SAMPLING METHODS AND APPROACHES USING RADIONUCLIDE TRACERS IN THE STUDY OF SEDIMENT RESUSPENSION AND CROSS MARGIN TRANSPORT IN NEARSHORE OF THE LAURENTIAN GREAT LAKES

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ABSTRACT
Preliminary results from an investigation of the U-238/Th-234 parent/daughter pair in the nearshore environments of Lake Michigan show that short-lived, naturally-occurring radionuclides are useful in determining the time scales of sediment transport processes on the orders of days to weeks. While the long term, depositional sink for some new particles entering the lake, either from rivers or shoreline erosion, has been well documented, the aggregate time scale of the processes by which these particles move from source to sink is not well quantified. These relatively “new” particles may undergo numerous episodes of resuspension and redeposition before making their way through the coastal margin and entering the long term focusing processes active in the depositional basins of the lake. These episodic events can be large in scale and magnitude, resuspending as much or more material than is permanently deposited in the lake on an annual basis. Th-234, a particle reactive radionuclide with a 24 day half-life, would appear to be well suited as a tracer for following rapid particle movement, particle residence times in the overlying water, and alongshore transport processes. The remotely operated vehicle (ROV) deployed technique for collection of surface sediments residing in non-depositional (hard bottom) lakebeds and some preliminary time series results for transient excess Th-234 inventories in the nearshore of Lake Michigan are described.

Key Words: Sediments, Resuspension, ROV, Radionuclides, Great Lakes

1 INTRODUCTION
Sediment resuspension, particularly in response to episodic storm events, is a common characteristic of coastal and nearshore environments. The resulting “benthic storms” may play a significant role in the dynamics of benthic systems, including structuring the physical characteristics of the bottom, providing a mechanism for the recycling of biogeochemically important materials to the water column, and driving horizontal and cross margin transport of suspended particulates (Klump et al., 1995). Such influences are not limited to shallow waters. Even at the deepest sounding in the Great Lakes evidence of major episodic physical disturbance and current flow exists. Extensive ripple marks on the bottom at over 400 m in Lake Superior, for example, are indicative of occasional, recurring bottom currents in excess of 10-40 cm s⁻¹ (Klump et al., 1989).

Coastal sediment resuspension in the Great Lakes is a common phenomenon. Using Coastal Zone Color Scanner images, Mortimer (1988), pointed out several major, basin-scale coastal hydrodynamic phenomena observable in southern Lake Michigan, including front formation and upwelling events, intermittent mobilization and surface transport of sediment resuspended by storms, and extensive temporary trapping of river and bluff derived materials between the shoreline and the offshore-migrating thermal front.

Satellite imagery compiled over the last few decades displays the frequent spin-up of high turbidity coastal plumes in response to wind driven wave action. These events occur throughout the year, but are dominated in magnitude by major annually recurrent plume events during the winter-spring transitional period, generally early February through early April, prior to the onset of thermal stratification in late May and early June (Fig. 1) (Chen et al., 2002).

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Note: Discussion open until June 2004.
Fig. 1 A satellite image of a major coastal “plume event” in the southern basin of Lake Michigan (Based on the image provided by the SeaWiFS Project, NASA/Goddard Space Flight Center, and ORBIMAGE). The nearshore bottom (inserted photographs) is characterized by rocky, hard clay and sandy substrates, well scoured by wave action.
The nearshore zone (< 40-m depth) within southern Lake Michigan is characterized by a diverse array of bottom types from cobble and boulder fields, to sand and gravel bottoms, to hard clay banks (Fig. 1). Despite a lack of permanently accumulating sediments within most of this region, it is an area of intense and episodic sediment resuspension. Storm fronts typically pass through this region every 5-7 days in the winter and roughly every 7 to 10 days in the summer (Roebber and Gehring, 2000). Depending upon the strength, direction and fetch of the winds, wave action and shore parallel currents can perturb bottom sediments to depths in excess of 30 m (Lou et al., 2000). These events can be very large in magnitude, resuspending as much material into the nearshore water column in a matter of days, as is annually deposited to the sediment sink in the entire permanent depositional zone of the lake (Eadie et al., 1996; Schwab et al., 2000).

It has long been known that permanent sediment accumulation in southern Lake Michigan is skewed to the eastern side of the basin (Robbins and Edgington, 1975). What has also been shown, however, using repeated measurements of Cs-137 inventories in the depositional environments of the lake, is that horizontal movement and focusing of sediments is occurring on a time scale of decades (Edgington and Robbins, 1990). On one hand, therefore, deposition and focusing at depth are long-term processes at or near steady state in the lake. The depositional imprint on the bottom in terms of thickness of the mass accumulated indicates that these processes have been relatively constant for at least the last 3,000 years. On the other hand, storm induced resuspension in the shallow coastal zone of the lake occurs very quickly, very episodically and has the potential to move a great deal of mass of material within a matter of days to weeks. The answer to the question of how “new” sediments are transported from source to sink (i.e. from where they enter in shallow waters to where they end up in deep waters) and the time scales of that process are fundamental to understanding the fate of riverine, shoreline, and atmospherically derived materials in the lake. The timing of these episodic events could have an influence on the ecology of the system, particularly during the winter – spring transitional period when the spring bloom is initiated, fueled by regenerated nutrients and increasing solar radiation (Ji et al., 2002).

The contrast in these observations led to the development of a major research program to study the influence of these episodic events in Lake Michigan, the EEGLE project (Episodic Events Great Lakes Experiment (http://www.glerl.noaa.gov/eegle/eegle.html)). Of particular interest was the fact that the largest of these events appeared to occur during late winter – early spring, time periods for which there were very few previous in situ observations. It was hypothesized that the nearshore bottom serves as a temporary repository for sediments entering the lake and that this resuspension initiates both alongshore and shore normal or cross margin transport of fine-grained particles. The nearshore region, however, does not contain bottom types amenable to conventional sediment sampling techniques. Ekman grabs, ponars, and gravity or box corers are incapable of operating on these bottoms where virtually no penetration by the sampler is possible. This “resuspendible pool” temporarily resides as a fine-grained dusting of material over otherwise hard, non-depositional substrates. Consequently, it was necessary to develop techniques to quantitatively measure the inventories of sediments and radionuclides residing in this transient particle pool. A quantitative measure on an areal basis was essential to establish the magnitude of the inventories of sediments and sediment associated constituents, and how those inventories vary in both space and time. Such information is critical for understanding the frequency and duration of resuspension, and on the residence time of particles in the nearshore benthic system.

2 SAMPLING METHODOLOGY

In order to sample materials which exist as a thin veneer or flocculent layer on these nearshore, wave and current impacted bottoms, it was necessary to devise a sampling technique that could both collect a representative sample with little or no disturbance prior to sampling, and do so quantitatively from a known surface area. The developed technique uses a pair of hydraulic vacuuming devices deployed by a small remotely operated vehicle (ROV). These vehicles are increasingly being used as sampling tools and platforms for benthic studies in large lake systems (Klump et al., 1992, 1997, 2001). The vehicle is powered from the surface through an umbilical which also serves to control the maneuvering of the ROV, light the sampling zone when ambient light is insufficient to view operations, transmit a video image of
the bottom area to be sampled, and control the operation of the sampling system. The ROV is capable of working at depths to > 300 m, but in this sampling configuration is limited to ~50 m (Fig. 2).

Fig. 2 Surficial sediment sampler and early prototype mounted on ROV (top and bottom right).
Bottom left: Construction, operation, and details of nose cone and flapper valve cap.

The sampler operates like a hydraulic lift, and hence, the length of sample lines and head between the water surface and the deck are the main limitations on depth of operation in the current configuration. The sampling chamber consists of a clear polycarbonate jacketed cylinder attached to an aluminum nose cone which serves as the sampling head and a PVC cap that holds a one-way flapper valve (Fig. 2). The inner sampling chamber is a cylinder ~40 cm in length and 6.7 cm in diameter (standard 3 inch plastic core liner). The outer jacket is a 10.2 cm inside diameter (ID), clear polycarbonate cylinder to which is attached an 3.6 cm long tapered aluminum nose cone with an opening equal to the inner sampling chamber diameter. Sampling from the inner chamber is accomplished through a 12.5 mm diameter standpipe centered in the chamber at ~ 2.5 cm above the nose cone. In practice this standpipe is at or near the sediment-water interface during sampling. Water from the outer chamber is drawn through the nose
Fig. 3 Typical sampling sequence on sandy bottom in water depth of 25 m: 1) sampling initiated; 2) mid sampling; 3) sampling completed and sampler raised

cone through a radial array of 16 fine water jets (~ 1.5 mm diameter) whose turbulent action resuspends surface sediments into the closed inner chamber (surface area sampled = 35.3 cm²). The sediment slurry created is pumped directly to the surface through 12 mm ID clear flexible plastic tubing attached to the stand pipe using air-driven Teflon diaphragm pumps (Wilden, Inc) on deck. Two independent samplers are raised and lowered as much as 15 cm by electrically driven slide tables controlled from the ROV operator’s control module on board the research vessel.

A typical sampling sequence includes: visual inspection of the bottom area to be sampled, moving the ROV into position and dropping it slowly to the bottom without disturbing the surrounding area. This is relatively easily achieved when the vehicle is nearly neutrally buoyant. Once the vehicle is resting on the bottom, one of the samplers is lowered until firmly engaged in the sediment surface. The upper end of the sampling chamber is fitted with a one-way flapper valve that allows water to exit the chamber as the sampler is lowered, but prevents inflow from the top during pumping. The valve consists of a 6.3 cm outside diameter (OD) circular piece of 0.5-mm thick silicon rubber held in place with a 3.8 cm OD stainless steel washer. The pump is activated drawing bottom water from the outer jacket through the aluminum nose cone causing the jetting action to vigorously resuspend the surface layer of sediments trapped within the sample chamber (Fig. 3). These sediments are entrained in the central standpipe and pumped to the surface. Flow rates are approximately 10 L/min. The sediment slurry generated is continuously sampled until the water clears. Typically, at these depths with an approximately 60 m sampling line, this takes less than 2 min. Once the resuspendible sediment has been exhausted the pumping ceases, the sampler is raised and the ROV is repositioned for the next sample or the sampling sequence is repeated with the opposite sampler. Since the entire operation is monitored by video cameras, samples that are not quantitative, due to loss, disturbance, or erosion from outside the cone, are discarded and the sequence restarted. Sediment slurries (4 to 12 L in total volume) are temporarily stored on deck in 4-L plastic jars.

In areas where fine-grained surface sediments are sparse, several samples may be combined in order to obtain sufficient mass for analysis. For subsequent analysis of radionuclides, principally Th isotopes (Waples et al., 2003), particles > 63 µm in size are removed from these slurries by settling and the remaining fraction (0.45-63 µm) is recovered via pressure filtration across tared, 293 mm, 0.45 µm nitrocellulose filters on board the research vessel within a few hours of collection. The activity of Th-234 in Lake Michigan is approximately an order of magnitude lower than in seawater, owing to the concomitantly lower U-238 concentration in lake water (Waples et al., 2000).

3 PHYSICAL CHARACTER OF SAMPLES COLLECTED: MASS INVENTORIES, ORGANIC MATTER CONTENT, AND PARTICLE SIZE DISTRIBUTION OF RESUSPENDIBLE MATERIALS

This sampling methodology was deployed throughout the nearshore of the southern basin of Lake Michigan during four cruises from September 1998 to August 1999. Over 200 samples were collected and analyzed. Water depths ranged from 10-50 m. The sub-63 µm particle size masses collected ranged from less than 0.0020 to over 1.0 g cm⁻² with an average of 0.25 g cm⁻² (Fig. 4). Particle size distribution (Jackson, 1985) of samples collected at seven typical stations from throughout the nearshore zone of the southern basin is given in Table 1 in terms of mass inventories (mg cm⁻²) for six fractions: sand (> 63 µm, discarded prior to analysis), coarse silt (62.5-31 µm), medium silt (31-16 µm), fine silt (16-8 µm), very fine silt (8-2 µm), and clay (< 2 µm); and in terms of weight percent for the less than 63 µm fraction retained for analysis. For this analytical sample, the clay fraction showed the greatest variability and ranged from a low of 8% to a high of 33% of the sand-eliminated sample on a dry weight basis, but in general the samples collected were relatively similar in composition. Organic matter content as determined by loss on ignition (500° C) averaged 3.7% for all samples (n = 195) and ranged from a low of <0.2% to a high in excess of 10%. There was no apparent correlation between organic matter content and the mass inventory collected (Fig. 5) or the water depth of the station (Fig. 4).
Fig. 4 Mass inventories (g m$^{-2}$) and loss on ignition (LOI, fraction by weight) for “resuspendible” sediment samples collected with the ROV deployed sampler from throughout the nearshore of the southern basin of Lake Michigan (plotted against water depth).

Table 1 Particle size distribution of surface sediment (“resuspendible particulates”) for a set of typical locations in the nearshore zone of southern Lake Michigan. Given are mass inventories (in mg cm$^{-2}$) for the total sample recovered, and the % by weight of the sand-free (< 63 µm) fraction utilized for analysis of sediment Th-234 activities and inventories.

<table>
<thead>
<tr>
<th>Station location</th>
<th>Depth meters</th>
<th>Sand &gt; 63 µm</th>
<th>Coarse silt 62.5-31 µm</th>
<th>Medium silt 31-16 µm</th>
<th>Fine silt 16-8 µm</th>
<th>V. fine silt 8-2 µm</th>
<th>Clay &lt;2 µm</th>
<th>% of &lt;63 um fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Whitefish Bay</td>
<td>26</td>
<td>2309</td>
<td>275</td>
<td>121.4</td>
<td>63.6</td>
<td>40.5</td>
<td>80.9</td>
<td>47</td>
</tr>
<tr>
<td>Milwaukee</td>
<td>23</td>
<td>3572</td>
<td>49</td>
<td>26.1</td>
<td>26.1</td>
<td>7.5</td>
<td>52.3</td>
<td>30</td>
</tr>
<tr>
<td>Racine</td>
<td>45</td>
<td>648</td>
<td>142</td>
<td>102.5</td>
<td>75.8</td>
<td>66.2</td>
<td>33.1</td>
<td>34</td>
</tr>
<tr>
<td>Waukegan</td>
<td>30</td>
<td>602</td>
<td>75</td>
<td>78.9</td>
<td>66.6</td>
<td>100.4</td>
<td>101.4</td>
<td>18</td>
</tr>
<tr>
<td>Gary</td>
<td>15</td>
<td>64</td>
<td>18</td>
<td>11.4</td>
<td>11.4</td>
<td>17.1</td>
<td>12.5</td>
<td>25</td>
</tr>
<tr>
<td>Indiana Dunes</td>
<td>20</td>
<td>82</td>
<td>59</td>
<td>83.9</td>
<td>45.7</td>
<td>45.7</td>
<td>61.6</td>
<td>20</td>
</tr>
<tr>
<td>St. Joseph</td>
<td>20</td>
<td>229</td>
<td>15</td>
<td>4.8</td>
<td>5.6</td>
<td>1.3</td>
<td>9.0</td>
<td>43</td>
</tr>
<tr>
<td>Mean</td>
<td>25.6</td>
<td>1073</td>
<td>90.3</td>
<td>61.3</td>
<td>42.1</td>
<td>39.8</td>
<td>50.1</td>
<td>31%</td>
</tr>
</tbody>
</table>
4 CHANGING INVENTORIES OF A SHORT-LIVED RADIONUCLIDE: A METHODOLOGY FOR ASSESSING SHORT-TERM SEDIMENT DEPOSITION AND RESUSPENSION RATES

The utility of Th-234 as a tracer for following short term particle dynamics in aquatic systems has been well described and extensively used in marine systems (Aller and Cochran, 1976; Bacon and Anderson, 1982; Bhat et al., 1969; Buesseler et al., 1992; Coale and Bruland, 1985; Gustafsson et al., 1997; Kaufman et al., 1981; Kershaw and Young, 1988; Moran and Buesseler, 1992; Wei and Murray, 1992). Its utility in freshwater systems, however, has been hindered by the low activity of the parent U-238 and the consequent analytical complexities of quantification of Th-234 at very low levels (~ 0.02 dpm per liter). These analytical complexities have been resolved using isotope dilution and low-level beta counting (Waples et al., 2003) and routine measurements have been made as part of the EEGLE program. The application of the U-238/Th-234 parent/daughter pair relies principally on the fact that the parent U-238 is a long-lived (1/2 life 4.5 x 10^9 y), soluble, conservative constituent in lake water with an activity in Lake Michigan at a constant 0.23 ± 0.02 dpm L^-1 (105 fCi L^-1) and its daughter Th-234 is a short lived (1/2 life 24.1 d), particle reactive constituent, which is rapidly scavenged by fine-grain particles and transported both horizontally and vertically along with the active particle pool. The presence of excess, i.e. unsupported Th-234, in sediments is a measure of the deposition of particles from the water column to the lake bottom. In nearshore areas, this deposition is temporary, with no net permanent accumulation occurring, and the amount of excess Th-234 on the bottom varies according to particle input, movement, settling, resuspension, and redeposition processes, all of which are highly dependent upon episodic storm and wave events. These conditions can change on a daily basis. The presence of excess Th-234 on the bottom indicates that particle removal from the water column to the sediments is sufficiently rapid, and the residence time of particles in the water column is sufficiently short to create a disequilibrium between production of Th-234 from its parent U-238 and removal by particle transport processes.

The simplest approach using short-lived, particle reactive radionuclide inventories for inferring particle residence times within the water column and particle transport dynamics activities in a one dimensional water column (i.e. vertical transport) applies an instantaneous calculation based upon the amount of activity (e.g. excess Th-234, Be-7, etc.) in the water column relative to the flux to the sediments \( J_{sed} \). The latter is calculated from the measured inventory in surface sediments \( I_{sed} \) as collected with the ROV deployed sampling system, i.e.

\[
J_{sed} = \lambda \cdot I_{sed}
\]
where $\lambda$ is the decay coefficient for the radionuclide in question. The residence time of the radionuclide
and, by inference, the particles with which it is associated is then simply the water column inventory ($I_w$) divided by the “loss” to the sediments, i.e.:

$$\tau_{\text{res}} = \frac{I_w}{J_{\text{sed}}} \quad (2)$$

A second method for calculating the flux to the sediment involves comparing measurements of excess Th-234 inventories (i.e. fluxes) repeatedly at a single location over time. A change in the inventory results from both decay of Th-234 previously deposited and the addition or loss of material in the intervening time interval and may be calculated (see e.g., Fitzgerald et al., 2001 and Canuel et al., 1990, in which this approach was used for the short lived radionuclide, Be-7). $I_1$ and $I_2$ are the inventories taken sequentially over the elapsed time interval $t$ ($t = t_2 - t_1$). The inventory at $t_2$ ($I_2$) is composed of the remaining initial existing inventory, less decay, ($I_{\text{residual}}$) plus the amount added (or substracted) during the time interval ($I_{\text{new}}$), i.e.:

$$I_2 = I_{\text{new}} + I_{\text{residual}} \quad (3)$$

where

$$I_{\text{residual}} = I_1 \exp(-\lambda t) \quad (4)$$

and

$$I_{\text{new}} = I_2 - I_1 \exp(-\lambda t) \quad (5)$$

The flux of new material ($J_{\text{new}}$) during the time interval $t$ is, therefore, given by:

$$J_{\text{new}} = \frac{\lambda [ I_2 - I_1 \exp(-\lambda t)]}{1 - \exp(-\lambda t)} \quad (6)$$

An example of this calculation for a station in Lake Michigan is as follows:

- Initial sample collected @ $t_1 = 14$ Apr 1999
- Second sample collected @ $t_2 = 18$ Apr 1999
- Elapsed time: $\Delta t = 4$ days
- Measured Sediment inventory @$t_1$: $I_1 = 0.189$ dpm cm$^{-2}$
- Measured Sediment inventory @$t_2$: $I_2 = 0.277$ dpm cm$^{-2}$

Therefore, the flux of Th-234 entering the bottom ($J_{\text{sed}}$) is equivalent to $289$ dpm m$^{-2}$ d$^{-1}$ (13.14 fCi cm$^{-2}$ d$^{-1}$). The water column inventory of Th-234, based upon the activity of the particle fraction and the depth of the water (23 m) is equal to $0.306$ dpm cm$^{-2}$ (139 fCi cm$^{-2}$). This leads to a residence time for particle associated Th-234 of ~ 11 days.

One may also calculate short term sediment deposition (or erosion) rates, $\omega$, as:

$$\omega = \frac{J_{\text{sed}}}{A_w} \quad (7)$$

where $A_w$ is the particulate activity in the water column, in this example equivalent to $115$ dpm g$^{-1}$ (52.3 pCi g$^{-1}$). The short-term mass sediment deposition, therefore was equivalent to $0.25$ mg cm$^{-2}$ d$^{-1}$ or $91.8$ mg cm$^{-2}$ y$^{-1}$. This is approximately 10 times the average sedimentation rate for the lake as a whole (Robbins and Edgington, 1975). The rationale for using the activity of the suspended particulate fraction in the water column in equation 7, as opposed to the activity of resuspendible sediments collected from the bottom, is that it is assumed that the activity within the water column better reflects the pool of particles actively involved in particle transport, and that given the vigor of the sampling technique, bottom sediments are variously diluted with older and normally unresuspended materials.

### 4.1 The Linnwood 2000 Time Series Example

Application of this method may be illustrated by a short time series at a single location in 20 m of water on the western side of the basin near Milwaukee (Linnwood 20-m station which is 2.5 km from shore). Samples were collected from the water column using a submersible pump and surface sediments using the ROV suction sampler on a daily basis for a period of 8 consecutive days from 21-28 March 2000. This station was one of four in a shore-normal transect sampled for both water column and sediment Th-234 activities and inventories. Empirical measures of water column inventories are calculated daily from the concentration of suspended particulates (TSS, mg L$^{-1}$) and the activity of Th-234 (dpm g$^{-1}$) on these particles. Sediment inventories are similarly calculated from mass inventories on the bottom (kg m$^{-2}$) and Th-234 activities in sediments. Water column inventories averaged $670 \pm 107$ dpm m$^{-2}$, whereas
sediment inventories were generally much greater, and more variable, ranging from 0 to nearly 5500 dpm m$^{-2}$.

Inventories and the flux of excess Th-234 calculated via equation 6 are given on a daily basis at this location in Table 2. Both positive (deposition) and negative (erosion) results are obtained, as sediment inventories fluctuate from day to day. Residence times for particles in the overlying water calculated from the water column inventory (I$_{w}$) and J$_{sed}$, i.e.:

$$T_{res} = |I_{w} / J_{sed}|$$  \hspace{1cm} (8)

are short, often less than a day (range ~ 0.1 to 4.2 days) in keeping with the rapid movement of particles in and out of the benthic system. Calculated mass deposition and erosion rates vary dramatically from day to day, from a depositional input of 75 g m$^{-2}$ d$^{-1}$ to an erosional loss of 90 g m$^{-2}$ d$^{-1}$. The sum of this very active particle movement to and from the sediment over the eight days, however, is close to zero (0.84 g m$^{-2}$ d$^{-1}$), i.e. no net sediment accumulation or loss, despite fluxes varying by two orders of magnitude in both directions. This result is consistent with the general observation and hypothesis that the nearshore benthic system provides a temporary repository for readily suspended particles, and that these particles are the source of turbidity observed in the frequently occurring, episodic coastal plumes in Lake Michigan.

5 CONCLUSIONS

An ROV deployed suction sampler is shown to be an effective tool for the quantitative recovery of sediments temporarily residing on non-depositional, hard clay and sandy bottoms, areas where conventional sampling gear is unsuitable. This sampling methodology is applied to the determination of inventories of short-lived radionuclides and other constituents that reside within this actively transported pool of materials. Th-234 is shown to be a useful tracer for tracking coastal resuspension and particle transport dynamics on the time scale of days in freshwater systems like the Laurentian Great Lakes.

ACKNOWLEDGEMENTS

This work has been supported by grants from the National Science Foundation (OCE 97-9727151) Coastal Ocean Process (CoOP) program; the NOAA National Undersea Research Program; the University of Wisconsin Sea Grant Institute, the NOAA National Sea Grant College Program, and the Department of Commerce (NA46R60481-R/GB-37); the State of Wisconsin; and the Great Lakes WATER Institute of the University of Wisconsin-Milwaukee. Special thanks to Paul Goudy for suggestions as to the design, to Greg Barske for fabrication and design, and to the crew of the R/V Neeskay and Dave Lovalvo of Eastern Oceanics, Inc. Great Lakes WATER Institute Contribution Number 438.
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