



# The removal of particle-reactive radionuclides in shallow water: Bottom scavenging versus particle settling of iodine-131 and beryllium-7



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## ABSTRACT

In pelagic waters, the removal of particle-reactive radionuclides is controlled by nuclide sorption to particles and subsequent settling by gravity. However, in shallow nearshore waters, the dominant mechanism of nuclide scavenging is not so clear. Understanding how particle-reactive radionuclides are scavenged from the water column is critical if these tracers are to be used as proxies of particle flux in shallow aquatic systems. In this study, we present evidence that the removal of particle-reactive radionuclides in nearshore and turbulent waters is primarily controlled by bottom scavenging. Specifically, we measured both water column and bottom sediment activities of sewage-sourced iodine-131 (<sup>131</sup>I,  $t_{1/2} = 8.02$  days) and atmospherically-sourced beryllium-7 (<sup>7</sup>Be,  $t_{1/2} = 53.3$  days) in a semi-enclosed harbor. We show that the water column <sup>7</sup>Be/<sup>131</sup>I flux ratio that is required to sustain observed harbor bottom inventories of both nuclides is incongruent with <sup>7</sup>Be/<sup>131</sup>I activity ratios on water column particles, and (2) <sup>131</sup>I and <sup>7</sup>Be derived mass fluxes of particulate matter to the harbor bottom are in concordance with each other and independently made estimates of river sediment loading to the harbor only when bottom scavenging of both particle-bound and dissolved (<0.7 μm) nuclide fractions are considered.

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“A man with a watch knows what time it is. A man with two watches is never sure.”

—Segal's Law

## 1. Introduction

In aquatic systems, naturally occurring radionuclides have been used as tracers of particle mass flux for half a century (e.g., Bhat et al., 1969). In pelagic waters, the removal of radionuclides from the water column is thought to be controlled by nuclide sorption to particles and subsequent settling by gravity. If the rate of supply of a nuclide is known, then the expected inventory of that nuclide ( $A_E$ , Bq m<sup>-2</sup>) can be calculated as the ratio of its supply rate and the

radioactive decay constant of the nuclide ( $\lambda = \text{d}^{-1}$ ). The difference between the expected and observed ( $A_O$ , Bq m<sup>-2</sup>) nuclide inventory (i.e., the missing inventory) times the nuclide decay constant gives the flux or removal rate of the nuclide ( $J_A$ , Bq m<sup>-2</sup> d<sup>-1</sup>) on settling particles.

$$J_A = \lambda(A_E - A_O) \quad (1)$$

The particle mass flux ( $J_{mass}$ , g m<sup>-2</sup> d<sup>-1</sup>) can then be calculated as the product of the nuclide flux and the particle mass/activity ratio ( $mass/A^{part}$ ).

$$J_{mass} = J_A \times \frac{mass}{A^{part}} \quad (2)$$

In shallow coastal waters, however, Waples et al. (2015, 2017) found that the removal of particle-reactive radionuclides is primarily controlled by bottom scavenging, where turbulent diffusion rapidly presents both particle-bound and dissolved fractions of the nuclide to bottom sediments. Any sorption of the dissolved nuclide fraction to bottom sediments will increase the estimate of mass flux if eq. (2) is used, which assumes that all the nuclide activity removed from the water column is solely mediated by settling

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particles.

Understanding how particle-reactive radionuclides are scavenged from the water column is critical if these tracers are to be used as proxies of particle flux in shallow aquatic systems. In this study, we present additional evidence that the removal of particle-reactive radionuclides in nearshore and turbulent waters is primarily controlled by bottom scavenging. Specifically, we measured both water column and bottom sediment activities of sewage-sourced iodine-131 ( $^{131}\text{I}$ ,  $t_{1/2} = 8.02$  days) (Smith et al., 2008; Rose et al., 2013) and atmospherically-sourced beryllium-7 ( $^7\text{Be}$ ,  $t_{1/2} = 53.3$  days) (e.g., Walling, 2013) in a semi-enclosed harbor (Montenero, 2015). We show that the water column  $^7\text{Be}/^{131}\text{I}$  flux ratio that is required to sustain observed harbor bottom inventories of both nuclides is incongruent with  $^7\text{Be}/^{131}\text{I}$  activity ratios on water column particles, and (2)  $^{131}\text{I}$  and  $^7\text{Be}$  derived mass fluxes of particulate matter to the harbor bottom are in concordance with each other and independently made estimates of river sediment loading to the harbor only when bottom scavenging of both particle-bound and dissolved ( $<0.7 \mu\text{m}$ ) nuclide fractions are considered.

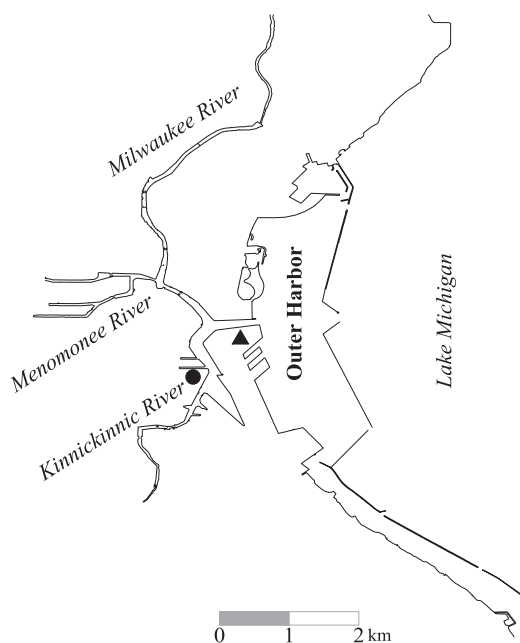
## 2. Materials and methods

### 2.1. Study site

This study was conducted in the Milwaukee Outer Harbor (area:  $4.4 \text{ km}^2$ ; average depth:  $\sim 8 \text{ m}$ ; Fig. 1). The harbor receives an average sewage effluent discharge of  $\sim 300 \times 10^6 \text{ L d}^{-1}$  from the Jones Island wastewater treatment plant (JI WWTP), and the confluent flow of the Milwaukee, Menomonee, and Kinnickinnic Rivers. Four gaps in the outer seawall provide open access to Lake Michigan ( $58,000 \text{ km}^2$ ).

### 2.2. Sediment sampling

Sediment samples were collected by gravity coring on four occasions in 2014 (16 July, 29 July, 26 August and 8 October). Sediment



**Fig. 1.** Study area showing the Milwaukee Outer Harbor in Milwaukee, Wisconsin ( $43.0^\circ \text{ N}$ ,  $87.9^\circ \text{ W}$ ). Outflow of Milwaukee, Menomonee, and Kinnickinnic Rivers conflate at entrance to Outer Harbor. Jones Island wastewater treatment plant (delta) discharges treated effluent directly to the Outer Harbor. UW School of Freshwater Sciences (circle).

cores (diameter =  $6.8 \text{ cm}$ ) were extruded and the top  $10 \text{ cm}$  were placed in medium-sized plastic jars (section 2.4) and analyzed for  $^{131}\text{I}$  and  $^7\text{Be}$  activity by gamma spectroscopy. Due to the short half-life of both nuclides, inventories of both nuclides below  $10 \text{ cm}$  depth were assumed to be negligible.

### 2.3. Water column sampling

Water from the Milwaukee Outer Harbor was collected on 29 July, 26 August and 8 October 2014. Approximately  $180 \text{ L}$  of water were collected during each sampling event from three sites spanning the mid-northern, -central, and -southern Outer Harbor water column (i.e.,  $\sim 60 \text{ L}$  per site). Water was collected using a  $30 \text{ L}$  Niskin bottle (General Oceanics, Inc.) and stored in  $20 \text{ L}$  plastic carboys. At each of the three stations, two carboys were filled with water from just below the surface, and one was filled with water taken from approximately  $5 \text{ m}$  deep.

On shore, water samples from all three sites were immediately combined in large plastic buckets and filtered through tared glass fiber filters ( $142 \text{ mm}$ ,  $0.7 \mu\text{m}$  pore size) for particulate matter. Next, the filters were analyzed by gamma spectroscopy to determine the particulate fraction of  $^{131}\text{I}$  and  $^7\text{Be}$  activity. The filters were then dried ( $60^\circ \text{ C}$ ,  $24 \text{ h}$ ) and weighed to calculate the concentration of suspended particulate material ( $\text{g SPM L}^{-1}$ ).

The dissolved ( $<0.7 \mu\text{m}$ ) nuclide fractions in the filtrate were scavenged in a sequential series of treatments involving the addition of anion resin ( $20 \text{ g}$  BioRad AG 1-X8; Montenero, 2015) and newly formed iron hydroxide precipitate (Waples et al., 2003), where each scavenging compound (i.e., resin or iron) was filtered from the sample before the next was added. Because this method evolved with time, the 29 July procedure involved one scavenging treatment with anion resin followed by one treatment with iron precipitate; the 26 August procedure involved two scavenging treatments with anion resin and one treatment with iron precipitate; and the 8 October procedure involved two scavenging treatments with anion resin followed by two treatments with iron precipitate. The collected resins and precipitates from each treatment were then placed in separate jars and analyzed by gamma spectrometry.

The total activity of dissolved  $^{131}\text{I}$  ( $A_{\text{total}}$ ) was calculated as:

$$A_{\text{total}} = \frac{A}{1 - (B/A)} \quad (3)$$

where  $A$  was the measured  $^{131}\text{I}$  activity from the first anion resin treatment and  $B$  was the measured  $^{131}\text{I}$  activity from the second anion resin treatment (Rutgers van der Loeff et al., 2006). The  $B/A$  ratio for the 29 July sample was taken as the average of the  $B/A$  ratios for 26 August and 8 October (i.e.,  $\sim 50\%$ ). Because some  $^7\text{Be}$  was scavenged on the anion resin, total dissolved  $^7\text{Be}$  activity was simply calculated as the sum of all  $^7\text{Be}$  activity measured on all collected scavenging material (i.e., anion resin and iron precipitate). Previous studies have assumed complete (e.g., Hawley et al., 1986; Jweda et al., 2008) or nearly complete ( $\sim 80\%$ ; Kadko et al., 2015) scavenging of  $^7\text{Be}$  by iron.

### 2.4. Gamma spectroscopy

Samples in this study were analyzed in medium sized jars (Parkway Plastics, Inc.,  $83 \text{ mm}$  diameter,  $55 \text{ mm}$  height,  $230 \text{ ml}$  volume) by gamma spectroscopy on one Canberra high-purity germanium (HPGe) well detector (model no. GCW4023) and two Ortec HPGe planar detectors (model no. GMX-23210-P and LO-AX-70450/30-S). Counting efficiencies on each gamma detector were determined using a europium-152 ( $^{152}\text{Eu}$ ) standard (Nichols et al.,

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