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Medically-Derived ¹³¹I as a Tool for Investigating the Fate of Wastewater Nitrogen in Aquatic Environments

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ABSTRACT: Medically derived ¹³¹I ($t_{1/2} = 8.04$ d) is discharged from water pollution control plants (WPCPs) in sewage effluent. Iodine's nutrient-like behavior and the source-specificity of ¹³¹I make this radionuclide a potentially valuable tracer in wastewater nitrogen studies. Iodine-131 was measured in Potomac River water and sediments in the vicinity of the Blue Plains WPCP, Washington, DC, USA. Dissolved ¹³¹I showed a strong, positive correlation with δ^{15} N values of nitrate (δ^{15} NO₃⁻) in the river, the latter being a traditional indicator of nutrient inputs and recycling. Surface water δ^{15} NO₃⁻ values ranged from 8.7 to 33.4‰; NO₃⁻ + NO₂⁻ concentrations were 0.39–2.79 mg N L⁻¹ (26–186 μ M). Sediment profiles of particulate ¹³¹I and δ^{15} N indicate rapid mixing or sedimentation and in many cases remineralization of a heavy nitrogen source consistent with wastewater nitrogen. Values of δ^{15} N in sediments ranged from 4.7 to 9.3‰. This work introduces ¹³¹I as a tool to investigate the short-



term fate of wastewater nitrogen in the Potomac River and demonstrates the general utility of ¹³¹I in aquatic research.

1. INTRODUCTION

Iodine-131 ($t_{1/2} = 8.04$ d) is widely used in nuclear medicine, primarily to treat hyperthyroidism and thyroid cancer, with an annual average of 188 treatments per million people in developed countries. The administered dose of ¹³¹I (~0.1–1 and 4–8 Gbq for hyperthyroidism and thyroid cancer, respectively) is metabolized by patients and eliminated from the body mostly in urine.^{1,2} In the United States, patient excreta are exempt from sewer discharge regulations and are released directly into sewerage.³

Several studies have shown that >75% of ¹³¹I entering a water pollution control plant (WPCP) leaves the plant in sewage effluent;^{4–12} the rest is presumably lost to sewage sludge or radioactive decay. Flow recycling (activated sludge) commonly used to maintain bacterial populations required for sewage treatment can integrate pulses of ¹³¹I entering a WPCP over many days and result in a relatively continuous discharge of ¹³¹I at varying concentrations to the environment in sewage effluent.¹³ It is not surprising then that medically derived ¹³¹I has been measured in surface waters, macroalgae, and sediments in multiple locations (ref 14 and references therein).

Distributions of naturally occurring iodine in aquatic environments have been well-studied and show that iodine exhibits nutrient-like behavior. In the water column, it is cycled among three major species: iodate (IO_3^-) , iodide (I^-) , and dissolved organic iodine (DOI).^{15,16} Most work supports dissimilatory and assimilatory biological reactions as the primary mechanisms for these transformations.^{17–19} Iodine is

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also incorporated into biogenic particulate matter and deposited in sediments where it is subject to diagenetic reactions. Solid-phase iodine generally shows decreasing concentrations with depth and preferential loss with respect to carbon in sediments. Experimental evidence indicates that iodine is released to pore water during organic matter decomposition.²⁰

Iodine-131 discharged to the Potomac River via sewage effluent in the vicinity of the Blue Plains WPCP in Washington, DC, behaves like its stable analog. It is incorporated into particulate organic matter, deposited in sediments, and remineralized during diagenesis; these reactions are rapid with respect to the 8-day half-life of ¹³¹I.

Nitrogen isotopes have been used extensively to study the fate and transport of wastewater in aquatic ecosystems. More specifically, many studies have used the distinct nitrogen isotopic signature of treated sewage from WPCPs (enriched in ¹⁵N relative to ambient water and sediment) to estimate the fraction of wastewater-derived nitrogen in food webs and subsequently assess the influence of sewage nutrients on eutrophication.²¹ There has been particular interest in the impacts of wastewater-derived nutrient inputs on water quality in the Chesapeake Bay and its tributaries. The Potomac River is

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Figure 1. Location of Potomac River and boundaries of the tidal river zone (inset). Sampling area in the tidal Potomac River showing locations of Blue Plains, Alexandria, and Arlington WPCPs (\times) and station locations (\bullet).

the second largest tributary of the Chesapeake Bay and receives sewage effluent from the Blue Plains WPCP, the largest nitrogen point source to the Bay.²² Improved sewage treatment has decreased total nitrogen discharges to the Potomac River²³ but there is continued interest in understanding the fate of sewage nitrogen discharged from the Blue Plains WPCP.

This work examined the relationship between ¹³¹I and nitrogen and the potential utility of $^{131}\ensuremath{I}$ as a tool to study the fate of wastewater nitrogen in the Potomac River in the vicinity of the Blue Plains WPCP. Potomac River surface water and sediment samples were collected on five separate days between April 2009 and April 2010. Surface water ¹³¹I concentrations measured in the Potomac River were <MDA to 3.77 ± 0.05 Bq L^{-1} , with the highest concentrations in samples collected from the Blue Plains outfall. Solid-phase surface sediment (0-1 cm)concentrations ranged from -MDA to 117 \pm 2 Bq kg⁻¹ dry weight. The highest values were measured in surface sediments and decreased with depth up to 5 cm. Specific activities of ¹³¹I in suspended solids and surface sediments suggested preferential association of this radionuclide with particulate organic matter, a behavior consistent with naturally occurring iodine. Solid-phase sediment profiles of ²¹⁰Pb ($t_{1/2} = 22.3$ y) and ⁷Be $(t_{1/2} = 53.2 \text{ d})$ indicated that surface sediments ($\leq 10 \text{ cm}$) in the sampling area are buried or replaced at least seasonally and cannot be used to characterize short-term deposition and transport in this system. However, solid-phase sediment ¹³¹I profiles suggested steady-state conditions on fortnightly to monthly time scales making this radionuclide useful to characterize processes occurring in this time frame.¹⁴

During three of the sampling events (April, June, and November 2009), surface water samples were collected for determination dissolved nitrate isotopic composition ($\delta^{15}NO_3^{-}$). Here we present a subset of the surface water ¹³¹I data from Rose et al.¹⁴ with the $\delta^{15}NO_3^{-}$ values. The solid-phase sediment ¹³¹I profiles from Rose et al.¹⁴ were used to

calculate the flux of ¹³¹I to the sediments and subsequently characterize reactive nitrogen deposition; isotopic and mass balances of sediment nitrogen estimated the fraction of wastewater nitrogen remineralized in sediments.

2. STUDY AREA

The Potomac River is a tributary of the Chesapeake Bay that discharges to the Atlantic Ocean. This study focused on a freshwater region of the tidal Potomac River from Reagan National Airport to the Woodrow Wilson Bridge (Figure 1). The mean tidal range in this area is ~0.9 m, and the average depth is ~3 m, with a narrow channel ranging in depth from 7 to 21 m.^{24–26} Maximum flood and ebb current velocities are ~35 cm s⁻¹ at Alexandria, VA.²⁷

There are three WPCPs discharging to this area: Blue Plains, Arlington, and Alexandria. Outside of this sampling area, the nearest WPCP discharging to the river is >11km away. The average flows at the Arlington (Arlington County) and Alexandria (Alexandria Sanitation Authority) WPCPs are $\sim 113 \times 10^6$ liters per day (MLD) and 135 MLD, respectively. Sewage effluent concentrations of ¹³¹I at each of the three plants showed that the Blue Plains WPCP (DC Water) is the predominant source of ¹³¹I to the sampling area. The Blue Plains WPCP is a tertiary treatment facility located in Washington, DC. The plant serves all of Washington, DC, and parts of Loudoun and Fairfax Counties in Virginia as well as parts of Montgomery and Prince George's Counties in Maryland. The average flow to the plant is \sim 1400 MLD. Sewage effluent is discharged to the Potomac River via two outfalls (001 and 002). Outfall 002 is the primary outfall for the plant and was the discharge point sampled in this investigation. A portion of the system is serviced by combined sewers (sanitary + storm sewers); outfall 001 is used only during times of bypass overflows and was not sampled in this investigation.¹ Blue Plains discharges $\sim 1.9 \times 10^6$ kg of nitrogen annually,²⁸

predominantly as nitrate (NO_3^-) , with lesser amounts of dissolved organic nitrogen (DON; DC Water, personal communication, 2011).

Water depths in this portion of the river are generally 2-3 m at mean lower low water (MLLW), and there is a channel running through the area that reaches depths of 10 m MLLW. Water and sediment samples were collected at four shallow water stations (2-3 m MLLW). Water samples were also collected from Blue Plains outfall 002. The Blue Plains outfall 002 was sampled at or near low water, and the outfall was clearly visible. Station 2 was within ~50 m of the visible outfall.¹⁴ Figure 1 shows the general sampling area, station locations, and locations of the WPCPs.

3. METHODS

3.1. Surface Water Nitrogen. Surface water samples for determination of dissolved inorganic nitrogen $(NO_3^- + NO_2^-)$ and NH4+) concentrations and for isotopic composition of dissolved nitrate ($\delta^{15}NO_3^{-}$) were collected in June, August, and November 2009 from stations 1-4 and Blue Plains outfall 002. Briefly, water samples for nutrient analyses were collected in HCl-cleaned, low-density polyethylene (LDPE) bottles. The samples were vacuum filtered through 0.7 μ m glass fiber filters. An aliquot of the filtrate was frozen for nutrient analysis. Dissolved inorganic nitrogen $(NO_3^- + NO_2^- \text{ and } NH_4^+)$ concentrations were determined by the Analytical Laboratory, School of Marine and Atmospheric Sciences, Stony Brook University, using a Lachat, Quickchem 8000 flow injection automated nutrient analyzer. Samples were run in triplicate and checked against a blank value determined using high resistance reverse osmosis. All results were corrected for the blank. Precision of the measurements was $\pm 5\%$.

Surface water was collected for isotopic composition of dissolved nitrate ($\delta^{15}NO_3^{-}$) in HCl-cleaned LDPE bottles and frozen after collection. Prior to analysis the samples were vacuum filtered through 0.7 μ m glass fiber filters and acidified with H₂SO₄. The isotopic composition of dissolved nitrate ($\delta^{15}NO_3^{-}$) was analyzed in triplicate at the Stable Isotope Laboratory at Boston University using a modified ammonia diffusion method described by Sigman et al.²⁹

3.2. Sediment. Sediment cores were collected from each of the four stations as described in detail by Rose et al.¹⁴ Cores were visually examined for quality and sectioned at 1 cm intervals to 10 cm, and approximately five cores were composited at each site. Sediments were dried at 80 °C and homogenized using a mortar and pestle. Concentration and stable isotopic composition of carbon and nitrogen in the sediments were determined using a Costech elemental analyzer in-line with a Thermo Delta Plus XP isotope ratio mass spectrometer and a Conflo III interface. Dry, homogenized sediment were analyzed in triplicate. Total nitrogen values were determined for the samples without further treatment. A concentration calibration curve was generated for each run using an acetanilide standard (Costech). Isotopic calibration curves were generated for δ^{15} N with isotopic standards: IAEA N2, IAEA NO3, USGS 25, and USGS 40. Check standards (acetanilide, USGS 40, and IAEA C8) were analyzed throughout each run. Nitrogen concentrations of the check standards were ≤6% of actual values. Check standards were within 0.2% of actual values for δ^{15} N. All stable isotope ratios are reported in units of per mil (%) using the standard notation

$$\delta^{15} \mathrm{N} = \left[\frac{R_{\mathrm{sample}}}{R_{\mathrm{standard}}} - 1\right] \times 1000 \tag{1}$$

where $R = {}^{15}\text{N}/{}^{14}\text{N}$ of the reference standard Vienna Pee Dee Belemnite. The standard deviation of nitrogen elemental and isotopic composition for replicate analyses were $\leq 0.02\%$ and $\leq 0.6\%$, respectively.

3.3. Gamma-Ray Spectrometry. Determination of ¹³¹I in the water and sediment samples by γ -ray spectrometry using the emission at 364.5 keV is described in detail by Rose et al.¹⁴ The limit of detection for ¹³¹I was ≤ 0.03 Bq L⁻¹ for surface water and ≤ 3.6 Bq kg⁻¹ for sediments. All activities are decay-corrected to time of collection and reported $\pm 1\sigma$ counting error. Specific activities for solids are reported in units of dry mass.

4. RESULTS

Nutrient concentrations in surface water showed little variation among the sampling events and no seasonal trend. NO₃⁻ + NO₂⁻ ranged from 0.39 ± 0.02 to 2.79 ± 0.13 mg N L⁻¹ (26 ± 1 to 186 ± 9 μ M). Surface water δ^{15} NO₃⁻ values ranged from 8.7 ± 0.3 to 33.4 ± 7.3% (Table 1).

Table 1. Nitrate + Nitrite Concentrations and Stable Isotopic Composition of Nitrate ($\delta^{15}NO_3^{-}$) in Surface Waters Collected from Stations 1–4 and Blue Plains Outfall 002 in the Potomac River

station no.	$NO_3^- + NO_2^- (mg N L^{-1})$	$\delta^{15} \text{NO}_3^{-}$ (%)				
June 2009						
1	1.06	8.7 ± 0.3				
2	1.26	18.7 ± 4.8				
3	1.07	11.6 ± 0.6				
4	0.97	16.3 ± 2.1				
outfall	2.79	27.1 ± 0.2				
August 2009						
1	0.76	15.5 ± 0.1				
2	1.10	26.3 ± 3.0				
3	0.95	24.5 ± 0.5				
4	0.92	25.4 ± 2.7				
outfall	1.11	27.7 ± 2.3				
November 2009						
1	1.06	9.8 ± 1.2				
2	0.50	33.4 ± 7.3				
3	1.37	15.9 ± 2.8				
4	1.20	17.7 ± 2.9				
outfall	0.39	31.6 ± 6.4				

Solid-phase sediment nitrogen concentrations were generally higher in surface sediments and ranged from 0.094 \pm 0.002 to 0.392 \pm 0.009%; lower at station 2 (~0.1%) compared to the other stations (~0.3%). δ^{15} N values were 2.76 \pm 0.14 to 9.31 \pm 0.19%. Station 2 exhibited the largest gradients in both (Figure 2).

Surface water concentrations and sediment profiles presented in Rose et al.¹⁴ showed the highest ¹³¹I concentrations nearest the outfall and decreasing with distance away from it. Upstream station 1 had the lowest sediment and surface water concentrations. Surface water ¹³¹I concentrations ranged from below detection to 0.68 \pm 0.02 Bq L⁻¹ in June, August, and November 2009. The highest concentrations were measured in samples collected from Blue Plains outfall 002. Low TSS and chlorophyll a concentrations in the outfall samples as well visual



Figure 2. Depth profiles of percent total nitrogen N_T (\bullet) and $\delta^{15}N_T$ (O) in Potomac River sediments collected from stations 1–4.

observations at time of sampling strongly suggested that these samples represent sewage effluent from Blue Plains with little dilution. $^{14}\,$

Iodine-131 was detected in sediments to 5 cm. The concentrations ranged from 2.8 \pm 0.3 to 80.0 \pm 0.3 Bq kg⁻¹ dry weight, with the highest values in surface sediments decreasing with depth. At station 1, ¹³¹I was restricted to the top 1–2 cm with lower concentrations than surface sediments at the other stations.¹⁴

5. DISCUSSION

5.1. lodine-131 and Nitrogen in Surface Water. The $\delta^{15}NO_3^{-}$ values in surface water collected from the Blue Plains outfall (mean = 29%, Table 1) were distinctly heavier

(enriched in ¹⁵N) than the mean $\delta^{15}NO_3^-$ value determined for the Potomac River watershed (7% $_{o}$).³⁰ Degradation of organic matter during sewage treatment and loss of isotopically light, gaseous nitrogen products causes sewage effluent to be enriched in ¹⁵N.^{31–34} Return flows, or activated sludge returns, used to maintain bacterial populations within a sewage treatment plant can further enrich the nitrogen pool. Sewage effluent $\delta^{15}NO_3^-$ values as high as 40% $_o$ have been measured in other WPCPs.³⁵

The strong positive correlation between $\delta^{15}NO_3^-$ values and ^{131}I concentrations in surface water collected at the sampling sites (Figure 3) can be explained by the flow pattern of Blue Plains' sewage effluent in the river. Hearn³⁶ determined that sewage effluent discharged from Blue Plains is mostly confined

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Figure 3. Iodine-131 versus $\delta^{15}NO_3^{-}$ in surface water collected from stations 1–4 and the Blue Plains outfall 002 in the Potomac River in June, August, and November 2009. Data points for station 1 and the outfall are shown in shaded areas. The relatively large $\delta^{15}NO_3^{-}$ errors (up to 6%c) in some cases may be attributed to high DON concentrations.

to the shallow regions of the river south of the outfall (stations 3 and 4). The lower $\delta^{15}NO_3^{-1}$ values and ^{131}I concentrations in surface water at station 1 (mean = 11%) relative to the outfall reflect a decreased wastewater influence at this site (Figure 3). The distributions of ¹³¹I in these sediments showed a similar pattern.¹⁴ The linear relationship ($r^2 = 0.874$) between ¹³¹I concentrations and $\delta^{15}NO_3^{-}$ values among the four stations implies that mixing time is short relative to the half-life of ¹³¹I. Deviations from the linear mixing line may be explained by changes in river flow, fluctuations in ¹³¹I inputs, incorporation of dissolved ¹³¹I into particulate phases and diagenetic release of ¹³¹I from sediments, which has been observed for naturally occurring iodine.^{37,38} Further, nonlinearity may be an artifact of sampling the stations at different points in the tidal cycle on a given day. Errors in the $\delta^{15} NO_3^{-1}$ values (up to 6 % in some cases) may be associated with the presence of DON and the interference of this species during nitrate extraction for isotopic analysis.²⁹ Despite these sources of variation, the linear relationship between $\delta^{15}NO_3^{-}$ and ^{131}I is strong and identified the $\delta^{15}NO_3^{-}$ signature of Blue Plains sewage effluent (mean = 29%); this value was used as the sewage end-member in isotopic and mass balance calculations presented in section 5.3. 5.2. Iodine-131 and Nitrogen in Sediments. While the

²¹⁰Pb and ⁷Be profiles demonstrated that the sedimentary deposits at these sites are not in steady state over longer time scales, sediment inventories of ¹³¹I suggested approximate steady-state conditions for this radionuclide on monthly time scales.¹⁴ With the steady-state assumption, the ¹³¹I flux to the sediments was calculated for each profile using the following equation,

$$J = I\lambda \tag{2}$$

where J is the flux to sediments (Bq cm⁻² d⁻¹), I is the inventory of radioisotope (Bq cm⁻²), and λ is the decay constant of radioisotope (d⁻¹). The average ¹³¹I flux to the sediments in June, August, and November 2009 was 2.1 × 10⁻³ Bq cm⁻² d⁻¹ (7.7 × 10³ Bq m⁻² y⁻¹). Using surface sediment (0–1 cm) ratios of nitrogen to ¹³¹I (mol Bq⁻¹), the average flux of nitrogen to the sediments was 9.1 × 10⁻⁶ mol N cm⁻² d⁻¹

(0.47 kg N $m^{-2}\ y^{-1};$ Table 2). Shultz^{26} estimated the flux of nitrogen to the sediments in a larger region of the Potomac

Table 2. Iodine-131 Flux to Sediments at Stations 1-4 in the
Potomac River as Determined from Solid-Phase Sediment
Profiles Presented by Rose et al. ¹⁴

station no.	131 I flux (Bq cm ⁻² d ⁻¹)	$\frac{\mathrm{N}:^{131}\mathrm{I}}{(\mathrm{mol}\ \mathrm{Bq}^{-1})^a}$	$\frac{\text{N flux}}{(\times 10^{-6} \text{ mol N cm}^{-2} \text{ d}^{-1})^{b}}$		
		June 2009			
1	0.15	0.0470	6.9		
2	1.6	0.0053	8.5		
3	1.8	0.0047	8.6		
4	1.6	0.0067	11		
		August 2009			
1	0.10	0.0491	4.8		
2	4.7	0.0019	8.9		
3	2.5	0.0051	13		
4	2.1	0.0047	9.7		
November 2009					
1	0.36	0.0228	8.3		
2	5.4	0.0025	13		
3	2.8	0.0029	8.0		
4	1.6	0.0054	8.8		

^{*a*}Ratio of total nitrogen to ¹³¹I in the top 1 cm of the sediment at the same locations. ^{*b*}Nitrogen flux to sediments at the same stations calculated from the ¹³¹I flux to the sediments.

Table 3. Estimates of Nitrogen Flux to Sediments in the Potomac River

study region	surface area (×10 ⁶ m ²)	estimated N flux to sediments (kg N m ⁻² y ⁻¹)	ref
National Airport – Woodrow Wilson Bridge	4.5	0.47	this study
Woodrow Wilson Bridge – Hallowing Point, VA	54.7	0.12-0.18	26
Lower Potomac River	722	0.013	39

River $(55 \times 10^6 \text{ m}^2)$ that is located south of the sampling area in the present study (Table 3). The estimate reported in Shultz²⁶ was 0.12–0.18 kg N m² y⁻¹. Boynton et al.³⁹ reported a burial rate of 9.32 × 10⁶ kg N y⁻¹ in sediments of the Lower Potomac River (surface area = 722 × 10⁶ m²), which translates into a flux of 0.013 kg N m² y⁻¹, substantially lower than our estimate. It is reasonable that the flux of nitrogen to the sediments is higher in the vicinity of Blue Plains relative to the Potomac River as a whole. However, direct comparison of these numbers is difficult due to the differences in spatial extent of these studies. Decreases in nitrogen loading from WPCPs in recent decades further complicate this comparison. Since the 1980s, nitrogen discharges from Blue Plains have decreased by ~70%.²⁸ Water column suspended particulate concentrations for both ¹³¹I and nitrogen would better constrain the flux estimates.

5.3. Mass and Isotopic Balances. Solid-phase sediment δ^{15} N profiles indicated a heavy nitrogen source, consistent with wastewater nitrogen (Figure 3). Isotopic fractionation during diagenesis cannot explain these observations. Such fractionation should enrich the solid-phase pool in ¹⁵N as the lighter isotope is preferentially lost during decomposition of organic matter. Furthermore, major net isotopic fractionation of carbon and nitrogen is not expected during early diagenesis of sediments.



Figure 4. $\delta^{15}N_TN_T$ versus N_T in Potomac River sediments collected from stations 1–4. The linear slopes (*m*) represent the net isotopic value of nitrogen lost (or gained) from the sediments during organic matter decomposition or transport.

Thus, sediment values largely reflect the isotopic composition of source materials. 40,41

Mass and isotopic balances suggested that the $\delta^{15}N$ profiles are explained by remineralization of wastewater nitrogen in these sediments. Assuming mixing between wastewater nitrogen (N_W) and the river end-member (no wastewater nitrogen; N_R), the mass and isotopic balances are given by

$$N_{\rm T} = N_{\rm W} + N_{\rm R} \tag{3}$$

$$\delta^{15} \mathrm{N}_{\mathrm{T}} \mathrm{N}_{\mathrm{T}} = \delta^{15} \mathrm{N}_{\mathrm{W}} \mathrm{N}_{\mathrm{W}} + \delta^{15} \mathrm{N}_{\mathrm{R}} \mathrm{N}_{\mathrm{R}} \tag{4}$$

where N_T is the sediment nitrogen concentration (%) and $\delta^{15}N$ is the stable isotopic composition of the respective components (‰). During remineralization of organic matter composed of N_W and N_R the mass and isotopic changes of nitrogen are represented by

$$\delta^{15} N_T N_T = \delta^{15} N_W N_W + \delta^{15} N_R N_R + \delta^{15} N_\Delta \Delta N_T$$
(5)

where ΔN_T is the net change in sediment nitrogen concentration and $\delta^{15}N_{\Delta}$ = the change in isotopic composition during remineralization. When $\delta^{15}N_WN_W$ (or $\delta^{15}N_RN_R$) = constant or zero and $\Delta N_T \neq 0$, eq 5 becomes

$$\frac{\mathrm{d}(\delta^{15}\mathrm{N}_{\mathrm{T}}\mathrm{N}_{\mathrm{T}})}{\mathrm{d}\mathrm{N}_{\mathrm{T}}} = \delta^{15}\mathrm{N}_{\Delta} + \frac{\mathrm{d}(\delta^{15}\mathrm{N}_{\Delta})}{\mathrm{d}\mathrm{N}_{\mathrm{T}}}\Delta\mathrm{N}_{\mathrm{T}}$$
(6)

where linear slopes of $\delta^{15}N_TN_T$ versus N_T estimate the net isotopic signal of nitrogen lost (or gained) from the sediments during organic matter decomposition or transport.^{42–47} In most cases, the plots demonstrate that the nitrogen being remineralized or added at these sites is heavy nitrogen, which is consistent with remineralization of organic matter directly derived from or recycled from a wastewater source (Figure 4).

The relative contribution of wastewater nitrogen was estimated using this two end-member approach. The sewage effluent end-member ($\delta^{15}N_W = 29\%$) was the average $\delta^{15}NO_3^-$ value measured in samples collected in the outfall (Table 1). The outfall samples were collected at low water, which nearly coincides with slack water in this region of the Potomac River. The outfall discharge was visibly distinguishable from ambient river water due to the relatively low suspended

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solids concentrations in the sewage effluent. Furthermore, low chlorophyll a concentrations measured in the oufall also indicate little mixing with ambient river water at the time of sample collection and support the use of the outfall sample as the wastewater end-member.¹⁴ The river end-member ($\delta^{15}N_R = 7\%$) represents the mean $\delta^{15}NO_3^-$ measured for the Potomac watershed.³⁰ Interpreting the mixing model slopes as representative of the weighted $\delta^{15}N$ value of remineralized substrate, then with the end-members of 7% (river) and 29% (sewage effluent) the results suggest that 9 to 59% of the nitrogen remineralized in the sediments at these stations is derived from wastewater discharged from Blue Plains. The relatively low nitrogen (~0.1–1.5%) and carbon (~2%) content¹⁴ of the sediments at station 2 relative to the other stations (~0.3\% N and 4% C) may explain the anomalous results at that station (Figure 4).

This work introduces medically derived ¹³¹I as a tool to study the short-term fate of wastewater nitrogen in this system. However, the utility of ¹³¹I is not limited to the Potomac River. Many studies have documented the presence of ¹³¹I in aquatic environments, ^{14,48–52} and continuous discharges of this radioisotope in sewage effluent are likely to be widespread in urban areas.

Solid-phase sediment profiles of 131 I provide a relatively simple means to estimate the reactive nitrogen flux to sediments on fortnightly to monthly time scales. Iodine-131 is particularly useful for assessing deposition of material to the river bed in a dynamic sedimentary environment where mass fluxes to the sediments cannot be determined using longer-lived radionuclides, such as 210 Pb and 7 Be.

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Notes

The authors declare no competing financial interest.

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