Environ. Sci. Technol. **2001**, *35,* 300−305

Beryllium-7 as a Tracer of Short-Term Sediment Deposition and Resuspension in the Fox River, Wisconsin

SHARON A. FITZGERALD,*,† J. VAL KLUMP,‡ PETER W. SWARZENSKI,§ RICHARD A. MACKENZIE,‡ AND KEVIN D. RICHARDS†

U.S. Geological Survey, 8505 Research Way, Middleton, Wisconsin 53562, Great Lakes Water Institute, Center for Great Lakes Studies, 600 East Greenfield Avenue, Milwaukee, Wisconsin 53204, and U.S. Geological Survey, 600 Fourth Street South, St. Petersburg, Florida 33701

Short-term (∼monthly) sediment deposition and resuspension rates of surficial bed sediments in two PCB-laden impoundments on the Fox River, WI, were determined in the summer and fall of 1998 using ^{7}Be, a naturally occurring radioisotope produced in the atmosphere. Decay-corrected activities and inventories of ^{7}Be were measured in bed sediment and in suspended particles. Beryllium-7 activities generally decreased with depth in the top 5−10 cm of sediments and ranged from undetectable to ∼0.9 pCi cm^{-3}. Inventories of ^{7}Be, calculated from the sum of activities from all depths, ranged from 0.87 to 3.74 pCi cm^{-2}, and the values covaried between sites likely reflecting a common atmospheric input signal. Activities of ^{7}Be did not correlate directly with rainfall. Partitioning the ^{7}Be flux into "new" and "residual" components indicated that net deposition was occurring most of the time during the summer. Net erosion, however, was observed at the upstream site from the final collection in the fall. This erosion event was estimated to have removed 0.10 g (cm of sediment)$^{-2}$, corresponding to ∼0.5 cm of sediment depth, and ∼6−10 kg of polychlorinated biphenyls (PCBs) over the whole deposit. Short-term accumulation rates were up to ∼130 times higher than the long-term rates calculated from ^{137}Cs profiles, suggesting an extremely dynamic sediment transport environment, even within an impounded river system.

Introduction

The Lower Fox River in Wisconsin flows to the northeast from Lake Winnebago to Green Bay, the largest freshwater "estuary" in the world, and from Green Bay into Lake Michigan (Figure 1). This highly industrialized watercourse also contains the world's highest density of paper and pulp mills (*1*). Historical releases of polychlorinated biphenyls (PCBs) from the paper and pulp industry have resulted in the accumulation of about 30 000 kg of these compounds in this portion of the Fox River, largely in association with organic-rich riverine bed sediments, and fish-consumption advisories throughout much of the Green Bay system (*2*). Point sources of PCB inputs to the river have been largely eliminated; continual and episodic resuspension of historically contaminated sediments is now the primary source of PCBs to the water column and Green Bay. This PCB release will maintain fish consumption advisories for a period of decades. The Fox River is also the source of the ∼8500 kg of PCBs estimated to be already present in the bed sediments of Green Bay (*3*) with similar potential persistent effects.

The short- and long-term fate of PCB-laden sediment in the Lower Fox River must be considered within any remediation scenario. Short-term particle and toxic contaminant transport modeling of PCBs in the Lower Fox River has been developed as an outgrowth of the Green Bay Mass Balance Program (*2, 4*). Although several key parameters were measured, this modeling effort relied only on assumptions of deposition and resuspension rates (*4*). The naturally occurring atmospherically derived radioisotope beryllium-7 (^{7}Be) has the potential of refining predictions of these processes by providing actual rates of short-term (month-scale) deposition and resuspension in sediments.

Beryllium-7 is produced by cosmic ray spallation of nitrogen and oxygen in the atmosphere (*5*). The half-life ($t_{1/2}$) of this isotope is 53.3 d (decay constant, $\lambda = 0.01299$ d^{-1}). The average lifetime (τ) of 77 d ($\tau = \lambda^{-1}$) establishes the effective application time period of just under 1 yr with monthly resolution. Beryllium-7 becomes associated with aerosols and is delivered to the terrestrial environment via wet and dry deposition. In aqueous environments, it becomes associated with particles and colloids largely via cation exchange. Measured distribution coefficients (K_d) are on the order of 10^4−10^5, and ^{7}Be strongly sorbs to suspended particles (*6*). Because most hydrophobic contaminants, including PCBs, are also particle reactive (i.e., similar or higher K_d values), ^{7}Be may be used to elucidate types and rates of processes that directly affect the cycling of these contaminants. On the basis of work in the Chesapeake Bay (*6*), there was no significant correlation between ^{7}Be activities on particles and particle composition. Under these conditions, interpretation of the ^{7}Be activities in sedimentary deposits is greatly simplified and is controlled largely by the residence time of the particles in the overlying water. In Galveston Bay, TX, 74−86% of the total ^{7}Be became associated with particles within 1 h of a rainfall event (*7*). Subsequent aggregation and settling of particles resulted in the removal of 70% of total ^{7}Be from the water column in less than 1 d. Because of this rapid scavenging of ^{7}Be onto particle surfaces once introduced into aqueous environments, this radioisotope becomes useful for determining particle resuspension and deposition.

Environmental Setting

The Fox River in Wisconsin is the largest tributary to Green Bay (Figure 1). Two sites on the Lower Fox River were selected at which depth distributions ^{137}Cs and total (Σ)PCB congener concentrations were previously determined (*4*), indicating depositional environments. Site LLDBM (44°11′36″ N, 88°28′19″ W) is in the southwest corner of Little Lake Butte des Morts, a relatively wide length of the river located immediately downstream of Lake Winnebago (Figure 1). The LLBDM site is located within a large depositional area (deposit A) that contains approximately 716 kg of PCB (*4*) and is a site targeted for a PCB removal remediation effort. Site DEPERE (44°26′44″ N, 88°03′50″ W) is located about 25 mi downstream of the LLBDM site and is the last impoundment of the Fox

* Corresponding author present address: U.S. Geological Survey, GSA Center, 651 Federal Dr., Guaynabo, Puerto Rico 00965; e-mail: safitzge@usgs.gov.

† U.S. Geological Survey, Middleton.

‡ Center for Great Lakes Studies.

§ U.S. Geological Survey, St. Petersburg.

10.1021/es000951c CCC: $20.00
Published on Web 12/13/2000

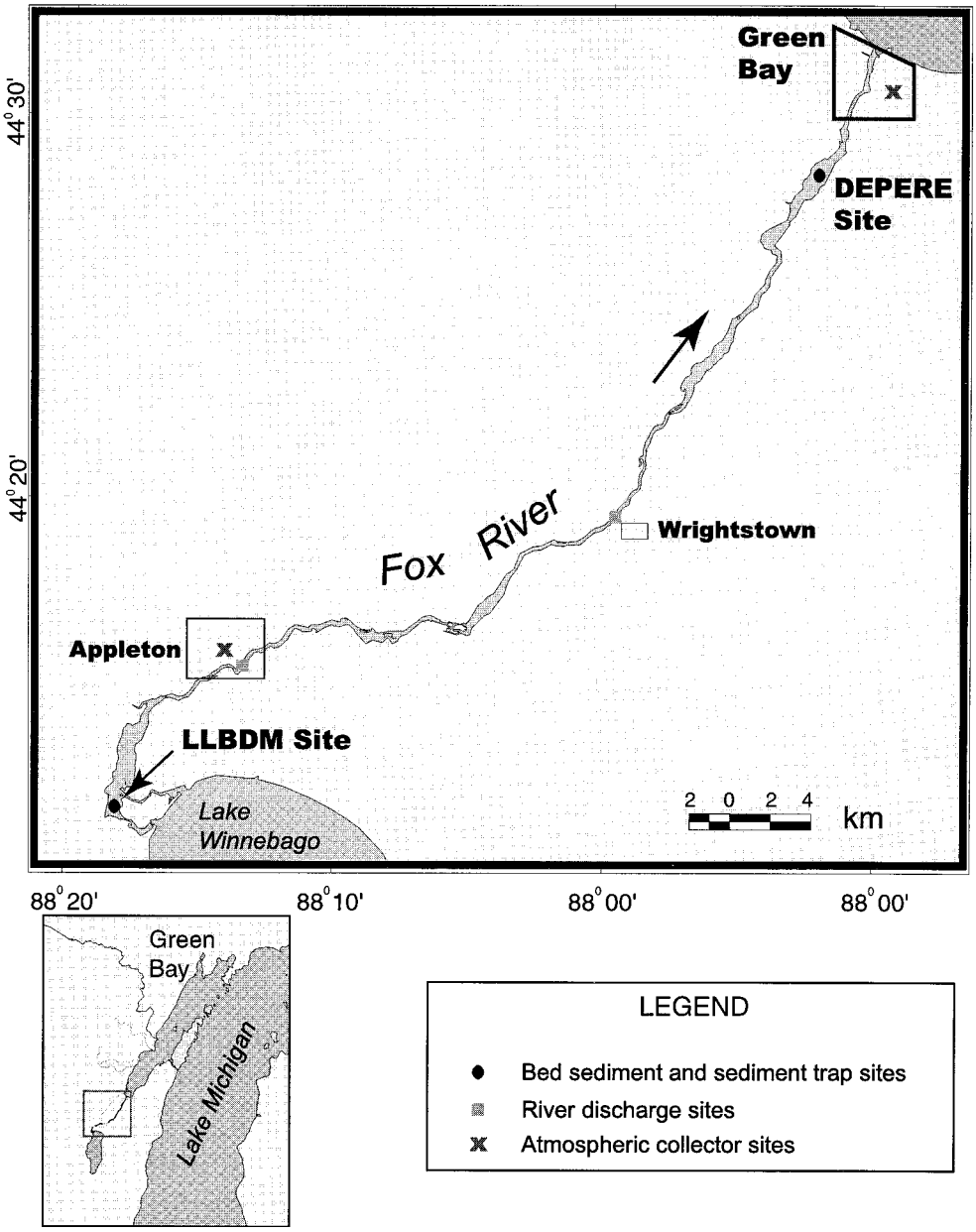

FIGURE 1. Map showing sediment sampling, rainfall collection, and river discharge sites.

River before it enters Green Bay. This site is located approximately 230 m upstream of the DePere Dam and is within a larger deposit (deposit HH) that contains approximately 103 kg of PCB (*4*). Sampling sites were located using GPS, and the horizontal errors were generally less than ~20 m. Sample depths at both sites varied and ranged from approximately 1 to 2 m during all sampling events.

Sediment traps were used at both sites to collect integrated samples of suspended fluvial material for determination of ^{7}Be. The trap at the LLBDM site was located about 1 km downstream of the bed sediment site sampling site. The trap at DEPERE was located about 1 km below the DePere Dam.

Discharge measurements for the entire sampling period (May−September 1998) were obtained at stream gages located near each site (*8*). These stations were located on the Fox River at Appleton, WI (USGS 04084450), ~9 km downstream of the LLBDM site, and on the Fox River at Rapide Croche Dam near Wrightstown, WI (USGS 04084500), ~20 km upstream of the DEPERE site. Continuous rainfall records for the sampling period were obtained at the weather stations at Appleton, WI, and Green Bay, WI (*9*).

Field Sampling and Laboratory Analyses

Bed sediment was sampled from May to September 1998 for the determination of ^{7}Be activities. Sediment cores were collected either with an Ekman box corer (area = 525 cm^2; first two samplings) or with a push corer (10.2 cm i.d.; last two samplings). All cores were sectioned in 1-cm increments. Sediment from three push cores at each site was combined to have adequate sediment for ^{7}Be determinations, generally between 50 and 100 g. The variability of measured porosity, bulk density, and solids density between replicate Ekman cores was previously determined to be less than 10% at sites in the Milwaukee and Manitowoc Rivers, WI (Table 1) (*10*). Bias associated with sampling from the Ekman corer was assessed by comparisons of these three parameters to push core samples collected at the same time. The relative standard deviations for these parameters were somewhat higher, up to 31%, reflecting the somewhat greater difficulty of accurately sectioning an Ekman core sample. All sediment samples were placed in clean, tared 1-L plastic bottles; placed in a cooler; and returned to the laboratory within 48 h for ^{7}Be analysis.

TABLE 1. Relative Standard Deviations (%) Comparing Replicate Ekman Cores and Ekman versus Push Cores for Estimated Porosity, Bulk Density, and Solids Density of Bottom Sediments from the Milwaukee and Manitowoc Rivers, WI[a]

	replicate Ekman cores		Ekman vs push cores	
constituent	Milwaukee River (site a) (%)	Manitowoc River (%)	Milwaukee River (site a) (%)	Milwaukee River (site b) (%)
porosity [cm^3 of water (cm of wet sediment)$^{-3}$]	1.9	1.0	3.5	1.5
bulk density [g (cm of wet sediment)$^{-3}$]	3.4	4.2	31	12
solids density [g (cm of dry sediment)$^{-3}$]	9.5	4.8	12	3.5

[a] S. A. Fitzgerald, unpublished data.

Sediment traps were used to collect suspended particles for measurement of average ^{7}Be activity in suspended fluvial material. Sediment traps consisted of clean, 1-L plastic bottles that were taped to a hollow metal rod (modified from refs *11* and *12*). The aspect ratio of the traps was 4:1, within the range suggested by Gardner (*11*) for effective trapping. This rod was fitted over a solid metal rod of slightly smaller diameter that was advanced about 0.5 m into the river bottom such that it would not move with the current. The hollow rod was held in place by the use of a set screw. Two bottles were taped to each hollow rod, one that sat on the bottom (bottom of the bottle flush with the sediment−water interface) to intercept bed load, and another taped such that the opening was near the middle the total water depth. Sample collection consisted of loosening the set screw, pulling the hollow rod off the solid one, and carefully capping the bottles. The capped bottles were removed from the rod and placed in a cooler. New bottles were taped on and the rod assembly was redeployed. ^{7}Be activities for suspended particles were taken as the average of the activities in both the top and bottom traps.

All sediment samples were dried at 60 °C to constant weight, ground, and homogenized using a mortar and pestle. Porosity was determined from the water content and an assumed average dry sediment density of 2.45 g cm^{-3}. Dried bed sediment was compressed into pellets using a pellet press, weighed (to the nearest 0.01 g), and measured (to the nearest 0.1 cm in height). The activity of ^{7}Be was determined using either an EG&G Ortec lithium-drifted germanium or EG&G Ortec intrinsic germanium detector coupled to a multichannel analyzer housed in a shielded clean room (*13* and modified from ref *14*). The detectors were calibrated with sediment samples spiked with a known amount of a traceable ^{7}Be standard. This yield-tracer sediment, pressed into pellets in 10-g multiples between 10 and 120 g, was counted to determine the absolute counting efficiency as a function of pellet height. Counts in the known region of interest in the γ-spectrum for ^{7}Be (477.6 keV) were recorded and compared with the known γ-activity of standards. A calibration curve was established based upon sample size in height (sample geometry). Detector efficiency as a function of both sample weight and geometry was determined. Measured counts per minute (cpm) were divided by detector efficiency yielding disintegrations per minute (dpm). This activity was then converted to picocuries per gram of dry weight (pCi g^{-1}), which was then divided into the decay-corrected value in the spiked standard sample to yield specific detector efficiency for a given pellet height. The detector stability was determined by monthly determinations of ^{137}Cs in a standard sample. The detection limit for Be^7 was ~0.2 pCi g^{-1}, assuming a counting time of 24 h. Counting errors, based on statistical analysis of the peak fits, averaged 6.7% (range = 4.1− 9.6%). Background counts were approximately 0.02 cpm at 477.6 keV. Activities were decay-corrected to the middle of each sampling period.

Activities and Inventories of ^{7}Be in Bed Sediments. Activities (*A*, in pCi cm^{-3}) of ^{7}Be at 1-cm intervals ranged from being undetectable to ~0.9 pCi cm^{-3} wet sediment (Figure 2). Depth profiles in bed sediments of ^{7}Be activity generally decreased with sample depth at both sites with the exception of the first sample period (May 28) at the DEPERE site where activities were highest at 4−5 cm depth. The presence of ^{7}Be at depth in the sediments generally results from rapid deposition and/or sediment mixing. Decreasing activity with sediment depth is expected due to the burial, isolation (from the atmospheric source), and decay of ^{7}Be associated with sediments at depth. The presence of a

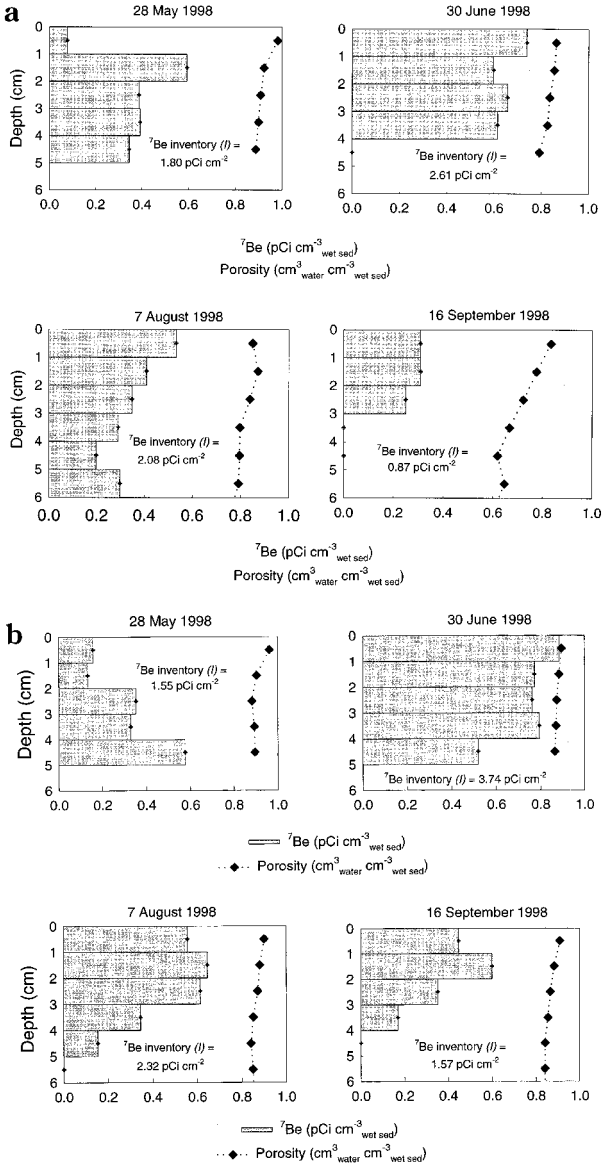

FIGURE 2. Profiles of ^{7}Be activity [*A*, in pCi (cm of wet sediment)$^{-3}$], total inventories (*I*, in pCi cm^{-2}), and porosity [cm^3 of water (cm of wet sediment)$^{-3}$] at the (a) LLBDM and (b) DEPERE sites.

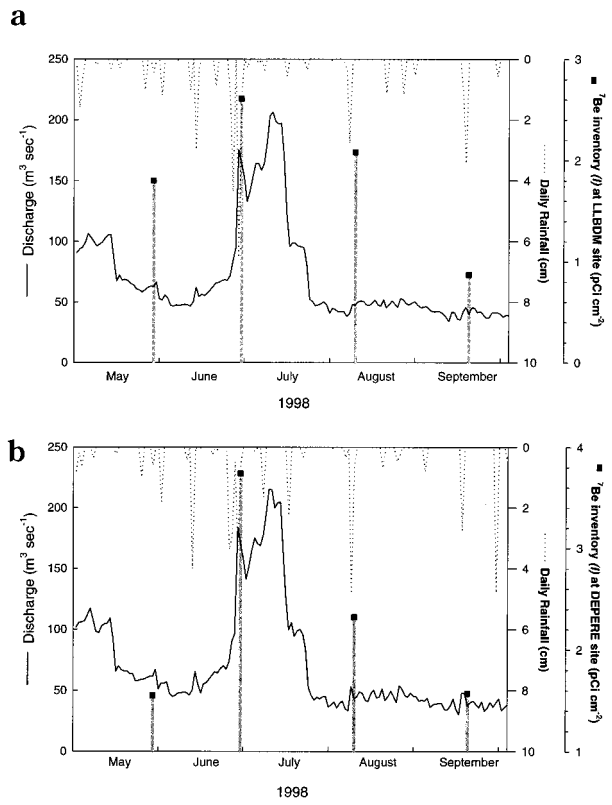

FIGURE 3. Inventories of ^{7}Be (in pCi cm^{-2}); Fox River Discharge (in ft^3 s^{-1}) at (a) Appleton, WI, and (b) Rapide Croche Dam near Wrightstown, WI; and rainfall (in in.) at (a) Appleton and (b) Green Bay, WI, from May 1998 through October 1998.

subsurface peak, like that observed in the initial sampling in May at the DEPERE site, is probably due to a combination of rapid deposition, erosion, and sediment mixing. A likely scenario is that a relatively quiescent period marked by slow sediment deposition and mixing is followed by a large deposition event that buries the sediments with higher ^{7}Be at depth. The increasing activities with sediment depth during the first sampling period could have resulted from a pulse of particles with relatively high ^{7}Be activities associated with sediments from the spring snowmelt prior to the May sampling.

Beryllium-7 inventories (I, in pCi cm^{-2}) were calculated by summing the activities for each 1-cm interval at each site ($I = \Sigma Az$ where z is the linear depth). Total ^{7}Be inventories ranged from 0.87 (LLBDM, September) to 3.74 pCi cm^{-2} (LLBDM, June) (see Figure 2). Note that the inventories for the first and third samples from LLBDM and the first two samples from DEPERE represent minimum values for the inventory because the cores were not sufficiently long to reach sediment with undetectable ^{7}Be activity. Values tended to covary between the two sites, apparently reflecting the atmospheric delivery of ^{7}Be via wet precipitation. The highest measured inventories occurred in the late June samples. These samples were collected immediately after a moderate rain event (~3.5 and ~6.5 cm) with a recurrence interval of less than 2 yr. The peak in river discharge after this rainfall event was not directly due to the rainfall event because flow in the Fox River is controlled by dams. This peak in flow must have been due to the opening of the upstream dam for a period of about 3 weeks shortly after the event.

Despite this peak in coincident ^{7}Be activities with a large rainfall event, measured inventories at other times did not correlate directly with rainfall events (Figure 3). For example, ^{7}Be inventories at both sites were higher during the first sampling (May 28) as compared to the fourth (September 16), although the latter had a larger antecedant rainfall event. Also, ^{7}Be activities in rainfall are known to vary over the year with the highest activities generally occurring in spring when atmospheric production of ^{7}Be peaks (*15*). Seasonal variations in the production rate, coupled with variations in rainfall patterns, could obscure correlation of rainfall events with activities in bed sediments. Similarly, activities of ^{7}Be in sediments from Cape Lookout Bight did not correlate well with measured inventories in sediments (*16*). It is possible that month to month variations in atmospheric input may be too small with respect to changes in the sediment inventory to show a direct correlation. In any case, ^{7}Be inventories in sediments may be assumed to result from the episodic deposition and resuspension (erosion) of sediment, which alters the inventory of ^{7}Be within the sediment bed over time.

TABLE 2. Total, Residual, and Net ^{7}Be Inventories and Net Time-Dependent ^{7}Be Fluxes Determined from Bed Sediments at the Two Sampling Sites

	^{7}Be Inventories (pCi cm^{-2})			elapsed	net time-dependent
date	total	residual	new[a]	time (d)	^{7}Be flux (fCi cm^{-2} d^{-1})
			LLBDM		
May 28	1.8				
Jun 30	2.6	1.2	1.4	33	42
Aug 7	2.1	1.6	0.5	38	13
Sep 16	0.87	1.2	0.37	40	−9.3
			DEPERE		
May 28	1.5				
Jun 30	3.7	1.0	2.7	33	82
Aug 7	2.3	2.3	0	38	0
Sep 16	1.6	1.4	0.2	40	5.0

[a] New = total − residual.

Short-Term Sediment Deposition and Resuspension Rates. Short-term sediment deposition and resuspension were estimated by mathematically separating the total ^{7}Be inventories (in pCi cm^{-2}) in bed sediments into residual and new inventories (after ref *16*). In this approach, the residual inventory is calculated from the previous sampling period and has been decay-corrected to the present sampling date. The new inventory (i.e., present sampling) is the total inventory minus the residual inventory. Sediment deposition is indicated when the total inventory is greater than the residual inventory, yielding a positive value that represents newly deposited ^{7}Be and sediment. Sediment resuspension (i.e., removal) is indicated when the calculated residual inventory is greater than the total inventory. The net ^{7}Be flux (in fCi cm^{-2} d^{-1}) is equivalent to the new inventory divided by the sampling interval.

Total, residual, and new inventories and net ^{7}Be fluxes are shown for all sampling dates (Table 2). Note that residual and new inventories are calculated from changing total inventories and therefore cannot be calculated for the first sampling period. Net deposition occurred between most sampling events as indicated by positive values for new ^{7}Be inventories and net ^{7}Be flux. The negative value calculated for the September sample from LLBDM indicates a resuspension event resulting in net removal of ^{7}Be and sediment erosion from this location under conditions of relative low and constant discharge (see Figure 3). Steuer et al. (*4*) showed that flow through the Neenah Slough, one of two dam-controlled conduits for outflow from Lake Winnebago, preferentially resuspended sediments from the LLDBM site, especially under conditions of low water level in Little Lake Butte des Morts. In keeping with this result, water level at the LLBDM site was noted as the lowest of all the sampling dates, about 1 m deep. This likely explains the somewhat counterintuitive result predicting erosion in the absence of

TABLE 3. Sediment Mass (g), Total ^{7}Be (pCi), and ^{7}Be Activity (pCi g^{-1}) in Sediment Traps at the Two Sampling Sites

date	sediment trap position	sediment mass (g)	^{7}Be (pCi)	^{7}Be acty (pCi g^{-1})
		LLBDM		
May 28	top			
	bottom			
Jun 30	top	33.13	129.8	3.92
	bottom	43.72	167.0	3.82
Aug 7	top	16.92	90.8	5.37
	bottom	23.06	224.4	9.73
Sep 16	top	17.34	66.8	3.85
	bottom	20.54	69.6	3.39
		DEPERE		
May 28	top	68.58	167.1	2.44
	bottom	342.62	842.1	2.46
Jun 30	top	40.84	253.6	6.21
	bottom	148.78	322.9	2.17
Aug 7	top	4.55		
	bottom	73.87	248.2	3.36
Sep 16	top	19.16	90.3	4.71
	bottom	39.96	105.1	2.63

a discharge peak. Moreover, this result highlights the control of dam operations at the LLDBM site on resuspension of contaminant-laden sediments.

Sediment Mass Accumulation (or Removal). The short-term deposition rate may be calculated from the time dependent ^{7}Be flux by assuming that the mean activity of the sediment particles entering or leaving the bottom is equivalent to the particle activity recorded in the sediment traps during the concurrent period of deployment:

$$\text{short-term deposition (erosion) rate} = \psi = \frac{^{7}\text{Be flux (fCi cm}^{-2}\text{ d}^{-1})}{\text{mean suspended particle acty (fCi mg}^{-1})} \quad (\text{mg cm}^{-2}\text{ d}^{-1}) \quad (1)$$

In general, suspended particles collected in near bottom traps were lower in activity than in near surface traps, but the average difference was less than 30% (range 0.4−29%) except for the June DEPERE sample (Table 3). Short-term deposition/erosion rates (ψ) were calculated using the mass-weighted average ^{7}Be activities per gram of the top and bottom traps at each site for each date (Table 4).

Long-term linear sedimentation rates in both these sediments are here estimated to be ~0.3 cm yr^{-1} at LLBDM and ~0.5 cm yr^{-1} at DEPERE based upon published ^{137}Cs profiles (*4*). These rates are generally 1−2 orders of magnitude lower than the short-term linear deposition rates at these sites which range from 0 to 65 cm yr^{-1} (Table 4). At average measured porosities, the long-term linear sedimentation rates translate to net sediment accumulation rates (ω) of ~60 and ~75 mg cm^{-2} yr^{-1} at the LLBDM and DEPERE sites, respectively. Resuspension (R), therefore, may be estimated as the ratio of the short-term deposition (erosion) rate to the long-term net sediment accumulation rate:

$$R = \psi/\omega \quad (2)$$

Effectively this ratio represents a measure of the non-steady-state, mean relative amount of short-term sediment movement into or out of the river deposit over time. The ratio varies from −16 (erosional episode) to >130 (depositional episode) and is an indication of the importance of transient particle resuspension and redeposition in these types of highly dynamic systems (Table 4).

The value of such data lies, in part, in the ability to link these fluxes to other constituents of interest. The Wisconsin Department of Natural Resources (*17*) gives an average value for (Σ)PCB concentration in surficial sediments in the LLBDM region of the river as ~3 μg g^{-1}, and surface sediment concentrations average ~2 μg g^{-1} in the Lower Fox River downstream of DePere. For example, a measured erosional flux using ^{7}Be of 2.6 mg cm^{-2} d^{-1} (LLBDM, August to September) translates into a (Σ)PCB flux of ~8 ng cm^{-2} d^{-1} back into the river. With a surface area of ~3 × 10^{6} m^{2} of PCB-contaminated sediments in the LLBDM region (*14*), PCB re-introduction into the water during this 40-d period is calculated to be ~10 kg. For comparison, the total annual estimated flux of PCBs from the Fox River to Green Bay is estimated to be on the order of 200 kg yr^{-1} (*18*). Hence, even relatively minor resuspension events may contribute significantly to this process, and major resuspension/erosion events will, in all likelihood, dominate mass transport downstream.

Acknowledgments

We thank Scott Cieniawski for assistance in administering this grant and for his help during field work. We also thank the management and support staff at the U.S. Geological Survey's Center for Coastal Geology (St. Petersburg, FL) for logistical support during preparation of this manuscript. This study was supported by a research grant (IAG DW14947829-01-0) from the Great Lakes National Program Office (GLNPO) of the U.S. Environmental Protection Agency and in part by the University of Wisconsin Sea Grant Institute under grants

TABLE 4. Net Time-Dependent ^{7}Be Fluxes, Mean ^{7}Be Suspended Particle Activities, and Calculated Short-Term Deposition/Erosion Rates and Resuspension Ratios at the Two Sampling Sites

station date	net time-dependent ^{7}Be flux (fCi cm^{-2} d^{-1})	mean ^{7}Be suspended particle acty[a] (fCi mg^{-1})	short-term deposition (+) or erosion (−) rates[b] (ψ) mass (mg cm^{-2} d^{-1})	equiv linear rates (mm/Δt^{-1})	equiv linear rates (cm yr^{-1})	resuspension ratio[c] (ψ/ω)
			LLBDM (ω = 60)			
May 28						
Jun 30	42	3.86	11	18	20	67
Aug 7	13	7.88	1.7	3.2	3.1	10
Sep 16	**−9.3**	3.60	**−2.6**	**−5.1**	**−4.7**	**−16**
			DEPERE (ω = 75)			
May 28						
Jun 30	82	3.04	26.9	59	65	131
Aug 7	0	3.17	0.0	0	0	0
Sep 16	5.0	3.31	1.5	4.0	3.7	7

[a] From suspended particles collected in sediment traps and mass weighted. [b] Calculated via eq 1; linear rate calculated using average porosities. [c] ω = long-term net sediment accumulation rate (mg cm^{-2} yr^{-1}).

from the National Sea Grant College Program, NOAA, and the State of Wisconsin (Grant NA46R60481-R/EC-2). Use of brand names does not constitute endorsement by the U.S. Geological Survey.

Literature Cited

(1) *Wisconsin Water Quality Assessment Report to Congress*: *1996*; Wisconsin Department of Natural Resources: Madison, WI, 1996; WT254-96.
(2) Velleux, M.; Endicott, D.; Steuer, J.; Jaeger, S.; Patterson, D. *J. Great Lakes Res.* **1995**, *21* (3), 359.
(3) Manchester-Neesvig, J. B.; Andren, A. W.; Edgington, D. N. *J. Great lakes Res.* **1996**, *22* (2), 444.
(4) Steuer, J. J.; Jaeger, S.; Patterson, D. *A Deterministic PCB Transport Model for the Lower Fox River between Lake Winnebago and DePere, Wisconsin*; Publication WR 389-95; Wisconsin Department of Natural Resources: Madison, WI, 1995.
(5) Lal, D.; Malhotra, P. K.; Peters, B. *J. Atmos. Terr. Phys.* **1958**, *12*, 306.
(6) Olsen, C. R.; Larsen, I. L.; Lowry, P. D.; Cutshall, N. H.; Nichols, M. M. *J. Geophys. Res.* **1986**, *91*, 896.
(7) Baskaran, M.; Santschi, P. H. *Mar. Chem.* **1993**, *43*, 95.
(8) *Water Resources Data for Wisconsin, 1998*; U.S. Geological Survey; U.S. Government Printing Office: Washington, DC, 1999; WI-98-1.
(9) Wisconsin State Climatology Office, Madison, WI, unpublished results.
(10) Fitzgerald, S. A. U.S. Geological Survey, Guaynabo, Puerto Rico, unpublished results.
(11) Gardner, W. D. *J. Mar. Res.* **1980**, *38* (1), 41.
(12) Sullivan, J. F. Wisconsin Department of Natural Resources, unpublished results.
(13) Klump, J. V. University of Wisconsin—Milwaukee, Great Lakes Water Institute, unpublished results.
(14) Larsen, I. L.; Cutshall, N. H. *Earth Planet. Sci. Lett.* **1981**, *54*, 379.
(15) Robbins, J. A.; Eadie, B. J. *J. Geophys. Res.* **1991**, *98*, 17,081.
(16) Canuel, E. A.; Martens, C. S.; Benninger, L. K. *Geochim. Cosmochim. Acta* **1990**, *54*, 237.
(17) *Polychlorinated Biphenyl (PCB) Contaminated Sediment in the Lower Fox River: Modeling Analysis of Selective Sediment Remediation*: *1997*; Wisconsin Department of Natural Resources, Bureau of Watershed Management: Madison, WI, 1997; WT482-97.
(18) Glenn, W. U.S. Environmental Protection Agency, personal communication, 2000.

Received for review February 2, 2000. Revised manuscript received October 24, 2000. Accepted October 25, 2000.

ES000951C