

Seasonal and spatial dynamics of $^{234}\text{Th}/^{238}\text{U}$ disequilibria in southern Lake Michigan

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[1] Measurements of $^{234}\text{Th}/^{238}\text{U}$ disequilibria were made on an approximately bimonthly basis over the course of a year in nearshore (10–40 m deep) and offshore (160 m deep) surface waters of the southern basin of Lake Michigan (18,100 km²). The mean activity of ^{238}U in Lake Michigan measured 230 ± 20 dpm m⁻³, approximately 1 order of magnitude lower than what is typically found in marine systems.

Measured median activities of dissolved (<0.45 μm) and excess particle-bound ^{234}Th were 8.0 and 60.6 dpm m⁻³, respectively. Using a simple one-dimensional model, median residence times for dissolved and particle-bound ^{234}Th were ~1 and ~14 days, respectively. ^{234}Th -based particle settling velocities had a median value of 0.4 m d⁻¹ (range: ~0.0–1.4 m d⁻¹), and instantaneous ^{234}Th -based estimates of the net vertical mass flux had a median value of 0.4 g m⁻² d⁻¹ (range: ~0.0–5.2 g m⁻² d⁻¹). Average particle settling velocities were generally constant over time and increased only slightly in the shallowest (10 m) sampling stations. Calculated mass fluxes showed a strong correlation with temporal and spatial changes in the concentration of total suspended matter, which, in the nearshore area, closely followed seasonal variations in wind-induced wave height. Using a simple two-box model, the ^{234}Th -based cross-margin mass export rate for the entire southern basin of Lake Michigan was equal to 1.35×10^9 kg yr⁻¹, which is in excellent agreement with a $^{210}\text{Pb} - ^{137}\text{Cs}$ based, basin-wide mass sedimentation rate of 1.28×10^9 kg yr⁻¹.

INDEX TERMS: 4808 Oceanography: Biological and Chemical: Chemical tracers; 3022 Marine Geology and Geophysics: Marine sediments—processes and transport; 0932 Exploration Geophysics: Radioactivity methods; 1806 Hydrology: Chemistry of fresh water; **KEYWORDS:** ^{234}Th , Lake Michigan, particle transport

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1. Introduction

[2] Satellite imagery and long-term turbidity records show that very large sediment resuspension events frequently occur during early spring in the nearshore zone of southern Lake Michigan [Eadie *et al.*, 1996]. The role of these events in cross-margin transport of biogeochemically important materials from terrestrial sources to lacustrine sinks is unclear. While long-term patterns of sediment deposition and focusing have been carefully detailed using ^{210}Pb and ^{137}Cs [Robbins and Edgington, 1975], the resolution of these radionuclide tracers is on the decade scale. The dynamics of resuspension and redeposition over the nearshore lakebed, however, vary on the timescales of hours to days, consistent

with observed storm front frequencies and meteorological forcing events. These nearshore zones exhibit no net permanent accumulation of sediment, but rather serve as temporary repositories for new inputs of suspended material deriving from coastal erosion processes, riverine inputs, and autochthonous production stimulated by associated nutrient introductions. Such processes are not unique to the Great Lakes, but are commonly observed phenomena in nearly all coastal systems. Understanding the temporal dynamics of these systems requires methods and tracers capable of tracking particles and their associated constituents on the day-to-month timescale.

[3] The $^{234}\text{Th}/^{238}\text{U}$ daughter-parent nuclide pair represents an ideal tracer for this application. Naturally occurring, particle reactive ^{234}Th (half-life, $t_{1/2} = 24.1$ days) and its generally soluble parent, ^{238}U ($t_{1/2} = 4.47 \times 10^9$ years), have been measured in marine systems for over 3 decades.

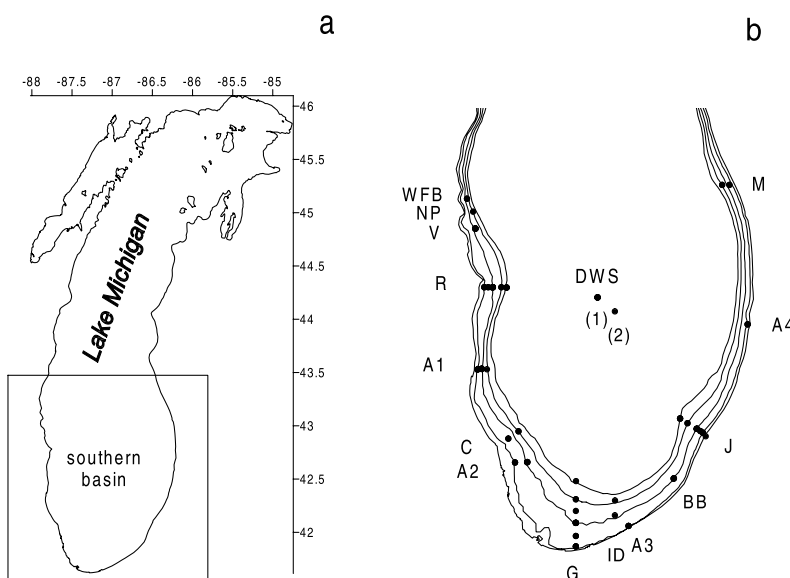


Figure 1. (a) Lake Michigan and the southern basin study site. (b) Sampling stations in southern Lake Michigan. Solid lines show the Lake Michigan shoreline and 10-m isopleths to a depth of 40 m. Geographic coordinates and sampling dates for each station are given in Table 1.

Measured disequilibrium between the ^{234}Th daughter and its parent ^{238}U and the distribution of ^{234}Th between particles and the dissolved phase provides an ideal tracer for the study of both short-term particle fluxes and particle reactive scavenging rates. With only a few exceptions [see, e.g., Vogler *et al.*, 1996; Morris *et al.*, 1994], however, $^{234}\text{Th}/^{238}\text{U}$ ratios have not been measured in freshwater systems because of limitations in the established gamma and beta counting methods used to measure the activity of ^{234}Th . Accurate measurements of ^{234}Th in freshwater systems are difficult to make because concentrations of the isotope are generally an order of magnitude or more lower than those found in marine systems. More importantly, activity ratios of ^{234}Th to other thorium isotopes are up to 10^4 times lower in fresh water than they are in marine systems [Broecker and Peng, 1982] and difficulties arise in separating the beta counts generated by these other thorium isotopes (particularly ^{228}Th) from those generated by ^{234}Th and its immediate daughter $^{234\text{m}}\text{Pa}$. However, by empirically correcting for the interferences from real and apparent betas that are emitted by other thorium isotopes and their progeny [Waples *et al.*, 2003], accurate measurements of ^{234}Th activity in many freshwater systems are possible.

[4] In this first large-scale survey of $^{234}\text{Th}/^{238}\text{U}$ disequilibrium in a freshwater environment, measurements of $^{234}\text{Th}/^{238}\text{U}$ disequilibrium were made in the southern basin of Lake Michigan in a series of experimental studies carried out over a 3-year period from 1998 to 2000. All of the studies were undertaken as part of the Coastal Ocean Processes (CoOP) Episodic Events Great Lakes Experiment (EEGLE). In this paper, we focus primarily on the study of surface water $^{234}\text{Th}/^{238}\text{U}$ disequilibrium measured on six separate cruises in 1999. Estimates of the residence time of dissolved and particle-bound ^{234}Th are calculated using a simple one-dimensional model, extrapolated to the calculation of settling velocities, and mass fluxes, and examined in the context of both time and space. Finally, a ^{234}Th -based

cross-margin mass export rate for the entire southern basin of Lake Michigan is discussed in the light of independent sediment budgets for southern Lake Michigan based on the allochthonous mass import estimates of Colman and Foster [1994] and the ^{210}Pb - ^{137}Cs geochronology of Edgington and Robbins [1976].

2. Methods

2.1. Study Area

[5] At 57,750 km², Lake Michigan is the sixth largest lake in the world [Beeton, 1984]. The lake is divided into two major basins (north and south) with the southern basin occupying approximately one third of the total lake area (18,100 km²) [Edgington and Robbins, 1976] (Figure 1). Morphologically, the southern basin of the lake may be characterized as a relatively simple, conical depression with gradual, sloped bathymetry leading to a maximum depth in the center of the basin of ~170 m. Particle exchange between the southern basin and the remainder of the lake is negligible, with a net mass export rate of less than 1% of local sedimentation [Colman and Foster, 1994]. Sediment accumulation, however, is highly skewed, with the most rapid rates of permanent deposition occurring, not in the deepest portion of the lake, but on the southeastern flank, presumably in response to general circulation patterns that transport material from the western shore to the eastern side of the basin in a counterclockwise direction [Edgington and Robbins, 1990].

[6] Because the focus of this study was to better understand the role of cross-margin transport processes and the behavior of suspended particulate matter in the nearshore and nondepositional area of southern Lake Michigan, surface water samples were primarily collected in the coastal margin in water depths ranging from 10 to 40 m (Figure 1b). Surface water samples were also collected, however, at ~160-m, deepwater stations (DWS) in the center of the

southern basin of Lake Michigan, which is located approximately 50 km from the nearest shore.

2.2. Sampling Procedure and Thorium Analysis

[7] A complete and in-depth discussion of the procedure we have developed to measure $^{234}\text{Th}/^{238}\text{U}$ disequilibria in Lake Michigan is presented by *Waples et al.* [2003]. A brief summary of this procedure follows.

[8] Water samples of up to ~ 200 L were collected using a submersible pump and stored briefly in 50-L plastic containers until separation of the phases by filtration (specific sampling depths, sampling locations and sampling dates are presented in Table 1). The particle-bound thorium fraction on suspended sediment was separated from the water sample by filtration through preweighed nitrocellulose filters ($0.45\ \mu\text{m}$, 293 mm, Millipore), usually within 2 hours of collection. The time of filtration was noted and the filter was stored at 4°C until it could be dried and weighed to determine the mass of total suspended solids. One hundred liters of the filtrate were placed in a 200-L plastic conical container for the recovery of the dissolved thorium fraction (operationally defined by the filter pore size). After adding a requisite aliquot portion of the isotopic diluent, ^{229}Th , the thorium was co-precipitated onto newly formed iron hydroxide (created by addition of 1 mg ferrous iron per liter of sample water) and separated by filtration.

[9] Thorium was isolated from uranium and all other elements by separation on an ion-exchange column. The separated thorium isotopes were next transferred to a stainless steel counting planchet by electrodeposition to provide an essentially weightless source. The sample planchet was first counted using a low background (~ 0.7 cpm) gas-flow proportional counter with 2.25-inch diameter detectors and anti-coincidence circuitry (G542 System, Gamma Products, Inc.) for four 500-min periods to obtain total (gross) beta counts. The planchet was then counted using a silicon surface barrier detector to determine the activities of ^{228}Th , ^{229}Th , ^{230}Th , and ^{232}Th by α -spectrometry. Total alpha count times were activity dependent, but typically ranged from 7000 to 14,000 min.

[10] Beta counts originating from the in-growing progeny of ^{228}Th and ^{229}Th and apparent beta contributions from internal conversion electrons were finally subtracted from the total beta count using correction algorithms for the beta count contribution from each α -active thorium isotope. The activities of dissolved ^{238}U in Lake Michigan water and particle-bound ^{238}U (on suspended particles and iron hydroxide precipitate) were measured using the method of *Edgington et al.* [1996].

[11] Where comparisons between single ^{234}Th sample analyses are made, propagated counting statistics for all ^{234}Th activities and ^{234}Th -based rate processes (i.e., residence times, settling velocities, and mass fluxes) are shown [*Knoll*, 1989]. We have found that the average one-sigma ($1s$) error based solely on propagated counting statistics is approximately one half the $1s$ error based on replicate sample analyses because sampling errors and other processing uncertainties are not factored into simple propagated counting statistics [*Waples et al.*, 2003; *Buesseler et al.*, 2001]. For clarity, propagated counting errors are not displayed in graphs showing temporal or spatial trends. Where appropriate, quartile values (25%, median, 75%) for

each data set are shown instead. We frequently chose median over mean values as a better estimate of the average due to the frequent presence of large outliers in each data set.

3. Results and Discussion

3.1. ^{238}U Activity in Lake Michigan Water

[12] Unlike the oceans where the uranium content has been well defined and may be generally deduced from measurements of salinity [*Chen et al.*, 1986], the uranium concentration in freshwater systems is only poorly correlated with salinity and must be independently determined [see *Waples et al.*, 2003]. However, because water in Lake Michigan has a hydraulic residence time of 62 years [*Quinn*, 1992], it can be assumed that many naturally occurring conservative tracers (e.g., ^{238}U) are uniformly mixed in the lake. Repeated measurements of dissolved ^{238}U ($n = 13$) in the nearshore zone of southern Lake Michigan agreed with this assumption and were tightly constrained between 0.20 and 0.26 dpm L^{-1} . For further calculations it was taken that the activity of ^{234}Th in secular equilibrium was equal to the mean activity of its parent ^{238}U at 0.23 ± 0.02 dpm L^{-1} (i.e., 230 ± 20 dpm m^{-3}) ($1s$, $n = 13$). Potential errors arising from the use of a mean ^{238}U activity in calculations of particle residence times and scavenging rates (discussed below) are directly related to the extent of disequilibria between ^{234}Th and ^{238}U (i.e., where 0% disequilibrium is defined as secular equilibrium between ^{234}Th and its parent, ^{238}U). In Lake Michigan, where disequilibrium between ^{234}Th and ^{238}U is generally always large (median: 66%), rate calculations based upon $^{234}\text{Th}/^{238}\text{U}$ disequilibria are relatively insensitive to small variations in the activity of ^{238}U .

[13] Particle-bound ^{238}U averaged 0.86 ± 0.49 dpm g^{-1} ($1s$, $n = 42$) and was used as the average supported activity of ^{234}Th bound to particles. This mean supported activity of ^{234}Th was subtracted from each measured total activity of particle-bound ^{234}Th , and subsequent discussion of particle-bound ^{234}Th , therefore, relates only to the excess ^{234}Th bound to particles. Excess ^{234}Th activity on particles typically exceeded the supported activity of ^{234}Th by 2 orders of magnitude.

3.2. ^{234}Th Activity in Lake Michigan Water: Nearshore Data

[14] In 1999, large volume water samples were collected in the nearshore region of the southern basin of Lake Michigan on six separate cruises during the months of February, March, April, May, August, and October. A complete list of the sample metadata, suspended particle concentrations, measured ^{234}Th concentrations, and ^{234}Th -derived rates are presented in Table 1. Quartile values of the entire nearshore data set are presented in Table 2. All sampling locations are shown in Figure 1.

[15] Nearshore samples were collected at sites with water depths that ranged from 10 m to 40 m. Samples were almost uniformly collected at a depth of 5 m below the water surface because we were most interested in the behavior both of particles in the photic zone and of those particles that could be observed by satellite imagery. Sampling depths labeled as “integrated” were collected from an

Table 1. Nearshore ²³⁴Th Measurements in Southern Lake Michigan^a

Zone ^b	Latitude	Longitude	Collected	Water Depth, ^c m	Sample Depth, ^d m	TSM, ^e mg L ⁻¹	P ²³⁴ Th, ^f dpm m ⁻³	D ²³⁴ Th, ^g dpm m ⁻³	res D, ^h days	res P, ⁱ days	V_{ps} ^j m d ⁻¹	J ^k g m ⁻² d ⁻¹
<i>Cruise G0299^l</i>												
R	42.750	87.725	15 Feb. 1999	15	5	4.3	65.3 ± 4.3	0.0 ± 3.7	0.0 ± 0.6	13.8 ± 2.0	0.4 ± 0.1	1.6 ± 0.2
R	42.750	87.650	15 Feb. 1999	30	5	1.0	53.0 ± 3.6	0.0 ± 3.7	0.0 ± 0.6	10.4 ± 1.4	0.5 ± 0.1	0.5 ± 0.1
C	42.102	87.610	16 Feb. 1999	15	5	1.9	38.4 ± 2.8	0.0 ± 3.9	0.0 ± 0.6	7.0 ± 0.9	0.7 ± 0.1	1.3 ± 0.2
C	42.133	87.552	16 Feb. 1999	30	5	0.4	60.3 ± 4.5	7.2 ± 4.0	1.1 ± 0.6	12.9 ± 1.9	0.4 ± 0.1	0.2 ± 0.0
G	41.685	87.223	16 Feb. 1999	15	5	0.9	49.6 ± 3.0	0.0 ± 3.5	0.0 ± 0.5	9.6 ± 1.2	0.5 ± 0.1	0.4 ± 0.1
G	41.842	87.223	17 Feb. 1999	30	5	0.4	53.9 ± 3.3	0.0 ± 3.9	0.0 ± 0.6	10.6 ± 1.4	0.5 ± 0.1	0.2 ± 0.0
J	42.134	86.511	17 Feb. 1999	15	5	5.8	95.3 ± 6.1	0.0 ± 3.4	0.0 ± 0.5	24.6 ± 4.2	0.2 ± 0.0	1.2 ± 0.2
J	42.168	86.584	18 Feb. 1999	30	5	0.7	54.2 ± 4.1	6.3 ± 3.8	1.0 ± 0.6	11.1 ± 1.6	0.4 ± 0.1	0.3 ± 0.0
M	43.188	86.344	18 Feb. 1999	15	5	1.8	97.7 ± 6.0	10.3 ± 3.5	1.6 ± 0.6	27.9 ± 5.1	0.2 ± 0.0	0.3 ± 0.1
M	43.188	86.386	19 Feb. 1999	30	5	2.0	72.8 ± 4.3	6.8 ± 3.3	1.1 ± 0.5	16.8 ± 2.5	0.3 ± 0.0	0.6 ± 0.1
<i>Cruise G0399</i>												
R	42.750	87.725	10 March 1999	15	5	6.9	124.8 ± 8.6	0.0 ± 4.0	0.0 ± 0.6	41.3 ± 9.1	0.1 ± 0.0	0.8 ± 0.2
R	42.750	87.650	10 March 1999	30	5	0.8	94.3 ± 5.0	6.7 ± 4.3	1.0 ± 0.7	25.4 ± 4.4	0.2 ± 0.0	0.2 ± 0.0
C	42.102	87.610	10 March 1999	15	5	22.4	88.1 ± 4.5	0.0 ± 3.7	0.0 ± 0.6	21.6 ± 3.4	0.2 ± 0.0	5.2 ± 0.8
C	42.133	87.552	11 March 1999	30	5	5.0	56.4 ± 4.0	0.0 ± 4.4	0.0 ± 0.7	11.3 ± 1.6	0.4 ± 0.1	2.2 ± 0.3
G	41.685	87.223	13 March 1999	15	5	8.6	66.3 ± 4.3	0.0 ± 3.3	0.0 ± 0.5	14.1 ± 2.0	0.4 ± 0.1	3.1 ± 0.4
G	41.842	87.223	13 March 1999	30	5	0.5	41.7 ± 3.2	8.6 ± 4.7	1.4 ± 0.7	8.1 ± 1.1	0.6 ± 0.1	0.3 ± 0.0
J	42.134	86.511	11 March 1999	15	5	2.4	75.8 ± 6.7	22.5 ± 7.7	3.8 ± 1.4	20.0 ± 3.8	0.3 ± 0.0	0.6 ± 0.1
J	42.168	86.584	12 March 1999	30	5	0.7	71.1 ± 5.6	8.8 ± 4.9	1.4 ± 0.8	16.5 ± 2.7	0.3 ± 0.0	0.2 ± 0.0
M	43.188	86.344	14 March 1999	15	5	0.5	70.4 ± 3.2	6.2 ± 4.0	1.0 ± 0.6	16.0 ± 2.3	0.3 ± 0.0	0.2 ± 0.0
M	43.188	86.386	14 March 1999	30	5	0.6	65.4 ± 2.9	17.2 ± 5.3	2.8 ± 0.9	15.4 ± 2.3	0.3 ± 0.0	0.2 ± 0.0
<i>Cruise N0499</i>												
V (I)	43.001	87.798	14 April 1999	23	5	0.8		14.4 ± 3.3				
V (II)	43.001	87.798	18 April 1999	23	5	0.9	154 ± 23	22.4 ± 4.3	3.7 ± 0.8	99 ± 58	0.1 ± 0.0	0.0 ± 0.0
NP	43.073	87.812	15 April 1999	30	5	0.8	204 ± 20	27.2 ± 2.2	4.7 ± 0.6	>100	0.0 ± 0.0	0.0 ± 0.0
R	42.750	87.699	18 April 1999	20	5	2.1	209 ± 18	18.4 ± 3.0	3.0 ± 0.6	>100	0.0 ± 0.0	0.0 ± 0.0
A1	42.398	87.787	19 April 1999	10	5	11.1	290 ± 35					
A1	42.400	87.764	19 April 1999	20	5	4.5	136 ± 15	4.4 ± 1.1	0.7 ± 0.2	53 ± 16	0.1 ± 0.0	0.4 ± 0.1
A1	42.399	87.733	19 April 1999	30	5	1.3	197 ± 34					
A2	42.000	87.503	19 April 1999	20	5	1.5	45.1 ± 2.3	7.4 ± 1.2	1.2 ± 0.2	8.8 ± 1.1	0.6 ± 0.1	0.8 ± 0.1
G	41.640	87.223	20 April 1999	10	5	6.4						
G	41.741	87.223	20 April 1999	20	5	2.1	55.5 ± 4.8	11.9 ± 2.2	1.9 ± 0.4	11.9 ± 1.8	0.4 ± 0.1	0.9 ± 0.1
ID	41.837	87.000	21 April 1999	30	5	0.5	80.0 ± 7.1	18.6 ± 1.6	3.1 ± 0.4	21.2 ± 3.9	0.2 ± 0.0	0.1 ± 0.0
<i>Cruise N0599</i>												
WFB (I)	43.129	87.847	26 May 1999	26	5	0.7	49.9 ± 5.9	32.0 ± 5.2	5.6 ± 1.1	11.7 ± 2.2	0.4 ± 0.1	0.3 ± 0.1
WFB (II)	43.129	87.847	3 June 1999	26	5	0.3	78.1 ± 3.2	39.5 ± 7.7	7.2 ± 1.6	24.2 ± 4.8	0.2 ± 0.0	0.1 ± 0.0
R	42.751	87.700	27 May 1999	20	5	0.5	49.6 ± 5.3	36.8 ± 4.9	6.6 ± 1.1	12.0 ± 2.2	0.4 ± 0.1	0.2 ± 0.0
A1	42.400	87.763	28 May 1999	20	5	0.7	64.2 ± 5.4	38.0 ± 4.9	6.9 ± 1.1	17.5 ± 3.3	0.3 ± 0.1	0.2 ± 0.0
A2	42.002	87.502	28 May 1999	20	5	0.5	56.6 ± 7.7	66.7 ± 6.7	14.2 ± 2.3	18.4 ± 4.6	0.3 ± 0.1	0.1 ± 0.0
G	41.741	87.223	29 May 1999	20	5	0.4	63.9 ± 6.9	47.7 ± 6.1	9.1 ± 1.6	18.8 ± 4.0	0.3 ± 0.1	0.1 ± 0.0
BB	41.932	86.662	30 May 1999	20	5	0.6	53.9 ± 5.9	24.9 ± 4.1	4.2 ± 0.8	12.4 ± 2.2	0.4 ± 0.1	0.2 ± 0.0
J	42.143	86.531	31 May 1999	20	5	0.6	57.2 ± 3.2	5.8 ± 3.6	0.9 ± 0.6	11.9 ± 1.6	0.4 ± 0.1	0.3 ± 0.0
<i>Cruise N0899</i>												
V (I)	43.002	87.800	26 Aug. 1999	23	5	1.3	48.6 ± 1.7	44.4 ± 3.1	8.3 ± 1.1	12.3 ± 1.9	0.4 ± 0.1	0.5 ± 0.1
V (II)	43.002	87.800	31 Aug. 1999	23	integrated	0.7	46.5 ± 1.2					
V (II)	43.002	87.800	31 Aug. 1999	23	5	0.7	57.6 ± 1.6	14.9 ± 1.7	2.4 ± 0.3	12.7 ± 1.7	0.4 ± 0.1	0.3 ± 0.0
R	42.749	87.701	26 Aug. 1999	20	5	0.9	76.9 ± 2.1	4.0 ± 1.5	0.6 ± 0.2	17.9 ± 2.5	0.3 ± 0.0	0.3 ± 0.0
A1	42.400	87.763	27 Aug. 1999	20	5	0.7	47.9 ± 1.2	31.4 ± 2.0	5.5 ± 0.7	11.0 ± 1.5	0.5 ± 0.1	0.3 ± 0.0
A1	42.400	87.763	27 Aug. 1999	20	integrated	1.3	60.6 ± 1.4					
A2	42.002	87.502	27 Aug. 1999	20	5	0.7	35.8 ± 1.0	12.9 ± 1.4	2.1 ± 0.3	6.9 ± 0.8	0.7 ± 0.1	0.5 ± 0.1
A2	42.002	87.502	27 Aug. 1999	20	integrated	1.2	45.1 ± 2.0					
G	41.741	87.223	28 Aug. 1999	20	5			19.0 ± 1.5				
G	41.741	87.223	28 Aug. 1999	20	integrated	1.0	54.5 ± 1.4					
ID	41.774	87.000	28 Aug. 1999	20	integrated	0.8	60.0 ± 1.5					
BB	41.931	86.664	28 Aug. 1999	20	5	0.7	62.4 ± 2.9	134 ± 14	49 ± 13	65 ± 48	0.1 ± 0.1	0.1 ± 0.0
BB	41.931	86.664	28 Aug. 1999	20	integrated	0.7	58.1 ± 1.5					
J	42.143	86.531	28 Aug. 1999	20	5	0.8	63.9 ± 2.5	23.5 ± 2.1	4.0 ± 0.5	15.6 ± 2.3	0.3 ± 0.0	0.3 ± 0.0
J	42.143	86.531	28 Aug. 1999	20	integrated	0.7	74.2 ± 2.9					
<i>Cruise L1099</i>												
R	42.750	87.748	16 Oct. 1999	10	5	5.8	53.1 ± 5.1	1.5 ± 2.4	0.2 ± 0.4	13.0 ± 2.2	0.5 ± 0.1	2.8 ± 0.4
R	42.749	87.701	16 Oct. 1999	20	5	2.0	40.5 ± 3.5	3.6 ± 2.2	0.6 ± 0.3	16.4 ± 2.3	0.7 ± 0.1	1.3 ± 0.2
R	42.749	87.620	16 Oct. 1999	40	5	0.9	32.7 ± 2.7	11.6 ± 2.8	1.8 ± 0.5	12.5 ± 1.6	0.8 ± 0.1	0.7 ± 0.1
A1	42.398	87.787	16 Oct. 1999	10	5	0.8	21.1 ± 2.8	9.9 ± 2.3	1.6 ± 0.4	12.6 ± 1.9	1.4 ± 0.2	1.1 ± 0.2
A2	42.000	87.572	16 Oct. 1999	10	5	2.6	30.5 ± 1.1	0.0 ± 1.2	0.0 ± 0.2	14.5 ± 2.5	0.9 ± 0.1	2.4 ± 0.3
G	41.640	87.223	16 Oct. 1999	10	5	7.7	65.5 ± 3.3	0.9 ± 1.5	0.1 ± 0.2	11.6 ± 1.5	0.4 ± 0.0	2.8 ± 0.4
G	41.741	87.223	16 Oct. 1999	20	5	1.5	61.2 ± 6.6	5.3 ± 4.6	0.8 ± 0.7	13.0 ± 2.2	0.4 ± 0.1	0.6 ± 0.1

Table 1. (continued)

Zone ^b	Latitude	Longitude	Collected	Water Depth, ^c m	Sample Depth, ^d m	TSM, ^e mg L ⁻¹	P ²³⁴ Th, ^f dpm m ⁻³	D ²³⁴ Th, ^g dpm m ⁻³	res D, ^h days	res P, ⁱ days	V_p^j m d ⁻¹	J_s^k g m ⁻² d ⁻¹
A3	41.728	86.922	16 Oct. 1999	10	5	9.7	73.7 ± 3.7	0.0 ± 1.8	0.0 ± 0.3	16.4 ± 2.3	0.3 ± 0.0	2.9 ± 0.4
J	42.127	86.496	18 Oct. 1999	10	5	4.0	60.6 ± 2.9	0.8 ± 1.2	0.1 ± 0.2	12.5 ± 1.6	0.4 ± 0.1	1.6 ± 0.2
J	42.143	86.531	18 Oct. 1999	20	5	2.1	60.2 ± 5.3	3.5 ± 3.0	0.5 ± 0.5	12.6 ± 1.9	0.4 ± 0.1	0.8 ± 0.1
J	42.188	86.627	18 Oct. 1999	40	5	0.3	63.2 ± 6.5	15.8 ± 3.4	2.6 ± 0.6	14.5 ± 2.5	0.3 ± 0.1	0.1 ± 0.0
A4	42.591	86.241	18 Oct. 1999	10	5	1.6	57.2 ± 3.0	1.1 ± 1.4	0.2 ± 0.2	11.6 ± 1.5	0.4 ± 0.1	0.7 ± 0.1

^aMeasured ²³⁴Th activities and associated rate calculations for all nearshore samples collected during the 1999 EGGLE cruises in southern Lake Michigan. Blank values indicate either no measurement was made or values could not be calculated. Residence times of >100 indicate very long residence due to near equilibrium between ²³⁴Th and its parent ²³⁸U (see equation (3)). Errors for each value reflect propagated counting errors only (±1s).

^bGeneral area in which the sample was collected as shown in Figure 1b.

^cWater depth is equal to the water column depth at the sampling site.

^dSample depth is equal to the depth at which the sample was collected. “Integrated” sample depths indicate water was collected over the entire water column.

^eTSM is total suspended matter, given in mg L⁻¹ dry weight.

^fActivity of ²³⁴Th bound to particles (>0.45 μm).

^gActivity of ²³⁴Th in the dissolved fraction (<0.45 μm).

^hCalculated residence time (in days) of newly formed ²³⁴Th before uptake onto a particle (see equation (2)).

ⁱCalculated residence time (in days) of particles at the depth of sample collection (see equation (3)).

^jThe ²³⁴Th derived particle settling velocity in meters per day.

^kCalculated mass flux of suspended matter at the depth of sample collection based on the measured ²³⁴Th activities.

^lCruise designator identifies the month and year of the cruise as well as the research vessel employed to collect samples (L is Laurentian; N is Neeska; G is Lake Guardian).

integrated depth where the submersible collection pump was raised and lowered through the water column as the sample was collected.

[16] For all samples collected, total suspended matter (TSM) concentrations ranged from 0.3 mg L⁻¹ to 22.4 mg L⁻¹ with a median concentration of 0.9 mg L⁻¹. Excess particle-bound ²³⁴Th activities (A_{Th}^p) for particles with diameters greater than 0.45 μm ranged from 21.1 ± 2.8 dpm m⁻³ to 290 ± 35 dpm m⁻³ with a median A_{Th}^p of 60.6 dpm m⁻³. Although the highest observed A_{Th}^p of 290 ± 35 dpm m⁻³ slightly exceeded the activity of ²³⁸U in the surrounding water (i.e., 230 ± 20 dpm m⁻³), activities of particle-bound ²³⁴Th in excess of the parent nuclide’s activity can be generated when particles are repeatedly resuspended into new water masses with relatively high activities of dissolved ²³⁴Th. In support of this hypothesis, A_{Th}^p values on the western shore of Lake Michigan were all high during the April 1999 cruise, with a median activity of 200 dpm m⁻³ (n = 6).

[17] Activities of dissolved ²³⁴Th (A_{Th}^d), operationally defined as any ²³⁴Th passing through a filter pore size of 0.45 μm, ranged from ~0 dpm m⁻³ to 134 ± 14 dpm m⁻³, with a median activity of 8.0 dpm m⁻³. Approximately 20% of the A_{Th}^d values were ~0 dpm m⁻³. These zero values tended to occur during the winter months (i.e., February, March, and October) when TSM concentrations were high. Calculated A_{Th}^d for all samples with no measurable A_{Th}^d , based on the A_{Th}^p of the sample and the mean distribution coefficient (K_d , discussed below) for all samples collected

during February, March, and October ($K_d = 10.7 \times 10^6$, n = 20), averaged 2.7 ± 3.1 dpm m⁻³ or approximately 1% of the potential ²³⁴Th activity in secular equilibrium with ²³⁸U, suggesting that the majority of these samples had A_{Th}^d close to 0 dpm m⁻³ at the time of sample collection.

[18] Disequilibrium between ²³⁸U and total ²³⁴Th (i.e., $A_{Th}^T = A_{Th}^p + A_{Th}^d$) ranged from ~0 to 87% with a median value of 66%, which is comparable to the extent of ²³⁴Th/²³⁸U disequilibria in very productive or shallow marine systems [see Moran *et al.*, 2003].

3.3. Nearshore Particle Residence Times and Scavenging Rates

[19] This suite of ²³⁴Th measurements allows the independent calculation of particle residence times and particle scavenging rates. In surface waters, disequilibrium between dissolved ²³⁴Th and its parent, ²³⁸U, is largely due to the absorption of the daughter nuclide onto particles suspended in the water. Disequilibrium between total ²³⁴Th and ²³⁸U, meanwhile, is primarily caused by the removal of ²³⁴Th on particles settling out of the water column. In a homogenous, steady state system, where

$$\frac{\partial A_{Th}^d}{\partial t} = \frac{\partial A_{Th}^p}{\partial t} = 0, \quad (1)$$

residence times of both dissolved and particle-bound ²³⁴Th (which are independent of radioactive decay) can be calculated using a simple one-dimensional model [see,

Table 2. Summary of All Nearshore Data Presented in Table 1^a

Sample	Water Depth, m	Sample Depth, m	TSM, mg L ⁻¹	P ²³⁴ Th, dpm m ⁻³	D ²³⁴ Th, dpm m ⁻³	Missing ²³⁴ Th, %	res D, days	res P, days	V_p , m d ⁻¹	J_s , g m ⁻² d ⁻¹
Minimum	10	5	0.3	21.1	0.0	0	0.0	3.7	0.0	0.0
25% quartile	15	5	0.7	53.0	1.1	57	0.1	11.1	0.3	0.2
Median	20	5	0.9	60.6	8.0	66	1.1	13.9	0.4	0.4
75% quartile	25	5	2.1	74.0	19.8	74	3.6	18.7	0.5	0.9
Maximum	40	integrated	22.4	290	134	87	49	> 100	1.4	5.2
n	66	66	65	63	56	54	54	54	54	54

^aSee Table 1 for column unit descriptors.

e.g., *Coale and Bruland*, 1985, 1987], where the residence time or mean life of dissolved ²³⁴Th (τ_d , days) is equal to

$$\tau_d = \frac{A_{\text{Th}}^d}{(A_U - A_{\text{Th}}^d)\lambda}, \quad (2)$$

and the residence time or mean life of particle-bound ²³⁴Th (τ_p , days) is equal to

$$\tau_p = \frac{A_{\text{Th}}^p}{(A_U - A_{\text{Th}}^p)\lambda}, \quad (3)$$

where A_U is the activity of dissolved ²³⁸U, and λ is the decay constant for ²³⁴Th (i.e., $\lambda = 0.028761 \text{ day}^{-1}$). While it cannot be assumed that steady state conditions are in effect at any particular point or time along the coast of Lake Michigan (a subject that is briefly addressed by *Klump et al.* [2003] and the focus of a forthcoming paper based on daily measurements of ²³⁴Th/²³⁸U disequilibria in nearshore Lake Michigan), average values of ²³⁴Th/²³⁸U disequilibria over time and space (i.e., annual and basin-wide averages) and the averaged process rates derived from equations (2) and (3) do provide a meaningful range of estimates and a means for comparison to one-dimensional ²³⁴Th studies in particle-rich marine systems [e.g., *Aller and Cochran*, 1976; *McKee et al.*, 1984; *Cochran et al.*, 1986].

[20] On the basis of these steady state assumptions, the residence time of dissolved ²³⁴Th in nearshore Lake Michigan surface water ranged from ~ 0 to ~ 49 days with a median value of 1.1 days (Table 2). These values of τ_d are similar to those reported for shallow or particle-rich marine systems. Here τ_d varied from 1.2 to 9.7 days in Mecklenburg Bay [*Kersten et al.*, 1998], < 1 to 10 days in Dabob Bay [*Wei and Murray*, 1992], 0.5 to 7.7 days in the eastern Irish Sea [*Kershaw and Young*, 1988], ~ 8 days in Buzzards Bay [*Cochran et al.*, 1986], 5.7 to 47 days in the California Current [*Coale and Bruland*, 1985], 0.3 to 4.0 days over the Yangtze continental shelf [*McKee et al.*, 1984], and ~ 1.4 days in Long Island Sound [*Aller and Cochran*, 1976].

[21] For particle-bound ²³⁴Th, residence times in Lake Michigan ranged from 3.7 days to greater than 100 days, with a median value of 13.9 days (Table 2). In marine systems, τ_p varied from 1.3 to 20.1 days in Mecklenburg Bay [*Kersten et al.*, 1998], 1 to 11 days in the coastal northwest Atlantic [*Gustafsson et al.*, 1997], 3 to 60 days in Dabob Bay [*Wei and Murray*, 1992], 7 to 50 days in the Eastern Irish Sea [*Kershaw and Young*, 1988], 2.4 to 18 days in the California Current [*Coale and Bruland*, 1985], and 0.5 to 11.0 days over the Yangtze continental shelf [*McKee et al.*, 1984].

[22] The net settling velocity of suspended particles (V_p , m d^{-1}) was simply calculated as the depth at which a water sample was collected (i.e., 5 m) divided by the mean residence time of particle-bound ²³⁴Th (τ_p) in that sample [*Wei and Murray*, 1992]. Settling velocities of suspended particles in nearshore southern Lake Michigan surface waters ranged from ~ 0.0 m d^{-1} to 1.4 m d^{-1} , with a median settling velocity of 0.4 m d^{-1} (Table 2). This compares favorably to previous estimates of mass settling velocities in Lake Michigan based on sediment trap studies. *Evans et al.* [1998] calculated a mean settling velocity of

0.45 m d^{-1} for small (< 4 μm), light (~ 2.5 g cm^{-3}) minerals, and a mean settling velocity of 0.40 m d^{-1} for inorganic aggregates. In an extensive seasonal study, *Shafer* [1988] found that particles between 0.4 and 8.2 μm in diameter make up 50 to 90% of the total suspended mass in Lake Michigan and that this material is composed primarily of a seasonally varying mix of organo-mineral aggregates, amorphous detritus, diatom fragments, allochthonous minerals (quartz, clays, feldspars), and calcite. When *Schwab et al.* [2000] subjected their numerical model of coastal circulation and sediment transport to the meteorological forcing that was observed during March of 1998, the sediment particle size was set to 15 μm with a settling velocity of 0.5 m d^{-1} . The model generally reproduced the timing, magnitude and site of sediment resuspension (as revealed by satellite imagery), offshore sediment transport, and the region of sediment deposition (based upon a long-term sediment accumulation map). Detailed structure of the modeled resuspended sediment plume, however, differed from satellite images of the actual resuspension event.

[23] Instantaneous estimates of the net mass flux ($\text{g m}^{-2} \text{d}^{-1}$) from surface waters were calculated as the product of the concentration of total suspended matter (g m^{-3}) and the net settling velocity of the suspended particles (m d^{-1}). At 5 m depth, the net vertical flux of particles out of surface waters (i.e., through the 5-m plane) in nearshore southern Lake Michigan ranged from ~ 0.0 $\text{g m}^{-2} \text{d}^{-1}$ to 5.2 $\text{g m}^{-2} \text{d}^{-1}$, with the median net particle flux equal to 0.4 $\text{g m}^{-2} \text{d}^{-1}$ (Table 2). The lowest flux estimates were not the result of low TSM concentrations but rather very low settling velocities based upon high A_{Th}^p values.

3.4. ²³⁴Th Activity in Lake Michigan Water: Deepwater Data

[24] As a means of observing baseline particle concentrations and ²³⁴Th/²³⁸U ratios more representative of autochthonous particle production, with (presumably) minimal interference from resuspended sediment, deepwater station (DWS) samples were collected on six separate cruises during the months of August and October of 1998 and February, March, August, and October of 1999. A comparison of the deepwater data in Table 3 and the nearshore data in Table 2 shows a general trend consistent with the reduced abundance of particulate matter in offshore locations. Compared to the nearshore zone, the median TSM concentration at DWS decreased by nearly 50% to 0.5 mg L^{-1} . With less particulate mass in the water, the median A_{Th}^d in offshore Lake Michigan more than doubled to 19.7 dpm m^{-3} , leading to a three-fold increase in the median τ_d to 3.3 days. The median offshore A_{Th}^p at DWS also increased by nearly 45% to 86.5 dpm m^{-3} , which nearly doubled the median τ_p to 22.9 days. A more detailed examination of the relationship between water depth and TSM concentrations, particle settling velocities, and mass fluxes is presented below.

3.5. Temporal Dynamics of ²³⁴Th/²³⁸U Disequilibria and Suspended Particle Behavior

[25] At a latitude of $> 42^\circ\text{N}$, Lake Michigan experiences large seasonal fluctuations in climatic forcing. Average surface water temperatures fluctuate nearly 20°C over an annual cycle and the water column generally stratifies between late May and mid-October. In the southern basin,

Table 3. The ²³⁴Th Measurements at the Deepwater Station (DWS) in Southern Lake Michigan^a

Cruise	Zone	Latitude	Longitude	Collected	Water Depth, m	Sample Depth, m	TSM, mg L ⁻¹	P ²³⁴ Th, dpm m ⁻³	D ²³⁴ Th, dpm m ⁻³	res D, days	res P, days	V _p , m d ⁻¹	J _s , g m ⁻² d ⁻¹
L0898	DWS (1)	42.706	87.099	22 Aug. 1998	160	5	1.0	37.6 ± 1.8	0.0 ± 0.9	0.0 ± 0.1	6.8 ± 0.8	0.7 ± 0.1	0.7 ± 0.1
L0898	DWS (1)	42.706	87.099	22 Aug. 1998	160	25	1.8	91.8 ± 3.8	46 ± 12	8.8 ± 2.5	34.8 ± 9.1	0.7 ± 0.2	1.3 ± 0.3
L1098	DWS (1)	42.706	87.099	23 Oct. 1998	160	20	0.5	87.5 ± 3.9	9.9 ± 3.5	1.6 ± 0.6	22.9 ± 3.7	0.9 ± 0.1	0.4 ± 0.1
G0299	DWS (1)	42.706	87.099	18 Feb. 1999	160	5	0.1	86.5 ± 5.7	25.0 ± 5.8	4.2 ± 1.1	25.4 ± 4.9	0.2 ± 0.0	0.0 ± 0.0
G0399	DWS (1)	42.706	87.099	14 March 1999	160	5	0.4	88.6 ± 6.6	32.1 ± 6.2	5.6 ± 1.2	28.2 ± 6.0	0.2 ± 0.0	0.1 ± 0.0
N0899	DWS (2)	42.646	87.001	30 Aug. 1999	160	5	0.7	57.9 ± 1.5	3.7 ± 1.0	0.6 ± 0.2	11.9 ± 1.5	0.4 ± 0.1	0.3 ± 0.0
L1099	DWS (1)	42.706	87.099	18 Oct. 1999	160	5	0.3	70.1 ± 5.4	19.7 ± 1.7	3.2 ± 0.4	17.4 ± 2.9	0.3 ± 0.0	0.1 ± 0.0

^aSee Table 1 for column unit descriptors.

average surface wind speeds peak during the winter months and gradually decrease by ~50% by summer. Seasonal changes in wind speed affect the mean significant wave height, which varies by nearly 80% between December and June (National Data Buoy Center, buoy 45007). In 1999, median TSM concentrations were highest when the water column was thermally unstratified, reflecting the impact of generally higher wind speeds during winter months and their respective impact on the mean wave height and sediment resuspension in shallow water (Figure 2a). Spatial variability in the suspended sediment load was also highest during the unstratified season (as shown by the range in quartile values), reflecting both the rapid and local nature of wind-driven sediment resuspension.

[26] Temporal variations in A_{Th}^d (Figure 2b, dotted lines) were generally inversely proportional to suspended sediment concentrations with median activities of nearly 0 dpm m⁻³ in February and October of 1999 and generally higher activities of ~30–40 dpm m⁻³ in the summer months. Median values of A_{Th}^p (Figure 2b, solid lines), however, were remarkably consistent at ~50–70 dpm m⁻³, with the exception of measurements made during April. In April 1999, values of A_{Th}^p on the west shore of the southern basin of Lake Michigan were nearly 4 times higher than A_{Th}^p on the south and southeast shore. The existence of such a gradient in excess particle-bound ²³⁴Th activities stands out in this data set, and cannot be completely explained with only surface water measurements. One explanation, as mentioned above, invokes horizontal advection of new water masses over a frequently resuspended particle pool on the western shore. In any case, with the greater part of total ²³⁴Th bound to particles, total ²³⁴Th/²³⁸U disequilibrium was remarkably consistent at ~60–75%, except for the month of April when the median A_{Th}^p increased thereby lowering the extent of disequilibrium to ~40% (Figure 2c). Barring April, approximately monthly changes in median A_{Th}^p were only ~2–12% of ²³⁴Th production (via ²³⁸U decay).

[27] The distribution or partition coefficient of ²³⁴Th (K_d) between dissolved and particle-bound states varied significantly over the seasons. K_d is typically described as

$$K_d = \frac{[Th]_{particle}}{[Th]_{solution}}, \quad (4)$$

where $[Th]_{particle}$ is the activity of particle-bound ²³⁴Th per kilogram of suspended matter and $[Th]_{solution}$ is the activity of dissolved ²³⁴Th per kilogram of water. In southern Lake Michigan, median K_d values for each cruise ranged from ~2 × 10⁶ to ~11 × 10⁶ (Figure 2d), which is consistent with the K_d range of 0.5 to 10 × 10⁶ deemed characteristic of coastal marine sediments [*International Atomic Energy Agency*, 1985; *Kershaw and Young*, 1988]. Values of K_d were higher during the unstratified winter months than they were during the summer months and tended to shift in relation to the concentration of total suspended matter. Reasons for the observed temporal oscillation in K_d can be explained in terms of changes in the number of particles in the water column as well as their size and composition. During winter months, increased resuspension of particles would lead to higher concentrations of small particles

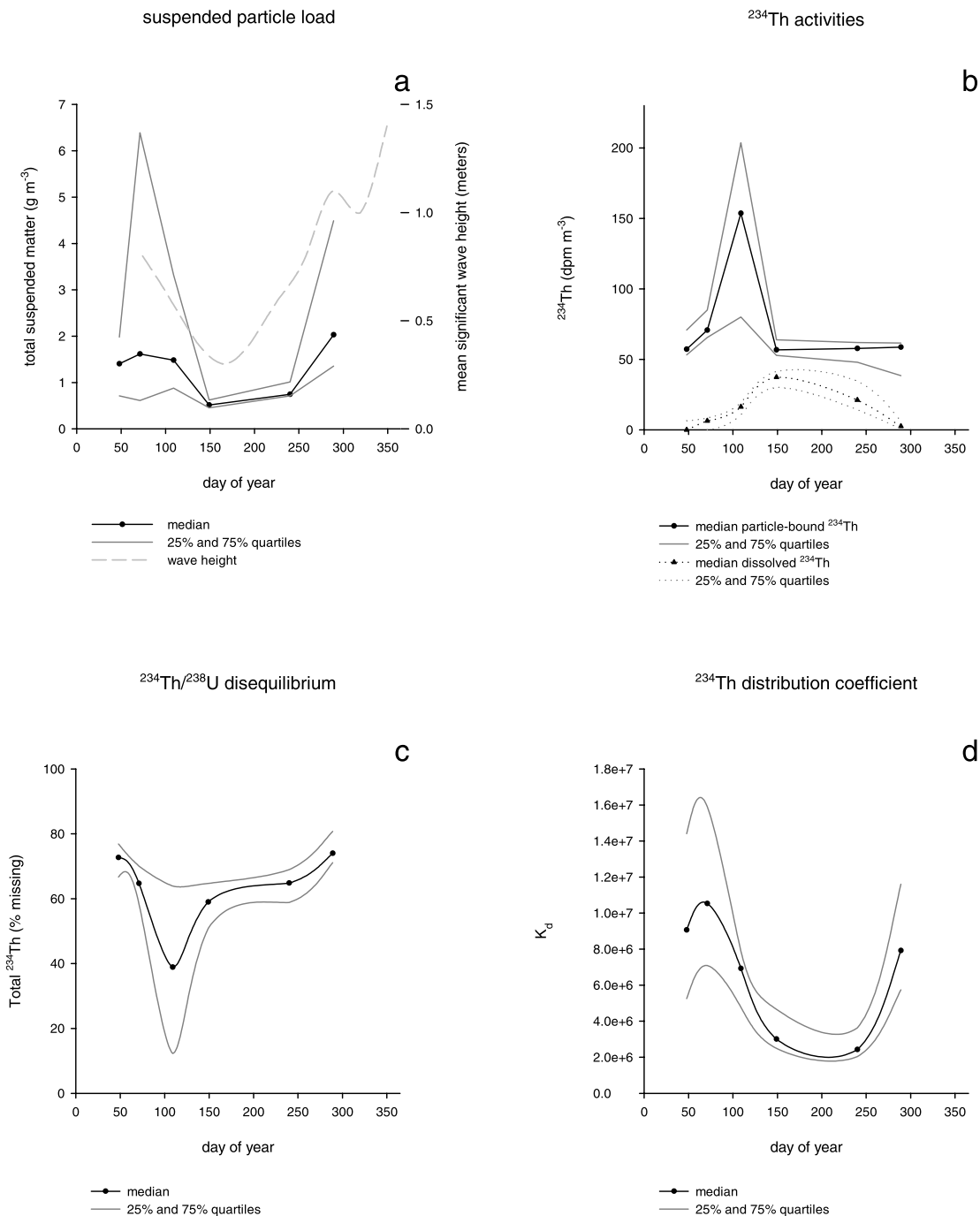


Figure 2. Temporal dynamics of (a) total suspended matter concentrations and wave height, (b) dissolved and particle-bound ^{234}Th activities, (c) total (dissolved + particle-bound) ^{234}Th disequilibria, and (d) distribution coefficients between particle-bound and dissolved fractions of ^{234}Th in nearshore (10–40 m deep) Lake Michigan. Monthly mean significant wave heights for 1999 (dashed line, Figure 2a) were obtained from the National Data Buoy Center (buoy 45007).

(>0.45 μm), effectively increasing the particle-binding surface area available to ^{234}Th . In effect, the amount of ^{234}Th bound to a given mass of particles would increase resulting in a higher K_d . Conversely, seasonal fluctuations in the concentration of organic colloids (<0.45 μm) could also explain the observed variations in K_d with a larger fraction of ^{234}Th bound to higher concentrations of organic colloids

during summer. In one of the few studies of seasonal $^{234}\text{Th}/^{238}\text{U}$ disequilibria, *Wei and Murray* [1992] found ^{234}Th distribution coefficients ranging between 10^6 and 10^8 in Dabob Bay, with K_d values below the euphotic zone generally an order of magnitude higher than in the euphotic zone. Moreover, at 10 m below the surface, ^{234}Th K_d values showed a seasonal oscillation similar to those observed in

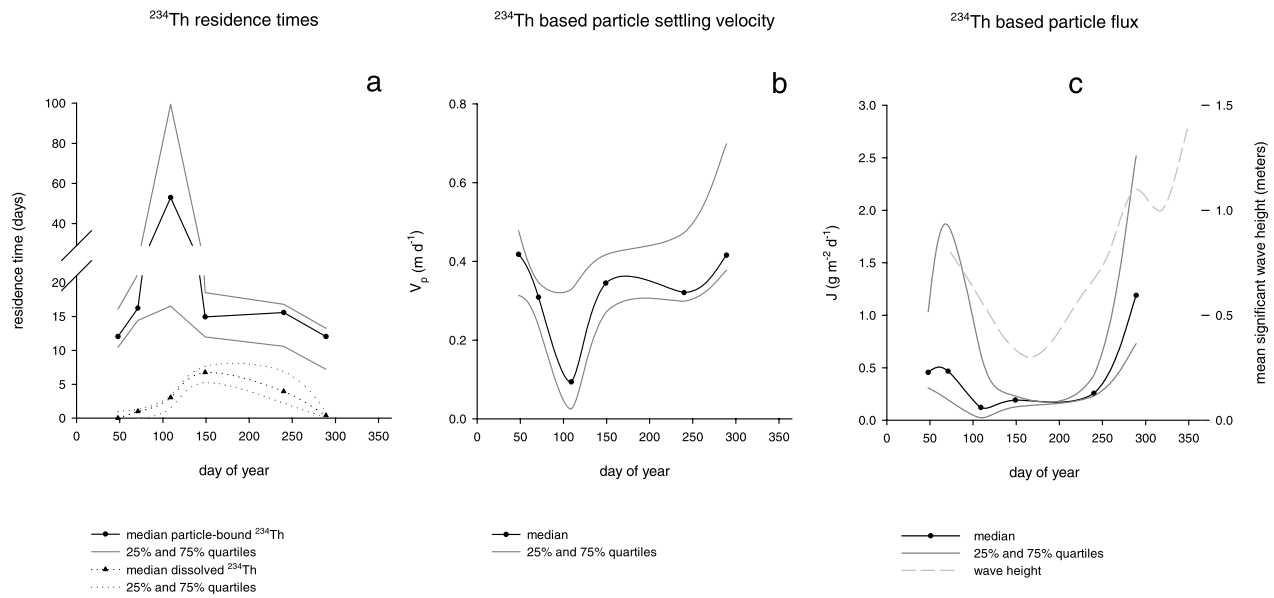


Figure 3. Temporal dynamics of (a) dissolved and particle-bound ^{234}Th residence times, (b) ^{234}Th -based particle settling velocities and (c) ^{234}Th -based mass fluxes and wave height in nearshore Lake Michigan.

Lake Michigan, with higher K_d values tending to occur during the winter months.

[28] Calculated particle process rates based upon measured $^{234}\text{Th}/^{238}\text{U}$ disequilibria, TSM concentrations, and the one-dimensional steady state model described by equations (2) and (3) also showed temporal trends consistent with seasonal changes in physical forcing related to wind-induced changes in wave height (and turbulence) and thermal stratification and its effect on isolating the lakebed from surface related processes, especially in nearshore Lake Michigan (Figure 3). The median τ_d (black dotted line, Figure 3a) was lowest during the winter months (~ 0.0 days in February, 0.4 days in October) and rose to a maximum of 6.8 days in late May in conjunction with the rise and fall of the mean wave height and concentrations of total suspended matter as shown in Figure 2a. The median τ_p ranged from 12.0 to 16.2 days, except during the month of April when very high values of A_{Th}^p along the western shore of southern Lake Michigan drove the calculated median particle residence time up to 53 days. Median particle settling velocities at 5 m below the surface (Figure 3b) were inversely proportional to the particle-bound ^{234}Th residence times, with most values (barring April) ranging between 0.31 m d^{-1} and 0.42 m d^{-1} . It is interesting to note, however, that settling velocities at the 75% quartile closely followed the temporal trend in mean wave height, suggesting that at the upper boundary, wave height and its associated turbulence played some role in the size of particles being resuspended. That is, presumably larger particles with faster settling velocities were more often measured during periods of high wave activity. Mass fluxes at 5 m depth (solid lines, Figure 3c) closely followed the temporal trend in TSM concentrations, with median mass fluxes during the winter months (e.g., $0.46 \text{ g m}^{-2} \text{ d}^{-1}$ in February, $1.19 \text{ g m}^{-2} \text{ d}^{-1}$ in October) at least twice as high as those of the summer (e.g., $0.19 \text{ g m}^{-2} \text{ d}^{-1}$ in late May, $0.26 \text{ g m}^{-2} \text{ d}^{-1}$ in August).

This is especially evident in the temporal trend of the 75% quartile, which, again, closely follows the temporal trend in mean wave height.

3.6. Spatial Dynamics of $^{234}\text{Th}/^{238}\text{U}$ Disequilibria and Suspended Particle Behavior

[29] Large spatial variations in suspended particle concentrations and $^{234}\text{Th}/^{238}\text{U}$ disequilibria were observed in March, April, and October as evidenced by the large spread in quartile values in Figures 2 and 3. However, with the exception of A_{Th}^p measured in April, spatial differences in $^{234}\text{Th}/^{238}\text{U}$ disequilibria and suspended particle concentrations tended to correlate more strongly with water depth rather than geographical location around Lake Michigan's southern basin. Total suspended matter concentrations clearly decreased with increasing water depth (Figure 4a), as would be expected given that new sediment inputs from coastal bluff erosion and riverine discharge would decrease in relative significance as a function of distance from shore as well as the fact that sediment resuspension for any given wind event would also decrease as a function of increasing water depth. The mean surface water particle concentration in 20 m of water was less than 25% of the mean concentration in 10 m of water (where horizontal distances between the 10- and 20-m isopleths ranged from <1 to 10 km). In water depths exceeding 20 m, however, average surface water TSM concentrations were relatively constant at ~ 0.5 to $\sim 1.1 \text{ g m}^{-3}$, depending on the location and season of sample collection (e.g., samples at all 30 m deep stations were collected only in February and March).

[30] Mean ^{234}Th derived particle settling velocities (Figure 4b) remained fairly constant, with increasing water depth suggesting a fairly uniform range of particle size and composition across the nearshore axis, although a slightly higher range of settling velocities in the shallowest waters (10 m) may have indicated the existence of a larger particle

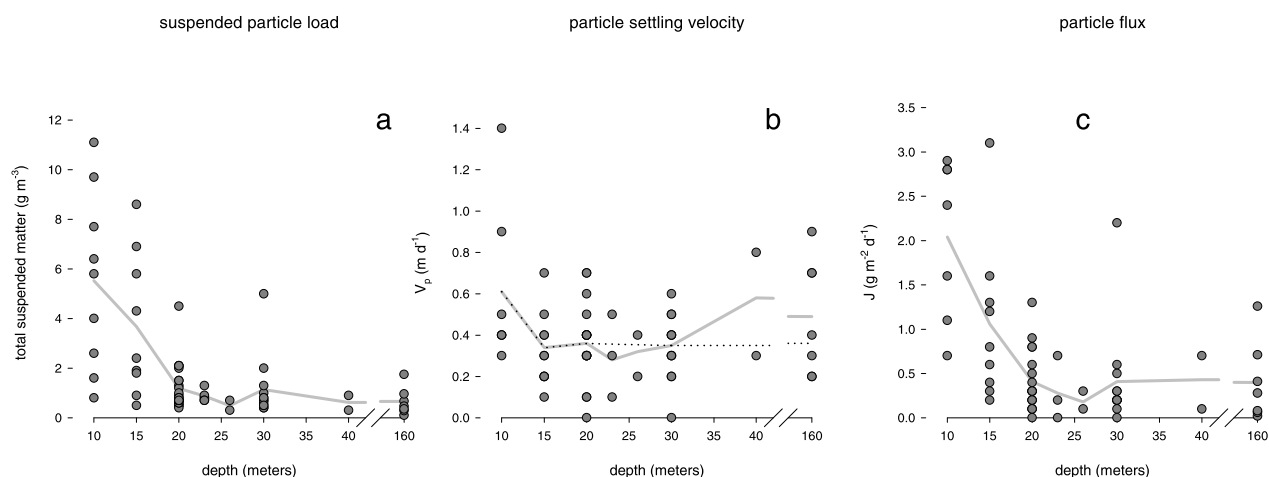


Figure 4. (a) Total suspended matter concentrations, (b) ²³⁴Th-based particle settling velocities, and (c) ²³⁴Th-based particle mass fluxes in relation to water depth in nearshore and offshore Lake Michigan. The solid shaded line in each panel represents the mean at each water depth. The dotted line in Figure 4b represents the mean settling velocity at a 5-m sampling depth at selected water depths of 10, 15, 20, 30, and 160 m. One sample (ID: C15, collected: 10 March 1999) contained 22.4 g m⁻³ TSM and was not included in either the graph or the calculated means.

class (e.g., resuspended sand). This trend is particularly evident if only frequently sampled (i.e., $n \geq 5$) water depths and uniform sampling depths (i.e., 5 m) are considered (Figure 4b, dotted line).

[31] ²³⁴Th derived mass fluxes (Figure 4c) also showed an inverse correlation with water depth. Driven predominantly by the onshore-offshore gradient in TSM concentrations, the mean surface water mass flux in 20 m of water was approximately 20% of the mean flux in 10 m of water. At water depths of 20–160 m, however, mean surface water mass fluxes were relatively constant at $\sim 0.4 \text{ g m}^{-2} \text{ d}^{-1}$.

3.7. Integrated Flux Estimates and Cross-Margin Transport

[32] Lake Michigan has one of the best constrained sediment budgets for any system of comparable scale. Using ²¹⁰Pb and ¹³⁷Cs geochronology, *Edgington and Robbins* [1976] measured sedimentation rates throughout the southern basin of Lake Michigan and determined that (1) 10,600 km² of the 18,100 km² lakebed of southern Lake Michigan is nondepositional with no net accumulation of sediment and (2) the spatially weighted, mean mass sedimentation rate over the 7500 km² depositional area is equal to 17.0 mg cm⁻² yr⁻¹ or $1.28 \times 10^9 \text{ kg yr}^{-1}$ (Table 4). Because the southern basin of Lake Michigan is essentially a closed system with limited exchange of particulate mate-

rial between the northern and southern basins [*Colman and Foster*, 1994], ²³⁴Th/²³⁸U disequilibria may also be used to estimate the mean mass sedimentation rate over the depositional area in the following way.

[33] ²³⁴Th-based mass fluxes in the nearshore zone (i.e., 10–40 m depth) were all measured over nondepositional areas. Because new materials (from eroding bluffs and riverine inputs) are constantly entering the lake via the nearshore zone, inputs to this nondepositional zone, on an approximately annual basis at least, must equal the export and cross-margin transport of these materials to the depositional zone if the nearshore is to remain nondepositional. Taking the temporally integrated average of the median ²³⁴Th-based nearshore mass flux (i.e., the median curve in Figure 3c) to be the “average” mass flux in the nearshore nondepositional zone of southern Lake Michigan requires an export of approximately $1.35 \times 10^9 \text{ kg yr}^{-1}$ from the nearshore nondepositional zone to the permanently accumulating sediment deposits. This focusing translates to a mean ²³⁴Th-based mass flux of 17.9 mg cm⁻² yr⁻¹ over the depositional area, which is within 5% of the ²¹⁰Pb derived mean mass sedimentation rate. A major assumption of this calculation, of course, is that the fine materials, while moving from nearshore to offshore, passed through the 5-m-deep plane from which we sampled.

Table 4. Particle Accumulation Rates in the Southern Basin of Lake Michigan

Zone	²¹⁰ Pb-Based Sedimentation ^a		²³⁴ Th-Based Mass Flux ^b		Source/Sink Mass Balance ^c		Lakebed Area, ^a km ²
	mg cm ⁻² yr ⁻¹	10 ⁹ kg yr ⁻¹	mg cm ⁻² yr ⁻¹	10 ⁹ kg yr ⁻¹	mg cm ⁻² yr ⁻¹	10 ⁹ kg yr ⁻¹	
Nondepositional	0.0		12.7 ^d	1.35			10,600
Depositional	17.0	1.28	17.9 ^e	1.35	24.0 ^f to 40.0 ^g	1.8 to 3.0	7500

^aSource is *Edgington and Robbins* [1976].

^bSource is this study.

^cSource is *Colman and Foster* [1994].

^dTemporally integrated median ²³⁴Th-based particle flux in nearshore Lake Michigan.

^eThe ²³⁴Th-based particle flux assuming offshore transport in nondepositional, nearshore Lake Michigan.

^fMud import budget from rivers, aerosols, and bluffs.

^gSouthern lake basin mud budget based on volume of Holocene lake sediment and average sand/mud ratio.

[34] Temporally integrated averages of the 25 and 75% quartile mass fluxes (i.e., the shaded lines in Figure 3c) translated to cross-margin mass export rates of 0.8 and 3.2×10^9 kg yr⁻¹, respectively. This range, based upon a 24.1-day half-life tracer, agrees remarkably well with independent sediment budgets developed by Colman and Foster [1994] for the southern basin of Lake Michigan based on allochthonous mass inputs from rivers, aerosols, and eroding bluffs (1.80×10^9 kg yr⁻¹) as well as estimates calculated from the distribution and thickness of Holocene lake sediment and an average sand/mud ratio of the surface sediments (3.0×10^9 kg yr⁻¹).

4. Conclusions

[35] The results of this study represent the first major survey of ²³⁴Th/²³⁸U ratios in a freshwater system. Although the dissolved ²³⁸U activity in Lake Michigan is only ~10% of what is normally found in a marine system, sufficient activity of the daughter nuclide ²³⁴Th could be measured in 100–200 L of water using the approach of Waples et al. [2003].

[36] Median residence times for dissolved and particle-bound ²³⁴Th in southern Lake Michigan were of the same order as those reported in the literature for shallow or productive marine systems. More importantly, median particle settling velocities and mass flux estimates based on ²³⁴Th/²³⁸U disequilibria in Lake Michigan were in agreement with previous estimates of these properties based on sediment trap studies [Evans et al., 1998; Shafer, 1988], basin-wide ²¹⁰Pb – ¹³⁷Cs geochronology [Edgington and Robbins, 1976], and allochthonous mass import estimates [Colman and Foster, 1994].

[37] This consistency is remarkable, considering the differences in both the techniques (mass accumulation versus particle removal) and the half-life of the tracers (24.1 days for ²³⁴Th versus 22.3 years for ²¹⁰Pb). Moreover, this also argues that on the basis of current assumptions, measurements of contemporary processes are relevant to historical deposition patterns in Lake Michigan and that the system is near steady state with respect to sediment inputs and burial over the last century (i.e., the time frame of ²¹⁰Pb geochronology).

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