse Bay at the top of the thermocline. We thank Prof. Janet Holden, School of Public Health, University of Illinois Medical Center, for making it available to us.

Table 3. Frequency distributions of fly ash particles per SEM field of view

Counts of flyash particles/field of view	Number of fields of view with that count		
	Sample 1	Sample 2	Sample 3
0	80	113	120
1	24	22	32
2	9	12	14
3	3	1	10
4	2	1	2
5	0	1	1
6	1	0	1
7	0	0	0
8	1	0	0
	120	150	180

Table 4. Statistical analysis of the data

	Sample 1	Sample 2	Sample 3
Fields of view	120	150	180
$\bar{x}$, particles per field	0.608	0.387	0.606
Standard deviation, s, particles/field	1.211	0.809	1.070
Variance, s^2	1.46716	0.65485	1.1452
$\chi^2 = (N - 1)s^2\sqrt{x}$	287.0	252.3	338.5

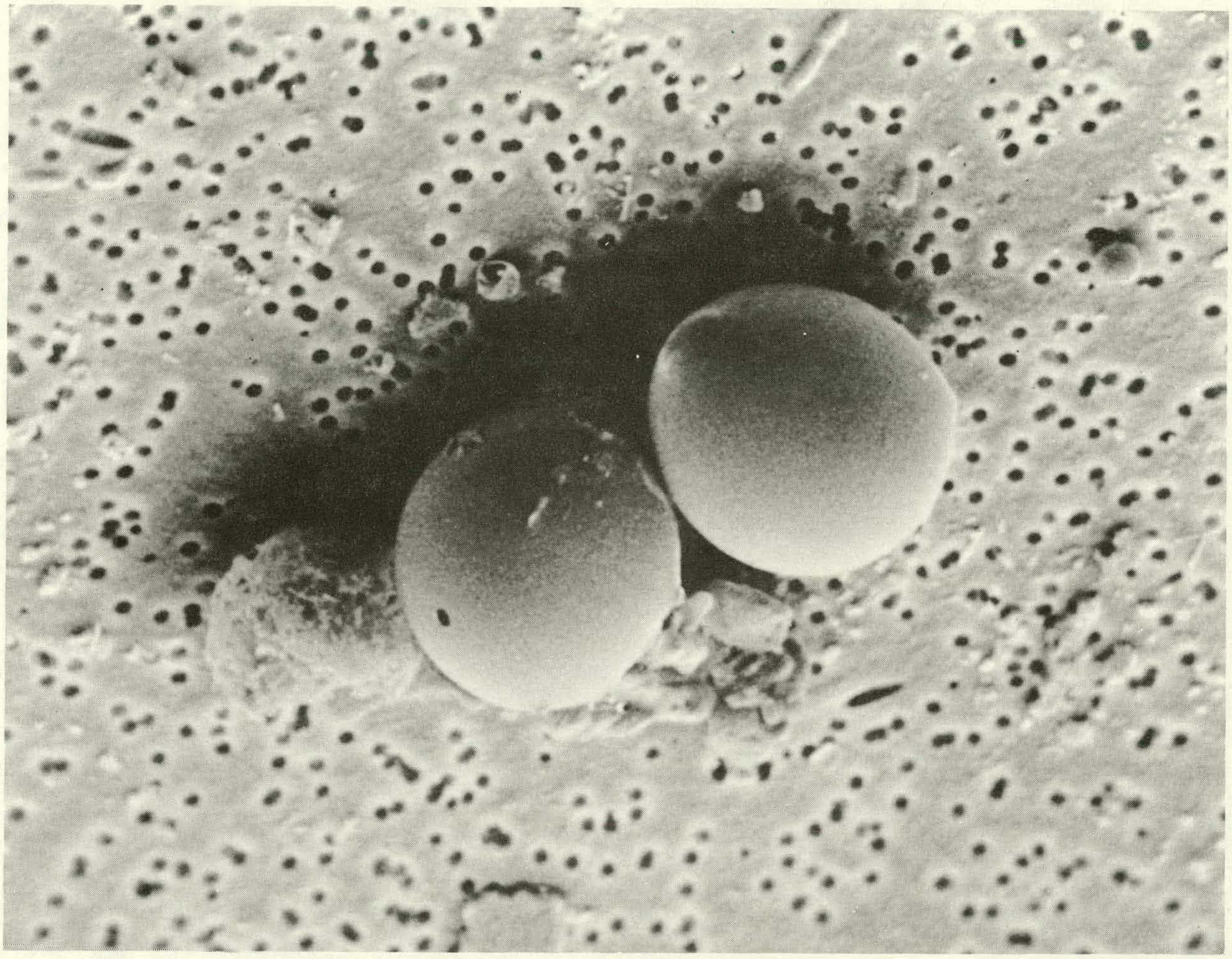

FIG. 2.--Cluster of fly ash spheres observed in sample T-40-T (same magnification as in Figure 1).

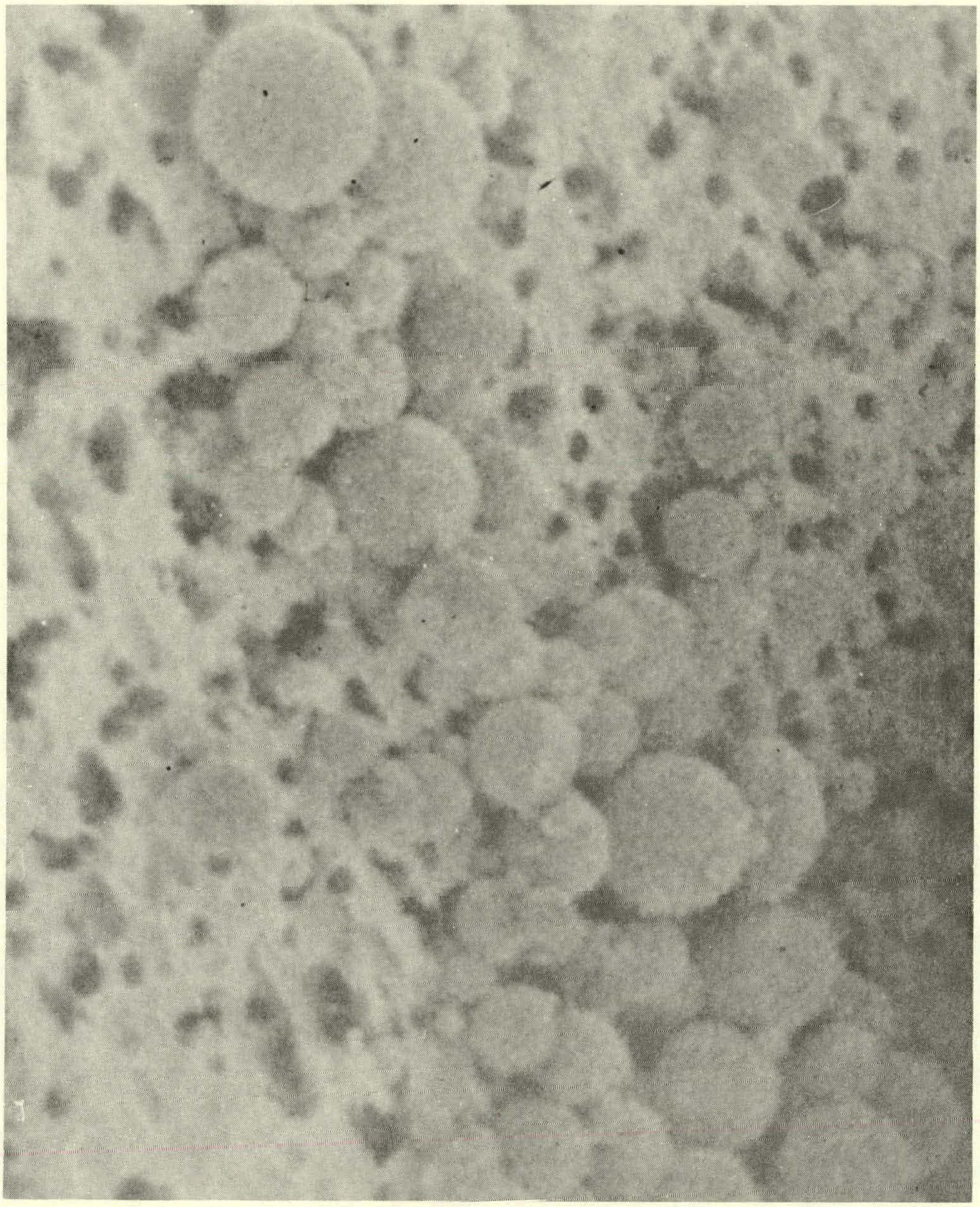

FIG. 3.--Cluster of spheres in fly ash collected downstream of electrostatic precipitators at a local power plant (the diameter of the membrane apertures is 0.45 μm).

Poisson distribution. Even without calculating the χ^2 statistic, the lack of agreement between the mean and the variance makes apparent the inapplicability of the Poisson distribution function.

This result is consistent with our qualitative observations that fly ash particles often occur in groups of two or three (Figure 2). There are two sound reasons to expect such clustering: a fraction of coal fly ash is magnetic and thus self-adhering; and particles collected immediately downstream from an electrostatic precipitator at a local power plant occurred in extensive clusters that showed signs of sintering (Figure 3). This clustering probably is responsible for our observation that the Coulter Counter gave consistently lower estimates of particle concentrations than did scanning electron micrography.

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04496

THE DISSOLUTION OF ASTERIONELLA FORMOSA FRUSTULES

E. M. Yaguchi and H. L. Conway

Kinetics of the dissolution of frustules of Asterionella formosa, an important Lake Michigan planktonic diatom, were studied to determine the role of this process in the cycling of silicon in Lake Michigan. The results of this preliminary study will be useful in evaluating field data, developing mathematical models for silicon cycling, and relating frustule dissolution to the cycling of various trace elements.

A uniform population was cultured in the laboratory in synthetic lake water and allowed to die from nutrient limitation (silicon). The cells were then removed by filtration, resuspended in 2 liters of low-silicon synthetic lake water in a Pyrex flask, and kept constantly stirred at 15°C. Such conditions approximate those found in the epilimnion of Lake Michigan. Samples were taken at varying time intervals for 30 days and analyzed for soluble reactive silicon and amorphous (particulate) silicon. A control flask containing the same medium, but without cells, was treated similarly.

The process of frustule dissolution appeared to follow first-order kinetics, with a rate constant of 0.12 days^{-1} (Figure 1). Nearly two-thirds of the frustules (measured as amorphous silicon) disappeared during the 30 days. The slope of the line labelled "Cells, total" was not significantly different from zero (Student's t-test, $\alpha = 0.05$), implying a material balance for silicon had been attained during the course of the experiment. If soluble unreactive silicon compounds (colloids or polymerized silicic acid molecules) were being formed as the dissolution proceeded, the sum of soluble-reactive plus amorphous silicon would have decreased owing to the presence of a third component in the system. This observation agrees with literature reports that colloidal silicon is unstable in natural waters and disappears spontaneously[1] and that monomeric silicic acid (H_4SiO_4) is the dominant chemical species in solution at pH <9.[2] These results facilitate interpretation of our studies on silicon cycling in Lake Michigan, i.e., they indicated no formation of unreactive intermediate silicon species in the dissolution process.

Results from the control flask suggest that soluble reactive silicon was being leached from the flask walls at a very slow rate, approximately 0.1 µm Si/day. This amount is negligible when compared with the amount of reactive silicon resulting from frustule dissolution. At no time was any amorphous silicon detected in the control flask.

Scanning electron micrographs were taken of frustules from the experimental flask on the 15th and 30th days of the experiment (Figure 2). An increase in the proportion of broken frustules was seen on the 30th day. This breakage would have resulted from the constant turbulence and the weakening of the frustule due to dissolution and thinning, and in turn, would have increased the surface area, further enhancing dissolution.

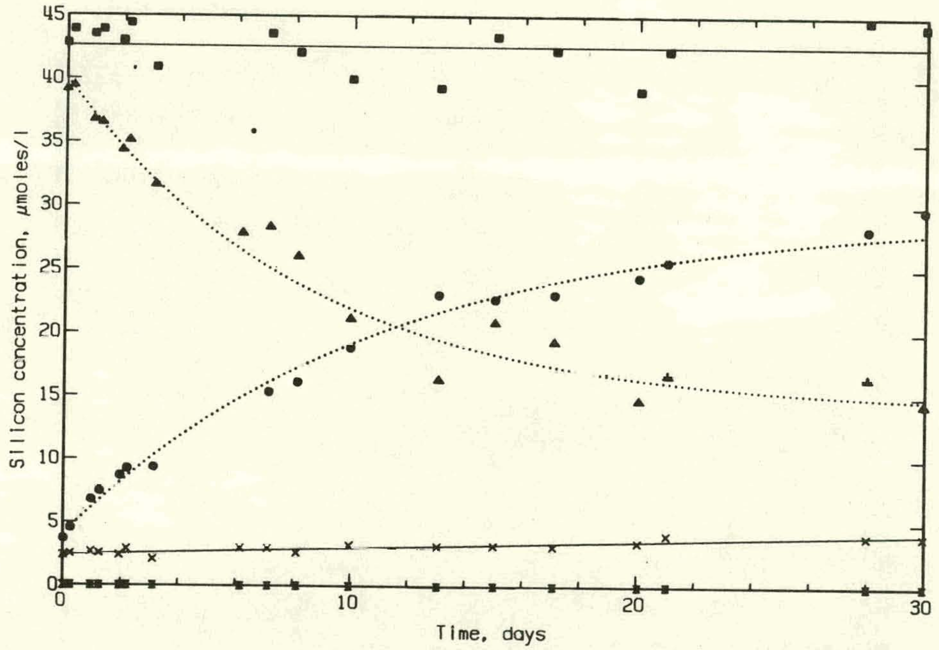

FIG. 1.--Soluble reactive silicon (test ●; control x) and amorphous silicon (test ▲; control ⧗) and the sum of soluble and amorphous silicon (test ■), during the dissolution experiment.

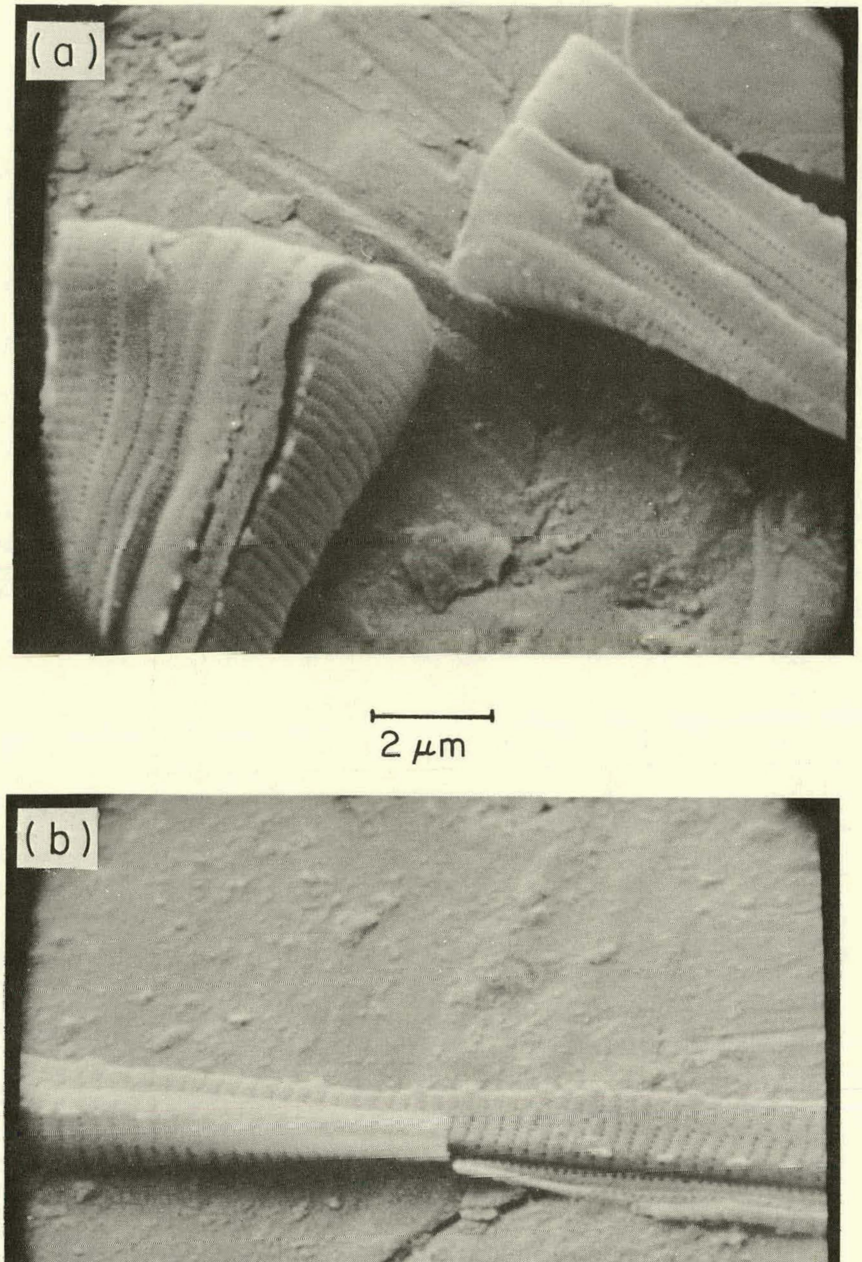

FIG. 2.--Scanning electron micrographs of Asterionella formosa frustules at (a) 15th day and (b) 30th day of the dissolution experiment.

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04497

REGENERATION OF SILICON FROM SEDIMENTS OF THE GREAT LAKES

J. A. Robbins,* K. Remmert,* and D. N. Edgington

The role of the sediments in the cycling of silicon in the Great Lakes may have been underestimated. It is commonly held that most, if not all, of the diatom frustules formed by conversion of soluble reactive silicon to amorphous silica in epilimnetic waters, dissolve within the water column. According to this view, recent sediments should have little or no influence on the seasonal transfer of silicon between soluble and biogenic forms. Yet recent investigations have shown for a core from Lake Michigan that intact diatom frustules occur in modern sediments[1] and that there is an appreciable gradient of dissolved silica in pore water near the surface of one sediment-water interface.[2] Similarly, Conway has shown that there is large increase in dissolved silica concentration in the bottom meter of the water column.[3] Such gradients imply significant fluxes of silicon to overlying waters. Thus, recent sediments would seem to be at once a sink and a source of silicon in the Great Lakes. To gain a better appreciation of the role recent deposits may have in the cycling process, we have undertaken a series of field and laboratory experiments.

In one study[4] the upper 60 cm of seven undisturbed cores from the southern basins of Lakes Michigan and Huron were sectioned and squeezed aboard ship at in situ temperatures (6 ± 0.2°C) in a nitrogen atmosphere (< 2 ppm O_2). Pore water samples were analyzed immediately for molybdate-complexable silica (reactive). Acidified samples were analyzed subsequently using flame atomic absorption spectrophotometry for total silicon. An example of the vertical distribution of dissolved silicon in cores at two locations in Lake Michigan is shown in Figure 1. There is essentially no difference in either core between total silicon and reactive silicon. The very sharp concentration gradients indicate fluxes as high as 4000 μg Si/cm^2/yr if interstitial transport of silicon occurs primarily by a diffusional mechanism. On

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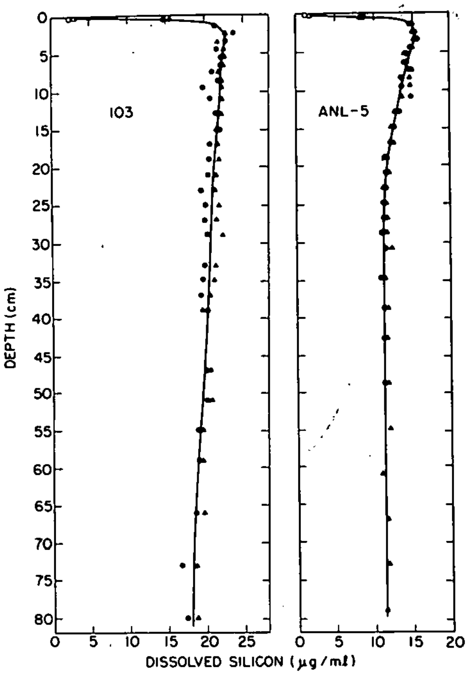

FIG. 1.--Vertical distribution of dissolved silica in cores from Lake Michigan.

the basis of such gradients in cores from each lake, we estimate that the annual flux from the fine-grained sediments comprises at least 5% of the total silicon in Lakes Huron and Michigan. This annual release from sediments is comparable to or exceeds annual tributary loadings of soluble silicon. Recently Nriagu[5] has reached a similar conclusion concerning the significance of the sediments as a source of soluble silicon in Lakes Superior, Erie, and Ontario.

Our flux estimates may even be conservative as it is known that benthic organisms inhabiting near-surface sediments can enhance silicon transfer across the sediment-water interface.[6] In the Great Lakes, large populations of oligochaete worms and amphipod Pontoporeia affinis occur widely. Thus it is expected that biological activity would have an effect on silica exchange in these lakes. We have some indirect evidence of benthic organism influence from comparison of interstitial silicon concentration gradients in these seven cores with radioactivity data. The value of the surficial silicon gradient is inversely correlated with the depth of sediment mixing inferred from either cesium-137 or lead-210 distributions. A recurrent feature in the distribution of silicon in these cores is the presence of a subsurface maximum. On the basis of a diagenic model of reaction and diffusion we have shown that this profile can arise if the concentration of silicon in pore water is controlled by the rate of dissolution of amorphous silica.[2] The solid line shown in Figure 1 represent a least-squares fit of the diffusion-reaction equation (this analysis) to the data.

In a subsequent series of experiments[7] we estimated the flux of silica directly rather than on the basis of interstitial gradients. Additional cores from Lakes Michigan and Huron, plus cores from Lake Erie, were stored at in situ temperatures (~6°C) in their plastic core liners with overlying water. Increases in concentration of reactive silica over periods of hours to weeks in occasionally stirred overlying water exposed to air provided estimates of exchange of silica across the sediment-water interface. In three cores from Lake Michigan rates ranged from 1460 to 2100 μg Si/cm^2/yr, averaging 1820 μg/cm^2/yr; in eight cores from Lake Huron rates ranged from 1070 to 2050, averaging 1340 μg/cm^2/yr. In thirteen cores from Lake Erie, collected at the end of June, rates ranged from 710 to 4790 averaging 2340 μg/cm^2/yr (see Table 1) with highest fluxes tending to occur in central parts of depositional basins where sediments are relatively higher in organic carbon. Provided measured fluxes represent annual regeneration rates, sediments in Lake Erie, like those of Lakes Michigan and Huron, also contribute substantially to the inventory of dissolved silica in the water column and constitute a source comparable to tributary rivers.

Silica release rates can be reduced to levels expected from molecular diffusion only by the addition of chlordane or rotenone in the overlying water or by the exposure of the entire core to intense gamma radiation (5 Mrad ^{60}Co) (see Table 2). Such treatments presumably remove the biological influence on silica movement in the cores. However, when benthic invertebrates (tubificids and Pontoporeia) were added to two cores from Lake Michigan, the silica release rate was not increased. The effects of mixing processes at the interface are being further investigated.

These studies raise many important questions concerning the role of the benthos, the nature of the sources of dissolved silicon, the seasonal variability of silica released, etc., but show that the recent sedimentary deposits cannot be neglected in considering the cycle of silicon in the Great Lakes.

Table 1. Silicon release from sediments.

Station	Location		Si flux from sediments, µg Si/cm^2/yr
Lake Michigan[a]			
5-1	43° 06' N	86° 22.8' W	2100
5-2	43° 06' N	86° 22.8' W	1460
5-3	43° 06' N	86° 22.8' W	1900
Northern Lake Huron[b]			
1-10	44° 20' N	82° 55.8' W	1070
2-11	44° 20' N	82° 20.0' W	1140
2-14	44° 20' N	82° 20.0' W	1016
4-3	44° 20' N	81° 50.0' W	1320
4-4	44° 20' N	81° 50.0' W	1240
43-2	45° 20' N	83° 00.0' W	1650
44-1	45° 20' N	82° 50.0' W	2050
46-2	45° 20' N	82° 30.0' W	1260
Lake Erie[c]			
6	41° 46.2' N	82° 58.9' W	4790
10	41° 31.0' N	82° 30.0' W	2900
13	41° 49.0' N	82° 06.3' W	3390
16	41° 54.0' N	81° 53.0' W	2340
20	42° 02.0' N	81° 36.0' W	2800
24	42° 03.8' N	81° 17.7' W	2390
28	42° 08.7' N	81° 00.4' W	2010
30	42° 24.8' N	80° 47.7' W	1980
31	42° 13.6' N	80° 42.6' W	710
33	42° 26.5' N	80° 15.0' W	1670
38	42° 29.1' N	80° 02.0' W	1730
42	42° 30.6' N	79° 53.6' W	1690
47	42° 32.8' N	79° 44.9' W	2040

[a] Cores taken 12 September 1976.
[b] Cores taken 20—30 July 1976.
[c] Cores taken 19—28 June 1976.

Table 2. Effects of selected treatments on silica release from sediments.

Core	Treatment	Release rate (µg Si/cm^2/hr) Before treatment	Release rate (µg Si/cm^2/hr) After treatment
NLH 2-11	Rotenone	0.126 ± 0.021	0.091 ± 0.020
NLH 2-14	Chlordane	0.116 ± 0.016	0.036 ± 0.010
NLH 4-4	Control	0.139 ± 0.012	0.086 ± 0.009
NLH 44-1	Gamma radiation	0.234 ± 0.010	0.116 ± 0.013
LM 5-1	Tubificids	0.241 ± 0.022	0.177 ± 0.012
LM 5-3	Pontoporeia	0.217 ± 0.015	0.202 ± 0.016
LM 5-2	Control	0.166 ± 0.017	0.120 ± 0.014
NLH 44-3	Sediment stirred at coring	0.032 ± 0.016	---

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04498

COMPARATIVE LEAD-210, CESIUM-137, AND POLLEN-GEOCHRONOLOGIES OF RECENT SEDIMENTS FROM LAKES ERIE AND ONTARIO

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Several methods have been employed to determine rates of accumulation of recent sediments in the Great Lakes. We have used the naturally-occurring radionuclide, lead-210, to measure sedimentation rates in Lakes Michigan[1] and Huron.[2] Geochronology with lead-210 is based on the radioactive decay of the isotope following burial in sediments. This method can reveal changes in sedimentation occurring over the past 100 years or so.[3] Although more limited in its application, the distribution of fallout cesium-137 can also be used to obtain estimates of recent sedimentation rates.[4,5] Geochronology with cesium-137 is based on the occurrence of a horizon in sediments corresponding to the onset of nuclear testing about 25 years ago. Kemp et al. have used ragweed pollen profiles to determine sedimentation rates in the Great Lakes.[6] The ragweed pollen method relies on the presence of a sedimentary horizon associated with a dramatic increase in ragweed vegetation following forest clearance in the mid-1800's. Both of these latter methods, relying as they do on the appearance of a horizon event, provide a measure of average rates of sedimentation over approximately the past 25 and 130 years, respectively. In several studies we have found that both ^{137}Cs and ^{210}Pb give consistent measures of sedimentation rates when the effects of steady-state mixing of surface sediments,[1,5] presumably by the activities of benthic organisms,[7] are properly taken into account. In one study we made a rough comparison of ^{210}Pb and ragweed pollen sedimentation rates[1] which indicated that the methods provide consistent results. The only other comparison of lead-210 with palynological methods in the Great Lakes by Bruland et al. was inconclusive.[8] Unfortunately, their cores from Lake Superior had sedimentation rates so low that both excess ^{210}Pb and Ambrosia pollen were confined to the

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upper few centimeters.

In this last year we have examined the distribution of lead-210, cesium-137, and Ambrosia (ragweed) pollen in five cores from Lakes Erie and Ontario, where sedimentation rates are sufficiently high (up to 1 cm/yr) to provide an accurate comparison. We have found that in two of the five cores (see Table 1), including a core (G16) from Lake Erie collected by diver, all three measures of sedimentation rates are in excellent agreement. In two other cores, rates based on lead-210 were significantly higher than those from cesium-137 or ragweed pollen. Lower average rates inferred from these latter methods appear to have resulted from occasional massive losses of sediments at some locations in the lakes. Such events are apparent in the distribution of lead-210 but not of pollen and correlate with the occurrence of major storms. An example is shown for the distribution of lead-210, cesium-137, and ragweed pollen in core M32 from the deepest part of the eastern basin of Lake Erie (Figure 1). The distribution of excess lead-210 is not strictly exponential as one would expect for a uniform sedimentation rate and constant flux of lead-210 to the sediment surface (R+E). Rather, the profile is exponential in pieces. The least squares fit to the data represented by the two linear portions indicate equal sedimentation rates above and below the break point at about 6 cm depth.

Table 1. Comparison of sedimentation rates determined by different methods

Station	Location	Water depth, m	Sedimentation rates, $g \cdot cm^{-2} \cdot yr^{-1}$ [a]		
			^{210}Pb	^{137}Cs	Ambrosia
Lake Ontario					
KB	44°05'N 76°25'W	26	0.057 ± 0.01 (0.20)	0.060 ± 0.01 (0.29)	0.064 (0.31)
WB	43°24'N 79°27'W	101	0.078 ± 0.01 (0.34)	0.072 ± 0.02 (0.31)	0.027 (0.12)
Lake Erie					
M32	42°32'N 79°40'W	58	0.441 ± 0.03 (1.4)	0.35 ± 0.04 (~1.2)	0.26 (0.85)
G16	42°00'N 81°36'W	24	0.083 ± 0.005 (0.46)	0.080 ± 0.016 (0.44)	≥ 0.073 (≥ 0.40)
	41°46'N 82°59'W	11	0.096 ± 0.01 (0.20)	0.047 ± 0.02 (~0.10)	0.059 (0.12)

[a] Values in cm/yr given in parentheses.

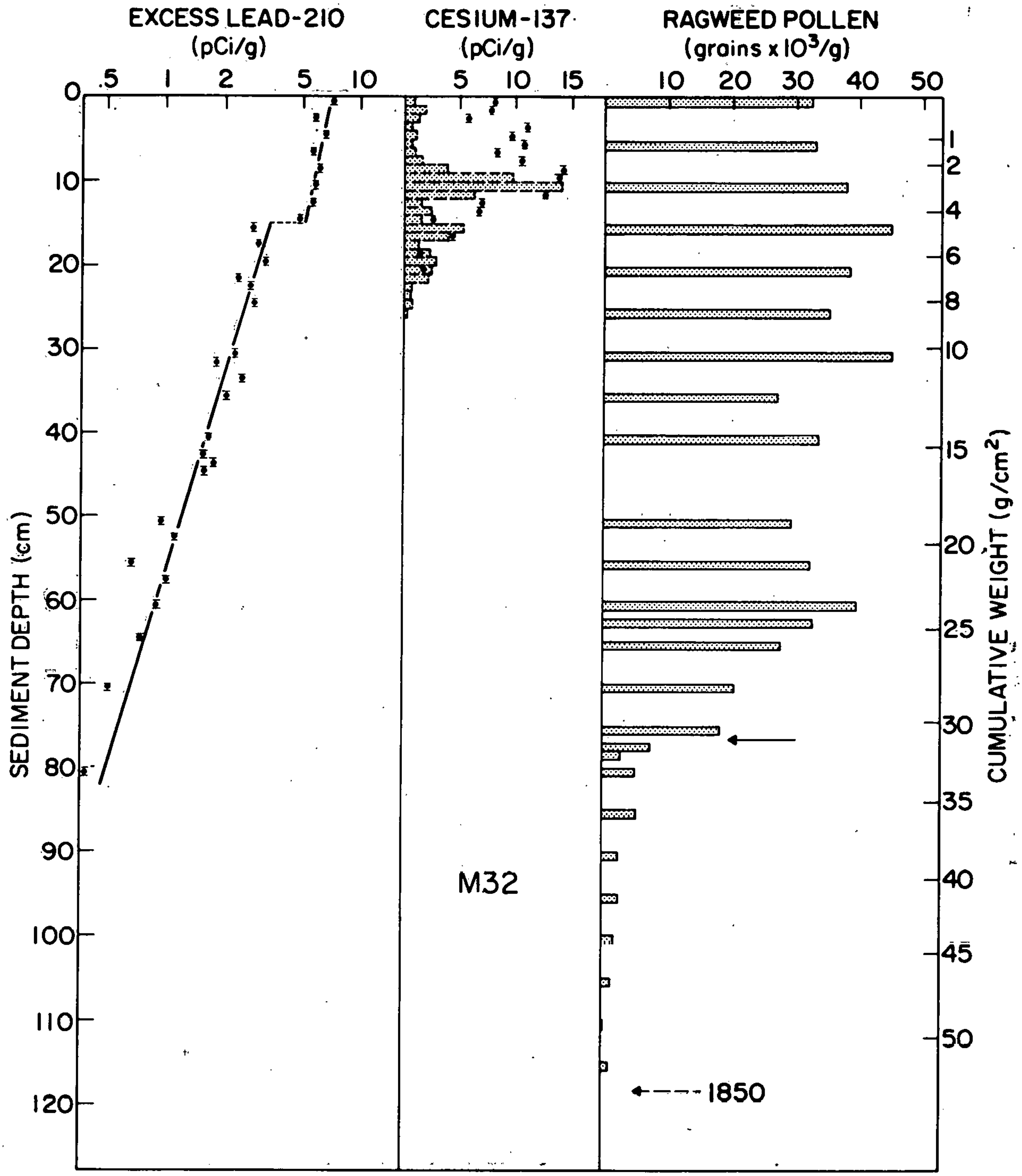

FIG. 1.--Distribution of excess lead-210, cesium-137, and ragweed pollen in Lake Erie sediments.

Such a profile could arise from loss of material at this depth. Assuming that the distribution of excess lead-210 would be exponential if sediment loss had not occurred, it is possible to infer that about 6 g/cm^2 must have been stripped from surface sediments about 11 years ago ($\sim$1960). This date corresponds closely to a major storm on this lake in November 1958. From our studies of lead-210 distributions in sediments from Lake Michigan, we had already found evidence in sedimentary records of major storm events.[3] Because pollen influx around this period was not changing markedly, and because of the statistical scatter in the data, an event such as loss of this much sediment around 1940 would not be apparent in the pollen profile. These results show that non-continuous sedimentation processes lead to significant differences between the long-term average and the short-term rates of sedimentation. Both measures may be valid but not equivalent. Rates based on lead-210 and ragweed pollen in core M32 are brought into closer accord by inclusion of the supposedly eroded piece of sediment. In one core from western Lake Erie, rates of sedimentation inferred from the three methods all disagree as can be seen from Table 1. The reasons for this are unclear, but it might be expected that in very shallow water (11 m) both physical mixing and biological reworking of sediments could be quite important, and the distributions of all three quantities could be artifacts.

The distribution of cesium-137 shown in Figure 1 is compared with the distribution expected on the basis of immediate transfer to underlying sediments of cesium-137 deposited on surface water (histogram). Location of the expected distribution is determined by the sedimentation rate computed from the excess lead-210 profile. While the initial rise of cesium-137 is consistent with the measured distribution, the recent activity in sediments (about 1960 to the present time) is higher than expected on the basis of known annual atmospheric inputs. A similar departure is found in the diver-collected core, and therefore, the excess ^{137}Cs cannot be attributed to a disturbance of surface material during coring. High surface values are evidently not maintained by surface mixing either, since excess lead-210 distributions are exponential in cores G16 and M32 up to the sediment-water interface. It is also unlikely that delayed contributions of cesium-137 originating from erosion or leaching of the radionuclide from the watershed can produce these elevated levels of cesium-137 in surface sediments.[9] A possibility that seems likely to us involves resuspension and lateral movement of surface sediments with high ^{137}Cs contents. Significant quantities of cesium-137 stored temporarily in low or zero net sedimentation areas of the lake may be resuspended and in time contribute to sediments accumulating in more permanent depositional basins.

These ongoing studies of recent sedimentation processes in the Great Lakes illustrate the value of using several independent methods of determining recent sedimentation rates. Such studies are of major value in assessing the role of the sedimentary deposits in the regulation of radioactivity and trace elements in lakes in reconstructing the geochemical history of the Great Lakes from sedimentary records and in understanding the behavior of recent sedimentary deposits as quasipermanent repositories for nonconservative substances.

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04499

SEDIMENTOLOGICAL AND MINERALOGICAL CHARACTERISTICS OF RECENT SEDIMENTS AT SELECTED SITES IN THE SOUTHERN BASIN OF LAKE MICHIGAN

G. T. Tisue and G. Merk*

During the 1976 field season, sediment traps and current meters were set out in the southern basin of Lake Michigan[1] to study the relationship between suspended material and currents. We determined the gross mineralogical and chemical characteristics of the bottom sediments at the locations of these experiments. The results are shown in Table 1.

The stations encompass environments ranging from the shallow inshore regime at station 16B, where sand predominates, to the midlake deep at station 18, where principally clay size materials deposit. Stations 5, 6, 19, and 10 are intermediate in nature and exhibit the poor size sorting typical of locations that receive periodic inputs of relatively coarse materials.

It is noteworthy that even the sediments at station 6, 25 km offshore in water 85 m deep, come under the influence of forces sufficient to advect coarse, dense mineral grains originating in nearshore environments. This effect vanishes in deeper water, and the sediments at station 18 are characterized by very fine material settling in a low energy environment.

Stations 6 and 19 exhibit lower mass sedimentation rates than station 5.[2] This difference probably accounts for the much higher Ca/Mg ratios at 6 and 19, where calcite of autochthonous origin is less "diluted" by inputs of allochthonous dolomite. Cation exchange capacity increases in the order $5 < 6 < 19$. This change closely parallels the increase in clay content, as one would expect.

Sediment from station 5 was selected for more detailed mineralogical characterization because, as a result of poor sorting, it might contain the entire suite of minerals likely to be encountered in the southern basin. Table 2

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Table 1. Mineralological and chemical characteristics of sediment at selected sites

Station No.	Wt % sand[a]	Wt % silt[b]	Wt % clay[c]	CEC[d]	Wt % organic matter	Ca, ppm	Mg, ppm
5	20.0	37.4	42.6	10.3	6.1	546	322
6	18.2	34.4	47.5	11.0	4.7	2340	350
10	8.1	38.4	53.5				
16B	77.4	10.4	12.1				
18	0.4	26.3	73.3				
19	7.7	37.3	54.9	14.9	5.02	3000	381

[a] Analyses of 5 replicate cores from station 5 had relative S.D. = 15.3%.

[b] Analyses of 5 replicate cores from station 5 had relative S.D. = 10.8%

[c] Analyses of 5 replicate cores from station 5 had relative S.D. = 22.5%.

[d] Cation exchange capacity (CEC) in milliequivalents/100 g.

Table 2. Mineralogy at station 5[a]

Size class	Mineral	Relative abundance
Sand	Quartz : Plagioclase : K-Feldspar ::	6 : 1.5 : 1
Silt	Quartz : Dolomite : Plagioclase : K-Feldspar : Calcite ::	14.5 : 6.5 : 3.5 : 1.8 : 1
Clay	Illite ≅ Quartz > Chlorite ≅ Kaolinite > Vermiculite	

[a] Based on 4 subsamples from each of 5 replicate cores.

shows the relative proportions of the major minerals comprising the three classes of grain size at this station. In addition to these major components identified by x-ray diffraction, examination by petrographic microscope revealed the accessory minerals zircon, tourmaline, rutile, and garnet. Quartz is the predominant mineral at this location; it constitutes a significant fraction of even the clay size material. The presence of vermiculite, an expandable clay mineral of high exchange capacity, is noteworthy. It has not been reported previously in Lake Michigan sediments.

Poor size sorting in sediments may arise from differences in the time course for deposition of materials of various grain sizes. For station 5, the sedimentary record indicates that deposition is episodic in the sense that materials of different size distribution are deposited at different times. This conclusion may be reached by direct inspection of sediment texture as a function of depth in core, but it is most easily visualized by means of core x-radiography. Figure 1 reproduces a positive print of such an x-radiograph from a 5" gravity core taken at station 5.

The variations in film optical density that produce the lamellar structures shown in the radiograph arise from corresponding variations in the bulk x-ray attenuation coefficient. These variations are attributable in part to changes in water content which derive in turn from variations in interstitial volume (packing efficiency) with grain size. They may also be due in part to changes in composition of the solid phase. Since at this location the sediment is largely quartz, the first of these two effects is likely the more important.

The deposition process thus is seen to result in alternating layers of coarser and finer material. It is tempting to attribute part of this layering to the annual cycle of tributary runoff which has a maximum in spring that brings increased burdens of coarser material into the lake at the time when isothermal conditions, violent weather, and onshore erosion maximize the opportunity for larger grains to be transported to sites well offshore. Thermal stratification and warm weather doldrums then combine to produce an interval of more nearly quiescent settling of finer grains.

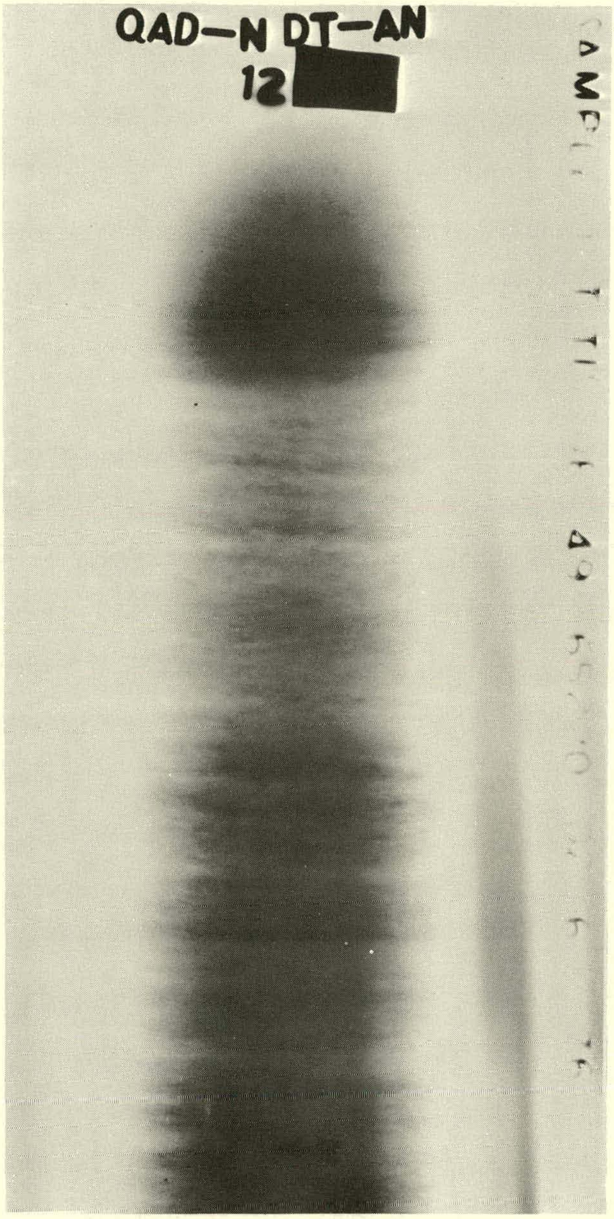

FIG. 1.--Positive print of x-radiograph from 5" core recovered at station 5. The lower edge of the opaque block under the identification code was placed at the sediment-water interface. The arrow was placed 25 cm below this point.

A second putative cause of the layering of coarse and fine materials is the "winnowing" that could result from periodic resuspension of bottom sediments by currents and/or bioturbation. Autumnal gales are commonplace on the Great Lakes, and current meter records show maximum near bottom current speeds from September onwards. Resuspension acts preferentially to erode finer and less dense material, leaving coarser and more dense material in place. Resuspended fines settling later under more quiescent conditions would result in alternating bands of coarse and fine particles.

Whatever the relative contributions of these two mechanisms, each is likely to be characterized by seasonal maxima. To test whether the observed layering might be associated with annual cycles, we determined the average depth interval at which maxima (or minima) recur in the radiograph film optical density. To do this, the core x-radiograph was reduced photographically and copied as a 35 mm black/white transparency. This transparency was scanned longitudinally with a sensitive microdensitometer. Output from the photomultiplier detector in this device was amplified and fed to a strip chart recorder. The microdensitometer scan rate and the recorder drive speed were adjusted until a trace was obtained whose length matched the distance between calibration points on the core at 0 and 25 cm depth. After correcting the ca. 15% enlargement introduced by the x-ray source-to-core-to-film geometry, direct measurements on this trace yielded a value for the average recurrence interval of 0.4 cm (N = 50, sigma = 0.13).

If the lamellae reflect annual deposition cycles, this interval ought to be comparable to the average annual accumulation depth of sediment at this station. The total thickness of the recent sediment approaches 7 m at this location,[3] corresponding to an average annual accumulation depth of about 0.1 cm over the 7,000 year age of the formation. This value refers to fully compacted sediment and thus represents the annual accumulation thickness observed at depth in the core, where compaction is virtually complete. Nearer the sediment-water interface, the water content rises as compaction decreases, reaching about 50% at 25 cm and 85% just below the surface.[2] The annual accumulation that eventually compacts to 0.1 cm thus could range in thickness from about 0.2 cm to about 0.4 cm in the upper 25 cm of the sediment column.

Using ^{210}Pb dating techniques, Robbins and Edgington[2] estimated a mass sedimentation rate at this station corresponding to an average annual accumulation thickness of 0.04 cm for fully compacted sediment. This value leads to predicted annual accumulations of 0.1 to 0.2 cm in the 0 to 25 cm interval, after correction for the observed porosity changes.

The stratification interval we observed in the upper 25 cm of sediment at station 5 (0.4 cm plus or minus 0.13) is sufficiently comparable to the annual accumulation depths inferred from these two independent measurements to reinforce strongly the notion that layering results from the occurrence of annual deposition cycles.

We are grateful to Argonne's QAD personnel for making the x-radiographs and to Dr. P. Groer for assistance with the scanning microdensitometer measurements.

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SETTLING BEHAVIOR OF LACUSTRINE ORGANIC MINERAL AGGREGATES

R. R. P. Chase* and G. T. Tisue

Organic mineral aggregates are a cohesive sediment type composed of an organic matrix sorbed onto mineral flocs. These aggregates often form the uppermost layer of the freshwater benthos, and are especially prevalent in relatively low energy environments. Typical aggregates range in size from a few to a few hundred micrometers and have a loosely articulated structure, often containing void spaces. This imparts to them a low bulk density (1.03501 ± 0.0114 g/cm^3 for Lake Michigan aggregates). Aggregates are deformable and susceptible to disruption under high shear conditions.

Despite their ubiquity, the hydrodynamic behavior of these particles has not been adequately characterized. We have investigated the kinematics of lacustrine organic mineral aggregates by means of a unique laboratory apparatus described earlier.[1] The results show striking departures from predictions of settling behavior based on classical theory.

We observed translational motion of the particles under three sets of conditions: (1) in doubly-distilled water; (2) in Lake Michigan water freed of organic matter by photo-oxidation; and (3) in untreated lake water. The first sequence furnished baseline data against which we compared the results in sequences 2 and 3. Sequence 2 revealed the effects of solution electrolytes on translation. Sequence 3 combined the effects of electrolytes with those of dissolved organic matter.

Figure 1 compares the experimental data from these three sequences to the prediction of the Basset-Boussinesq-Oseen (BBO) equation of motion. (For a discussion of this formulation and earlier tests of its validity, see Ref. 2.) In the distilled water sequence, aggregate behavior deviates sharply from predicted values, especially for the smaller aggregates (small L'). This deviation is nonlinear and may arise from skin friction reduction by the surface layer

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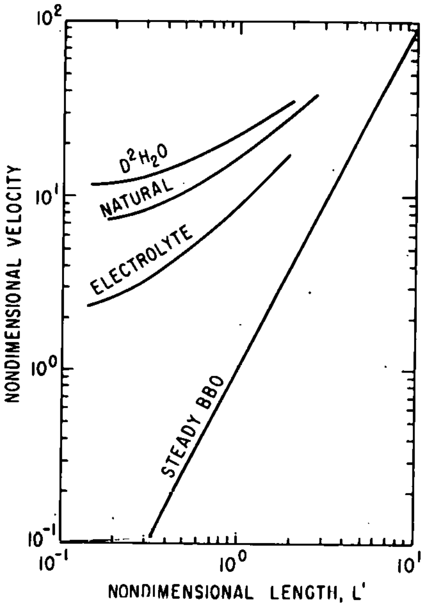

FIG. 1.--Settling velocities (nondimensional units) of lacustrine organic mineral aggregates vs. aggregate size (nondimensional units) in three different media. The straight line represents the prediction of the BBO equation.
(ANL neg. 149-77-317)

of sorbed organic matter. The second sequence (electrolytes only) also shows a nonlinear deviation from predicted behavior, but the overall skin friction reduction is less pronounced than in the absence of electrolytes. Inclusion of naturally occurring dissolved organic substances (sequence 3) caused an increase in settling velocities relative to the electrolyte-only experiments, again indicating a substantial reduction in skin friction.

In all three cases the observed deviations are nonlinear functions of aggregate size. As aggregate size increases, the observed settling velocities approach asymptotically the values predicted by classical theory. This asymptotic behavior suggests a shift in the balancing of the governing equation of motion. A general formulation of this equation is:

$$\left[C_D\rho - C_M\rho\hat{s} + C_E\rho\hat{s} - C_O\rho\hat{s}\right]\tfrac{1}{2}U^2A = V(\delta\rho)g \ ,$$

where

C_D = classical drag coefficient
C_M = surface layer coefficient
C_E = solution electrolyte coefficient
C_O = dissolved organics coefficient
$\rho\hat{s}$ = natural water density
ρ = aggregate density

U = aggregate settling velocity

A = aggregate cross-sectional area

V = aggregate characteristic volume

g = gravitational constant.

One may conclude that organic mineral aggregates experience surface forces altered by interactions of sorbed species (water molecules and organic moieties) with electrolytes and organic solutes. The net effect of these interactions is an increase in translational velocity over that predicted by theory—a nonlinear increase that becomes more pronounced as aggregate size decreases. This result has important implications in natural water systems.

For example, aggregate formation greatly alters the rate at which gravitational settling removes mineral matter of various sizes from the water column. Figure 2 emphasizes this point by showing the ratio of observed aggregate settling velocity to settling rates for individual mineral grains calculated from the BBO equation. This ratio is plotted as a function of mineral grain size for three different aggregate dimensions.

The "line of no influence" shown on the plot illustrates one of the singular features of aggregate dynamics. For any given aggregate size there is a critical grain size (defined by the intersections) above which a particle's

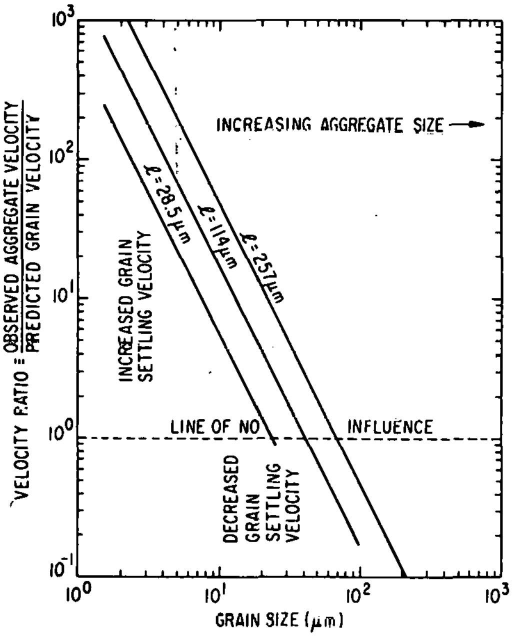

FIG. 2.--Effect on settling velocity of incorporation of a mineral grain in an organic mineral aggregate. The aggregate/grain velocity ratio is plotted as a function of grain size for aggregates of three sizes. (ANL neg. 149-77-316)

settling rate is reduced through incorporation in the aggregate, and below which its settling is accelerated. The accelerations and retardations may assume large values: ~ 65-fold acceleration for a 5 μm grain incorporated in a 114 μm aggregate, ~ 3-fold retardation for a 100 μm grain incorporated in a 257 μm aggregate.

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04501

FACTORS AFFECTING THE COLLECTION EFFICIENCY OF SEDIMENT TRAPS IN LAKE MICHIGAN

M. A. Wahlgren and D. M. Nelson

As a tool for limnological studies on large lakes, sediment traps offer a unique opportunity to obtain samples of particulate material integrated over time intervals as long as 1 or 2 months. Although sediment traps have been used for more than 20 years, the majority of previous studies have been carried out in shallow water and may not be applicable to the conditions of Lake Michigan. In addition, there has been little in situ study of factors affecting collection efficiency. Different studies often show apparently contradictory results; for example, on the question of the influence of trap diameter on collection efficiency, a recent summary[1] lists three studies in which no diameter influence was observed and two others where diameter dependence was found. In principle, sediment trap samples can provide data on sedimentation rates, fluxes of fecal pellets and other transient particulates, and sinking rates of settling material. However, if these measurements are to have quantitative significance, they must be characteristic of properties of the aquatic system, and independent of trap design parameters.

Three sets of experimental traps, which have been deployed for portions of two field years on Lake Michigan, have provided useful data on the effect of design factors on the collection efficiency of traps in this environment. The data also suggest effects of seasonal limnological factors. The test station was located in 67 meters of water (slightly less than the mean depth of Lake Michigan) 12 km SW of Grand Haven, Michigan. All traps are of basically similar design, being right circular cylinders with a collection funnel and bottle assembly at the bottom, which prevents resuspension of settled material and also yields a compact and easily stored sample. Set I had a constant height-to-diameter ratio of 7:1, with diameters of 4.7, 9.6, 15, and 20 cm. Set II consisted of 14.5 cm diameter traps having height-to-diameter ratios in the range of 1 to 7. Set III consisted of duplicate traps with diameters of 4.2, 9.6, and 15 cm, with one set of duplicates hung separately and the other set nested in a concentric arrangement to test the "snow fence" effect proposed by Kleerekoper.[2] For additional data on this effect, one of each pair of standard (H:D=3:1) or test traps was provided with a set of baffles (right cylindrical tubes 15 cm deep) arranged centrically in, and level with, the top of the sediment trap at various times and depths throughout the season.

Before discussing the results, it should be noted that most or all of the time, the rate of horizontal flow of water over the trap far exceeds the settling rate of particulate matter, and also that the composition of the settling particulate material is a continually varying mixture of resuspended mineral sediments, diatom frustules, fecal pellets, calcite, and various detrital aggregates. Despite the complexity of the system under study, relatively consistent trends are noted in all three test sets. The results are summarized briefly as follows.

Set I. Trap Diameter Dependence

During the period of lake thermal stratification, the collection per unit area in the 4.7, 9.6, and 15 cm diameter traps was the same within a few percent, but the 20-cm diameter trap collected from 40 to 180% more than the smaller traps. The collection efficiency was reported by Kirchner[1] to be independent of trap diameter in Crawford Lake (Ontario) for a trap set ranging from 3 to 40 cm in diameter and with a constant height of 25 cm, but the sampling was done under conditions of nearly complete absence of turbulence. The increased collection efficiency of the 20 cm trap observed in the present study is thought to represent trap response to the eddy size distribution in the water column. Further study of this interesting hydrodynamic phenomenon is difficult because of the expense of construction and the difficulty of shipboard handling of even larger traps.

Set II. Trap Height:Diameter Dependence

The results from these tests appear to fall into two similar but somewhat different categories illustrated in Figure 1. In all cases, the least dependence of trap collection efficiency on the height-to-diameter ratio is observed for the ratio of 5:1. During the stable summer stratification period (June through August), the collection efficiency is essentially constant over the range of height-to-diameter ratios from 3 to 7. A modified response in which collection

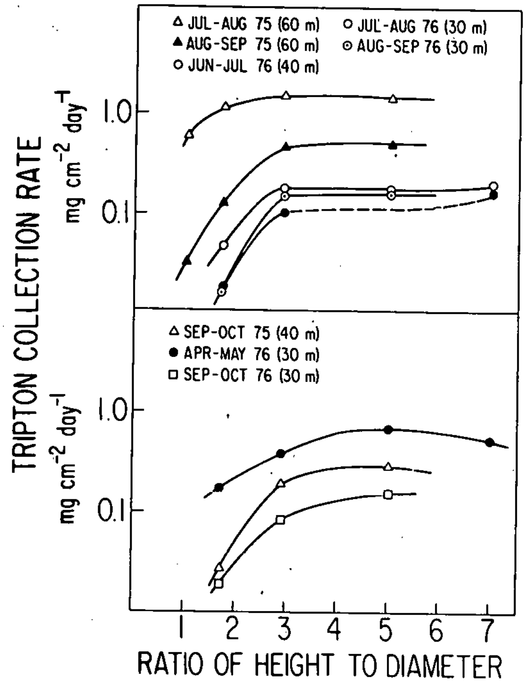

FIG. 1.--Tripton collection rate as a function of height-to-diameter ratio of sediment traps. (ANL neg. 149-77-300)

efficiencies increase more or less continuously with increasing height-to-diameter ratio appears to occur during the spring convective mixing period (March through May) and during breakup of the thermocline (September through November).

Set III. Concentric versus Single Traps

During each of the four test periods, all three annuli of the concentric trap collected more efficiently than any of the corresponding single traps, but the pattern changed throughout the season. Traps with baffles also collected more efficiently than open traps but also showed seasonal changes.

Based on these studies, the recommended design for sediment traps for Lake Michigan has a height:diameter ratio of 5:1 and a diameter in the range between 5 and 15 cm. Although baffles increase the collection of material, the response of baffled traps to the varied current and particulate characteristics of the lake is so complex that the use of baffles cannot be recommended except perhaps as a method of studying physical properties. Preliminary estimates of the fluxes of diatom frustules and of calcite, derived from observed changes in the concentrations of silica and of calcium in surface waters, give good agreement with the observed trap collections. Independent settling rates[3] estimates fall in the same range as those calculated from trap and filter data. These results confirm the usefulness of sediment traps in the study of cycling and transport of trace material in Lake Michigan.

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04502

LAKE MICHIGAN SEDIMENT TRAP STUDY: PRELIMINARY ASSESSMENT OF RESULTS

M. A. Wahlgren and D. M. Nelson

During 1975, a sediment trap and current meter array was maintained at ANL station 75-5. Samples collected at monthly intervals from these traps provided data at two depths which indicated considerable resuspension of surface sediment flocs, but only under the physical conditions of a relatively near-shore, high-energy environment.[1] In order to establish whether resuspension is a general, lakewide phenomenon, even in lower energy environments, and whether its rate is variable in different regions of the lake, arrays of sediment traps were installed during 1976 at 5 stations across the southern basin of Lake Michigan. The locations and characteristics of these stations are summarized in Table 1. At four of the stations a current meter was located 1.5 m off bottom. K. D. Saunders[2] presents a preliminary assessment of the ANL current meter data. Three of the stations (EPA-19, 18, and 16B) were located on the transect utilized during 1976 by NOAA for a comprehensive study of Lake Michigan surface currents.

Tripton-depth profiles which are typical of those obtained in this study are illustrated in Figure 1 for the May-June 1976 interval. Each profile apparently consists of a settling and resuspended particulate component. At each station

Table 1. Description of 1976 trans-basin sediment trap stations

Station	Location	Water dept. m	Sedimentary regime	Trap depth, m	Current meter,[a] m
RER 5	43°00'20" 86°22'12"	67	silt-clay	10, 20, 30, 40 53, 61, 65	1.5
EPA 19	42°44'00" 86°36.0'	93	silt-clay	30, 50, 85, 89, 90.5	1.5
EPA 18	42°44'00" 87°00'00"	159	post-glacial clay	30, 50, 80, 110, 151, 155, 156.5	1.5
EPA 16B	42°44'25" 87°31'40"	76	near silt-sand-clay junction	30, 50, 68, 72, 73.5	1.5
EPA 10	42°23'00" 87°25'00"	98	glacial clay	30, 50, 90, 94, 95.5	1.5

[a] Off bottom.

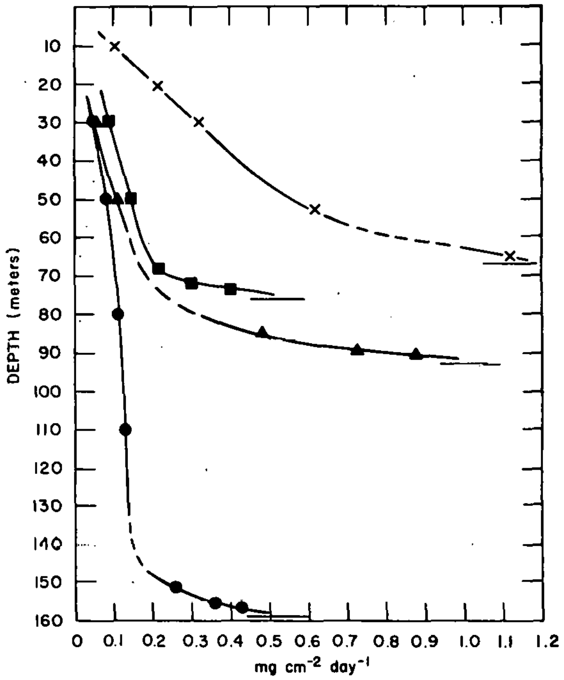

FIG. 1.--Tripton-depth profiles for the May-June 1976 collection period. Station 5, x; Station 19, ▲; Station 18, ●; Station 16, ■ . (ANL neg. 149-77-155)

the flux near bottom exceeds the local mass sedimentation rate[3] (based on radiometric dated cores) by a factor of the order of 50. From concurrent seston measurements, the apparent settling rates are estimated to range from 1 to 5 m day^{-1}. The data in Figure 1 suggest that very significant amounts of the floc layer are resuspended to at least 10 m off bottom at all 4 stations. It also appears that there is considerably greater resuspension at the 2 stations off the eastern shore, although the resuspension height is about the same. The slopes of the profiles > 10 m off bottom must reflect decomposition—aggregation processes in the water column, since for station 18 the profile is almost vertical between 70 and 140 meters depth.

It is also observed that the resuspension component of the tripton-depth profile at each station is surprisingly constant throughout the season. The data for station 5 are presented in Figure 2. Although the tripton collected in the mid-depth traps varies by a factor of 10, that within 10 meters of the bottom varies by less than a factor of 2 from April through October. Current meter records from this station during 1975 showed that from June through August, for periods of a week or longer, currents at one meter off bottom were below the threshold for meter response (2 cm sec^{-1}); even during the intermittent current

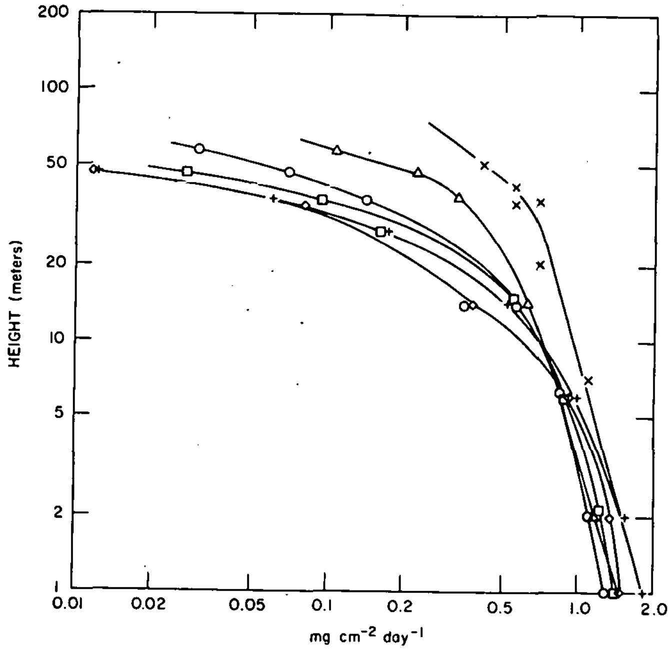

FIG. 2.--Tripton-depth profiles for station 5 for all collection intervals, 1976. 5 April–18 May, x; 18 May–4 June, Δ; 4 June–22 July, +; 22 July–16 August, □ ; 16 August–11 September, O; 11 September–3 October, ◇ . (ANL neg. 149-77-154)

episodes, the current rarely exceeded 5 cm sec^{-1}. During September and October, the currents were nearly continuous and ranged up to 10 cm sec^{-1}. These data suggest that physical erosion processes are not the primary factor in causing the observed resuspension, but rather that it results from a combination of biological and chemical processes occurring at the sediment-water interface, and wind and pressure-induced bottom oscillations.[4]

For plutonium, the results to date demonstrate that there is an essentially constant distribution coefficient (K_d) for the partition between near-bottom seston, tripton, or floc, and the hypolimnetic water for samples from all three stations. The benthic particulates at the nearshore station 5 show a somewhat lower K_d than do those from the offshore stations which may result from the higher proportion of quartz in sediments of this region[1] as well as from dilution by tributary inputs of low-activity suspended solids along the eastern shore of the lake.

Samples have been preserved for other biological and chemical analyses. However, the interpretation of such data will depend on the availability of results from the concurrent EPA and NOAA studies.

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04503

NEAR-BOTTOM CURRENTS MEASURED DURING THE 1976 MID-LAKE DYNAMICS EXPERIMENT

K. D. Saunders

This study was prompted by the need to obtain information on the resuspension and transport of sediment in the deep lake and on the residence time of conservative properties in the water of the Southern Basin of Lake Michigan. Current meters were deployed one meter off the bottom at the four locations shown in Table 1. These measurements were taken to determine the kinetic energy available for sediment transport and resuspension, the strengths and directions of the mean currents for sediment redistribution, and, in conjunction with concurrent current observations taken by the Great Lakes Environmental Research Laboratory, NOAA, the estimated residence time of water in the Southern Basin and the exchange of water across the mid-lake sill.

Mechanical and electronic problems prevented obtaining any data from station 10; and from the other stations, about 25% of the data are an absolute loss, about 25% of the data can be recovered by further processing, and about 50% of the data are acceptable. The current meters used at stations 16B, 18, and 19 were Braincon, model 381, Savonius-rotor, mechanical, histogram-recording current meters.

Because of the paucity of good data at stations 16B and 18, most of the discussion will deal with results from station 19. The early data (May) from station 19 show several events with strong currents, separated by longer intervals of very weak current. During the events of strong currents, the magnitude

Table 1. Station Data, Mid-lake Dynamics Experiment, 1976

Station No.	Latitude (North)	Longitude (West)	Depth, m
10	42°23'00"	87°25'00"	97
16B	42°44'25"	87°31'40"	79
18	42°44'00"	87°00'00"	161
19	42°44'00"	86°35'00"	92

of the currents is of the order $(0.05 - 0.1 \text{ m s}^{-1})$. No significant inertial frequency activity was seen during May and early June. Net transport, according to progressive vector diagrams, was toward the northwest.

From June through October, inertial frequency activity was generally present. This was apparent both from the spectra and from the directional plots of the currents. The energetic episodes were weaker, shorter, and separated by longer periods of quiescence than in the late spring. With the exception of the periods of days 154–174 and 201–221, the general movement of the water of this location was toward the west or northwest. In late summer through early autumn, the energetic events tended to merge, with very few periods of quiescence separating them.

Average directions, speeds, fluctuating component of the total kinetic energy densities, and the total kinetic energy densities for a number of averaging periods are given in Table 2. Under stratified conditions, the fluctuating kinetic energy at station 19 constitutes about 90% to 98% of the total kinetic energy, while under the more well-mixed spring conditions, the percentage is between 73% and 80% of the total.

At present, the period covering days 221–239 is the only period during which there are good data from all the mid-lake stations. The total kinetic energy density averaged over this period is greatest at station 16B and least at station 19. If this geographic variation of kinetic energy density is typical

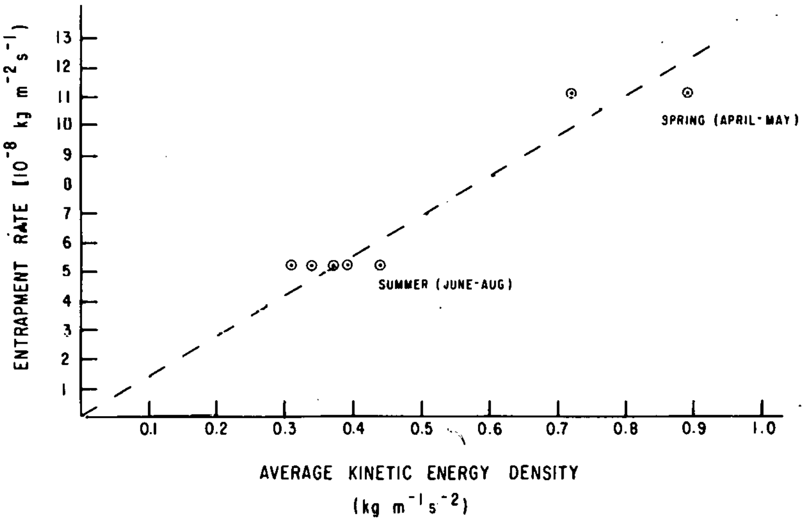

FIG. 1.--Average sediment entrapment rates (4 m above bottom) vs. average kinetic energy densities at Station 19 during the Mid-lake Dynamics experiment, 1976. The sediment entrapment rates were supplied by M. A. Wahlgren.[1]

Table 2. Summary of current meter data, Mid-lake Dynamics experiment, 1976

Station	Period (Julian days)	02–112	112–132	154–174	174–193	193–200	201–221	221–239	239–259	259–277
16B	$\bar{s}$[a]							0.0126	0.0081	0.0014
	$\bar{\theta}$[b]							-1.2°	7.7°	19.7°
	$\frac{1}{2}\rho\sigma^2$[c]							0.402	2.862	0.142
	K.E.[d]							0.480	2.895	0.221
18	$\bar{s}$			0.025	0.040		0.0164	0.0204	0.0125[e]	
	$\bar{\theta}$			18.6°	3.0°		87.3°	147.6°	-31.8°[e]	
	$\frac{1}{2}\rho\sigma^2$			2.283	4.825		0.338	0.191	0.316[e]	
	K.E.			2.597	5.642		0.472	0.400	0.394[e]	
19	$\bar{s}$	0.0171	0.0211	0.0090	0.0034	0.0025	0.0038	0.0036	0.0110	0.0058
	$\bar{\theta}$	41.9°	-12.3°	52.8°	-112.0°	-15.8°	-0.5°	-88.7°	-84.8°	-71.9°
	$\frac{1}{2}\rho\sigma^2$	0.577	0.652	0.393	0.384	0.305	0.361	0.334	0.753	0.946
	K.E.	0.723	0.887	0.433	0.390	0.309	0.369	0.340	0.814	0.963

[a] Average speed (m s^{-1}).

[b] Average direction in degrees clockwise from north.

[c] Average fluctuating kinetic energy density (kg m^{-1} s^{-2}).

[d] Total average kinetic energy density (kg m^{-1} s^{-2}).

[e] Averaging period 241–248.

of each station over the year, it is consistent with the sedimentation occurring on the eastern side of the lake, as there appears to be more energy available to keep material resuspended in the western and central regions of the lake.

Sediment entrapment rates determined from traps placed on the same mooring lines as the current meters are plotted versus average kinetic energy density in Figure 1 for station 19 during periods when both sediment entrapment rate and current data were available. There is an indication that the sediment entrapment rates may be directly proportional to the available kinetic energy of the currents, which would be suspected in situations involving resuspension of sediment by the currents. The average direction of water motion at one meter at the bottom indicates that resuspended sediment will be transported toward the west.

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Distribution of ANL-76-88 Part III

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