Medically-derived ^{131}I as a tracer in aquatic environments

A Dissertation Presented

by

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to

The Graduate School

in Partial Fulfillment of the

Requirements

for the Degree of

Doctor of Philosophy

in

Marine and Atmospheric Sciences

Stony Brook University

August 2011

Stony Brook University

The Graduate School

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Abstract of the Dissertation

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Marine and Atmospheric Sciences

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Medically-derived ^{131}I ($t_{½}$ = 8.04 d) has been measured in aquatic environments receiving sewage effluent discharges, yet few published data exist for the radioisotope in sewage effluent; most work has focused on sewage sludge. This work presents ^{131}I concentrations detected in sewage effluent from a small water pollution control plant (WPCP) serving a regional thyroid cancer treatment facility in Stony Brook, NY, USA. The concentrations detected in the Stony Brook WPCP ranged from 1.8 ± 0.3 to 227 ± 2 Bq L^{-1} in sewage effluent and 61 ± 12 to 2801 ± 32 Bq g^{-1} in suspended solids > 0.7 µm in the effluent. The primary source of ^{131}I is excreta from thyroid cancer inpatients treated at the Stony Brook University Medical Center (SBUMC). Time series measurements following known inputs indicated that ^{131}I is discharged for many days following an inpatient treatment. The sewage half-life, analogous to a radioactive half-life, describes the time it takes for half of a wastewater component to be removed from a WPCP. The sewage half-life of ^{131}I in this plant is 2.0 d. Due to the frequency of patients treated at the SBUMC and retention in the plant, sewage effluent discharges of ^{131}I are fairly continuous.

The behavior of medically-derived ^{131}I was investigated in the tidal Potomac River in the vicinity of Blue Plains, the world's largest advanced wastewater treatment plant. This plant serves all of Washington, DC, treats an average of 1.4 x 10^9 L d^{-1} and has a maximum capacity of > 4 x 10^9 L d^{-1}. Sewage effluent concentrations detected in samples collected from Blue Plains on different days ranged from 0.9 ± 0.1 to 8.1 ± 0.2 Bq L^{-1}. Concentrations of ^{131}I detected in sewage effluent and in the river suggest a continuous discharge of the isotope from Blue Plains. Surface water ^{131}I ranged from 0.076 ± 0.006 to 6.07 ± 0.07 Bq L^{-1}. Partitioning in sewage effluent and river water suggests that ^{131}I is associated with colloidal and particulate organic material. Iodine-131 was detected in sediments to depths of 5 cm with specific activities between 1.3 ± 0.8 and 117 ± 2 Bq kg^{-1} dry weight. The behavior of ^{131}I in the Potomac River is consistent with the cycling of natural iodine in aquatic environments. It is discharged to the river via sewage effluent, incorporated into particulate matter and deposited in sediments where it is subject to diagenetic remineralization.

Dissolved ^{131}I showed a strong, positive correlation with δ^{15}N values of nitrate in the river. Surface water $\delta^{15}NO_3^-$ values ranged from 8.7 ± 0.3 to 33.4 ± 7.3‰ with dissolved inorganic nitrogen (NO_3^- + NO_2^-) concentrations between 0.38 ± 0.02 and 2.79 ± 0.13 mg N L^{-1} (26 ± 1 and 186 ± 9 μM). δ^{15}N in sediments ranged from 4.7 ± 0.1 to 9.3 ± 0.1‰. Sediment profiles of particulate ^{131}I and δ^{15}N indicate rapid mixing or sedimentation and in many cases remineralization of a heavy nitrogen source consistent with wastewater nitrogen. Iodine-131 concentrations in sediments ranged from 2.8 ± 0.3 to 80.0 ± 0.3 Bq kg^{-1} dry weight. Values of δ^{15}N in sediments ranged from 4.7 ± 0.1‰ to 9.3 ± 0.1‰. This work introduces ^{131}I as a potentially valuable tool to study the short-term fate of wastewater nitrogen in this system, but its utility as a tracer is not limited to use in the Potomac River.

The presence of medically-derived ^{131}I has been documented in several aquatic environments. Continuous discharges of this radioisotope in sewage effluent are likely to be widespread. Further study of ^{131}I in receiving waters can provide valuable insight into the fate and transport of this radioisotope in the context of large scale accidental releases.

This work is dedicated to my Uncle Ben

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Acknowledgements

The following pages represent a significant portion of my life and something I am grateful to have had the opportunity to complete. I begin by dedicating this work to my Uncle Ben, who not only got me started but helped to keep me going all these years. I promise to enjoy this.

It is not possible to acknowledge all of the people who helped me bring this together but there are a few people that I would like to thank here. My advisors, Larry Swanson and Kirk Cochran encouraged me to continue this work after finishing my Master's degree. They stuck with me on this project for many years even though they weren't always sure what I was doing or where this was going to end up. I have learned a great deal from you and I thank you both. Thanks to my committee members, George Luther and Nick Fisher for their guidance and support. I would especially like to thank Bob Aller for his unwavering enthusiasm for this work and the direction he has given me over the years. Thank you all for your patience and encouragement.

None of the work in the Potomac River would have been possible without the support of my friends and colleagues at the Naval Research Laboratory. I owe never-ending thanks to Joe Smith for setting me up to complete the work in the Potomac River and helping me see it through. I am especially thankful to Joe Smith for being a great friend, making me laugh hard (but not snarf) and for teaching me to be froggy. I am very grateful to Tom Boyd for taking us on his fishing trips so that we could collect my samples. Rick Coffin generously provided me with the facilities and support to complete the work in the Potomac River. Thank you to Curt Millholland, Mike Montgomery and Anna Miller for their help with various aspects of this work. A special thank you to Becca Plummer for teaching me how to run the instruments I used at the Naval Research Laboratory.

David Hirschberg, Christina Heilbrun and Alisha Renfro at Stony Brook University taught me everything I know and love about γ-ray spectrometry. Without the help of Alisha Renfro and Christina Heilbrun, I would not have any of the ^{210}Pb data presented in this dissertation. I thank Jeronimo Pan for his help with the chlorophyll analyses and Darcy Lonsdale for the use of her lab. Thank you to Kamazima Lwiza for his friendship and teaching me how to make the maps contained in this document. I must acknowledge all that I learned from Mark Wiggins working with him on other projects.

Joe Daley at Stony Brook University, Environmental Health and Safety provided me with years of patient treatment data from the Stony Brook University Medical Center and answered countless questions about the medical use of ^{131}I. Thank you to Curtis Olsen at the University of Massachusetts, Boston for the use of his XRF. Milton Cruz and Gene Brewer at Suffolk County Department of Public Works patiently answered all of my questions about the Stony Brook Water Pollution Control Plant and provided me with data from the plant.

I would like to recognize the support of this work by DC Water through their internship program. Thank you to Sudhir Murthy, Elaine Wilson, Rudy Rimando at the Blue Plains Water Pollution Control Plant for their help with questions and sampling. Thanks to Phil Loar and Larry Slattery at the Arlington Water Pollution Control Plant and Leulu Gebremedhin at Alexandria Sanitation Authority for their help collecting samples.

My friends Zosia Baumann, Alyssa Hamann, Christine O'Connell and George Rieu-Sicart were generous with their love, support and laughter through some very difficult times. A very special acknowledgement to Bonnie Stephens whose friendship has made me a better person and whose editing has made me a better writer. My dear friend Karen Puiu recently reminded me that good friends aren't always near, but they are there when you need them most. Finally, Jodie Lumbrazo has often been the voice in my head these past 20 years helping me to pick myself up, brush myself off and start all over again.

"It's true; life really is generous to those who pursue their Personal Legend…"

~Paolo Coehlo, The Alchemist

Chapter 1

Introduction

Iodine-131 ($t_{½}$ = 8.04 d) is a fission product released from nuclear power plants, during nuclear weapons tests, nuclear fuel reprocessing and weapons production. Medical use is perhaps the more widespread source of ^{131}I to the environment. Iodine-131 has been administered to patients for treatment of thyroid disorders since the 1940s (Chapman and Evans, 1946; Hertz and Roberts, 1946; ICRP, 2004; Mattsson et al., 2006; Rasmuson, 2006). Today it is the most widely used radiopharmaceutical in nuclear medicine for therapeutic purposes, commonly used to treat hyperthyroidism and thyroid cancer. Both treatments utilize unsealed radioactive sources metabolized by the patient and therefore render the patient and his excreta radioactive. The number of diagnostic procedures using ^{131}I far exceeds the number of therapeutic treatments; however, the latter represents a greater potential source to sewerage. The standard protocol for treating thyroid cancer is removal of the whole thyroid gland followed by administration of ^{131}I to destroy any remaining cells. Thyroid cancer patients are typically given 4000 to 8000 MBq compared to 100 to 1000 MBq of ^{131}I for hyperthyroid treatments and most of the initial dose is eliminated from the body in urine (ICRP, 2004) In the United States, patient excreta are exempt from sewer regulations and are therefore released into sewerage (Martin and Fenner, 1997).

The discharge of ^{131}I from water pollution control plants (WPCPs) to the environment has long been recognized (Moss, 1973). Many studies have documented the presence of ^{131}I in sewage sludge (Barci-Funel et al., 1993; Dalmasso et al., 1997; Erlandsson et al., 1989; Erlandsson et al., 1983; Erlandsson and Mattsson, 1978; Martin and Fenner, 1997; Prichard et al., 1981; Puhakainen, 1998; Rose, 2003; Stetar et al., 1993). However, few values of ^{131}I in sewage effluent have been reported (Erlandsson et al., 1989; Fischer et al., 2009; Kleinschmidt, 2009; Puhakainen, 1998; Rose, 2003; Smith et al., 2008; Sodd et al., 1975). The results of several investigations indicate greater than 75% of ^{131}I entering WPCPs, leaves in the effluent (Barci-Funel et al., 1993; Dalmasso et al., 1997; Erlandsson et al., 1989; Erlandsson et al., 1983; Erlandsson and Mattsson, 1978; Martin and Fenner, 1997; Prichard et al., 1981; Puhakainen, 1998; Stetar et al., 1993). It is not surprising then that ^{131}I has been measured in various environments: surface waters (Howe and Lloyd, 1986; Smith et al., 2008; Sodd et al., 1975), macroalgae (Howe and Hunt, 1984; Howe and Lloyd, 1986; Marsh et al., 1988; Puhakainen, 1998; Rose, 2003; Waller and Cole, 1999) and sediments (Fischer et al., 2009; Smith et al., 2008). Furthermore, ^{131}I was measured in the atmosphere near municipal sewage incinerators (Kitto et al., 2005a; Kitto et al., 2006; Kitto et al., 2005b). While the occurrence and concentrations of ^{131}I in sewage effluent have been poorly characterized, these studies suggest continuous discharges of the radioisotope may be common and therefore useful as a tracer in receiving waters. Recently, Smith et al. (2008) proposed ^{131}I as a wastewater-specific particle tracer and suggested its use as a tracer for short-term sediment dynamics.

Iodine-129, the longest lived radioisotope of iodine with a half-life of 1.7 x 10^7y, has been used as a tracer of nuclear fuel reprocessing discharges (Keogh et al., 2007) and to study iodine geochemistry and terrestrial organic matter cycling in coastal environments (Oktay et al., 2001; Santschi and Schwehr, 2004; Schwehr et al., 2005). However, the extent to which isotopes of iodine can be used as tracers is currently limited by our knowledge of its geochemistry.

Iodine is a redox-sensitive element that can exist in several oxidation states at the Earth's surface. Iodate (IO_3^-) is the thermodynamically favored species of iodine in oxygenated water (Sillen, 1961), however iodide (I^-) is found ubiquitously. Non-volatile dissolved organic iodine (DOI) has been found to constitute between 9 and 85% of the total iodine pool in freshwater, estuarine and coastal waters (Abdel-Moati, 1999; Gilfedder et al., 2009; Gilfedder et al., 2010; Luther et al., 1991; Oktay et al., 2001; Truesdale, 1975; Wong and Cheng, 1998; Wong and Cheng, 2001), but does not appear to have significant concentrations in the open oceans. Wong et al. (2004) suggested that rivers may be a source of DOI to the ocean.

Freshwater concentrations of total iodine range from 0.004 to > 1 μM, but are generally < 0.1 μM (Fuge and Johnson, 1986; Gilfedder et al., 2009; Moran et al., 2002; Neal et al., 2007; Vought et al., 1970; Whitehead, 1984). Total iodine concentrations in seawater are typically 0.45 μM, with much less variability than observed in freshwater. In fact, much of what is known about the aquatic geochemistry is derived from marine and estuarine studies, where iodine has been well studied.

Iodine shows slight depletions in surface waters of the open ocean with increasing depletions shoreward (Truesdale, 1994; Truesdale and Upstill-Goddard, 2003; Wong, 1991; Wong, 1995; Wong and Zhang, 1992). Generally, I^- concentrations increase in areas of higher primary productivity (Campos et al., 1999; Elderfield and Truesdale, 1980; Truesdale, 1994; Truesdale et al., 2000; Truesdale and Upstill-Goddard, 2003; Tsunogai and Henmi, 1971; Waite et al., 2006; Wong, 1995; Wong and Zhang, 1992). Reduced forms of iodine have been shown to dominate estuarine systems at lower salinities (Abdel-Moati, 1999; Luther and Cole, 1988; Luther et al., 1991; Smith and Butler, 1979; Ullman et al., 1988; Zic and Branica, 2006). Most studies have demonstrated that transformations among iodine species occur within estuaries (Beck and Bruland, 2000; Butler and Smith, 1985; Cook et al., 2000; Edwards and Truesdale, 1997; Luther and Cole, 1988; Luther et al., 1991; Wong and Zhang, 2003b; Zic and Branica, 2006).

Conservative behavior during estuarine mixing has been observed for total iodine (Luther and Cole, 1988; Luther et al., 1991; Takayanagi and Cossa, 1985) and for inorganic iodine (Smith and Butler, 1979; Zic and Branica, 2006). Iodine enrichments observed in some estuaries may be due to sediment - water interactions where iodine is released from the sediments under anoxic conditions (Abdel-Moati, 1999; Butler and Smith, 1985; Edwards and Truesdale, 1997; Luther and Cole, 1988; Ullman et al., 1988; Wong and Zhang, 2003b).

Other sources of iodine may need to be considered in estuarine, coastal and freshwater systems. Sewage effluent discharges may be responsible for enrichments. Tap water iodine in excess of typical seawater concentrations has been measured (Andersen et al., 2002; Pedersen et al., 1999; Richardson et al., 2008). X-ray contrast agents may also contribute to total iodine concentrations in sewage effluent (Drewes et al., 2001). Vought (1970) measured higher concentrations of iodine in the Potomac River downstream of a WPCP, relative to upstream. Additionally, agricultural runoff may contribute to the total iodine pool in these environments (Moran et al., 2002; Whitehead, 1979; Whitehead, 1984). Soil and minerals have a low iodine content but in some areas weathering can contribute to iodine content of lakes and rivers (Fuge and Johnson, 1986; Whitehead, 1984).

The presence of I^- in aquatic environments may be explained by the prevailing redox state under low oxygen (Chapman, 1983; Farrenkopf et al., 1997b; Rue et al., 1997; Truesdale and Bailey, 2000) and anoxic conditions (Emerson et al., 1979; Luther and Campbell, 1991; Ullman et al., 1990; Wong and Brewer, 1977; Wong et al., 1985) and by interaction with sediments in coastal systems. The chemical oxidation of I^- in oxygenated aquatic environments is not thermodynamically favorable. Although photochemical and biological reactions can oxidize I^-, the reactive intermediates such as hypoiodous acid (HOI) formed in these reactions react with organic matter to re-form I^-. Therefore, I^- can persist under oxygenated conditions (Kupper et al., 2008; Luther, 2011; Luther and Cole, 1988; Luther et al., 1995).

Distributions of iodine support biological cycling as the primary mechanism for transformations among its species. However, previous investigations have not led to a definitive explanation for the role of organisms in the cycling of iodine in aquatic environments. Early work suggested that the enzyme nitrate reductase, found in both aerobic and anaerobic organisms, may be responsible for IO_3^- reduction in the surface oceans (Tsunogai and Sase, 1969). Indeed, it was established that the enzyme extracted from phytoplankton was able to reduce IO_3^- to I^- (Hung et al., 2005). Field data indicated a possible relationship between nitrate reductase and IO_3^- reduction (Campos et al., 1999; Wong and Hung, 2001). However, Waite and Truesdale (2003) determined that while nitrate reductase can reduce IO_3^-, it is not necessary for IO_3^- reduction in phytoplankton cultures.

The relationship between primary production and iodine cycling has been investigated in both laboratory and field studies. The experimental results are quite variable and lead to no consensus on the role of phytoplankton in the cycling of iodine (Bluhm et al., 2010; Butler et al., 1981; de la Cuesta and Manley, 2009). Several studies have demonstrated that phytoplankton are able to reduce natural concentrations of IO_3^- to I^- (Chance et al., 2007; Moisan et al., 1994; Waite and Truesdale, 2003; Wong et al., 2002) and this reaction has been inferred from indirect measurements (Campos et al., 1999; Hung et al., 2005; Wong and Hung, 2001). The question remains whether measured rates IO_3^- reduction are fast enough to sustain the concentrations of I^- observed in the field (Chance et al., 2007; Waite and Truesdale, 2003; Wong et al., 2002).

Some work has shown correlations between iodine and nutrient concentrations throughout the world oceans (Elderfield and Truesdale, 1980; McTaggart et al., 1994; Truesdale et al., 2000; Wong and Brewer, 1974). However, a general lack of evidence for seasonal variations among the iodine species further complicates the link to a biological mechanism for its cycling (Farrenkopf and Luther, 2002; Jickells et al., 1988; Tian et al., 1996; Truesdale, 1978; Truesdale and Bailey, 2002; Truesdale et al., 2003; Truesdale and Jones, 2000; Waite et al., 2006; Wong and Zhang, 2003a) . Wong (2001) argues that the accumulation of I^- takes weeks to months, whereas the phytoplankton biomass turnover time is hours to days. Therefore, the processes should not be expected to be in phase.

Respiration, not photosynthesis, has been used to explain iodine distributions in some studies (De Luca Rebello et al., 1990; Tian and Nicolas, 1995). The participation of bacteria in the cycling of iodine has been investigated. Bacteria have been shown to reduce IO_3^- in the laboratory (Amachi et al., 2007; Councell et al., 1997; Farrenkopf et al., 1997a). It has been proposed that the use of IO_3^- as an electron acceptor for the oxidation of organic matter may be as important as NO_3^- reduction under low oxygen conditions since the free energy yields for the reactions are similar (Farrenkopf et al., 1997b). Marine bacteria can produce volatile inorganic and organic iodine (Amachi et al., 2001; Amachi et al., 2004; Amachi et al., 2005; Fuse et al., 2003; Gozlan and Margalith, 1973). These volatile products are not likely to represent a significant loss to the atmosphere. Surface depletions of total iodine in the surface oceans are small (Campos et al., 1999; Elderfield and Truesdale, 1980; Truesdale et al., 2000; Waite et al., 2006). While it has been suggested that the greatest exchange of iodine among its reservoirs occurs between the atmosphere and hydrosphere, the calculated exchanges are 5×10^{11}g y^{-1}, or greater as proposed by Garland and Curtis (1981), and are small with respect to the hydrosphere's reservoir 7.1×10^{16} g (Fuge and Johnson, 1986).

Seaweeds also produce volatile organic iodine compounds (Giese et al., 1999) and are known to concentrate iodine with some species accumulating nearly 5% iodine dry weight (Kupper et al., 1998). The IAEA (2004) reported an iodine concentration factor of 1×10^4 for macroalgae. Concentrations in seaweeds have been found as high as 100,000 times that in seawater (Martinelango et al., 2006). Kupper et al. (2008; 1998) proposed a concentrating mechanism for iodine in seaweeds whereby I^- in seawater is first oxidized to hypoiodous acid (HIO) and I_2. The oxidized iodine may then enter the cell where it is reduced to I^- or incorporated into organic compounds. Recent work by Truesdale (2008) indicates that seaweeds may take up both I^- and IO_3^-. Furthermore, *Fucus serratus* was found to release IO_3^-. Truesdale suggested that seaweeds may be a possible mechanism for IO_3^- oxidation in the coastal zone.

Iodine's association with particulate organic matter is well established (Alvarado-Quiroz et al., 2002; Bojanowski and Paslawska, 1970; Bors et al., 1991; Brewer et al., 1980; Calvert et al., 1993; Francois, 1987; Harvey, 1980; Lieser and Steinkopff, 1989; Malcolm and Price, 1984; Pedersen and Price, 1980; Price et al., 1970; Sheppard and Hawkins, 1995; Sheppard and Thibault, 1992; Shishkina and Pavlova, 1965; Ullman and Aller, 1980; Upstill-Goddard and Elderfield, 1988; Vinogradov, 1939; Whitehead, 1973a; Whitehead, 1973b; Whitehead, 1974a;

Whitehead, 1974b; Whitehead, 1981). Several authors have presented experimental evidence for biologically-mediated incorporation into particulate phases (Bors et al., 1991; Lieser and Steinkopff, 1989; Muramatsu et al., 1990a; Muramatsu et al., 1990b; Sheppard and Hawkins, 1995; Whitehead, 1974b). While the mechanisms responsible for these transformations are largely unknown, there is general agreement that iodine is incorporated into particulate matter and subsequently deposited in sediments.

Sediments in various marine environments have been found to be enriched in iodine relative to overlying seawater (Bennett and Manuel, 1968; Bojanowski and Paslawska, 1970; Kennedy and Elderfield, 1987; Mackin et al., 1988; Malcolm and Price, 1984; Martin et al., 1993; Pedersen and Price, 1980; Price and Calvert, 1973; Price and Calvert, 1977; Price et al., 1970; Shishkina and Pavlova, 1965; Ullman and Aller, 1985; Upstill-Goddard and Elderfield, 1988; Vinogradov, 1939; Wakefield and Elderfield, 1985). Generally, there is a decrease in solid phase iodine content with depth in sediment (Kennedy and Elderfield, 1987; Mackin et al., 1988; Price et al., 1970; Shishkina and Pavlova, 1965; Upstill-Goddard and Elderfield, 1988). Data indicate that pore water iodine is an organic matter decomposition product (Ullman and Aller, 1980; Ullman and Aller, 1983; Ullman and Aller, 1985; Upstill-Goddard and Elderfield, 1988). Solid phases I/C ratios in surface sediments tend to be higher than the typical marine plankton molar ratio of $\sim 1 \times 10^{-4}$ (Elderfield and Truesdale, 1980) and show higher values in oxidizing and low sulfide sediments. Decreasing I/C ratios with depth in sediments indicate a preferential loss of iodine with respect to carbon during organic matter decomposition (Pedersen and Price, 1980; Price et al., 1970; Ullman and Aller, 1983).

Iodine enrichments in surface sediments have been explained by reaction with organic matter (Francois, 1987; Kennedy and Elderfield, 1987; Price and Calvert, 1973; Price and Calvert, 1977; Wakefield and Elderfield, 1985) and by adsorption onto metal oxides near the sediment water interface (Mackin et al., 1988; Ullman and Aller, 1980; Ullman and Aller, 1985). Both mechanisms are consistent with the higher surface I/C ratios measured in oxidizing sediments than those found in reducing sediments by Price and Calvert (1973; 1977). Ullman and Aller (1985) argue that surface enrichments are due to interaction with metal oxides and that the relationship observed between organic matter and iodine in sediments (Bojanowski and Paslawska, 1970; Calvert et al., 1993; Francois, 1987; Malcolm and Price, 1984; Pedersen and Price, 1980; Price et al., 1970; Shishkina and Pavlova, 1965; Vinogradov, 1939) is consistent with their hypothesis. They explain that the decomposition of fresh organic matter generates I^- which may be rapidly oxidized to IO_3^- by bacterially-mediated reactions near the sediment-water interface. Organic matter decomposition also generates reactive metal hydroxides, which precipitate in oxic surface sediments (Ullman and Aller, 1985). Metal hydroxides and minerals have been shown to sorb iodine species (Fuhrmann et al., 1998; Muramatsu et al., 1990b; Music et al., 1980; Whitehead, 1973a; 1974b; Yoshida et al., 1992). Some work indicates that iodine adsorption to soils and mineral phases is dependent on pH (Whitehead, 1973a; 1974b; Yoshida et al., 1992), redox state (Sheppard et al., 1995), iodine speciation and mineral composition (Fox et al., 2009; Fuhrmann et al., 1998). Anschutz and coworkers (2000) presented data to support reactions between iodine species and manganese in sediments. While the initial source of iodine to sediments may be organic matter, it may not be responsible for its recycling near the sediment-water interface.

Experimental evidence indicates that iodine in sediments is primarily organic (Harvey, 1980; Upstill-Goddard and Elderfield, 1988). Results from sediment traps in the North Atlantic showed correlations between iodine and organic matter (Brewer et al., 1980). Several studies demonstrated a relationship between organic matter and iodine in surface sediments (Bojanowski and Paslawska, 1970; Calvert et al., 1993; Francois, 1987; Malcolm and Price, 1984; Pedersen and Price, 1980; Price et al., 1970; Shishkina and Pavlova, 1965; Vinogradov, 1939). Francois (1987) presented evidence for the involvement of humic material in iodine enrichment in surface sediments. Laboratory experiments indicated that IO_3^- is reduced by sedimentary humic acids under slightly acidic pHs where it is possibly converted to electrophilic species that are incorporated in organic matter by addition reactions. However, I^- did not react with humics. It was suggested that this further explains the lack of surface enrichments in reducing sediments since I^- is the predominant species of iodine found under such conditions. Furthermore, similarities between sediment and humic material I/C profiles supported his suggestion that iodine enrichments in surface sediments are controlled by organic matter. Decreases in the I/C ratio in sediments were proposed to result from the displacement of iodine by nucleophilic species such as sulfide and thiosulfate produced in anaerobic decomposition of organic matter. It is important to note that the two proposed mechanisms for the enrichment of iodine in surface sediments (i.e., sorption onto mineral phases or sorption onto organic matter) are not mutually exclusive, as pointed out by Ullman and Aller (1985) and described above.

It is clear that questions still remain regarding the aquatic geochemistry of iodine. More specifically, the rates of the interconversions among the major species and the mechanisms for these reactions are largely unknown. Iodine is subject to recycling, transport, and uptake by aquatic organisms. Further investigation of iodine's geochemistry will contribute to a greater understanding of both natural elemental cycling processes as well as anthropogenic inputs, including accidental releases of radioactive iodine. It is known that iodine and its compounds influence atmospheric chemistry, including ozone destruction and particle formation (Carpenter, 2003) which play a role in Earth's radiative balance. Additionally, iodine is an essential nutrient element for animals, including humans. Diet is the primary source of this element for humans, yet millions of people worldwide are affected by iodine deficiency disorders. In many parts of the world, dietary intake is directly related to the concentration and speciation of iodine in the environment (Fuge, 1996). Iodine's aquatic geochemistry, particularly the marine component, is the dominant reservoir controlling its distribution in the atmosphere and terrestrial environments (Fuge, 1996; Fuge and Johnson, 1986; Whitehead, 1984).

This is the first investigation using medically-derived ^{131}I as a tracer to study the biogeochemistry of iodine in aquatic environments. The major objectives of this work were to: 1) determine the occurrence and concentrations of ^{131}I in sewage effluent at a relatively small WPCP with known inputs of the radioisotope from thyroid cancer inpatient treatments, 2) determine the occurrence and concentrations of ^{131}I in sewage effluent at three WPCPs in the Washington, DC metropolitan area discharging to the tidal Potomac River, 3) examine the behavior of ^{131}I in the tidal Potomac River and 4) evaluate the potential utility of medically-derived ^{131}I as a tracer of biogeochemical processes in aquatic environments receiving sewage effluent discharges.

Chapter 2

Medically-derived ^{131}I in municipal sewage effluent and implications for the study of other wastewater constituents

1. Abstract

Iodine-131 ($t_{½}$ = 8.04 d) has been measured in aquatic environments receiving sewage effluent discharges, yet few published data exist for the radioisotope in sewage effluent; most work has focused on sewage sludge. This work presents ^{131}I concentrations detected in sewage effluent from a small water pollution control plant (WPCP) serving a regional thyroid cancer treatment facility in Stony Brook, NY, USA. The concentrations detected in the Stony Brook Water Pollution Control Plant (SBWPCP) ranged from 1.8 ± 0.3 to 227 ± 2 Bq L^{-1} in sewage effluent and 61 ± 12 to 2801 ± 32 Bq g^{-1} in suspended solids > 0.7 µm in the effluent. The primary source of ^{131}I is excreta from thyroid cancer inpatients treated at the Stony Brook University Medical Center (SBUMC). Based on several time series measurements following known inpatient treatments, the mean sewage half-life of iodine is 2.9 d in this plant. It is widely recognized that sludge residence times are long relative to the hydraulic retention time in a WPCP. This work suggests that sewage half-life is an important consideration in studies of wastewater constituents. Flow recycling, or activated sludge returns, is commonly used to maintain bacterial populations required to achieve sewage treatment. In activated sludge plants, the hydraulic retention time describes only a portion of the effluent leaving the plant. While the sewage half-life determined here is characteristic of the SBWPCP, this parameter can be estimated for any plant if the flows are known.

2. Introduction

Iodine-131 is released from nuclear power plants, during nuclear weapons tests, nuclear fuel reprocessing and weapons production. Medical use is perhaps the more widespread source of ^{131}I to the environment. It is the most widely used radiopharmaceutical in nuclear medicine for therapeutic purposes, commonly used to treat hyperthyroidism and thyroid cancer. The number of diagnostic procedures using ^{131}I far exceeds the number of therapeutic treatments; however, the latter represents a greater potential source to sewerage. The standard protocol for treating thyroid cancer is removal of the whole thyroid gland followed by administration of ^{131}I to destroy any remaining tissue or cells. Thyroid cancer patients are typically given 4000 to 8000 MBq compared to 100 to 1000 MBq of ^{131}I for hyperthyroid treatments and most of the initial dose is eliminated from the body in urine (ICRP, 2004) In the United States, patient excreta are exempt from sewer discharge regulations and are therefore released into sewerage (Martin and Fenner, 1997).

Many studies have documented the presence of ^{131}I in sewage sludge. However, few values of ^{131}I in sewage effluent have been reported (Erlandsson et al., 1989; Fischer et al., 2009; Kleinschmidt, 2009; Puhakainen, 1998; Rose, 2003; Smith et al., 2008; Sodd et al., 1975). The results of several investigations indicate greater than 75% of ^{131}I entering WPCPs leaves in the effluent (Barci-Funel et al., 1993; Dalmasso et al., 1997; Erlandsson et al., 1989; Erlandsson et al., 1983; Erlandsson and Mattsson, 1978; Martin and Fenner, 1997; Prichard et al., 1981; Puhakainen, 1998; Stetar et al., 1993). It is not surprising then that that medically-derived ^{131}I is readily measurable in the environment (Fischer et al., 2009; Howe and Hunt, 1984; Howe and Lloyd, 1986; Kitto et al., 2005a; Kitto et al., 2006; Kitto et al., 2005b; Marsh et al., 1988; Puhakainen, 1998; Rose, 2003; Smith et al., 2008; Sodd et al., 1975; Waller and Cole, 1999). While the occurrence and concentrations of ^{131}I in sewage effluent have been poorly characterized, these studies suggest discharges of the radioisotope may be widespread and therefore useful as a tracer in receiving waters to study biogeochemical processes occurring on the time scale of approximately one month.

Distributions of naturally occurring iodine have been well studied and indicate that iodine is biologically cycled and remineralized similar to other nutrient elements. The nutrient-like behavior of iodine suggests that wastewater-derived ^{131}I may be used to study the short-term fate of wastewater nitrogen in aquatic systems. Wastewater discharges of ^{131}I would also be useful to investigate the natural cycling of iodine in receiving waters. More specifically, the rates and mechanisms governing the transformations of natural iodine in aquatic systems are not well known. Additionally, Smith et al. (2008) proposed ^{131}I as a wastewater-specific particle tracer and suggested its use as a tracer for short-term sediment dynamics.

The objective of this study was to determine the occurrence and concentrations of ^{131}I in sewage effluent at a relatively small WPCP with known inputs of the radioisotope from thyroid cancer inpatient treatments. The data presented here add to the small existing set of data of ^{131}I in sewage effluent.

3. Study Site

The SBWPCP, Suffolk County Sewer District #21, is a tertiary treatment facility located on the campus of Stony Brook University, Stony Brook, NY, USA. The plant's service area includes the campus, the SBUMC and a small number of private homes. The estimated size of the population served is 20,000. At the time of sampling, tertiary treatment was achieved via an oxidation ditch with both activated sludge and mixed liquor returns. Sodium hypochlorite was added to sewage effluent for disinfection prior to discharge from the plant. Travel time from the plant to its outfall in Port Jefferson Harbor, NY is four to six hours. The oxidation ditch no longer receives mixed liquor return and sewage effluent is disinfected using ultraviolet irradiation at the Port Jefferson WPCP (Suffolk County Sewer District #1) just prior to its discharge into Port Jefferson Harbor (E. Brewer, Suffolk County Department of Public Works, personal communication, 2010).

The design capacity of the plant is approximately 9.5×10^6 liters per day (MLD) or 2.5×10^6 gallons per day (MGD). Average flow is approximately 6.8 MLD (1.8 MGD). Average daily maximum and minimum flows are approximately 7.6 MLD (2.0 MGD) and 3.8 MLD (1.0 MGD), respectively. During the summer months and weekends, when school is not in full session, average flows decrease as much as 20%. The average daily maximum to minimum ratio of daily flow is approximately 2, but maximum flow in a given day can exceed 4 times the minimum daily flow. Minimum flows generally occur between 7 and 8 AM, after which there is a rapid increase in flow until it peaks around 12 PM. Flow then decreases slowly until about 2 AM, then more rapidly until about 7 AM (M. Cruz, Suffolk County Department of Public Works, personal communication, 2008).

The hydraulic retention time (HRT) of sewage in the SBWPCP is approximately 24 to 36 hours, depending on the University schedule as mentioned above. The mean cell residence time (MCRT), or the residence time of organic matter in the system, is about 36 days (Rose, 2003).

4. Methods

Sample Collection and Preparation

Sewage effluent was collected as a grab sample from the final effluent stream before discharge from the plant using a 1 L HDPE bottle. More than half of the samples were analyzed with no further treatment. The remaining samples were vacuum filtered through a 0.7 µm glass fiber filter (GFF). Aliquots of the filtrate were retained and analyzed with no further treatment. Sample volumes were 150 mL and 170 mL. Straight-side polypropylene jars (64 mm height; 64 mm diameter) were used for counting. Pre-weighed GFFs that were retained for γ-ray spectrometry determinations were dried at 40 ˚C overnight and re-weighed before analysis. More than 80% of the samples were collected between 12 PM and 3 PM. Sampling replicates were also collected at the SBWPCP on six additional days.

Determination of ^{131}I

The activity of ^{131}I was determined by γ-ray spectrometry (364.5 keV peak; branching ratio = 0.812) using Canberra low energy germanium detectors. Previous work determined that there is no self-absorption of γ-rays emitted at 364.5 keV by concentrated sludge samples and therefore no self-absorption correction was applied to the data (Rose, 2003). Generally, samples were counted for one day. Due to the relatively short half-life of ^{131}I, activities were corrected to account for decay during data acquisition as described in Hoffman and Van Camerik (1967). All concentrations of ^{131}I are reported for time of collection and with a 1σ counting error.

The counting efficiency for each geometry and each detector used in this investigation was determined using a certified ^{131}I standard solution. For the 150 mL and 170 mL sewage effluent samples, deionized water was spiked with the ^{131}I standard solution and counted three times on each detector to determine the counting efficiency of these geometries. The ^{131}I standard solution was applied to three GFFs with a pipette. Each filter was counted three times on each detector to determine the counting efficiency of the suspended solids samples. In each case, the mean counting efficiency was used to calculate sample activities. Counting efficiencies are reported in Appendix A.

Multi-day continuum background counts were determined for each detector with three ^{131}I-free sewage effluent samples. The mean background count for each detector was used to determine limit of detection (L_D) as described by Currie (1968). The detection limits were ≤ 1.7 Bq L^{-1} for sewage effluent samples and ≤ 50 Bq g^{-1} for suspended solids.

5. Results

Unfiltered Effluent

Iodine-131 concentrations detected in unfiltered sewage effluent collected on 77 different days between June 2006 and March 2009 ranged from 1.8 ± 0.3 to 217 ± 1 Bq L^{-1} (median = 11 Bq L^{-1}); nine samples < L_D (Table 2.1). Percent differences among ^{131}I concentrations in replicate samples were 0.9 to 7% (Table 2.2). The variation among the replicates is within the counting error.

Filtered Effluent

Iodine-131 concentrations detected in filtered sewage effluent collected on 46 different days between January 2007 and March 2009 ranged from 2.5 ± 0.3 to 227 ± 2 Bq L^{-1} (median = 18 Bq L^{-1}); three samples < L_D (Table 2.3).

Suspended Solids

Iodine-131 concentrations detected in suspended solids > 0.7 µm in sewage effluent collected on 36 different days between March 2007 and March 2009 ranged from 61 ± 12 to 2801 ± 32 Bq g^{-1} (median = 304 Bq g^{-1}); seven samples < L_D (Table 2.4). Suspended solids concentrations for these samples ranged from approximately 0.3 to 5 mg L^{-1}. Sample information, γ-ray spectrometry data and L_D values for sewage effluent samples and suspended solids are reported in Appendix B.

6. Discussion

Previous studies have reported values up to 32 Bq L^{-1} of ^{131}I in sewage effluent from WPCPs ranging in size from 30 to 645 MLD (8 to 170 MGD) (Erlandsson et al., 1989; Fischer et al., 2009; Puhakainen, 1998; Smith et al., 2008; Sodd et al., 1975). The highest concentrations in sewage effluent from the SBWPCP were measured in unfiltered sewage effluent (Table 2.1) collected on August 12, 2006; 217 ± 1 Bq L^{-1} following five inpatient treatments and 227 ± 2 Bq L^{-1} in filtered effluent (Table 2.2) collected on March 15, 2007 following four inpatient treatments. Dates of inpatient treatments at the SBUMC are listed in Table 2.5. It should be noted that while the suspended solids have a high specific activity of ^{131}I (Table 2.4), the contribution of the suspended solids to the total activity of the unfiltered effluent is minimal because the suspended solids concentrations are low. Additionally, no settling of solids was observed during counting and therefore homogeneity of the sample was maintained during counting. The following discussion assumes there is no difference between the concentrations of ^{131}I measured in whole effluent and the filtered effluent collected at the SBWPCP.

These concentrations can be primarily attributed to the frequency of thyroid cancer inpatient treatments performed at the SBUMC, the size of the plant and its sewage half-life. There are approximately 60 inpatient treatments per year at the SBUMC with about an equal number of outpatients. While outpatients treated at SBUMC and other facilities are possible sources to the plant, this is not a significant source to the SBWPCP. Outpatients at SBUMC leave the hospital following administration of the ^{131}I (J. Daley, Stony Brook University, Environmental Health and Safety, personal communication, 2010). Frequent or large inputs from persons treated at another facility are unlikely considering the size and composition of the population, which consists mainly of University students and employees. The SBUMC is a regional thyroid cancer treatment facility. These treatments require specialized rooms and personnel, and therefore are not performed at all hospitals. Medical use is the only known source of ^{131}I to this system (Rose, 2003).

Typically, thyroid cancer inpatients remain in the hospital 24 to 30 hours following treatment. Length of hospital stay is dependent on the time it takes for the patient's external dose rate, measured at 1 m, to decrease to less than 10 mrem h^{-1} from 20 to 30 mrem h^{-1} after administration of the dose (J. Daley, Stony Brook University, Environmental Health and Safety, personal communication, 2008). This is consistent with the work of Driver and Packer (2001) who found that 55% of the initial activity administered to thyroid cancer patients is excreted in the first 24 hours.

The data presented here clearly indicate that ^{131}I is discharged from the plant for many days following a patient treatment. Larsen and coworkers (1995) found elevated ^{131}I concentrations in digested sewage sludge at the Oak Ridge WPCP (Oak Ridge, TN) for more than one month following a single patient treatment. Previous work in the SBWPCP indicated that ^{131}I can be detected in sewage sludge for at least two weeks after known inputs (Rose, 2003). Retention of sewage sludge is dependent on plant design and sludge removal practices. More specifically, the recycling of biomass within a sewage treatment plant, or activated sludge, helps maintain a standing stock of bacteria necessary for sewage treatment. Organic matter may remain in a WPCP for a few weeks due to this recycling of solids. The residence time of organic matter in a system is well known and described by the MCRT. Martin and Fenner (1997) examined ^{131}I concentrations in primary sewage sludge at the Ann Arbor WPCP (Ann Arbor, MI) following an input of ^{131}I from a thyroid cancer treatment. They determined the effective sewer half-life for ^{131}I in that plant to be 1.6 days.

In the same way, the concentrations of ^{131}I in sewage effluent can be used to determine its sewage half-life. The term *sewage* half-life refers to the half-life of a wastewater constituent in a WPCP and is used to distinguish it from the *sewer* half-life described by Martin and Fenner (1997) derived from primary sludge data. In this study, there are twelve time series with sufficient data following an inpatient treatment, in which there was no further known input of ^{131}I. Each time series follows an exponential decay

$$C_t = C_o\, e^{-\lambda t} \qquad (1)$$

where C_t is the concentration of ^{131}I at time t, C_o is the initial concentration of ^{131}I and

$$\lambda = \frac{\ln 2}{T_{I-131}} \qquad (2)$$

is the decay constant. The natural log of the concentration of ^{131}I in sewage effluent plotted versus time results in a line with slope λ from which the sewage half-life of ^{131}I ($T_{I\text{-}131}$) can be derived (Figure 2.1). The $T_{I\text{-}131}$ determined from these plots ranged from 0.9 to 3.5 d (mean = 2.0 ± 0.7 d). This is reasonable if we consider that these data indicate ^{131}I can be measured in sewage effluent for two weeks following a patient treatment with no further input. The $T_{I\text{-}131}$ represents removal from the plant as sewage effluent discharges, losses to sludge as well as through radioactive decay.

The sewage half-life of non-radioactive iodine (T_s) can be estimated by accounting for radioactive decay. For each time series, the ^{131}I concentrations were decay-corrected using the relationship in Equation 1 and the known decay constant for ^{131}I, where C_o = the concentration of ^{131}I measured on the first day of the time series and t = time elapsed since the first day of the time series. Decay-corrected data plotted as described above for each time series are shown in Figure 2.2. The range of values for T_s is from 1.0 to 6.2 d (mean = 3.0 ± 1.0 d). The time series beginning June 14, July 27 and May 23 are the longest time series containing the most data. The T_s values for these three time series are in good agreement: 3.9, 3.6 and 3.9 d, respectively. Construction in the Stony Brook plant may have affected the flow in March 2009 and may account for the comparatively short T_s for that sampling period (E. Brewer, Suffolk County Department of Public Works, personal communication, 2010). The time series beginning June 21, 2007 results in a relatively long T_s that does not seem reasonable. Rather, it is possible that there was an input of ^{131}I between June 22 and June 28, the dates of the second and third samples taken in the time series. Daily fluctuations in flow are likely to be the primary source of variation in these estimates.

The mean λ of 0.30 d^{-1} (or 30% removal per day) is consistent with the fraction of sewage removed from the plant each day (Figure 2.2). Activated sludge and mixed liquor returns in the SBWPCP return approximately 69% of the total flow to the oxidation ditch. Flow out of the oxidation ditch is 21.9 MLD (5.8 MGD) and 15.1 MLD (4.0 MGD) of that flow is returned via mixed liquor and activated sludge returns (Figure 2.3). This results in a 31% removal of sewage as sewage effluent per day, suggesting that only a small fraction of iodine is lost to sewage sludge.

Partitioning of ^{131}I in sewage can be characterized assuming that incorporation into particles is rapid with respect to particle recycling and removal from the plant. The partition coefficients (K_d) determined in this study (Table 2.6) suggest that iodine is associated with the solid phase but the extent is likely overestimated. Experimental data and field data from various environments indicate that iodine is associated with particulate organic matter (Bors et al., 1991; Brewer et al., 1980; Calvert et al., 1993; Francois, 1987; Sheppard and Thibault, 1992; Ullman and Aller, 1980; Upstill-Goddard and Elderfield, 1988; Whitehead, 1973a; Whitehead, 1973b). K_d values determined by other investigators in laboratory experiments and natural settings range from 1 to 10^3 L kg^{-1} (Alvarado-Quiroz et al., 2002; Bird and Schwartz, 1997; Cochran et al., 2000; Muramatsu et al., 1990a; Yoshida et al., 1992). The K_d values calculated in this study range from 4.1 x 10^3 to 3.4 x 10^4 L kg^{-1} (mean = 1.5 x 10^4 L kg^{-1}; n = 29). These values are somewhat higher than those calculated previously for ^{131}I in the SBWPCP by Rose (2003) using sewage sludge and effluent concentrations (3 x 10^2 to 8 x 10^3 L kg^{-1}).

The higher K_d values determined here may be suggestive of DOI retention on filters (Karl et al., 1998; Maske and Garciamendoza, 1994; Moran et al., 1999). However, the higher specific activity of ^{131}I in the filtered particulates is more likely due to a preferential association of iodine with finer grained organic matter in suspended material. Iodine's association with particulate organic matter is well established (Alvarado-Quiroz et al., 2002; Bojanowski and Paslawska, 1970; Bors et al., 1991; Brewer et al., 1980; Calvert et al., 1993; Francois, 1987; Harvey, 1980; Lieser and Steinkopff, 1989; Malcolm and Price, 1984; Pedersen and Price, 1980; Price et al., 1970; Sheppard and Hawkins, 1995; Sheppard and Thibault, 1992; Shishkina and Pavlova, 1965; Ullman and Aller, 1980; Upstill-Goddard and Elderfield, 1988; Vinogradov, 1939; Whitehead, 1973a; Whitehead, 1973b; Whitehead, 1974a; Whitehead, 1974b; Whitehead, 1981). The decrease in K_d observed after patient treatments may be due to a lag time in bacterial uptake and incorporation into biomass.

The extent to which ^{131}I is lost to the sewage sludge is likely to be minimal. The frequency of patient treatments and the time it takes for ^{131}I to be removed from the system precludes any reasonable estimate of total discharges leaving in the effluent from a single patient treatment.

7. Conclusions

Known inputs of ^{131}I to the SBWPCP provided an opportunity to study the behavior of this radioisotope in sewage. Concentrations of ^{131}I in the sewage effluent discharged from the SBWPCP are a function of thyroid cancer inpatient treatments in the SBUMC, the size of the plant and its sewage half-life. Iodine-131 remains in the SBWPCP for at least two weeks following inpatient treatments. Due to the frequency of patients treated and retention in the plant, sewage effluent discharges of ^{131}I are fairly continuous.

These results add a significant amount of data to the small, existing set of ^{131}I concentrations in sewage effluent. Iodine-131 is a potentially valuable tracer of wastewater-derived material in surface waters receiving sewage effluent discharges. However, data from more WPCPs are needed to determine the extent to which ^{131}I can be used.

This work suggests that estimates of sewage half-life are important for investigations involving the behavior, degradation and removal of wastewater constituents during sewage treatment, and in assessing their environmental fate. Sludge retention time and MCRT are common terms used to describe the residence time of solids in a WPCP. The HRT describes the time it takes for a parcel of water to travel through a WPCP. HRT does not necessarily represent the time it takes for the entire parcel of water to travel through a plant. The data presented here clearly show that ^{131}I will remain in an activated sludge plant longer than the HRT. The primary purpose of activated sludge is to return biomass solids to the system to maintain a healthy microbial population for biological treatment. What may be overlooked is that the returns are primarily liquid and are therefore returning a portion of the total plant flow, which may be substantial in some WPCPs.

The sewage half-life, analogous to a radioactive half-life, describes the time it takes for half of a wastewater component to be removed from a WPCP. The fraction remaining after n half-lives is 2^{-n}. For iodine in the SBWPCP, it requires 5 half-lives or approximately 15 days to remove about 97% of the material. This is significantly longer than the 24-hour HRT of this plant. Studies in sewage treatment plants often consider HRT, but a wastewater constituent may remain in a WPCP far longer than its HRT. The sewage half-life determined here is characteristic of the SBWPCP; however this parameter can be estimated for any WPCP if its flows are known.

Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})	Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})
061206E	6/12/06 1:59 PM	48.4 ± 0.8	083006E	8/30/06 1:46 PM	46.8 ± 0.8
061306E	6/13/06 1:55 PM	21.8 ± 0.6	083106E	8/31/06 2:35 PM	30.5 ± 0.4
061406E	6/14/06 11:28 AM	93 ± 1	091406E	9/14/06 2:22 PM	8.8 ± 0.2
061506E	6/15/06 2:20 PM	65 ± 1	091506E-A	9/15/06 2:34 PM	108 ± 3
061606E	6/16/06 10:03 AM	60 ± 1	091806E-A	9/18/06 2:25 PM	23.1 ± 0.2
061906E	6/19/06 2:25 PM	21.4 ± 0.6	092006E-A	9/20/06 1:44 PM	14.0 ± 0.4
062006E	6/20/06 2:34 PM	17.6 ± 0.5	092206E-A	9/22/06 12:41 PM	7.2 ± 0.2
062106E	6/21/06 10:47 AM	16.1 ± 0.5	092906E-E	9/29/06 1:31 PM	187 ± 1
062206E	6/22/06 2:35 PM	9.6 ± 0.3	101906E-A	10/19/06 2:05 PM	83.2 ± 0.6
062306E	6/23/06 11:50 AM	9.0 ± 0.4	120606E	12/6/06 2:08 PM	10.6 ± 0.2
062606E	6/26/06 12:15 PM	11.9 ± 0.5	120706E	12/7/06 1:37 PM	8.5 ± 0.2
062706E	6/27/06 2:22 PM	8.8 ± 0.3	120806E	12/8/06 12:30 PM	6.7 ± 0.1
062806E	6/28/06 2:05 PM	5.5 ± 0.4	121106E	12/11/06 12:43 PM	10.9 ± 0.2
062906E	6/29/06 2:46 PM	2.0 ± 0.3	121206E	12/12/06 2:22 PM	7.9 ± 0.2
063006E	6/30/06 10:48 AM	4.0 ± 0.3	121306E	12/13/06 1:00 PM	6.2 ± 0.2
071706E	7/17/06 2:08 PM	$< L_D$	121406E	12/14/06 1:47 PM	4.9 ± 0.2
071806E	7/18/06 1:21 PM	$< L_D$	121506E	12/15/06 12:29 PM	4.3 ± 0.1
071906E-2	7/19/06 2:41 PM	65 ± 1	121906E	12/19/06 2:05 PM	7.1 ± 0.2
072006E-2	7/20/06 2:32 PM	50.2 ± 0.8	122006E	12/20/06 11:22 AM	7.3 ± 0.2
072106E	7/21/06 1:13 PM	24.1 ± 0.4	010307E	1/3/07 1:48 PM	$< L_D$
072406E	7/24/06 1:21 PM	7.2 ± 0.4	010507E	1/5/07 12:42 PM	$< L_D$
072506E	7/25/06 12:45 PM	7.5 ± 0.5	010807E	1/8/07 12:00 PM	$< L_D$
072606E	7/26/06 11:18 AM	49 ± 2	010907E	1/9/07 1:35 PM	$< L_D$
072706E	7/27/06 9:44 AM	60 ± 2	011107E	1/11/07 12:00 PM	$< L_D$
072806E	7/28/06 7:01 AM	22.2 ± 0.7	011507E	1/15/07 2:18 PM	$< L_D$
073106E	7/31/06 1:53 PM	6.4 ± 0.5	011607E	1/16/07 1:00 PM	$< L_D$
080106E	8/1/06 1:37 PM	5.4 ± 0.5	012307E	1/23/07 12:53 PM	4.9 ± 0.2
080206E	8/2/06 1:34 PM	6.1 ± 0.3	012407E	1/24/07 12:35 PM	5.3 ± 0.2
080406E	8/4/06 2:13 PM	4.9 ± 0.4	012507E	1/25/07 1:25 PM	52.2 ± 0.5
080706E	8/7/06 1:54 PM	1.8 ± 0.3	012607E	1/26/07 10:05 AM	59.4 ± 0.6
080806E-2	8/8/06 1:05 PM	43 ± 1	012809E[1]	1/28/09 12:00 PM	3.9 ± 0.2
080906E	8/9/06 12:47 PM	46 ± 1	031409E[1]	3/14/09 1:07 PM	67 ± 1
081006E-2	8/10/06 2:30 PM	96 ± 1	031509E[1]	3/15/09 1:31 PM	20.4 ± 0.6
081106E-2	8/11/06 2:26 PM	107 ± 1	031609E[1]	3/16/09 12:36 PM	7.1 ± 0.3
081206E	8/12/06 9:13 AM	217 ± 1	031709E[1]	3/17/09 1:30 PM	4.0 ± 0.2
081706E	8/17/06 1:35 PM	190 ± 2	031809E[1]	3/18/09 12:30 PM	3.0 ± 0.2
081806E	8/18/06 12:29 PM	121 ± 2	031909E[1]	3/19/09 12:34 PM	2.9 ± 0.3
082506E	8/25/06 11:44 AM	11.1 ± 0.3	032009E[1]	3/20/09 1:00 PM	3.4 ± 0.2
082906E	8/29/06 1:35 PM	77 ± 1			

[1] sample volume = 0.15 L

Table 2.1. Iodine-131 concentrations in **unfiltered** effluent samples collected from the SBWPCP. Sample volume = 0.17 L. $< L_D$ = activity below detection limit.

Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})
091506E-A	9/15/06 2:34 PM	108 ± 3
091506E-C	9/15/06 2:35 PM	109.6 ± 0.4
091806E-A	9/18/06 2:25 PM	23.1 ± 0.2
091806E-C	9/18/06 2:26 PM	23.3 ± 0.4
092006E-A	9/20/06 1:44 PM	14.0 ± 0.4
092006E-C	9/20/06 1:45 PM	14.2 ± 0.3
092006E-E	9/20/06 1:46 PM	14.0 ± 0.2
092206E-A	9/22/06 12:41 PM	7.2 ± 0.2
092206E-B	9/22/06 12:41 PM	7.3 ± 0.2
092206E-E	9/22/06 12:43 PM	7.7 ± 0.3
092906E-C	9/29/06 1:30 PM	193 ± 4
092906E-D	9/29/06 1:30 PM	183 ± 1
092906E-E	9/29/06 1:31 PM	187 ± 1
101906E-A	10/19/06 2:05 PM	83.2 ± 0.6
101906E-D	10/19/06 2:06 PM	83.7 ± 0.7
101906E-E	10/19/06 2:07 PM	81.1 ± 0.7

Table 2.2. Iodine-131 concentrations in sampling replicates of unfiltered effluent samples collected from the SBWPCP.

Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})	Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})
013107E[1]	1/31/07 12:27 PM	5.6 ± 0.3	032907E	3/29/07 11:35 AM	136 ± 2
020107E	2/1/07 12:30 PM	34.1 ± 0.5	052307E-A	5/23/07 11:46 AM	102 ± 1
020207E	2/2/07 12:40 PM	24.8 ± 0.9	052407E-A	5/24/07 10:33 AM	85 ± 1
020507E	2/5/07 12:29 PM	5.0 ± 0.3	052507E-A	5/25/07 1:31 PM	42.5 ± 0.5
020707E	2/7/07 10:52 AM	4.0 ± 0.3	053007E	5/30/07 12:12 PM	18.0 ± 0.6
020807E	2/8/07 12:47 PM	43.0 ± 0.8	053107E	5/31/07 1:00 PM	12.1 ± 0.3
020907E	2/9/07 12:26 PM	42 ± 1	060407E	6/4/07 11:39 AM	4.6 ± 0.3
021207E	2/12/07 1:25 PM	8.4 ± 0.4	060507E	6/5/07 2:09 PM	2.5 ± 0.3
030707E	3/7/07 12:00 PM	$< L_D$	060607E	6/6/07 12:59 PM	17.7 ± 0.4
030807E	3/8/07 12:38 PM	$< L_D$	060707E	6/7/07 2:18 PM	24.4 ± 0.3
030907E	3/9/07 11:33 AM	$< L_D$	060807E	6/8/07 2:25 PM	13.6 ± 0.6
031007E	3/10/07 11:15 AM	51.6 ± 0.9	061107E	6/11/07 1:33 PM	3.1 ± 0.3
031107E	3/11/07 10:42 AM	37.5 ± 0.7	061207E	6/12/07 2:33 PM	2.5 ± 0.3
031207E	3/12/07 1:07 PM	24.2 ± 0.6	061307E	6/13/07 1:00 PM	27.8 ± 0.8
031307E	3/13/07 12:48 PM	29.7 ± 0.8	061407E	6/14/07 2:30 PM	16.4 ± 0.7
031407E	3/14/07 12:43 PM	23.3 ± 0.6	061507E	6/15/07 2:30 PM	55.3 ± 0.9
031507E	3/15/07 12:46 PM	227 ± 2	061907E	6/19/07 2:31 PM	4.6 ± 0.3
031607E	3/16/07 12:41 PM	175 ± 2	062007E	6/20/07 12:58 PM	2.9 ± 0.5
032007E	3/20/07 11:12 AM	8.6 ± 0.4	062107E	6/21/07 2:15 PM	14.5 ± 0.8
032107E	3/21/07 1:12 PM	4.6 ± 0.3	062207E	6/22/07 1:13 PM	11.7 ± 0.6
032307E	3/23/07 12:42 PM	32.9 ± 0.5	062807E	6/28/07 1:55 PM	3.6 ± 0.3
032707E	3/27/07 12:43 PM	5.1 ± 0.4	071207E	7/12/07 1:35 PM	97 ± 1
032807E	3/28/07 1:12 PM	93 ± 1	071407E	7/14/07 1:00 PM	115 ± 1

[1] sample volume = 0.17 L

Table 2.3. Iodine-131 concentrations in **filtered** effluent samples collected from the SBWPCP. Sample volume = 0.15 L. $< L_D$ = activity below detection limit.

Sample #	Sample Collection Date & Time	^{131}I (Bq g^{-1})	Sample #	Sample Collection Date & Time	^{131}I (Bq g^{-1})
030707E	3/7/07 12:00 PM	$< L_D$	053007E	5/30/07 12:12 PM	345 ± 7
030807E	3/8/07 12:38 PM	$< L_D$	053107E	5/31/07 1:00 PM	383 ± 10
030907E	3/9/07 11:33 AM	$< L_D$	060407E	6/4/07 11:39 AM	153 ± 6
031007E	3/10/07 11:15 AM	651 ± 17	060507E	6/5/07 2:09 PM	61 ± 12
031107E	3/11/07 10:42 AM	341 ± 12	060607E	6/6/07 12:59 PM	126 ± 10
031207E	3/12/07 1:07 PM	221 ± 8	060707E	6/7/07 2:18 PM	260 ± 18
031307E	3/13/07 12:48 PM	295 ± 11	060807E	6/8/07 2:25 PM	164 ± 17
031407E	3/14/07 12:43 PM	312 ± 8	061107E	6/11/07 1:33 PM	72 ± 9
031507E	3/15/07 12:46 PM	2801 ± 32	061207E	6/12/07 2:33 PM	$< L_D$
031607E	3/16/07 12:41 PM	1289 ± 19	061307E	6/13/07 1:00 PM	369 ± 30
032007E	3/20/07 11:12 AM	91 ± 5	061407E	6/14/07 2:30 PM	203 ± 35
032107E	3/21/07 1:12 PM	80 ± 4	061507E	6/15/07 2:30 PM	722 ± 55
032307E	3/23/07 12:42 PM	227 ± 6	061907E	6/19/07 2:31 PM	154 ± 21
032707E	3/27/07 12:43 PM	76 ± 6	062007E	6/20/07 12:58 PM	$< L_D$
032807E	3/28/07 1:12 PM	779 ± 11	062207E	6/22/07 1:13 PM	$< L_D$
052307E-A	5/23/07 11:46 AM	1797 ± 34	062807E	6/28/07 1:55 PM	$< L_D$
052407E-A	5/24/07 10:33 AM	353 ± 5	071207E	7/12/07 1:35 PM	1282 ± 27
052507E-A	5/25/07 1:35 PM	916 ± 19	071407E	7/14/07 1:00 PM	1157 ± 28

Table 2.4. Iodine-131 concentrations in suspended solids > 0.7µm from sewage effluent samples collected at the SBWPCP. $< L_D$ = activity below detection limit.

Year	Month	Inpatient Treatment Date
2006	June	6, 8*, 9, 13, 23
	July	6, 18, 20, 25, 26
	August	7, 9*, 10, 11, 15, 16, 26
	September	6, 8*, 14, 15, 27, 28
	October	5, 6, 11, 18, 20, 24, 25, 31
	November	1, 2, 14, 17
	December	1, 8, 12, 15, 20, 28
2007	January	6, 24, 25, 31
	February	7
	March	9*, 12, 14, 21, 27, 28, 29, 30*
	April	17
	May	2, 15, 17, 18, 21, 22
	June	5, 12, 14, 20, 28, 29
	July	2, 5, 11, 12, 13, 18, 25
2009	January	14, 15, 23
	February	20, 25, 27
	March	13, 18

*Indicates two inpatient treatments

Table 2.5. Thyroid cancer inpatient treatment dates at the Stony Brook University Medical Center between June 2006 and July 2007 and January through March 2009 (J. Daley, Stony Brook University, Environmental Health and Safety, personal communication, 2010).

Sample #	Sample Collection Date & Time	$^{131}I_{Solids}$ (Bq g^{-1})	$^{131}I_{Effluent}$ (Bq L^{-1})	K_d (L kg^{-1})
031007E	3/10/07 11:15 AM	651 ± 17	51.6 ± 0.9	1.3×10^4
031107E	3/11/07 10:42 AM	341 ± 12	37.5 ± 0.7	9.1×10^3
031207E	3/12/07 1:07 PM	221 ± 8	24.2 ± 0.6	9.1×10^3
031307E	3/13/07 12:48 PM	295 ± 11	29.7 ± 0.8	1.0×10^4
031407E	3/14/07 12:43 PM	312 ± 8	23.3 ± 0.6	1.3×10^4
031507E	3/15/07 12:46 PM	2801 ± 32	227 ± 2	1.2×10^4
031607E	3/16/07 12:41 PM	1289 ± 19	175 ± 2	7.4×10^3
032007E	3/20/07 11:12 AM	91 ± 5	8.6 ± 0.4	1.1×10^4
032107E	3/21/07 1:12 PM	80 ± 4	4.6 ± 0.3	1.7×10^4
032307E	3/23/07 12:42 PM	227 ± 6	32.9 ± 0.5	6.9×10^3
032707E	3/27/07 12:43 PM	76 ± 6	5.1 ± 0.4	1.5×10^4
032807E	3/28/07 1:12 PM	779 ± 11	93 ± 1	8.4×10^3
052307E-A	5/23/07 11:46 AM	1797 ± 34	102 ± 1	1.8×10^4
052407E-A	5/24/07 10:33 AM	353 ± 5	85 ± 1	4.1×10^3
052507E-A	5/25/07 1:35 PM	916 ± 19	42.5 ± 0.5	2.2×10^4
053007E	5/30/07 12:12 PM	345 ± 7	18.0 ± 0.6	1.9×10^4
053107E	5/31/07 1:00 PM	383 ± 10	12.1 ± 0.3	3.2×10^4
060407E	6/4/07 11:39 AM	153 ± 6	4.6 ± 0.3	3.3×10^4
060507E	6/5/07 2:09 PM	61 ± 12	2.5 ± 0.3	2.5×10^4
060607E	6/6/07 12:59 PM	126 ± 10	17.7 ± 0.4	7.1×10^3
060707E	6/7/07 2:18 PM	260 ± 18	24.4 ± 0.3	1.1×10^4
060807E	6/8/07 2:25 PM	164 ± 17	13.6 ± 0.6	1.2×10^4
061107E	6/11/07 1:33 PM	72 ± 9	3.1 ± 0.3	2.3×10^4
061307E	6/13/07 1:00 PM	369 ± 30	27.8 ± 0.8	1.3×10^4
061407E	6/14/07 2:30 PM	203 ± 35	16.4 ± 0.7	1.2×10^4
061507E	6/15/07 2:30 PM	722 ± 55	55.3 ± 0.9	1.3×10^4
061907E	6/19/07 2:31 PM	154 ± 21	4.6 ± 0.3	3.4×10^4
071207E	7/12/07 1:35 PM	1282 ± 27	97 ± 1	1.3×10^4
071407E	7/14/07 1:00 PM	1157 ± 28	115 ± 1	1.0×10^4

Table 2.6. Partition coefficients (K_d) calculated for ^{131}I from the suspended solids > 0.7 µm and sewage effluent concentrations in the SBWPCP.

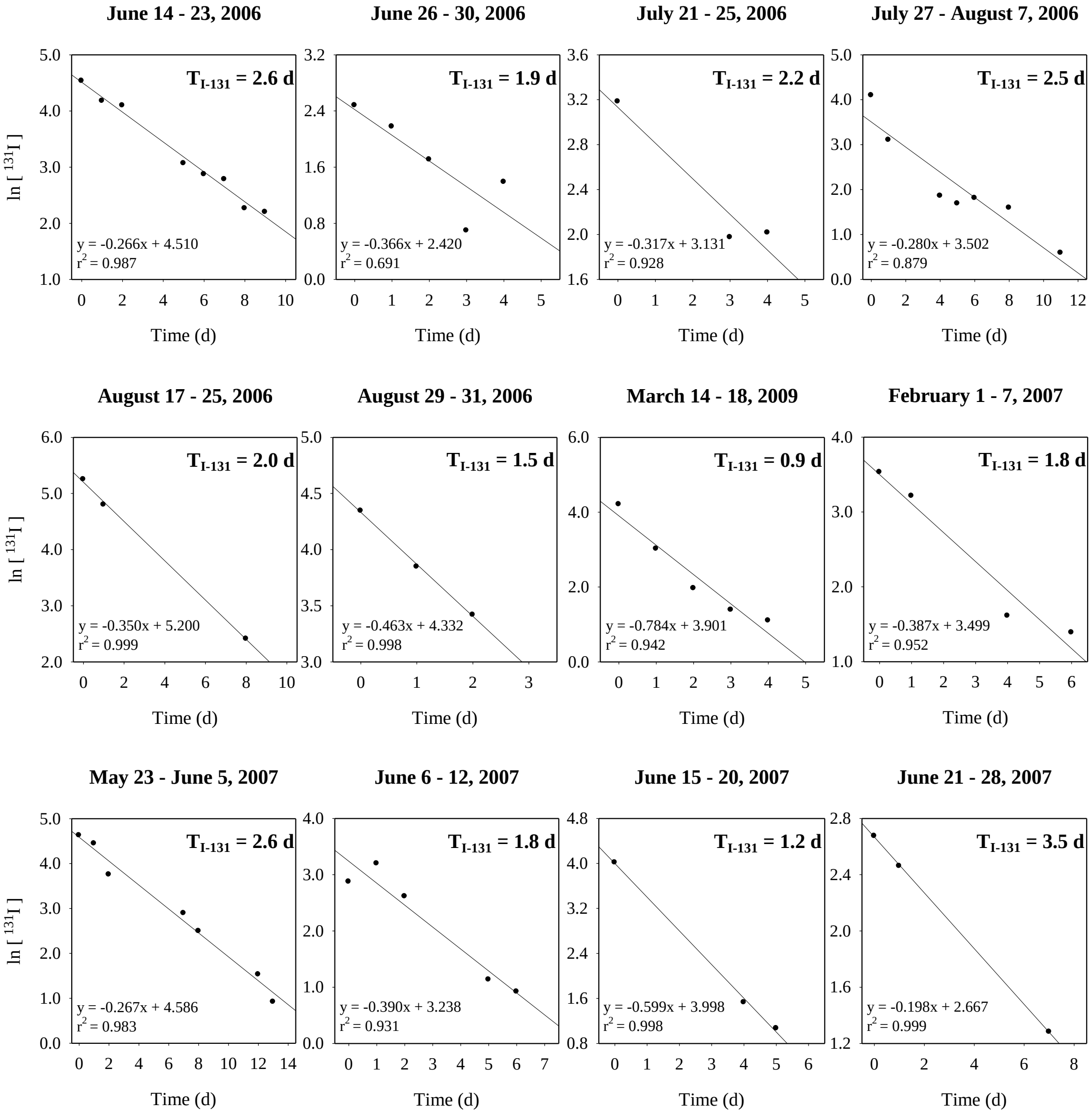

Figure 2.1. Natural log of ^{131}I concentrations in sewage effluent versus time in the SBWPCP for twelve sampling periods between inpatient treatments. **$T_{I\text{-}131}$** represents the sewage half-life of ^{131}I for each time series (mean $T_{I\text{-}131}$ = 2.0 ± 0.7 d).

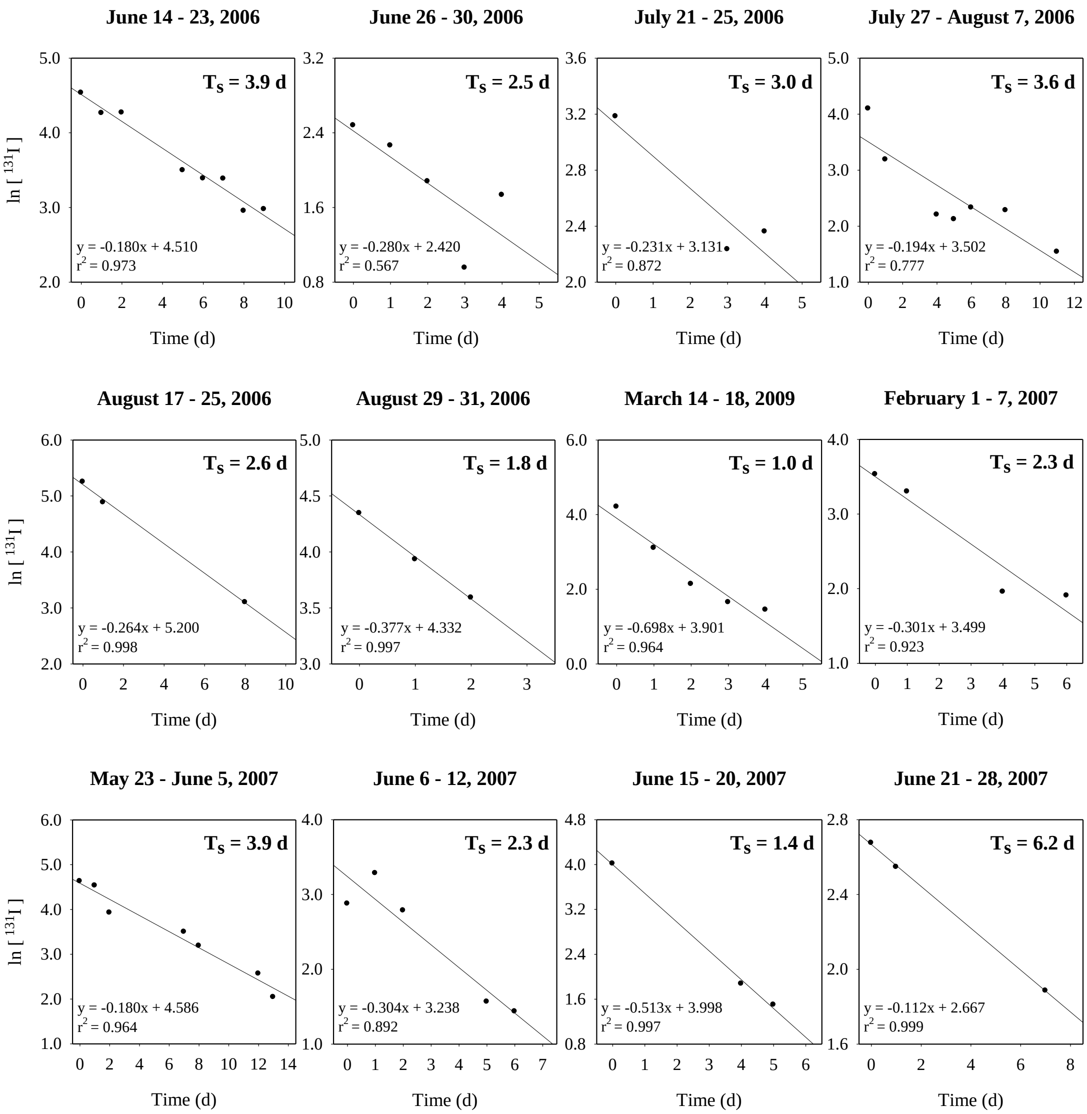

Figure 2.2. Natural log of decay-corrected ^{131}I concentrations in sewage effluent versus time in the SBWPCP for twelve sampling periods between inpatient treatments. **T_s** represents the sewage half-life for each time series (mean $T_s = 3 \pm 1$ d).

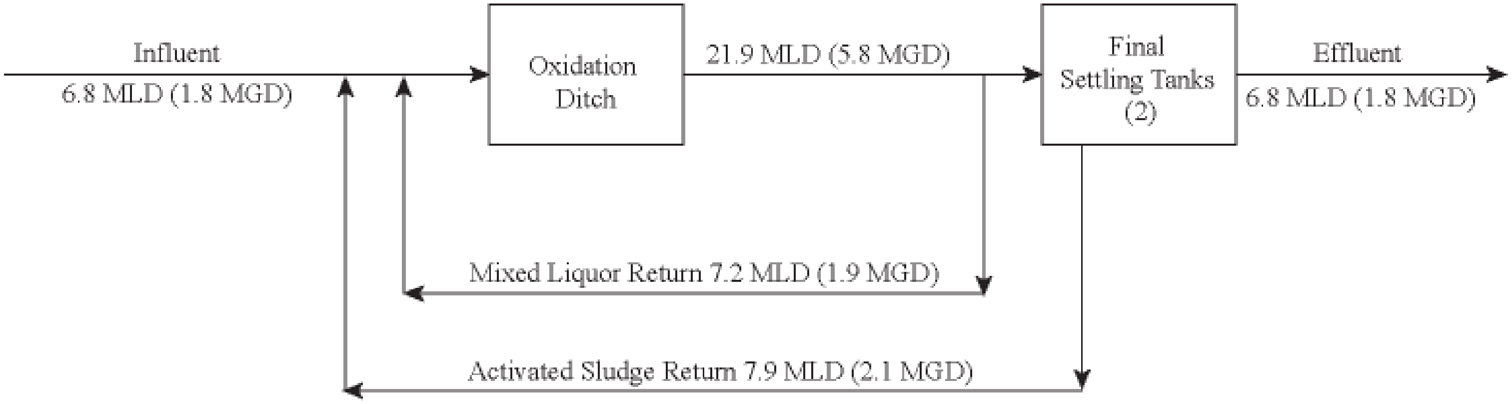

Figure 2.3. Flow schematic of the SBWPCP. The flow values represent average flows, rather than design flows. Average flows for the activated sludge and mixed liquor returns were determined in a June 2006 survey of the plant and represent the most accurate data available. Changes have been made since the plant was designed (E. Brewer, Suffolk County Department of Public Works, personal communication, 2010). Sludge removal processes are not included in this diagram.

Chapter 3

Medically-derived ^{131}I in the tidal Potomac River

1. Abstract

Iodine-131 ($t_{½}$ = 8.04 d) has been measured in Potomac River water and sediments in the vicinity of the Blue Plains Water Pollution Control Plant (WPCP), Washington, DC, USA. Concentrations measured in sewage effluent from Blue Plains WPCP and in the Potomac River suggest a relatively continuous discharge of this isotope. The range of ^{131}I concentrations detected in surface water was 0.076 ± 0.006 to 6.07 ± 0.07 Bq L^{-1}. Iodine-131 concentrations in sediments ranged from 1.3 ± 0.8 to 117 ± 2 Bq kg^{-1} dry weight. Partitioning in the sewage effluent from Blue Plains and in surface waters suggests that ^{131}I is associated with colloidal and particulate organic material. The behavior of medically-derived ^{131}I in the Potomac River is consistent with the behavior of natural iodine in aquatic environments. It is discharged to the river via sewage effluent, incorporated into particulate material and deposited in sediments. Sediment profiles of solid phase ^{131}I indicate rapid mixing or sedimentation and remineralization on short time scales.

2. Introduction

Iodine-131 is a fission product released from nuclear power plants, during nuclear weapons tests, nuclear fuel reprocessing and weapons production. In most environments, medical use is the more widespread source of ^{131}I. It is the most commonly used radiopharmaceutical in nuclear medicine for therapeutic purposes, used to treat hyperthyroidism and thyroid cancer. The number of diagnostic procedures using ^{131}I far exceeds the number of therapeutic treatments; however the latter represents a greater potential source to sewerage due to the nature of the treatments (ICRP, 2004). In the United States, patient excreta are exempt from sewer discharge regulations and are therefore released into sewerage (Martin and Fenner, 1997).

The discharge of ^{131}I from WPCPs to the environment has long been recognized (Moss, 1973). Few values of ^{131}I in sewage effluent have been reported (Erlandsson et al., 1989; Fischer et al., 2009; Puhakainen, 1998; Rose, 2003; Smith et al., 2008; Sodd et al., 1975). However, medically-derived ^{131}I has been measured in various environments: surface waters (Howe and Lloyd, 1986; Kleinschmidt, 2009; Smith et al., 2008; Sodd et al., 1975), macroalgae (Howe and Hunt, 1984; Howe and Lloyd, 1986; Marsh et al., 1988; Morita et al., 2010; Puhakainen, 1998; Rose, 2003; Waller and Cole, 1999), sediments (Fischer et al., 2009; Smith et al., 2008). Furthermore, ^{131}I was measured in the atmosphere near municipal sewage incinerators (Kitto et al., 2005a; Kitto et al., 2006; Kitto et al., 2005b). While the occurrence and concentrations of ^{131}I in sewage effluent have been poorly characterized, these data suggest that continuous discharges of the radioisotope may be common. Recent work has suggested the use of medically-derived ^{131}I introduced to aquatic systems via sewage effluent as a tracer (Fischer et al., 2009; Rose, 2003; Smith et al., 2008).

Distributions of naturally occurring iodine in aquatic environments have been well studied and indicate that although iodate (IO_3^-) is the thermodynamically favored species of iodine (Sillen, 1961), iodide (I^-) is found ubiquitously. Furthermore, non-volatile dissolved organic iodine (DOI) has been found to constitute between 9 and 85% of the total iodine pool in freshwater, estuarine and coastal waters (Abdel-Moati, 1999; Gilfedder et al., 2009; Gilfedder et al., 2010; Luther et al., 1991; Oktay et al., 2001; Truesdale, 1975; Wong and Cheng, 1998; Wong and Cheng, 2001). Most work supports biological cycling as the primary mechanism for transformations among the species (Campos et al., 1999; Elderfield and Truesdale, 1980; Truesdale, 1994; Truesdale et al., 2000; Truesdale and Upstill-Goddard, 2003; Waite et al., 2006; Wong, 1995; Wong and Zhang, 1992) and incorporation into organic phases, including particulate matter (Bors et al., 1991; Lieser and Steinkopff, 1989; Muramatsu et al., 1990a; Muramatsu et al., 1990b; Sheppard and Hawkins, 1995; Whitehead, 1974b). While the mechanisms responsible for these transformations are largely unknown, there is general agreement that is incorporated into particulate matter and subsequently deposited in sediments. Once in sediments, it is subject to diagenetic reactions. Solid phase iodine generally shows decreasing concentrations with depth (Kennedy and Elderfield, 1987; Mackin et al., 1988; Price et al., 1970; Shishkina and Pavlova, 1965; Upstill-Goddard and Elderfield, 1988) and preferential loss with respect to carbon (Pedersen and Price, 1980; Price et al., 1970; Ullman and Aller, 1983). Experimental evidence indicates that it is released to pore water during organic matter decomposition (Ullman and Aller, 1980; Ullman and Aller, 1983; Ullman and Aller, 1985; Upstill-Goddard and Elderfield, 1988).

The objectives of this study were: 1) to characterize the occurrence and concentrations of ^{131}I in sewage effluent at three WPCPs in the Washington, DC metropolitan area discharging to the tidal Potomac River, 2) to examine the behavior of ^{131}I in the tidal Potomac River and 3) to evaluate the potential utility of ^{131}I as a tracer of biogeochemical processes in aquatic environments receiving sewage effluent discharges. This work presents concentrations of medically-derived ^{131}I in sewage effluent, surface waters and sediments in the tidal Potomac River in the vicinity of Blue Plains WPCP. A suite of geochemical data is used to understand the geochemistry of ^{131}I in aquatic environments in the context of natural iodine behavior.

3. Study Sites

Tidal Potomac River

The Potomac River is a tributary of the Chesapeake Bay that discharges to the Atlantic Ocean. The tidal Potomac River stretches from approximately Chain Bridge in Washington, DC to the Chesapeake Bay, a distance of about 185 km. The tidal Potomac River has three divisions: 1) estuary, 2) transition zone and 3) tidal river (Figure 3.1). The Potomac Estuary extends about 74 km from the Chesapeake Bay to Morgantown, MD. It is classified as a partially mixed estuary and is typically brackish. The range of the transition zone is approximately 56 km from Morgantown, MD to Quantico, VA. The salinity transition occurs in this region with salinities from 0 to 18 PSU. The tidal river zone spans a distance of approximately 56 km from the head-of-tides, near Chain Bridge to approximately Quantico, VA. The mean tidal range in this area is approximately 0.9 m and is fresh throughout. The average depth is about 3 m with a narrow channel ranging in depth from 7 to 21 m (Callender et al., 1984; Glenn, 1988; Shultz, 1989). Maximum flood and ebb current velocities are ~ 35 cm s^{-1} at Alexandria, VA (Callender and Hammond, 1982).

This study focused on the region of the tidal river from Reagan National Airport to the Woodrow Wilson Bridge (Figure 3.2). There are three WPCPs discharging to this area: Blue

Plains, Arlington and Alexandria. Water depths in the sampling area are generally 2 to 3 m at mean lower low water (MLLW) and there is a channel running through it that reaches depths of 10 m MLLW. Salinities are zero. Water and sediment samples were collected from four shallow water stations (2 to 3 m MLLW). Water samples were also collected from Blue Plains outfall 002 (Figure 3.2). For most sampling events, the Blue Plains outfall 002 was sampled at or near low water and the sewage outfall discharge was clearly visible. Station 2 was within approximately 50 m of the visible outfall.

Blue Plains WPCP

The Blue Plains WPCP (DC Water) is a tertiary treatment facility located in Washington, DC (Figure 3.2). The plant's service area includes 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. There are 24 hospitals in the Blue Plains service area (E. Wilson, DC Water, personal communication, 2010). The average flow is approximately 1,400 x 10^6 liters per day (MLD) or 370 x10^6 gallons per day (MGD). 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 sewers, which combine sanitary and storm sewers. Outfall 001 is used only during times of bypass overflows and was not sampled in this investigation.

Arlington WPCP

The Arlington WPCP (Arlington County) is a tertiary treatment facility located in Arlington, VA and serves most of Arlington County, parts of Fairfax County and the cities of Alexandria and Falls Church, VA. There is one hospital in the service area (P. Loar, Arlington County, personal communication, 2009). The average flow is approximately 113 MLD (30 MGD). During the sampling period, the plant was undergoing construction for increased capacity and upgrades in treatment. Sewage effluent is discharged to Four Mile Run, a tributary of the Potomac River (Figure 3.2).

Alexandria WPCP

The Alexandria WPCP (Alexandria Sanitation Authority) is a tertiary treatment facility located in Alexandria, VA and serves the City of Alexandria and parts of Fairfax County, VA. There are two hospitals in the service area (L. Gebremedhin, Alexandria Sanitation Authority, personal communication, 2010). The average flow is approximately 136 MLD (36 MGD). The effluent is discharged to Hunting Creek, a tributary of the Potomac River (Figure 3.2).

4. Methods

Sewage Effluent

Sewage effluent was collected as grab samples at the WPCPs just before discharge from the plant. Samples from Blue Plains were collected from outfall 002 just beyond the effluent monitoring station. At the Arlington plant, the samples were collected from head of the sluice just before discharge. At the Alexandria plant, all samples were collected from the end of the chlorine contact tank just before it flows over the weir.

All samples were vacuum filtered through 0.7 µm glass fiber filters (GFF). 150 mL of the filtrate were placed in a straight-side polypropylene (PP) jar (64 mm height; 64 mm diameter) for γ analysis with no additional treatment.

Surface Water

Surface water samples for determination of ^{131}I were collected in April, June, August and November 2009 from stations 1 through 4 and Blue Plains outfall 002, and in February 2009 from station 2 only. All surface water samples were collected in 10 L PP carboys. Initial sample volume was recorded. Sample volume was reduced by heating in evaporating dishes for approximately 12 hours. The volume of concentrated sample was recorded. Any residue remaining in the dish was rinsed into the sample using 2.5% HNO_3. The added volume was accounted for when calculating the concentration. Typical concentration factors were ~ 20 – 40 (Appendix D). The sample was vacuum filtered through a 0.7 µm GFF. 150 mL of the filtrate was placed in a straight-side PP jar (64 mm height; 64 mm diameter) for γ analysis.

Three 20 L surface water samples were collected in June 2010 at station 4. Various pre- and post-heating filtration steps indicated that differences in the treatments were small and can mostly be accounted for by counting errors. The data suggest that, if there is any effect, the values measured in this work represent minimum values (6 to 8% lower). See Appendix D for the results and a detailed description of the treatments.

Surface water collected for total suspended solids (TSS) determination was vacuum filtered through a pre-weighed 0.4 μm polycarbonate membrane filter, dried and re-weighed. The weight of the solids was obtained by difference.

Water samples for nutrient analyses were collected in HCl cleaned, low density polyethylene (LDPE) bottles and vacuum filtered through 0.7 µm GFF filters. An aliquot of the filtrate was frozen for nutrient analysis. Dissolved inorganic nitrogen (NO_3^- +NO_2^- and NH_4^+) and PO_4^{3-} 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 5%.

A 0.7 μm GFF filter was retained for chlorophyll a analysis. Several drops of saturated $MgCO_3$ solution were applied to the filter and frozen until analysis. After a 24 hour acetone extraction, chlorophyll a concentrations were determined with a Turner 10-AU fluorometer.

All materials used to collect and process samples for dissolved organic carbon (DOC) analysis were pre-combusted at 500 ˚C. Surface water samples were collected in glass Wheaton jars. The samples were vacuum filtered using 0.7 µm GFF filters and placed in glass vials fitted with Teflon coated septa, acidified using H_3PO_4 and refrigerated until analysis. Prior to analysis, the samples were sparged using He gas.

Concentration of DOC was determined with an OI Analytical 1010 TOC analyzer using the wet chemical oxidation method described by Osburn and St-Jean (2007). DOC concentrations were normalized to a calibration curve generated for each run using potassium hydrogen phthalate (KHP) standard solutions.

Surface waters and suspended solids were collected for ^{131}I determination from stations 1 through 4 and Blue Plains outfall 002 in April 2010 and from station 4 in June 2010. Pre-cut (82 mm height, 62 mm outer diameter and 27 mm inner diameter), PP Absolute.Za™ cartridge filters were fit into a filter housing using a spacer. More than 100 L of surface water were pumped through the filters using a sump pump. Approximately 20 L of the filtrate were pumped into pre-cleaned (NaOH and HCl) PP carboys. 500 mL of the filtrate were further filtered through 0.2 µm membrane filters to insure that no significant leakage had occurred.

The water samples collected in April 2010 were further partitioned using a custom-made large volume tangential flow ultrafiltration (TFF) system, which uses a polyethersulfone membrane and recirculation to separate and concentrate DOC > 1kDa (Boyd and Osburn, 2004). Before running the samples, the system was cleaned with 20 L of 0.5 N HCl and 0.5 N NaOH. Rinses between samples using low DOC water were at background DOC concentrations. DOC concentrations were determined as described above. Iodine-131 was measured in the permeate (< 1 kDa) and retentate (0.5 µm – 1kDa).

Sediment

Sediments were collected using a hand-deployed, messenger-activated gravity corer with a 7 cm inner diameter polycarbonate barrel (Smith, 2007). Cores were sectioned onboard. Approximately five cores were composited at each site to obtain sufficient mass for γ analysis. In February 2009, only the top 2 cm of sediment from station 2 were collected. Sediment cores were collected from each of the four stations in April, June, August and November 2009 and April 2010. In April 2009, sediments were collected to 5 cm depths at 1 cm intervals. In June, August and November, sediments were sectioned at 1 cm intervals to 10 cm. In April 2010, the cores from stations 2 and 4 were sectioned at 2 cm intervals to 10 cm. At stations 1 and 3, the top 2 cm were taken and then the cores were sectioned at 1 cm intervals to 10 cm. All sediments were dried at 80˚C and homogenized using a mortar and pestle. Spike experiments indicated that there was no loss of ^{131}I (added in the form of ^{131}I^{-}) during heating at 80 ˚C. Approximately 30 g of sediment were packed into pre-weighed, straight-side 30 mL PP jars (41 mm height; 31 mm diameter) for γ analysis. Wet and dry masses of sediments collected in April 2010 were obtained to determine dry bulk density of these sediments. In April 2010, a pore water sample was separated from the top 2 cm of sediment collected from station 3 by centrifugation and vacuum filtered through a 0.7 µm GFF filter. 150 mL of the filtrate were counted as described above for water samples with no further treatment.

Sediment organic carbon (OC) concentrations were determined using a Costech Elemental Analyzer. 15 to 20 mg of dry, homogenized sediment were analyzed in triplicate. Samples were acidified with excess 10% HCl and dried in a drying oven at 60˚C prior to analysis. A concentration calibration curve was generated for each run using an acetanilide standard. Check standards (acetanilide, USGS 40 and IAEA C8) were analyzed throughout each run. Carbon concentrations of the check standards were no greater than 3% difference from the actual value. Values are reported with 1σ errors of replicate measurements.

Gamma-ray Spectrometry

Iodine-131, ^{7}Be ($t_{½}$ = 53.2 d) and ^{210}Pb ($t_{½}$ = 22.3 y) were determined by γ-ray spectrometry using Canberra low energy germanium detectors. The activity of ^{131}I was measured using the emission at 364.5 keV. The sediment sample collected in February 2009 was counted twice to confirm that the source of 364.5 keV peak was ^{131}I. Previous work demonstrated that there is no self-absorption of γ-rays emitted at 364.5 keV by concentrated sewage sludge samples and therefore no self-absorption correction was applied to the data (Rose, 2003). Samples were counted for one day or more depending on activity of the samples. Due to the relatively short half-life of ^{131}I, activities were corrected to account for decay during data acquisition as described in Hoffman and Van Camerik (1967).

The counting efficiency of each detector at 364.5 keV for each geometry used in this investigation was determined using a certified ^{131}I standard solution. For the jar geometries (30 and 150 mL), deionized water was spiked with a ^{131}I standard solution and counted three times on each detector. For the suspended solids collected on the cartridge filters, the ^{131}I standard solution was applied to three pre-cut PP (82 mm height, 62 mm outer diameter and 27 mm inner diameter) cartridge filters and each filter was counted three times on each detector. In each case, the mean counting efficiency of replicate analyses was used to calculate sample activities.

All sediment samples were recounted after two weeks or more to allow sufficient counting time and to ensure secular equilibrium between ^{222}Rn and ^{214}Pb, precursors to ^{210}Pb in the natural ^{238}U decay series. Lead-210 activities supported by the decay of its effective parent isotope ^{226}Ra (supported ^{210}Pb) were measured by the activity of ^{214}Pb using the emission at 352.0 keV. Total ^{210}Pb activities were determined using the 46.5 keV peak. Self-absorption of γ-rays at 46.5 keV by a sample is a function of sample matrix (i.e., density) and requires a correction to be applied to the counting efficiency (Cutshall et al., 1983). The correction applied to the data here was determined for each detector by counting ^{210}Pb standards of known activity and varying density in the same geometry as the samples. The transmission of an ^{241}Am (59.5 keV) standard source through the standard relative to an empty jar (transmission ratio) was determined for each standard. The linear relationship between efficiency (C/A) versus the transmission ratio is specific to each detector and is described by:

$$\frac{C}{A} = m\left(\frac{T}{T_o}\right) + b \qquad (1)$$

where C = the measured count rate of standard or sample (counts per second), A = activity of the standard or sample (Bq), m = linear slope, T = transmission through the standard or sample (counts), T_o = transmission through empty counting jar (counts) and b = y-intercept. Equation 1 was applied to the data to calculate the activity of ^{214}Pb and therefore the supported ^{210}Pb activity using the transmission ratio and count rate determined for each sample (Cochran et al., 1998; Cutshall et al., 1983; Feng et al., 1998). The ^{210}Pb standards were used to determine the efficiency of each detector at 46.5 and 352.0 keV. Excess ^{210}Pb activities were estimated by difference between total and supported values. The reported errors for excess ^{210}Pb reflect the 1σ counting errors for both the 46.5 and 352.0 keV peaks.

The counting time for ^{131}I analysis was not always sufficient to obtain an analytically significant peak for ^{7}Be (477.6 keV). In these cases, the ^{7}Be data were obtained during the recounts for ^{210}Pb. The counting efficiency at 477.6 keV of each detector in the sample geometry was determined using the γ-ray emissions of radioisotopes in IAEA-300 and IAEA-375 reference standards by interpolation using efficiencies of the standards between 200 and 662 keV (Renfro, 2010).

Multi-day continuum background counts of the sample matrix were determined for each detector at 364.5 keV and 477.6 keV. Counting time for each sample and the mean background were used to determine limit of detection (L_D) as described by Currie (1968). The L_D for ^{131}I was ≤ 0.4 Bq L^{-1} for sewage effluent, ≤ 0.03 Bq L^{-1} for surface water, ≤ 0.09 Bq kg^{-1} for suspended solids and ≤ 3.6 Bq kg^{-1} for sediments. The L_D for ^{7}Be was ≤ 3.6 Bq kg^{-1} for sediments. All activities are reported for time of collection ± 1σ counting error. Specific activities for solids are reported in units of Bq kg^{-1} dry mass.

5. Results

Sewage Effluent

Iodine-131 concentrations measured in sewage effluent collected at the Blue Plains WPCP on different days between January 2009 and May 2010 ranged from 0.9 ± 0.1 to 8.1 ± 0.2 Bq L^{-1} (n = 11); one sample < L_D.

Iodine-131 concentrations measured in sewage effluent collected at the Arlington WPCP on different days between October 2009 and May 2010 ranged from 0.5 ± 0.1 to 2.7 ± 0.1 Bq L^{-1} (n = 8); five samples < L_D.

Iodine-131 was detected in one sample of sewage effluent collected at the Alexandria WPCP on different days between October 2009 and June 2010 (0.61± 0.09 Bq L^{-1}), with four samples < L_D. Sewage effluent concentrations are shown in Table 3.1. Sample information, γ-ray spectrometry data and L_D values for sewage effluent samples are reported in Appendix C.

Surface Water

Iodine-131 concentrations measured in surface water collected on five different days between February and November 2009 ranged from 0.076 ± 0.006 to 6.07 ± 0.07 Bq L^{-1} (n = 21); two samples < L_D. The highest concentrations were detected in the Blue Plains outfall 002 and at station 2, adjacent to the outfall (Table 3.2).

Specific activities of ^{131}I measured in suspended solids in April 2010 ranged from 1004 ± 26 to 4178 ± 104 Bq kg^{-1} and < L_D at station 1. The mean concentration of ^{131}I for the three replicates collected in June 2010 was 261 ± 34 Bq kg^{-1}(Table 3.3). Sample information, γ-ray spectrometry data and L_D values for surface water and suspended solids are reported in Appendix D.

Nutrient concentrations in surface water showed little variation among the sampling events and no seasonal trend. NH_4^+ concentrations ranged from 0.013 ± 0.001 to 2.850 ± 0.140 mg N L^{-1} (0.87 ± 0.04 to 172 ± 9 µM); NO_3^- + NO_2^- ranged from 0.39 ± 0.02 to 2.79 ± 0.13 mg N L^{-1} (26 ± 1 to 186 ± 9 µM) and PO_4^{3-} ranged from < 0.001 to 0.045 ± 0.002 mg P L^{-1} (< 0.03 to 1.45 ± 0.07 µM). Chlorophyll a and TSS concentrations were lowest in the outfall and ranged from 0.02 to 16.38 µg L^{-1} and 0.4 to 16.0 mg L^{-1}, respectively. DOC concentrations ranged from 2.3 to 5.2 ppm. The water column characteristics are shown in Table 3.4.

The results of the TFF separations of the April 2010 samples indicate that approximately 70% of the DOC is associated with the colloidal (0.5 µm to 1 kDa) fraction and less so in the sewage outfall. Estimates suggest that a substantial portion (≥ 45%) of ^{131}I in the river and sewage effluent is associated with the colloidal fraction (Table 3.5).

Sediments

Organic carbon concentrations in sediments at station 1 ranged from about 3 to 5%. Station 2 has lower values with most approximately 1 to 2%, but was as high as 5% at 10 cm depth in June 2009. Station 3 and 4 organic carbon concentrations were about 3 to 4% with less variation than observed at stations 1 and 2 (Figure 3.3 and Appendix E).

Iodine-131 was detected in sediments to 5 cm. The concentrations ranged from 1.3 ± 0.8 to 117 ± 2 Bq L^{-1}, with the highest values in surface sediments decreasing with depth (Figure 3.4). At station 1, ^{131}I was restricted to the top 1 to 2 cm and generally at lower concentrations than surface sediments at the other stations. The concentration of ^{131}I in the pore water extracted from the top 2 cm of sediment collected from station 3 in April 2010 was 5.3 ± 0.3 Bq L^{-1}.

Supported ^{210}Pb concentrations are approximately 50 Bq kg^{-1} at each of the stations. Excess ^{210}Pb concentrations show little variation with depth for a given sampling event in the sediments and are generally 100 to 200 Bq kg^{-1}, with lower concentrations at station 2 (Figure 3.5).

Beryllium-7 was detected in sediments at station 1 in April 2010 to 10 cm. For other sampling events, ^{7}Be was restricted to the top 4 cm. Depth profiles generally showed higher concentrations in surface sediments and decreasing values with depth. Concentrations of ^{7}Be ranged from 107 ± 6 to 12 ± 3 Bq kg^{-1} (Figure 3.6). Sample information and γ-ray spectrometry data for sediment samples are reported in Appendix F.

6. Discussion

Sewage effluent is the only known source of ^{131}I to the Potomac River in the sampling area. Iodine-131 was detected in the sewage effluent from all three WPCPs. However, the data presented here indicate that the primary source of ^{131}I to the sampling area is the Blue Plains

WPCP. This is not surprising given the relative size of the drainage area, population and the number of hospitals it serves. While number of hospitals and plant size do not translate into a source of ^{131}I (as was observed in the SBWPCP, a small plant serving a single hospital), the average daily flow of Blue Plains is an order of magnitude higher than the other two plants, and concentrations of ^{131}I detected at Blue Plains suggest a more consistent source.

The distributions of ^{131}I in the surface waters are consistent with a dye study of the Blue Plains sewage effluent plume completed by USGS (Hearn, 1985). The results of their study indicated that the sewage effluent from Blue Plains is mostly confined to the shallow areas just south of the outfall (Figure 3.2), with residence times on the order of days, with little effluent entering the main channel. The low concentrations of ^{131}I measured in surface waters at station 1 agree well with this observation.

Partitioning experiments with TFF indicate that the ^{131}I in Blue Plains sewage effluent and Potomac River surface water is associated with colloidal material (> 1 kDa). The fraction of DOC in the colloidal phase (Table 3.5) was calculated using the following relationship

$$\% \text{ Colloidal} = \left[1 - \frac{\text{DOC}_\text{P}}{\text{DOC}_\text{T}}\right] \text{x } 100 \qquad (2)$$

where DOC_P = concentration of DOC in the permeate (< 1 kDa) and DOC_T = ambient concentration of DOC (< 0.5 µm) (Boyd and Osburn, 2004). The total ^{131}I concentrations were not obtained for this sampling event. However, the fraction of ^{131}I associated with the colloidal phase can be estimated in two ways. First, if DOI is concentrated to the same extent as DOC, the following values emerge for total concentrations of ^{131}I: 2.8, 0.5, 1.3 and 1.8 Bq L^{-1} for stations 2 though 4 and the outfall, respectively. These values suggest that more than half (56 to 72%) of the ^{131}I in the Potomac River is colloidal. This is consistent with the behavior of naturally occuring iodine, where DOI has been shown to constitute between 9 and 85% of the total iodine pool in aquatic environments (Abdel-Moati, 1999; Gilfedder et al., 2009; Gilfedder et al., 2010; Luther et al., 1991; Oktay et al., 2001; Truesdale, 1975; Wong and Cheng, 1998; Wong and Cheng, 2001), drinking water (Andersen et al., 2002) and precipitation (Gilfedder et al., 2008). Furthermore, Radlinger and Heumann (1997; 2000) found iodine associated with humic substances in waste water.

The surface water total ^{131}I concentrations estimated above are reasonable. However, the concentration estimated for station 2 does not reflect previous observations, where surface water concentrations at station 2 were less than the values measured in the outfall. Alternatively, the fraction of ^{131}I in the colloidal phase can be estimated assuming the concentration of ^{131}I in the outfall was the same as in the sewage effluent collected at Blue Plains on the same day (1.4 Bq L^{-1}). The low TSS and chlorophyll a concentrations measured in the outfall indicate little mixing with ambient river water at the time of sample collection (Table 3.4) and support this assumption. Furthermore, the Blue Plains outfall discharge was visibly distinguishable from ambient river water due to the relatively low susupended solids concetrations in the sewage effluent. The outfall sample was collected at low water, which nearly coincides with slack water in this region of the Potomac River. The result of this calculation suggests 45% of the ^{131}I in the outfall is associated with the colloidal phase. The calculation was extended to station 4 assuming a dilution factor of 2 from the outfall. This represents the average dilution between the two stations from the four previous cruises and agrees well with the USGS survey of the sewage outfall plume in the Potomac River (Hearn, 1985). This results in an estimated concentration of 0.7 Bq L^{-1} in the surface water at station 4 (47% ^{131}I associated with the colloidal fraction). Using either approach, calculations suggest that greater than 45% of the ^{131}I in the river and in sewage effluent from Blue Plains is colloidal. Further experiments where ambient ^{131}I concentrations are determined in addition to that which passes through a 1kDa filter would better constrain these values.

Iodine's association with particulate organic matter is well established (Alvarado-Quiroz et al., 2002; Bojanowski and Paslawska, 1970; Bors et al., 1991; Brewer et al., 1980; Calvert et al., 1993; Francois, 1987; Harvey, 1980; Lieser and Steinkopff, 1989; Malcolm and Price, 1984; Pedersen and Price, 1980; Price et al., 1970; Sheppard and Hawkins, 1995; Sheppard and Thibault, 1992; Shishkina and Pavlova, 1965; Ullman and Aller, 1980; Upstill-Goddard and Elderfield, 1988; Vinogradov, 1939; Whitehead, 1973a; Whitehead, 1973b; Whitehead, 1974a; Whitehead, 1974b; Whitehead, 1981). The partition coefficients (K_d) calculated in this investigation suggest an association of ^{131}I with the particulate organic phases in the Potomac River. K_d values estimated using surface sediment (top 1 cm) and surface water concentrations of ^{131}I (Table 3.6) are within the range of values (1 to 10^3 L kg^{-1}) determined by other investigators in laboratory experiments and natural soil and sedimentary environments

(Alvarado-Quiroz et al., 2002; Bird and Schwartz, 1997; Cochran et al., 2000; Muramatsu et al., 1990a; Yoshida et al., 1992).

Similar K_d values were obtained in April 2010 (0.13×10^2 to 2×10^2 L kg^{-1}) using surface sediment (top 2 cm) concentrations and dissolved (< 1 kDa) ^{131}I concentrations. However, the K_d values calculated using the suspended solids concentrations of ^{131}I for the same sampling event are somewhat higher (31×10^2 to 64×10^2 L kg^{-1}). This has been observed by others when using suspended particulate compared to surface sediment concentrations to calculate K_d values (Bird et al., 1995; Milton et al., 1992). While it may be suggestive of DOI retention on filters (Karl et al., 1998; Maske and Garciamendoza, 1994; Moran et al., 1999), DOC concentrations in small volume 0.7 µm GFF filtered samples compared to the large volumes filtered using the 0.5 µm cartridge filters show no evidence of significant DOC loss (Table 3.5). Rather, the higher specific activity of ^{131}I in the suspended particulates suggests a preferential association of iodine with the finer grained, organic material and subsequent dilution by mineral phases upon deposition in the sediments. The lower K_d values at station 2, where the organic carbon content is relatively low, further supports iodine's association with organic material or dilution by a different sediment pool.

Consistent with the behavior of naturally occurring iodine, ^{131}I is deposited in Potomac River sediments. Sediment inventories of ^{131}I (Figure 3.4) were calculated using the relationship

$$I = \sum A_i \rho_i x_i \qquad (3)$$

where I = inventory of radioisotope (Bq cm^{-2}), A_i = concentration of radioisotope of the *i*th interval (Bq g^{-1}), ρ_i = dry bulk density of interval (g cm^{-3}) and x_i = thickness of interval (cm). Assuming steady state, the flux of ^{131}I to the sediments was calculated for each of the profiles using the following equation

$$J = I \lambda \qquad (4)$$

where J = flux to sediments (Bq cm^{-2} d^{-1}), I = inventory of radioisotope (Bq cm^{-2}) and λ = decay constant of radioisotope (d^{-1}). The inventories support the steady state assumption for ^{131}I in the sediments. The average flux of ^{131}I to the sediments was 2.3×10^{-3} Bq cm^{-2} d^{-1}. For the sampling area, this results in 1.1×10^8 Bq d^{-1} of ^{131}I deposited in sediments. The average concentration of ^{131}I detected in sewage effluent from Blue Plains was 3 Bq L^{-1}. Using this value and the average flow of 1400 MLD, an estimated 4.2 x 10^{9} Bq ^{131}I d^{-1} is discharged from Blue Plains. Therefore, approximately 3% of the ^{131}I discharged to the river from Blue Plains is lost to the sediments in the sampling area.

Sediment profiles of ^{131}I observed here indicate that sediments in this region are rapidly accumulating, are mixed by infauna, or that ^{131}I is undergoing diagenetic reactions on weekly to fortnightly time scales. Radioisotopes of varying half-lives can be used to understand the depositional environment by determining sediment accumulation rates and the depth and rate of particle mixing either by physical or biological processes (Cochran, 1985; DeMaster and Cochran, 1982). The distributions of excess ^{210}Pb and ^{7}Be are used here to elucidate the behavior of ^{131}I. Excess ^{210}Pb refers to the atmospherically-derived component that is scavenged by suspended particles and deposited in sediments above that which is supported by decay of its effective parent ^{226}Ra, a member of the ^{238}U decay series (Appleby and Oldfield, 1983; Cutshall et al., 1983; Koide et al., 1972). Beryllium-7 is a cosmogenic radioisotope produced in the atmosphere that is also delivered to Earth's surface via precipitation, sorbed to particles and deposited in sediments (Canuel et al., 1990; Dibb, 1989; Olsen et al., 1986).

The near-constant excess ^{210}Pb through the top 10 cm of the sediments is suggestive of rapid accumulation or mixing relative to the half-life of ^{210}Pb (Figure 3.5). However, in some cases the excess ^{210}Pb concentrations vary substantially among the sampling events at a given station. For example, the excess ^{210}Pb increased from ~ 100 Bq kg^{-1} in August 2009 to > 120 Bq kg^{-1} in November 2009 at station 4. These results indicate that sediment redistribution may be a feature of these sediments on seasonal time scales.

Beryllium-7 at depths of 3 to 4 cm in the sediments further supports rapid mixing or sedimentation on seasonal time scales (Figure 3.6). A comparison of ^{7}Be and ^{131}I gives us insight into both the processes affecting the distributions of iodine in this environment as well as the general physical sedimentary environment.

In particular, mixing can typically be approximated as a diffusion process in surface sediments. Assuming a constant sedimentation rate, porosity and mixing intensity with depth in the sediments, radioisotope distributions can be represented by the general advection-diffusion equation:

$$\frac{\partial A}{\partial t} = D_B \frac{\partial^2 A}{\partial x^2} - \omega \frac{\partial A}{\partial x} - \lambda A \qquad (5)$$

where A = specific activity of the radioisotope (Bq g^{-1}), D_B = sediment mixing coefficient ($cm^2\ d^{-1}$), *x* = depth in the sediments (cm), ω = sediment accumulation rate (cm d^{-1}) and λ = decay constant of the radioisotope (d^{-1}). Assuming steady state, Equation 5 becomes

$$A(x) = A_o \exp\left(\frac{\omega - \sqrt{\omega^2 + 4D_B\lambda}}{2D_B}\ x\right) \qquad (6)$$

When sediment accumulation is small relative to mixing, Equation 6 is further simplified:

$$A(x) = A_o \exp\left(-\sqrt{\frac{\lambda}{D_B}}\ x\right) \qquad (7)$$

(Aller et al., 1980; Guinasso and Schink, 1975; Lecroart et al., 2010). Thus, the D_B can be calculated from the linear relationship between the natural log concentrations of the radioisotope versus depth. Figure 3.7 shows the plots for stations 3 and 4 in June, August and November 2009, when there was sufficient data for both ^{131}I and ^{7}Be. The mixing coefficients derived for these sediments are in the range of those calculated for other environments (Boudreau, 1994; Burdige, 2006; Tromp et al., 1995).

An inverse relationship between half-life and D_B for conservative tracers has generally been observed in sediments (DeMaster and Cochran, 1982; Lecroart et al., 2010; Pope et al., 1996). These profiles may be complicated by size and age selective mixing by deposit feeders, particularly because the presence of ^{131}I represents fresh sedimentary material. However, these phenomena would increase the apparent D_B values, which is not observed here for ^{131}I relative to ^{7}Be. Non-steady state conditions would underestimate D_B values (Cochran, 1985; DeMaster et al., 1991; DeMaster and Cochran, 1982; DeMaster et al., 1985b) and may be the case for ^{7}Be, where inventories show greater variation over the sampling period than ^{131}I, which shows less variation.

If mixing is the dominant process over monthly time scales, the D_B values for ^{131}I are likely underestimated. The time required to generate the observed profiles by mixing alone was estimated using the Einstein-Smoluchowski equation:

$$L^2 = 2D_B t \qquad (8)$$

where L = the mean squared displacement (cm) , D_B = the mixing coefficient (cm^2 d^{-1}) and t = elapsed time (d) (Smith et al., 1993; Wheatcroft et al., 1990). In most cases, more than 40 days (> 5 times the half-life of ^{131}I) would be required to mix ^{131}I to the depths in which it was detected. This suggests the need for another explanation of observed profiles.

A further complication to comparing the ^{7}Be and ^{131}I profiles is that naturally occurring iodine is not conservative upon deposition in sediments. Pore water (Bojanowski and Paslawska, 1970; Kennedy and Elderfield, 1987; Mackin et al., 1988; Martin et al., 1993; Pedersen and Price, 1980; Shishkina and Pavlova, 1965; Ullman and Aller, 1985; Upstill-Goddard and Elderfield, 1988) and solid phase enrichments in surface sediments have been explained by release of iodine to pore water during diagenesis and subsequent interaction with particulate organic matter and mineral phases (Francois, 1987; Kennedy and Elderfield, 1987; Mackin et al., 1988; Price and Calvert, 1973; Price and Calvert, 1977; Ullman and Aller, 1980; Ullman and Aller, 1985; Wakefield and Elderfield, 1985). Elevated pore water concentrations above either estimate for total ^{131}I in surface water in sediments collected from station 3 in April 2010, demonstrate that ^{131}I is being remineralized and perhaps concentrated in surface sediments or lost to the overlying water. In either case, the solid phase ^{131}I profile would appear to have more rapid loss of the isotope and result in underestimates of mixing rates. Such behavior of ^{131}I would be consistent with observed and inferred behavior of natural iodine in sediments. So while rapid mixing or sedimentation may be occurring, ^{131}I is undergoing diagenesis on weekly to fortnightly timescales.

If accumulation dominates over mixing, Equation 6 becomes:

$$A(x) = A_o \exp\left(-\frac{\lambda x}{\omega}\right) \qquad (9)$$

where the terms in the equation are defined above. Therefore, ω can be derived from the linear relationship between the natural log concentration of the radioisotope versus depth in the sediments (DeMaster et al., 1985a; Nittrouer et al., 1984). The plots for both ^{131}I and ^{7}Be are shown in Figure 3.7. Sediment accumulation rates for same profiles were calculated using Equation 9. The values are listed in Table 3.7 and suggest rapid sediment deposition (0.02 to 0.15 cm d^{-1}) at these sites on monthly time scales.

Additionally, the ^{7}Be sediment inventories are elevated above values supported by atmospheric inputs alone. For this region, the atmospherically-supported inventory is expected to be approximately 2 x 10^{-2} Bq cm^{-2} (Canuel et al., 1990; Dibb, 1989; Olsen et al., 1986; Renfro, 2010). In this study, the ^{7}Be inventories exceed the expected input in most cases, with inventories as high as 9.0 x 10^{-2} Bq cm^{-2} (Figure 3.6). The inventories at these sites may be explained by sediment focusing (Dibb and Rice, 1989). Alternatively, direct runoff or combined sewer overflows during heavy rainfall events can deliver additional ^{7}Be to the sediments. In these cases, ^{7}Be accumulated on land surfaces can reach the river during a heavy rainfall, thus, increasing the ^{7}Be input above that which is delivered by the precipitation alone (Renfro, 2010). X-radiographs or benthic community data would help to determine if the sedimentary environment in this region of the Potomac River is dominated by accumulation, physical mixing or mixing by infauna.

7. Conclusions

Iodine-131 is discharged to the tidal Potomac River from the Blue Plains, Arlington and Alexandria WPCPs. Concentrations measured in sewage effluent, surface water and sediment reflect the predominance of Blue Plains as a source of ^{131}I to the study area, the region bound by Reagan National Airport and the Woodrow Wilson Bridge. The data presented here also suggest a continuous source. Iodine-131 measured in surface waters and sediments is consistent with the flow of Blue Plains sewage effluent in the Potomac River as determined by Hearn (1985).

The behavior of ^{131}I in the Potomac River agrees well with the known biogeochemical cycling of naturally occurring iodine in aquatic environments. A significant portion ($\geq$ 45%) of

^{131}I in sewage effluent and Potomac River water is associated with colloidal material. This has been observed for stable iodine in coastal, estuarine and riverine environments.

Iodine-131 is incorporated into particulate phases and deposited in sediments. Flux estimates indicate that 3% of the ^{131}I discharged from Blue Plains is lost to the sediments in the sampling area. Elevated pore water concentrations demonstrate that ^{131}I is remineralized during diagenesis.

Near-constant sediment profiles of excess ^{210}Pb suggest rapid mixing or sedimentation relative its half-life. However, variations in the excess ^{210}Pb profiles among the sampling events indicate that at least the top 10 cm of sediment is likely replaced or removed seasonally. ^{7}Be profiles further constrain the time scale of sedimentation or mixing to monthly time scales. While the ^{210}Pb and ^{7}Be indicate a dynamic sedimentary environment, the ^{131}I profiles indicate an approximate steady state for this radioisotope on monthly time scales.

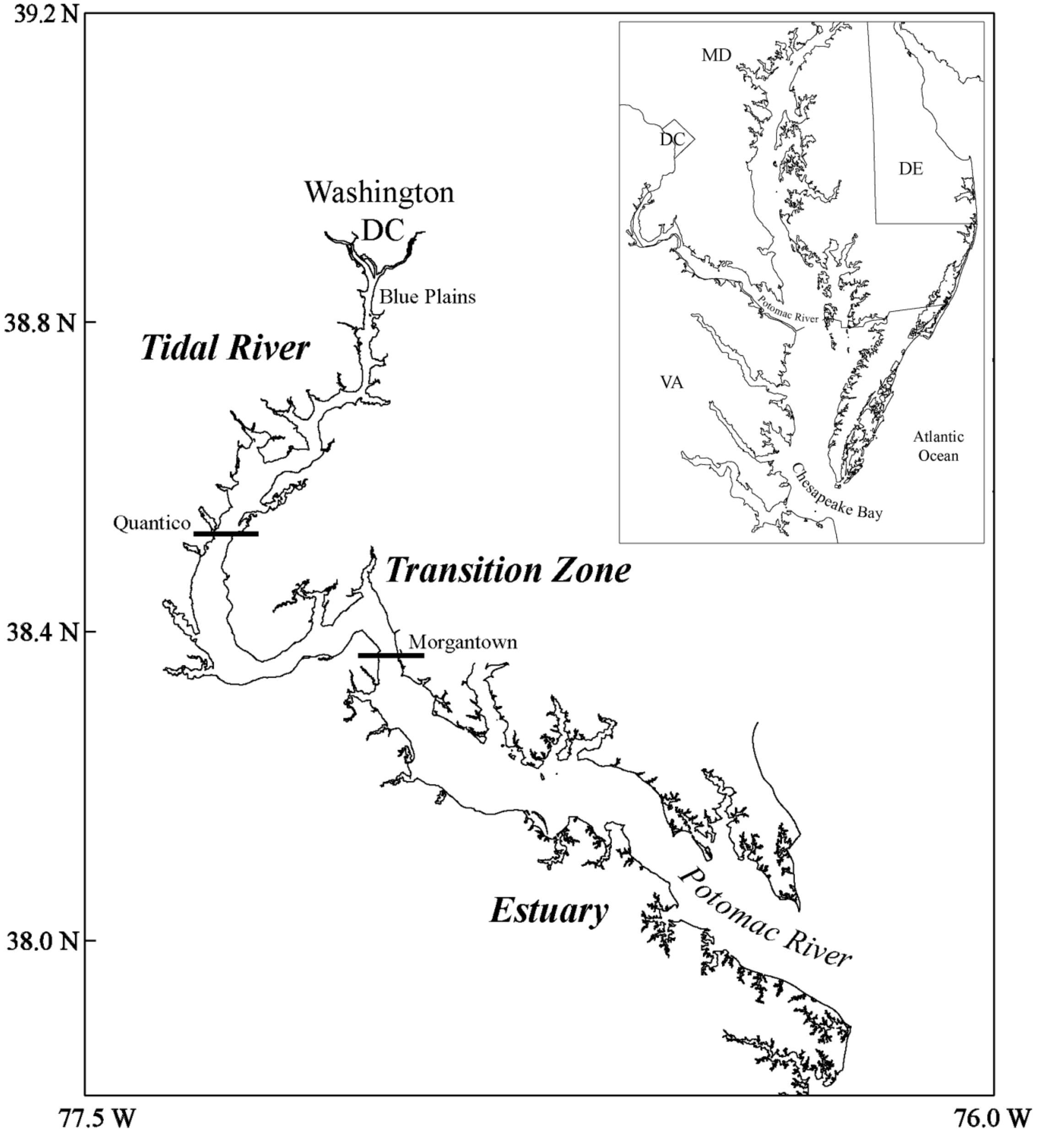

Figure 3.1. Hydrologic divisions of the tidal Potomac River (adapted from Glenn, 1988). The Potomac River is a tributary of the Chesapeake Bay, which discharges to the Atlantic Ocean (inset).

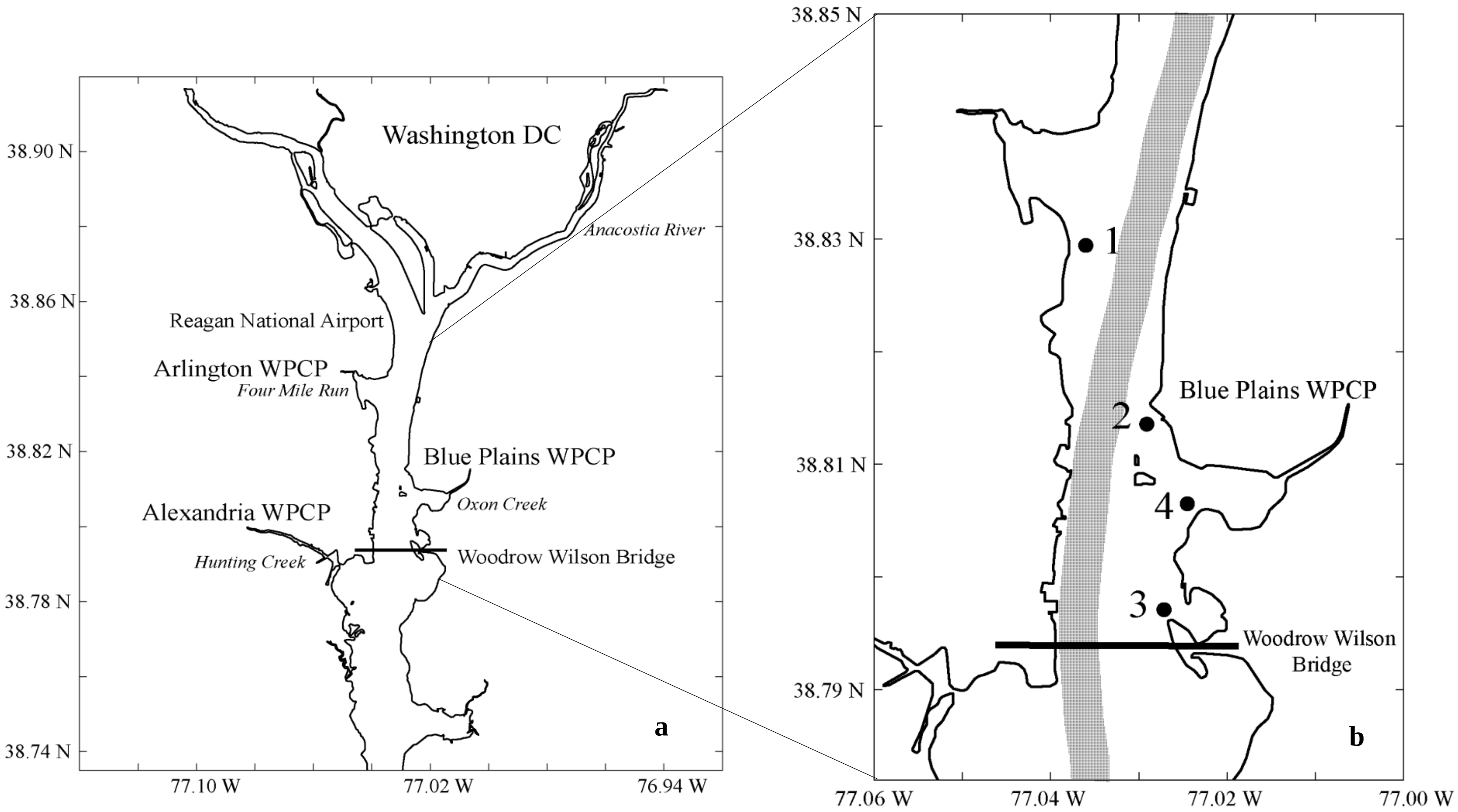

Figure 3.2. a) General sampling area in the tidal Potomac River showing the locations of Blue Plains, Arlington and Alexandria WPCPs. b) Station locations. Shaded area represents the approximate location of the main channel.

Sample #	Sample Collection Date & Time	^{131}I (Bq L^{-1})
Blue Plains		
BP_EFF_012909B	1/29/09 10:15 AM	4.3 ± 0.2
BP050109E-7	5/1/09 8:10 AM	8.1 ± 0.2
BP071509E	7/15/09 2:30 PM	3.9 ± 0.1
BP072209E	7/22/09 1:00 PM	3.7 ± 0.1
BP072909E	7/29/09 2:30 PM	3.1 ± 0.1
BP080509E	8/5/09 2:55 PM	2.3 ± 0.1
BP081209E	8/12/09 2:25 PM	0.9 ± 0.1
BP100709E	10/7/09 10:00 AM	$< L_D$
BP101409E	10/14/09 11:30 AM	1.4 ± 0.1
BP041210E	4/12/10 4:30 PM	1.4 ± 0.2
BP052610E	5/26/10 11:30 AM	3.4 ± 0.2
Arlington		
AR071509E	7/15/09 9:15 AM	$< L_D$
AR072209E	7/22/09 9:30 AM	1.0 ± 0.1
AR072909E	7/29/09 10:30 AM	$< L_D$
AR080509E	8/5/09 9:30 AM	2.7 ± 0.1
AR081209E	8/12/09 9:30 AM	$< L_D$
AR100709E	10/7/09 8:00 AM	$< L_D$
AR101409E	10/14/09 10:00 AM	0.5 ± 0.1
AR052610E	5/26/10 9:30 AM	$< L_D$
Alexandria		
AX052610E	5/26/10 10:30 AM	$< L_D$
AX061610E	6/16/10 9:30 AM	$< L_D$
AX062110E	6/21/10 2:30 PM	$< L_D$
AX100709E	10/7/09 2:00 PM	0.61 ± 0.09
AX101509E	10/15/09 8:30 AM	$< L_D$

Table 3.1. Iodine-131 concentrations in sewage effluent samples collected from Blue Plains, Arlington and Alexandria WPCPs. $< L_D$ = less than detection limit.

Station #	**February 2009**	**April 2009**	**June 2009**	**August 2009**	**November 2009**
1	-	0.100 ± 0.007	< L_D	0.076 ± 0.006	< L_D
2	6.07 ± 0.07	1.51 ± 0.02	0.18 ± 0.01	0.46 ± 0.03	0.59 ± 0.03
3	-	0.61 ± 0.03	0.18 ± 0.03	0.34 ± 0.03	0.35 ± 0.03
4	-	1.52 ± 0.04	0.28 ± 0.01	0.25 ± 0.01	0.37 ± 0.01
Outfall	-	3.77 ± 0.05	0.68 ± 0.02	0.54 ± 0.02	0.62 ± 0.02

Table 3.2. Iodine-131 concentrations (Bq L^{-1}) in surface water collected from stations 1 through 4 and the Blue Plains outfall 002 in the Potomac River. < L_D = less than detection limit. Dash indicates no data were collected.

Sampling Date	**Sample #**	**Mass (g)**	**Volume Filtered (L)**	**^{131}I (Bq kg^{-1})**
April 2010	1	1.221	105.6	$< L_D$
	2	0.408	255.0	3587 ± 93
	3	0.811	107.3	1004 ± 26
	4	1.155	103.5	1139 ± 24
	Outfall	0.332	518.1	4178 ± 104
June 2010	4A	1.538	132.8	300 ± 14
	4B	1.576	136.1	237 ± 12
	4C	1.761	152.1	246 ± 14

Table 3.3. Iodine-131 concentrations in suspended solids collected from stations 1 through 4 and Blue Plains outfall 002 in the Potomac River in April 2010. Replicate samples collected from station 4 in June 2010. Mass of suspended solids on the filters was estimated by multiplying the suspended solids concentration by the volume filtered.

Station #	**NH_4^+ (mg N L^{-1})**	**$NO_3^- + NO_2^-$ (mg N L^{-1})**	**PO_4^{3-} (mg P L^{-1})**	**Chl a (µg L^{-1})**	**TSS (mg L^{-1})**	**DOC (ppm)**
April 2009						
1	0.239	1.17	0.013	1.25	9.2	3.0
2	2.580	1.56	0.014	0.81	13.0	3.6
3	0.714	1.24	0.011	2.32	7.5	3.4
4	1.540	1.33	0.012	2.84	7.5	3.4
June 2009						
1	0.048	1.06	0.023	3.96	12.0	4.3
2	0.064	1.26	0.019	3.98	4.5	4.3
3	0.050	1.07	0.015	5.47	7.3	4.1
4	0.128	0.97	0.006	6.02	11.3	4.2
Outfall	0.036	2.79	0.006	0.56	0.8	4.6
August 2009						
1	0.114	0.76	0.013	12.32	12.7	3.9
2	0.116	1.10	0.006	9.52	14.7	3.9
3	0.069	0.95	0.002	15.54	14.7	5.1
4	0.013	0.92	0.005	16.38	16.0	3.8
Outfall	0.110	1.11	0.003	10.92	0.5	5.2
November 2009						
1	0.047	1.06	0.025	0.48	7.3	4.7
2	0.030	0.50	0.024	0.18	4.7	4.3
3	0.167	1.37	0.028	1.34	9.3	4.5
4	0.148	1.20	0.017	1.13	14.0	4.8
Outfall	0.021	0.39	0.019	0.02	0.4	4.0
April 2010						
1	0.088	1.10	< 0.001	12.6	11.6	2.3
2	0.040	2.26	0.032	1.6	1.6	3.8
3	0.081	1.12	< 0.001	12.5	7.6	2.8
4	0.092	1.10	< 0.001	10.3	11.2	3.1
Outfall	0.029	2.69	0.045	0.2	0.6	-

Table 3.4. Nutrient, chlorophyll a (Chl a), total suspended solids (TSS), and dissolved organic carbon (DOC) concentrations in surface waters collected from stations 1 though 4 and Blue Plains outfall 002 in the Potomac River. Dash indicates no data were collected.

Station #	Permeate (< 1 kDa)		Retentate (0.5 µm – 1 kDa)		Total < 0.5 µm (Cartridge Filter)		< 0.7 µm (GFF)		% Colloidal*	
	^{131}I (Bq L^{-1})	DOC (ppm)	^{131}I (Bq L^{-1})	DOC (ppm)	^{131}I (Bq L^{-1})	DOC (ppm)	^{131}I (Bq L^{-1})	DOC (ppm)	^{131}I (Bq L^{-1})	DOC (ppm)
1	< L_D	0.7	< L_D	7.3	-	2.3	-	2.3		70
2	0.97 ± 0.03	1.4	2.1 ± 0.2	7.3	-	4.0	-	3.8		65
3	0.16 ± 0.01	0.9	1.4 ± 0.2	8.3	-	2.9	-	2.8		69
4	0.37 ± 0.02	0.9	1.3 ± 0.2	8.1	-	3.2	-	3.1	47	72
Outfall	0.77 ± 0.02	1.8	2.2 ± 0.2	10.3	-	4.1	-	-	45	56

*Percent colloidal values were calculated using Equation 2 as described in the discussion on page 39

Table 3.5. Iodine-131 and dissolved organic carbon (DOC) concentrations in Potomac River surface water collected in April 2010 at stations 1 through 4 and in the Blue Plains outfall 002. The colloidal fraction was not calculated for stations 1 though 3. Dash indicates no data were collected.

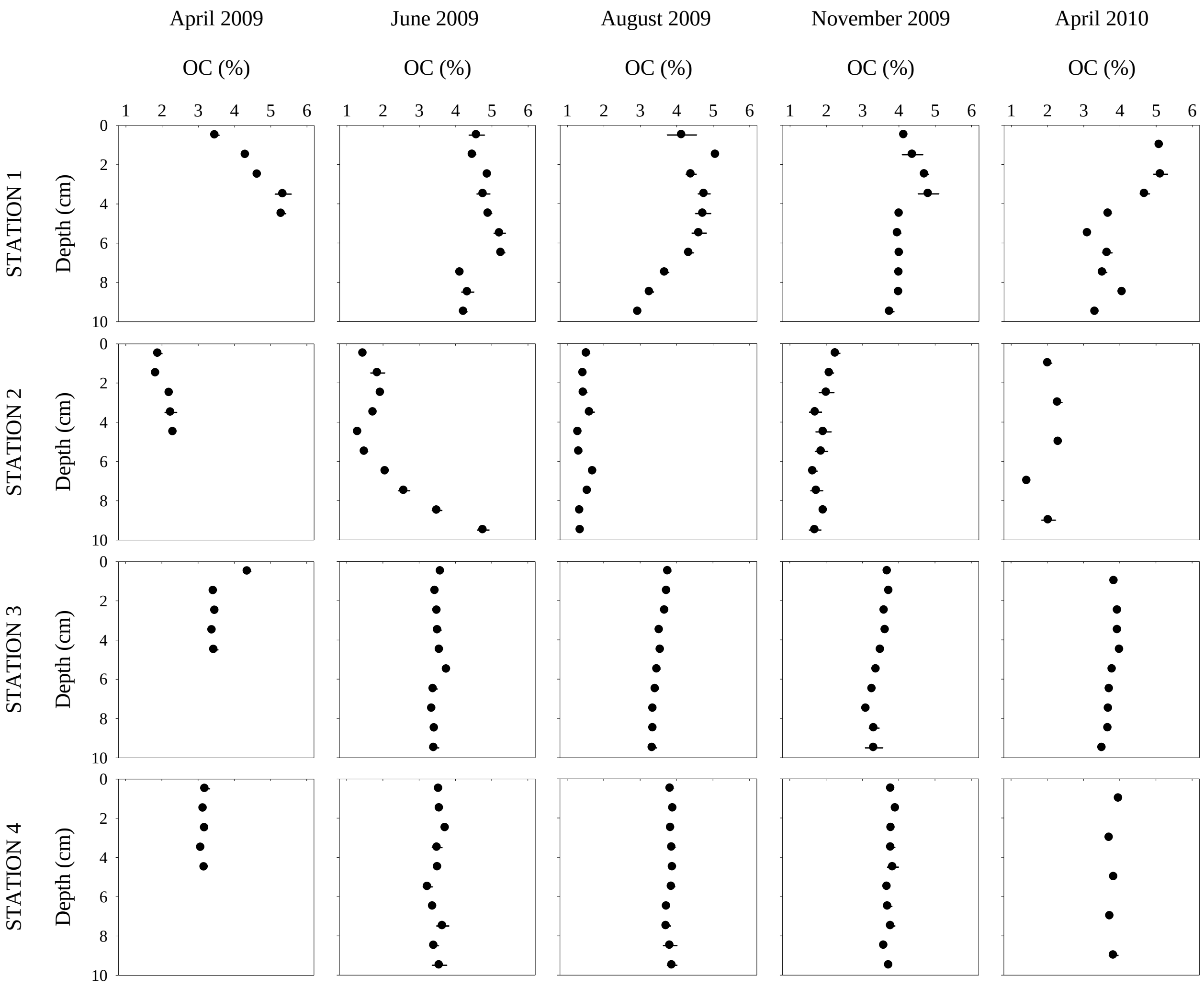

Figure 3.3. Depth profiles of organic carbon content (OC) in sediments collected from stations 1 through 4 in the Potomac River.

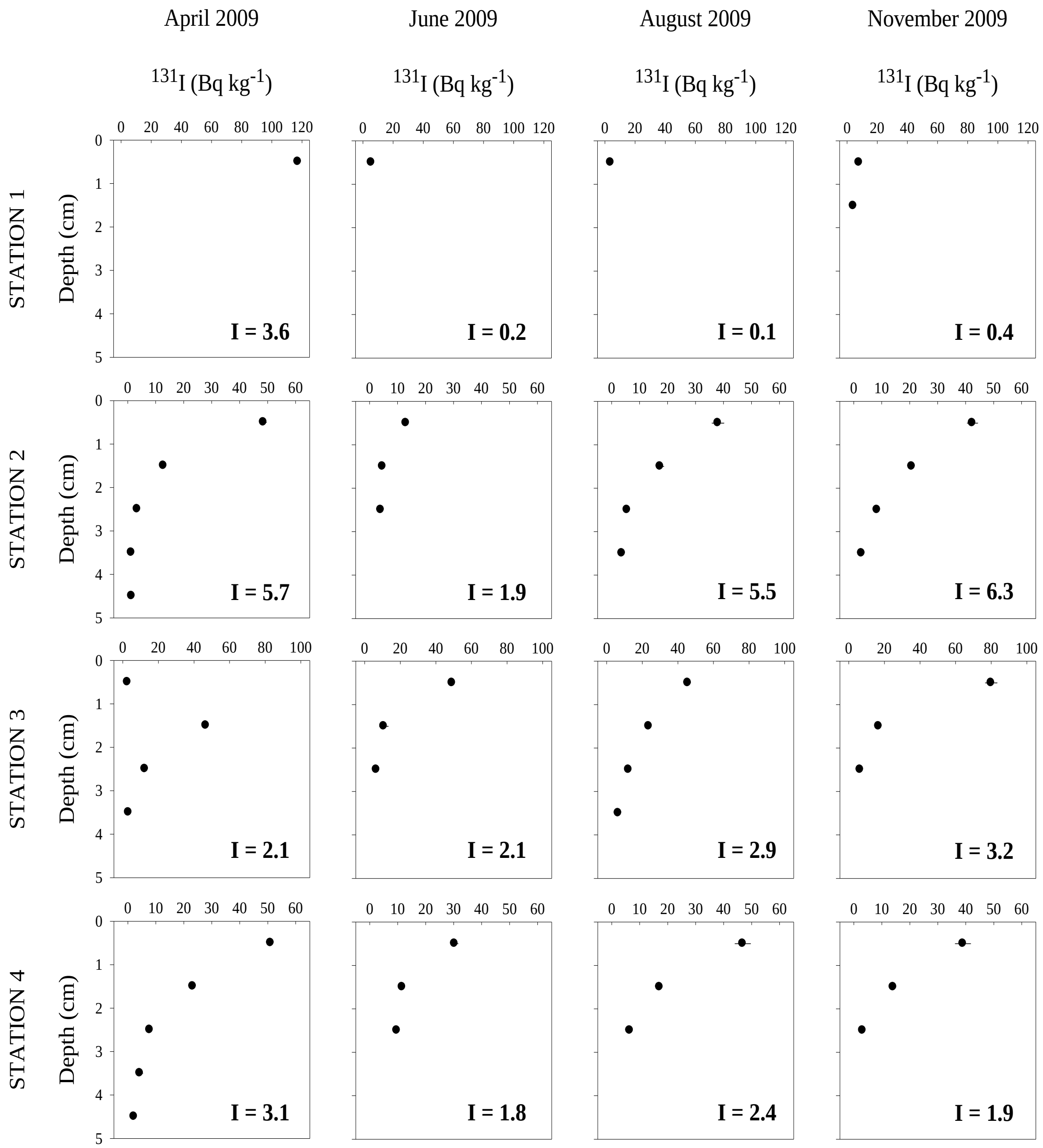

Figure 3.4. Depth profiles of ^{131}I in sediments collected from stations 1 through 4 in the Potomac River. Inventories (I) are shown for each profile in units of 10^{-2} Bq cm^{-2}.

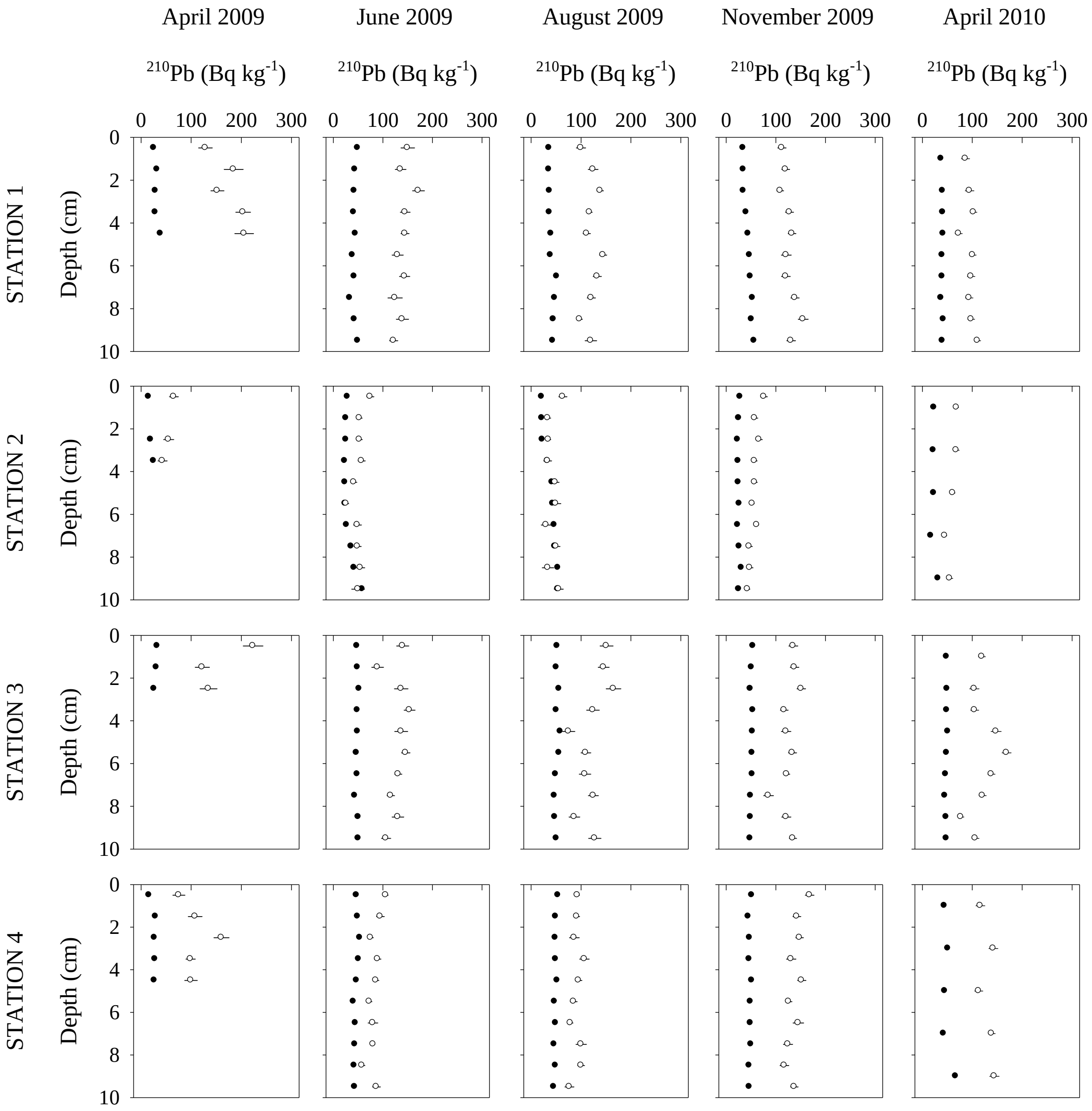

Figure 3.5. Depth profiles of supported ^{210}Pb (●) and excess ^{210}Pb (○) in sediments collected from stations 1 through 4 in the Potomac River.

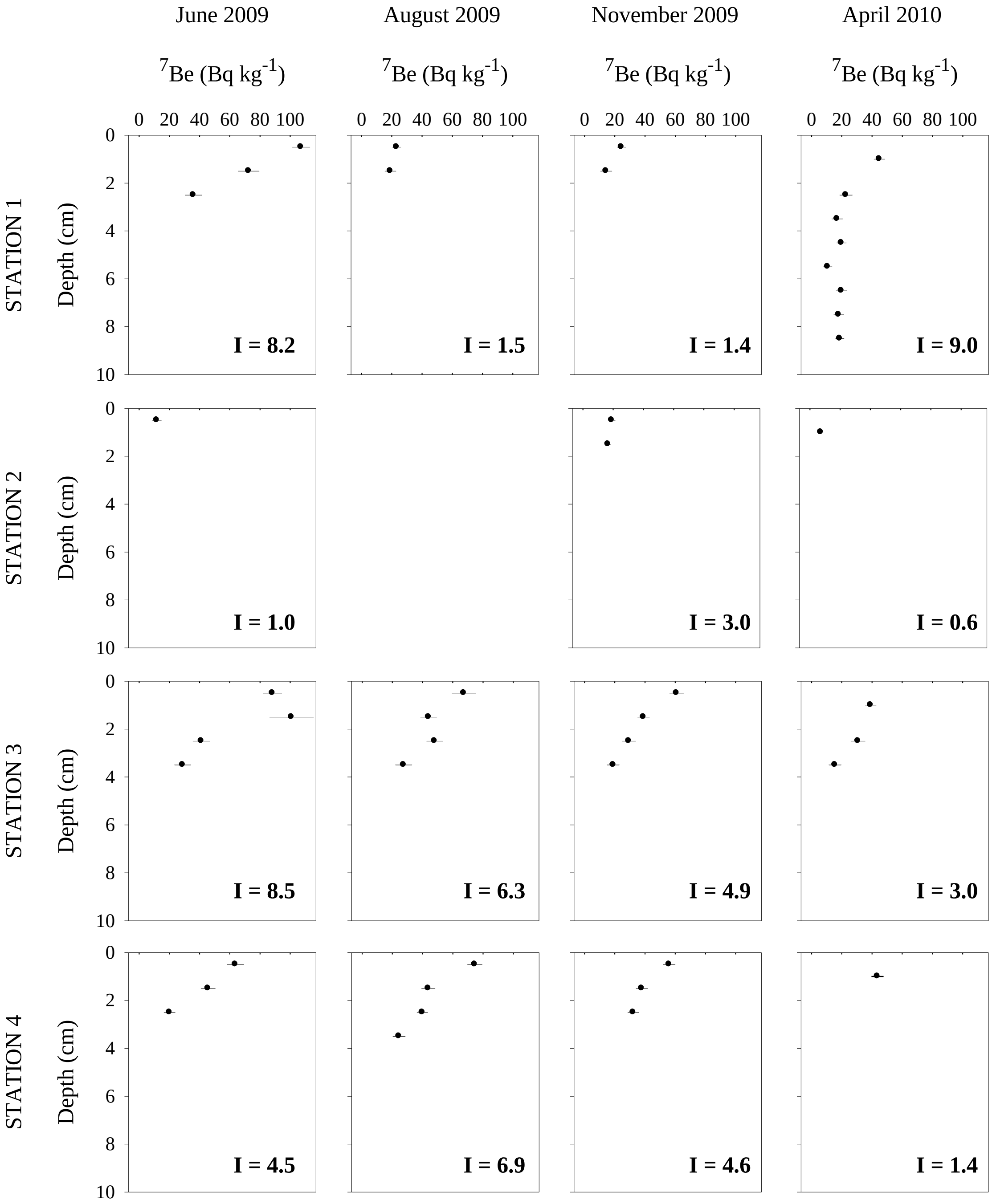

Figure 3.6. Depth profiles of ^{7}Be in sediments collected from stations 1 through 4 in the Potomac River. Inventories (I) are shown for each profile in units of 10^{-2} Bq cm^{-2}. No data are available from station 2 in August 2009.

Date	Station #	^{131}I Surface Sediment (Bq kg^{-1})	^{131}I Surface Water (Bq L^{-1})	K_d (L kg^{-1})
April 2009	**1**	117 ± 2	0.100 ± 0.007	12 x 10^2
	2	49 ± 1	1.51 ± 0.02	0.32 x 10^2
	3	3 ± 1	0.61 ± 0.03	0.04 x 10^2
	4	51.0 ± 0.8	1.52 ± 0.04	0.34 x 10^2
June 2009	**1**	5.5 ± 0.8	$< L_D$	-
	2	13.0 ± 0.8	0.18 ± 0.01	0.71 x 10^2
	3	49 ± 1	0.18 ± 0.03	2.7 x 10^2
	4	30 ± 1	0.28 ± 0.01	1.1 x 10^2
August 2009	**1**	3.7 ± 0.6	0.076 ± 0.006	0.49 x 10^2
	2	38 ± 2	0.46 ± 0.03	0.83 x 10^2
	3	46 ± 2	0.34 ± 0.03	1.3 x 10^2
	4	47 ± 3	0.25 ± 0.01	1.8 x 10^2
November 2009	**1**	7.9 ± 0.7	$< L_D$	-
	2	42 ± 2	0.59 ± 0.03	0.72 x 10^2
	3	80 ± 3	0.35 ± 0.03	2.3 x 10^2
	4	39 ± 3	0.37± 0.01	1.1 x 10^2

Table 3.6. Iodine-131 concentrations in surface sediments (0 to 1cm) and surface water collected from stations 1 through 4 in the Potomac River. $< L_D$ = less than detection limit. Dash indicates no data were collected.

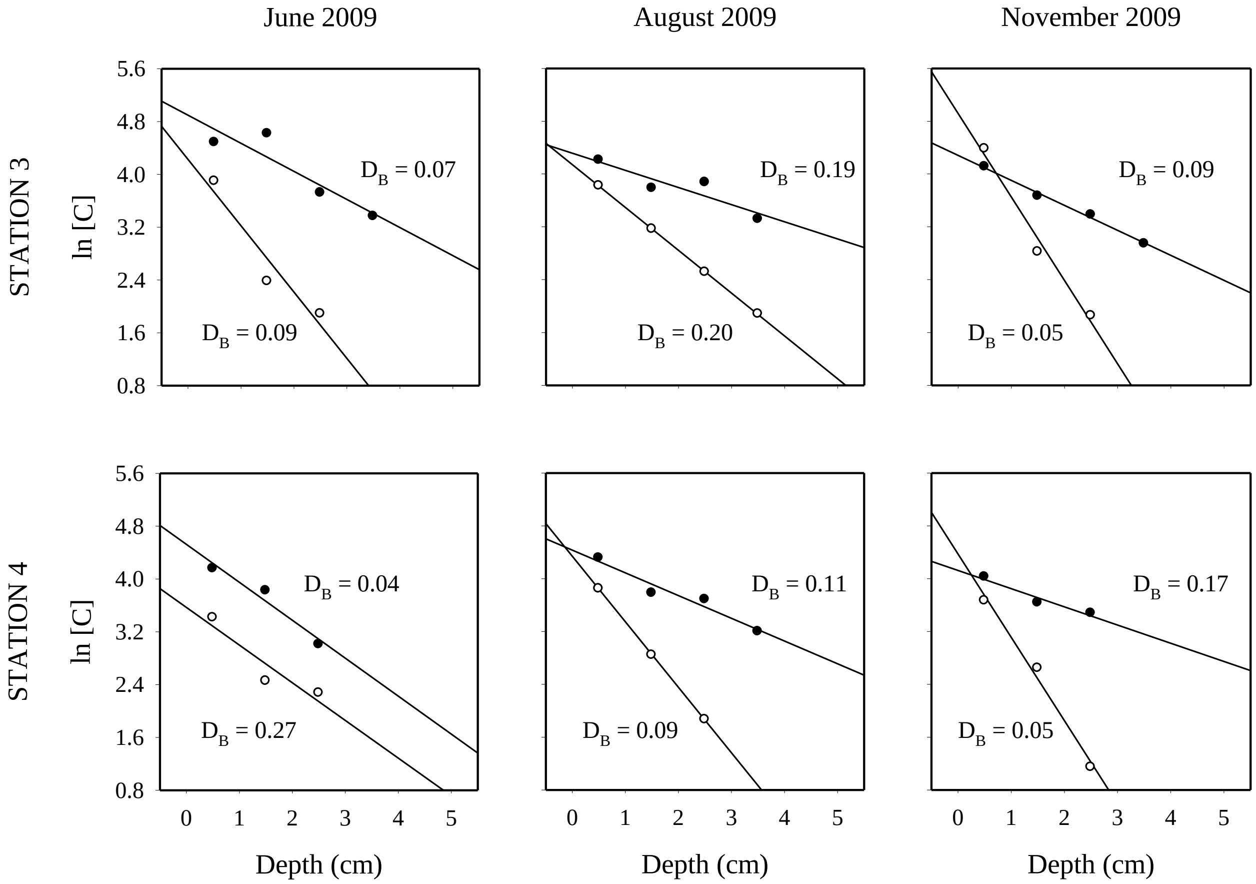

Figure 3.7. Natural log concentrations of ^{7}Be (●) and ^{131}I (○) versus depth in sediments collected from stations 3 and 4 in the Potomac River. Mixing coefficients (D_B) calculated from linear plots are shown in units of cm^2 d^{-1}.

Station #	**June 2009**		**August 2009**		**November 2009**	
	^{131}I	**^{7}Be**	**^{131}I**	**^{7}Be**	**^{131}I**	**^{7}Be**
3	0.09	0.03	0.13	0.05	0.07	0.03
4	0.15	0.02	0.09	0.04	0.07	0.05

Table 3.7. Sediment accumulation rates (cm d^{-1}) at stations 3 and 4 in June, August and November 2009 determined using ^{131}I and ^{7}Be profiles. These values were calculated using Equation 9 and the corresponding natural log concentration versus depth profiles shown in Figure 3.7.

Chapter 4

Medically-derived ^{131}I: A potential tool for understanding the fate of wastewater nitrogen in aquatic environments

1. Abstract

Iodine-131 ($t_{½}$ = 8.04 d) has been measured in Potomac River water and sediments in the vicinity of the Blue Plains water pollution control plant (WPCP), Washington, DC, USA. Dissolved ^{131}I showed a strong, positive correlation with δ^{15}N values of nitrate in the river. The range of ^{131}I concentrations detected in surface waters was 0.18 ± 0.01 to 0.68 ± 0.02 Bq L^{-1}. Surface water $\delta^{15}NO_3^-$ values ranged from 8.7 ± 0.3 to 33.4 ± 7.3‰; NO_3^- + NO_2^- concentrations were 0.39 ± 0.02 to 2.79 ± 0.13 mg N L^{-1} (26 ± 1 to 186 ± 9 µM). Sediment profiles of particulate ^{131}I and δ^{15}N indicate rapid mixing or sedimentation and in many cases remineralization of a heavy nitrogen source consistent with wastewater nitrogen. Iodine-131 concentrations in sediments ranged from 2.8 ± 0.3 to 80.0 ± 0.3 Bq kg^{-1}dry weight. Values of δ^{15}N in sediments ranged from 4.7 ± 0.1 to 9.3 ± 0.1‰. This work introduces ^{131}I as a potentially valuable tool to study the short-term fate of wastewater nitrogen in this system, but its utility as a tracer is not limited to use in the Potomac River.

2. Introduction

Iodine-131 is a fission product released from nuclear power plants, during nuclear weapons tests, nuclear fuel reprocessing and weapons production. However, in most aquatic systems, the source of ^{131}I is medical use. It is the most widely used radiopharmaceutical in nuclear medicine for therapeutic purposes, commonly used to treat hyperthyroidism and thyroid cancer. The number of diagnostic procedures using ^{131}I far exceeds the number of therapeutic treatments (ICRP, 2004); the latter represents a greater potential source to sewage systems. In the United States, patient excreta are exempt from sewer discharge regulations and are therefore released into sewerage (Martin and Fenner, 1997).

Pulses of ^{131}I can remain in a WPCP many days following initial input and can result in a relatively continuous source of this radioisotope to aquatic environments via sewage effluent. Flow recycling, or activated sludge returns, is commonly used to maintain bacterial populations required to achieve sewage treatment. In these plants, the hydraulic retention time (HRT) describes only a portion of the effluent leaving the plant. The sewage half-life, analogous to a radioactive half-life, describes the time it takes for half of a wastewater component to be removed from a WPCP. Sewage half-life, like the HRT, is characteristic of a WPCP. It can be estimated if flow volumes are well characterized and may be far longer than the HRT (Chapter 2).

It is not surprising then that ^{131}I has been measured in various environments: surface waters (Howe and Lloyd, 1986; Smith et al., 2008; Sodd et al., 1975), macroalgae (Howe and Hunt, 1984; Howe and Lloyd, 1986; Marsh et al., 1988; Puhakainen, 1998; Rose, 2003; Waller and Cole, 1999) and sediments (Fischer et al., 2009; Smith et al., 2008). Smith et al. (2008) proposed ^{131}I as a tracer in aquatic systems. With a half-life of 8.04 d, it is useful to study biogeochemical processes occurring on the time scale of up to one month.

In receiving waters, like stable iodine, ^{131}I is biologically cycled and remineralized similar to nitrogen. While the mechanisms for its transformations are not well known, iodine is cycled among three major species: iodate (IO_3^-), iodide (I^-) and dissolved organic iodine (DOI). It is incorporated into particulate matter and deposited in sediments. Iodine generally shows a decrease in solid phase content with depth in sediments (Kennedy and Elderfield, 1987; Mackin et al., 1988; Price et al., 1970; Shishkina and Pavlova, 1965; Upstill-Goddard and Elderfield, 1988) and is released to pore water during organic matter decomposition (Ullman and Aller, 1980; Ullman and Aller, 1983; Ullman and Aller, 1985; Upstill-Goddard and Elderfield, 1988). Decreasing solid phase I/C ratios with depth indicate preferential loss of iodine with respect to carbon during organic matter decomposition (Pedersen and Price, 1980; Price et al., 1970; Ullman and Aller, 1983).

The objective of this work was to investigate the potential of ^{131}I as a tool to study the fate of wastewater nitrogen in the Potomac River in the vicinity of the Blue Plains WPCP. The flux of ^{131}I to the sediments is used to estimate nitrogen deposition in sediments. Isotopic and mass balances of sediment nitrogen are used to estimate the fraction of remineralized nitrogen that is derived from waste water.

3. Study Area

The Potomac River is a tributary of the Chesapeake Bay which discharges to the Atlantic Ocean. The tidal Potomac River stretches from approximately Chain Bridge in Washington, DC to the Chesapeake Bay, a distance of about 185 km. The tidal Potomac River has three divisions: 1) estuary, 2) transition zone and 3) tidal river (Figure 4.1). The Potomac Estuary extends about 74 km from the Chesapeake Bay to Morgantown, MD. It is classified as a partially mixed estuary and is typically brackish. The range of the transition zone is approximately 56 km from Morgantown, MD to Quantico, VA. The salinity transition occurs in this region with salinities from 0 to 18 PSU. The tidal river zone spans a distance of approximately 56 km from the head-of-tides, near Chain Bridge to approximately Quantico, VA. The mean tidal range in this area is about 0.9 m and the salinities are zero. The average depth is about 3 m with a narrow channel ranging in depth from 7 to 21 m (Callender et al., 1984; Glenn, 1988; Shultz, 1989). Maximum flood and ebb current velocities are ~ 35 cm s^{-1} at Alexandria, VA (Callender and Hammond, 1982).

This study focused on a region of the tidal river from Reagan National Airport to the Woodrow Wilson Bridge (Figure 4.2). There are three WPCPs discharging to this area: Blue Plains, Arlington and Alexandria. Sewage effluent concentrations of ^{131}I at each of the three plants indicate that Blue Plains is the predominant source of ^{131}I to the sampling area (Chapter 3). The Blue Plains WPCP (DC Water) is a tertiary treatment facility located in Washington, DC. The plant's service area includes 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. There are 24 hospitals in the Blue Plains service area (E. Wilson, DC Water, personal communication, 2010). The average flow is approximately 1,400 x 10^6 liters per day (MLD) or 370 x 10^6 gallons per day (MGD). 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 sewers, which combine sanitary and storm sewers. Outfall 001 is used only during times of bypass overflows and was not sampled in this investigation.

The Arlington WPCP (Arlington County) is a tertiary treatment facility located in Arlington, VA and serves most of Arlington County, parts of Fairfax County and the cities of Alexandria and Falls Church, VA. The average flow is approximately 113 MLD (30 MGD). During the sampling period the plant was undergoing construction for increased capacity and upgrades in treatment. Sewage effluent from this plant is discharged to Four Mile Run.

The Alexandria WPCP (Alexandria Sanitation Authority) is a tertiary treatment facility located in Alexandria, VA and serves the City of Alexandria and parts of Fairfax County, VA. The average flow is approximately 136 MLD (36 MGD). The effluent is discharged to Hunting Creek.

Water depths in the sampling area are generally 2 to 3 m at mean lower low water (MLLW) and there is a channel running through it that reaches depths of 10 m MLLW. Water and sediment samples were collected at four shallow water stations (2 to 3 m MLLW). Water samples were also collected from Blue Plains outfall 002. For most sampling events, the Blue Plains outfall 002 was sampled at or near low water and the outfall was clearly visible. Station 2 was within approximately 50 m of the visible outfall. Figure 4.2 shows the general sampling area, station locations and locations of the WPCPs.

4. Methods

Surface Water

Surface water samples for determination of ^{131}I were collected in June, August and November 2009 from stations 1 through 4 and Blue Plains outfall 002. Water samples were collected in 10 L PP carboys. The initial sample volume was recorded. Sample volume was reduced by heating in evaporating dishes for approximately 12 hours. The volume of concentrated sample was recorded. Any residue remaining in the dish was rinsed into the sample using 2.5% HNO_3. The added volume was accounted for when calculating the concentration factor. Typical concentration factors were ~ 20 – 40 (Appendix D). The sample was vacuum filtered through a 0.7 µm glass fiber filter (GFF). 150 mL of the filtrate was placed in a straight-side PP jar (64 mm height; 64 mm diameter) for γ analysis.

For total suspended solids (TSS) concentrations in water, the samples were vacuum filtered through a pre-weighed 0.4 µm polycarbonate membrane filter, dried and re-weighed. The weight of the solids was obtained by difference.

Water samples for nutrient analyses were collected in HCl cleaned, low density polyethylene (LDPE) bottles. The samples were vacuum filtered through 0.7 µm GFF filters. An aliquot of the filtrate was frozen for nutrient analysis. Dissolved inorganic nitrogen ($NO_3^- + NO_2^-$ and NH_4^+) and PO_4^{3-} 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 ± 5%. A 0.7 µm GFF filter from preparation of the nutrient samples was retained for chlorophyll a analysis. Several drops of saturated $MgCO_3$ solution were applied to the filter and frozen until analysis. After a 24-hour acetone extraction, chlorophyll a concentrations were determined with a Turner 10-AU fluorometer.

Water was collected for isotopic composition of dissolved nitrate ($\delta^{15}NO_3^-$) in HCl cleaned LDPE bottles and was frozen after collection. Prior to analysis the samples were thawed, vacuum filtered through 0.7 µm GFF filters and acidified with H_2SO_4. The isotopic composition of dissolved nitrate ($\delta^{15}NO_3^-$) was analyzed at the Stable Isotope Laboratory at Boston University using a modified ammonia diffusion method described by Sigman et al. (1997).

Sediment

Sediments were collected using a hand-deployed, messenger-activated gravity corer with a 7 cm inner diameter polycarbonate barrel (Smith, 2007). Sediment cores were collected from each of the four stations in June, August and November 2009. Approximately five cores were composited at each site to obtain sufficient mass for γ analysis. Cores were sectioned onboard at 1 cm intervals to 10 cm. Sediments were dried at 80˚C and homogenized using a mortar and pestle. Spike experiments indicated that there was no loss of ^{131}I (added in the form of $^{131}I^-$) during heating at 80 ˚C. Approximately 30 g of sediment were packed into pre-weighed, straight-side 30 mL PP jars (41 mm height; 31 mm diameter) for γ analysis. Wet and dry mass of sediments collected in April 2010 were obtained to determine dry bulk density of these sediments.

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 (IRMS) and a Conflo III interface. 15 to 20 mg of dry, homogenized sediment were analyzed in triplicate. Total carbon and nitrogen samples were analyzed without treatment. For organic carbon, the samples were acidified with excess 10% HCl and dried in a drying oven at 60˚C prior to analysis. A concentration calibration curve was generated for each run using acetanilide standard. Isotopic calibration curves were generated for $\delta^{15}N$ with isotopic standards: IAEA N2, IAEA NO3, USGS 25 and USGS 40 and for $\delta^{13}C$ with isotopic standards: IAEA C8, IAEA CH6 and USGS 40. Check standards (acetanilide, USGS 40 and IAEA C8) for both concentration and isotopic composition were analyzed throughout each run. Carbon concentrations of the check standards were no greater than 3% difference and $\delta^{13}C$ values were within 0.4‰ for all analyses. Nitrogen concentrations of the check standards were no greater than 6% of actual value. Check standards for $\delta^{15}N$ were within 0.2‰ for all analyses. All stable isotope ratios are reported in units of per mil (‰) using the standard notation

$$\delta^{15}\mathrm{N} \text{ or } \delta^{13}\mathrm{C} = \left[\frac{\mathrm{R_{sample}}}{\mathrm{R_{standard}}} - 1\right] \times 1000 \qquad (1)$$

where R = $^{15}N/^{14}N$ or $^{13}C/^{12}C$ the reference standards are atmospheric nitrogen and Vienna Pee Dee Belemnite, respectively. Values are reported with 1σ errors of replicate measurements.

Gamma-ray Spectrometry

Iodine-131 was determined by γ-ray spectrometry using Canberra low energy germanium detectors. The activity of ^{131}I was measured using the emission at 364.5 keV. A sediment sample collected in February 2009 was counted twice to confirm that the source of 364.5 keV peak was ^{131}I. Previous work demonstrated that there is no self-absorption of γ-rays emitted at 364.5 keV by concentrated sewage sludge samples and therefore no self-absorption correction was applied to the data (Rose, 2003). Samples were counted for one day or more depending on activity of the samples. Due to the relatively short half-life of ^{131}I, activities were corrected to account for decay during data acquisition as described in Hoffman and Van Camerik (1967).

The counting efficiency of each detector at 364.5 keV for each geometry used in this investigation was determined using a certified ^{131}I standard solution. For the jar geometries (30 and 150 mL), deionized water was spiked with a ^{131}I standard solution and counted three times on each detector. In each case, the mean counting efficiency of replicate analyses was used to calculate sample activities.

Multi-day continuum background counts of the sample matrix were determined for each detector at 364.5 keV. Counting time for each sample and the mean background was used to determine limit of detection (L_D) as described by Currie (1968). The L_D for ^{131}I was ≤ 0.03 Bq L^{-1} for surface water and ≤ 3.6 Bq kg^{-1} for sediments. All activities are reported for time of collection ± 1σ counting error. Specific activities for solids are reported in units of dry mass.

5. Results

Surface water ^{131}I concentrations detected range from 0.076 ± 0.006 to 0.68 ± 0.02 Bq L^{-1}. 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 indications at time of sampling strongly suggest that these samples represent sewage effluent from Blue Plains with little dilution.

Nutrient concentrations in surface water showed little variation among the sampling events and no seasonal trend. NH_4^+ concentrations ranged from 0.013 to 0.167 mg N L^{-1} (0.87 ± 0.04 to 172 ± 9 μM); NO_3^- + NO_2^- ranged from 0.39 ± 0.02 and 2.79 ± 0.13 mg N L^{-1} (26 ± 1 to 186 ± 9 μM) and PO_4^{3-} ranged from < 0.002 to 0.028 ± 0.001 mg P L^{-1} (< 0.03 to 1.45 ± 0.07 μM). Chlorophyll a and TSS concentrations were lowest in the outfall and ranged from 0.02 to 16.38 μg L^{-1} and 0.4 to 16.0 mg L^{-1}, respectively. Surface water $\delta^{15}NO_3^-$ values ranged from 8.7 ± 0.3 to 33.4 ± 7.3‰ (Table 4.1).

Iodine-131 was detected in sediments to 5 cm. The concentrations ranged from 2.8 ± 0.3 to 80.0 ± 0.3 Bq kg^{-1} dry weight, with the highest values in surface sediments decreasing with depth. At station 1, ^{131}I was restricted to the top 1 to 2 cm with lower concentrations than surface sediments at the other stations (Figure 4.3). Sample information and γ-ray spectrometry data for sediment samples are reported in Appendix F.

Organic carbon concentrations in sediments at station 1 ranged from about 3 to 5%. Station 2 had lower values with most approximately 1 to 2%, but was as high as 5% at 10 cm in June 2009. Station 3 and 4 organic carbon concentrations were about 3 to 4% with less variation than observed at stations 1 and 2 (Chapter 3, Figure 3.3). $\delta^{13}C$ values of sediment organic carbon ranged from -28.23 ± 0.21‰ and -25.23 ± 0.02‰. The range of values is greater at stations 1 and 2, with little variation at stations 3 and 4 (Figure 4.4).

Solid phase sediment nitrogen concentrations were generally higher in surface sediments and ranged from 0.094 ± 0.002 to 0.392 ± 0.009% (Figure 4.5). $\delta^{15}N$ values were 2.76 ± 0.14 to 9.31 ± 0.19‰ (Figure 4.6). Station 2 exhibited the largest gradients in both cases. Concentration and isotopic composition of carbon and nitrogen are listed in Appendix E.

6. Discussion

Wastewater discharges are the only known source of ^{131}I to the Potomac River in the region bound by Reagan National Airport and the Woodrow Wilson Bridge. Sewage effluent and surface water ^{131}I concentrations detected in this investigation and discussed in Chapter 3 indicate that the Blue Plains WPCP is the primary source of this radioisotope to the sampling area. The relative distributions of ^{131}I at the four stations indicate there is less influence from Blue Plains at station 1 on monthly time scales. This is consistent with the flow pattern of sewage effluent from Blue Plains determined by Hearn (1985). The distinctly heavy nitrogen signal in the Blue Plains outfall 002 (mean $\delta^{15}NO_3^-$ = 29‰) and the strong positive correlation between ^{131}I and $\delta^{15}NO_3^-$ in surface waters collected at these stations further support this conclusion (Figure 4.7). Degradation of organic matter during sewage treatment and loss of isotopically light, gaseous nitrogen products causes sewage effluent to be enriched in ^{15}N (Costanzo et al., 2005; Heaton, 1986; Macko and Ostrum, 1994; Peterson and Fry, 1987). Furthermore, return flows, or activated sludge returns, used to maintain bacterial populations within a sewage treatment plant can further enrich the nitrogen pool. Sewage effluent values of $\delta^{15}NO_3^-$ as high as 40‰ have been measured in other WPCPs (Jordan et al., 1997).

Iodine-131's source specificity and its nutrient-like behavior make it a potentially valuable tool for studying the fate of sewage nitrogen in aquatic environments. Distributions of naturally occurring iodine support biological cycling as the primary mechanism for transformations among the species (Campos et al., 1999; Elderfield and Truesdale, 1980; Truesdale, 1994; Truesdale et al., 2000; Truesdale and Upstill-Goddard, 2003; Waite et al., 2006; Wong, 1995; Wong and Zhang, 1992) and incorporation into organic phases, including particulate matter (Bors et al., 1991; Lieser and Steinkopff, 1989; Muramatsu et al., 1990a; Muramatsu et al., 1990b; Sheppard and Hawkins, 1995; Whitehead, 1974b). While the mechanisms responsible for these transformations are largely unknown, iodine is incorporated into particulate matter and subsequently deposited in sediments where it is subject to diagenetic remineralization.

Solid phase profiles of ^{210}Pb ($t_{½}$ = 22.3 y) indicate that at least 10 cm of these sediments are exchanged or replaced on seasonal time scales. Shorter-lived, ^{7}Be ($t_{½}$ = 53.2 d) detected in the top 3 to 4 cm of these sediments further constrains the rate of mixing or rapid sedimentation to monthly or seasonal time scales. While the sedimentary deposits are not in steady state over long time scales, for the short half-life of ^{131}I such an assumption is reasonable. Furthermore, ^{131}I follows the expected behavior for this element (Chapter 3).

Thus, ^{131}I can be used to further understand the fate of wastewater nitrogen in this system. Sediment inventories of ^{131}I were calculated using the relationship

$$I = \sum A_i \rho_i x_i \qquad (2)$$

where I = inventory of radioisotope (Bq cm^{-2}), A_i = concentration of radioisotope of the *i*th interval (Bq g^{-1}), ρ_i = dry bulk density of interval (g cm^{-3}) and x_i = thickness of interval (cm). Assuming steady state, the flux of ^{131}I to the sediments was calculated for each of the profiles using the following equation

$$J = I \lambda \qquad (3)$$

where J = flux to sediments (Bq cm^{-2} d^{-1}), I = inventory of radioisotope (Bq cm^{-2}) and λ = decay constant of radioisotope (d^{-1}). The average flux of ^{131}I to the sediments is 2.1 x 10^{-3} Bq cm^{-2} d^{-1}

(7.7×10^3 Bq m^{-2} y^{-1}) (Figure 4.3). Surface sediment (0 to 1 cm) ratios of nitrogen to ^{131}I (mol Bq^{-1}) were used to estimate the flux of nitrogen to the sediments. The resulting average is 9.1×10^{-6} mol N cm^{-2} d^{-1} (0.47 kg N m^{-2} y^{-1}). Boynton and coworkers (1995) estimated a burial rate of 9.32×10^6 kg N y^{-1} in sediments of the Lower Potomac River (surface area = 722×10^6 m^2). This translates into a flux of 0.013 kg N m^2 y^{-1}, substantially lower than my estimate. Shultz (1989) estimated the flux of nitrogen to the sediments in a smaller area of the Potomac River (55×10^6 m^2) that encompassed the sampling area of the present study. The estimate reported in Shultz (1989) was 0.18 kg N m^2 y^{-1}, more similar to the estimate in this study. 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. Direct comparison of these numbers is difficult due to the decreases in the nitrogen loading from WPCPs in recent decades. Since the 1980s, nitrogen discharges from Blue Plains have decreased by about 70% (DC Water, 2011). Differences in spatial extent of these studies further complicate this comparison. However, it is clear that ^{131}I profiles can be used to estimate the flux of reactive nitrogen flux to sediments. Iodine-131, due to its short-half life, is expected to be confined to the surface mixed layer. It is within this region that ^{131}I is particularly useful. The sedimentary environment appears sufficiently dynamic that a radioisotope like ^{210}Pb is not useful in terms of a decay tracer but is still useful as a stable tracer.

Sediment δ^{15}N profiles indicate a heavy nitrogen source, consistent with wastewater nitrogen (Figure 4.6). Isotopic fractionation during diagenesis cannot explain these observations. Such fractionation should enrich the solid phase pool in ^{15}N as the lighter isotope is preferentially lost during decomposition of organic matter. Furthermore, net isotopic fractionation of carbon and nitrogen is not expected during early diagenesis of sediments. Thus, sediment values reflect the isotopic composition of source materials (Bickert, 2006; Burdige, 2006). Solid phase organic carbon δ^{13}C profiles support this conclusion (Figure 4.4).

Mass and isotopic balances suggest that the δ^{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 (N_R), the mass and isotopic balances are given by

$$N_T = N_W + N_R \qquad (4)$$

$$\delta^{15}N_TN_T = \delta^{15}N_WN_W + \delta^{15}N_RN_R \qquad (5)$$

where N_T = the sediment nitrogen concentration (%) and $\delta^{15}N$ = 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

$$\delta^{15}N_TN_T = \delta^{15}N_WN_W + \delta^{15}N_RN_R + \delta^{15}N_\Delta\Delta N_T \qquad (6)$$

where ΔN_T = the net change in sediment nitrogen concentration and $\delta^{15}N_\Delta$ = the isotopic composition during remineralization. When $\delta^{15}N_WN_W$ (or $\delta^{15}N_RN_R$) = constant or zero and $\Delta N_T \neq 0$, Equation 6 becomes

$$\frac{d\,(\delta^{15}N_TN_T)}{dN_T} = \delta^{15}N_\Delta + \frac{d\,(\delta^{15}N_\Delta)}{dN_T} \qquad (7)$$

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 (Aller and Blair, 2004; Aller and Blair, 2006; Aller et al., 2008; Martin et al., 2000; Sayles and Curry, 1988; Zhu et al., 2002). 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 influenced by a wastewater source (Figure 4.8).

The relative contribution of wastewater nitrogen was estimated using this two end-member approach. The average $\delta^{15}NO_3^-$ measured in the outfall was used as the sewage effluent end-member: $\delta^{15}N_W$ = 29‰ (Table 4.1). The Blue Plains outfall samples were collected at low water, which nearly coincides with slack water in this region of the Potomac River. The outfall discharge was visably distinguishable from ambient river water due to the relatively low suspended solids concentrations in the sewage effluent. Furthemore, low TSS and chlorophyll a concentrations measured in the oufall indicate little mixing with ambient river water at the time of sample collection (Table 4.1) and support this assumption. The river end member, $\delta^{15}N_R$ = 7‰, represents the mean $\delta^{15}NO_3^-$ measured for the Potomac watershed (Mayer et al., 2002). That end-member is consistent with the y-intercept of the plot of ^{131}I versus $\delta^{15}NO_3^-$ (Figure 4.7).

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 waste water discharged from Blue Plains.

7. Conclusions

The source of ^{131}I in the sampling area is sewage effluent, predominantly from the Blue Plains WPCP. The strong positive correlation between $\delta^{15}NO_3^-$ and ^{131}I identified the isotopic signature of sewage effluent discharged from Blue Plains and was further used to trace wastewater nitrogen in this system. Iodine-131 exhibits nutrient-like behavior, similar to nitrogen, where it is incorporated particulate matter and deposited in sediments. Upon deposition, it is subject to remineralization.

Solid phase sediment profiles of ^{131}I provided a means to estimate the flux of reactive nitrogen to sediments. Estimates were reasonable. However, water column suspended particulate concentrations for both ^{131}I and nitrogen would better constrain these estimates. Iodine-131 is particularly useful in a dynamic sedimentary environment where mass fluxes to the sediments cannot be easily determined. The long-lived radioisotope, ^{210}Pb, helped to characterize the physical setting but did not provide information regarding short-term deposition of material to the river bed.

Solid phase sediment $\delta^{15}N$ profiles indicate that heavy nitrogen is deposited in the sediments in the sampling area. The heavy nitrogen signal is consistent with a wastewater source. The average $\delta^{15}N$ value of nitrate measured in the Blue Plains sewage outfall was 29‰, compared to the riverine end-member of ~ 7%, (Boyer et al., 2002; this study). Mass and isotopic balances for nitrogen indicate that organic matter influenced by a wastewater source is remineralized in these sediments. Furthermore, wastewater nitrogen can constitute up to 59% of the sediment nitrogen pool in this region of the Potomac River.

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

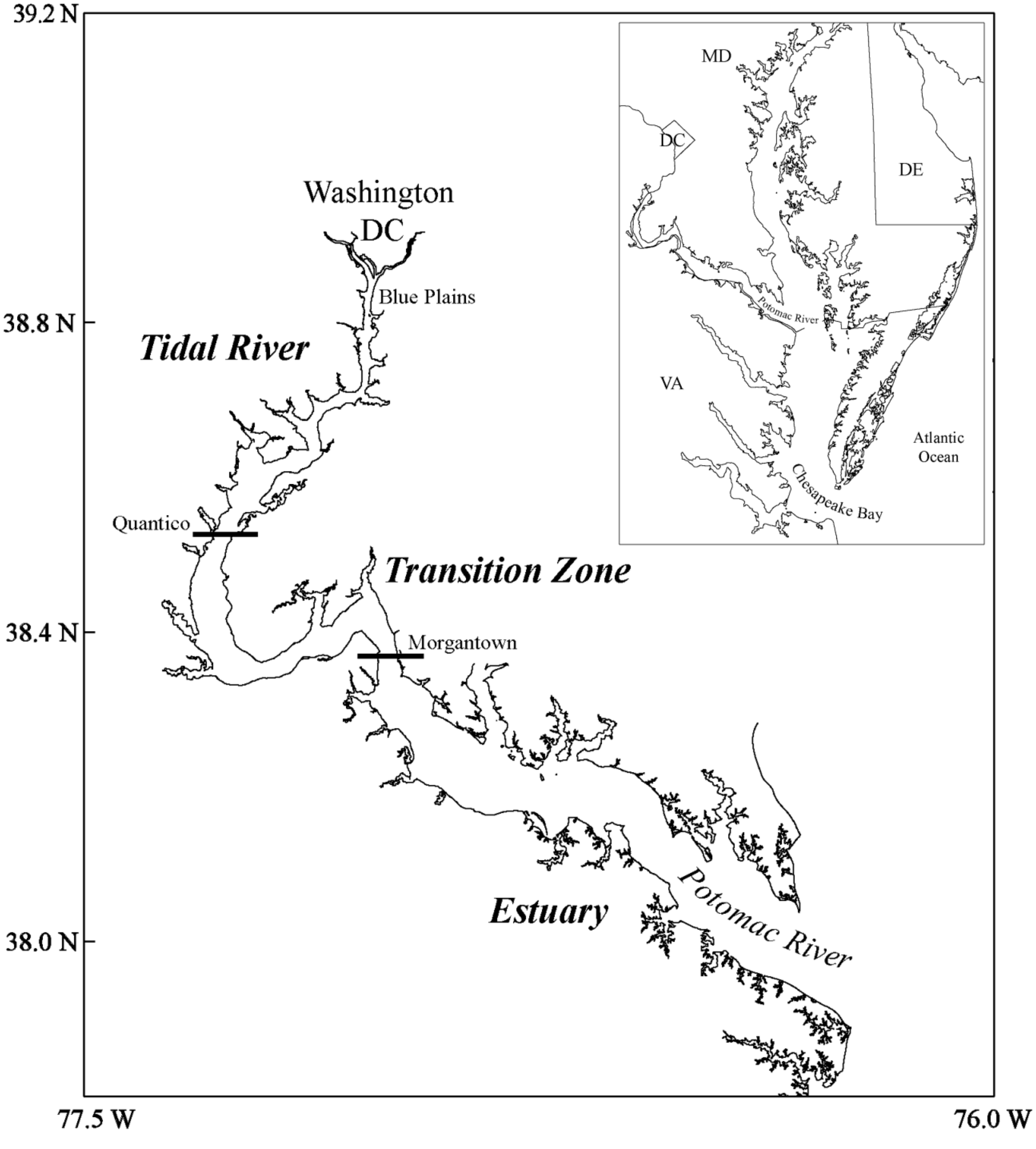

Figure 4.1. Hydrologic divisions of the tidal Potomac River (adapted from Glenn, 1988). The Potomac River is a tributary of the Chesapeake Bay, which discharges to the Atlantic Ocean (inset).

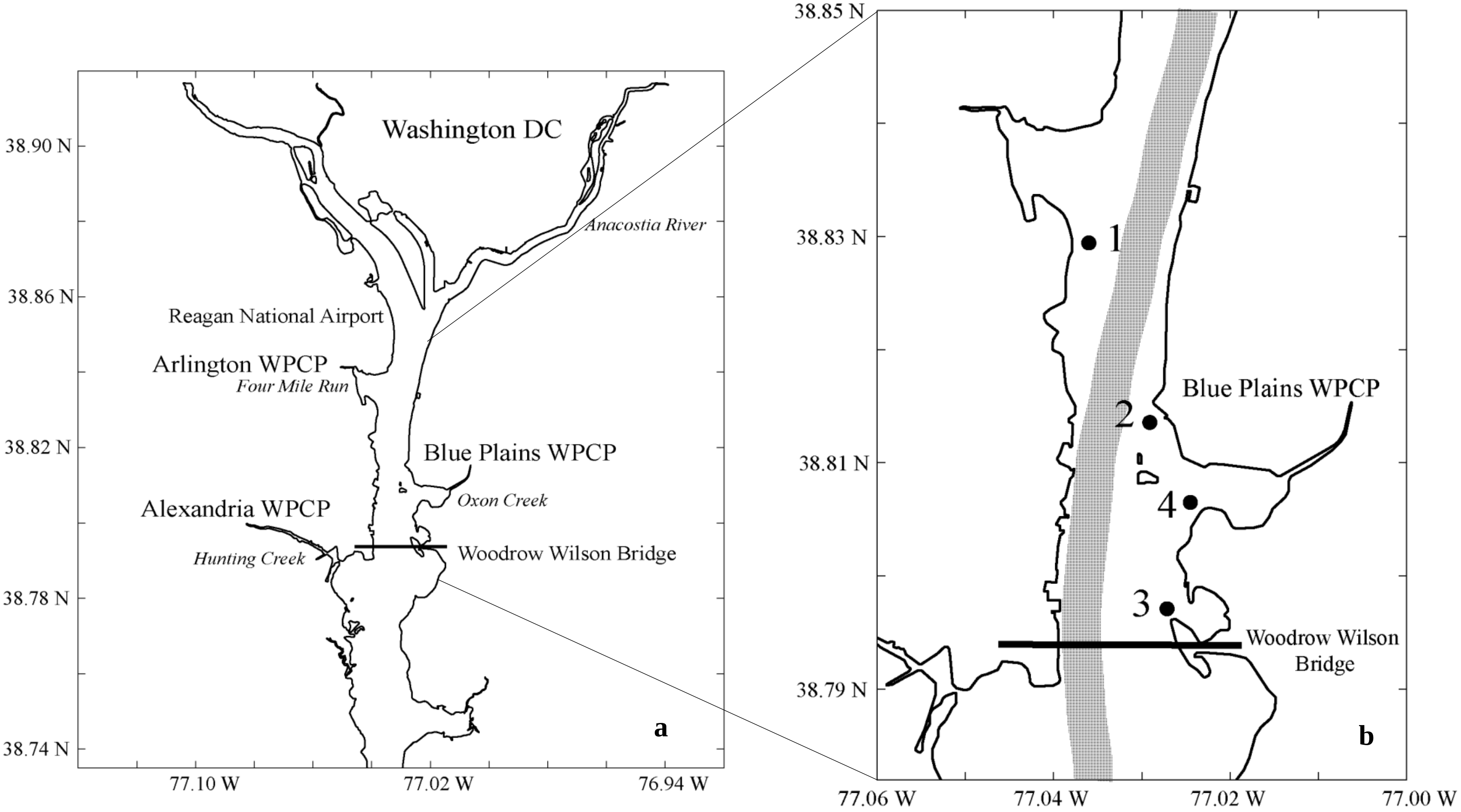

Figure 4.2. a) General sampling area in the tidal Potomac River showing the locations of Blue Plains, Arlington and Alexandria WPCPs. b) Station locations. Shaded area represents the approximate location of the main channel.

Station #	**NH_4 (mg N L^{-1})**	**NO_3^- + NO_2^- (mg N L^{-1})**	**PO_4^+ (mg P L^{-1})**	**Chl a (µg L^{-1})**	**TSS (mg L^{-1})**	**$\delta^{15}NO_3^-$ (‰)**	**^{131}I (Bq L^{-1})**
June 2009							
1	0.048	1.06	0.023	3.96	12.0	8.7 ± 0.3	$< L_D$
2	0.064	1.26	0.019	3.98	4.5	18.7 ± 4.8	0.18 ± 0.01
3	0.050	1.07	0.015	5.47	7.3	11.6 ± 0.6	0.18 ± 0.03
4	0.128	0.97	0.006	6.02	11.3	16.3 ± 2.1	0.28 ± 0.01
Outfall	0.036	2.79	0.006	0.56	0.8	27.1 ± 0.2	0.68 ± 0.02
August 2009							
1	0.114	0.76	0.013	12.32	12.7	15.5 ± 0.1	0.076 ± 0.006
2	0.116	1.10	0.006	9.52	14.7	26.3 ± 3.0	0.46 ± 0.03
3	0.069	0.95	0.002	15.54	14.7	24.5 ± 0.5	0.34 ± 0.03
4	0.013	0.92	0.005	16.38	16.0	25.4 ± 2.7	0.25 ± 0.01
Outfall	0.110	1.11	0.003	10.92	0.5	27.7 ± 2.3	0.54 ± 0.02
November 2009							
1	0.047	1.06	0.025	0.48	7.3	9.8 ± 1.2	$< L_D$
2	0.030	0.50	0.024	0.18	4.7	33.4 ± 7.3	0.59 ± 0.03
3	0.167	1.37	0.028	1.34	9.3	15.9 ± 2.8	0.35 ± 0.03
4	0.148	1.20	0.017	1.13	14.0	17.7 ± 2.9	0.37 ± 0.01
Outfall	0.021	0.39	0.019	0.02	0.4	31.6 ± 6.4	0.62 ± 0.02

Table 4.1. Nutrient, chlorophyll a (Chl a), total suspended solids (TSS), ^{131}I concentrations and the stable isotopic composition of nitrate ($\delta^{15}NO_3^-$) in surface waters collected from stations 1 though 4 and Blue Plains outfall 002 in the Potomac River.

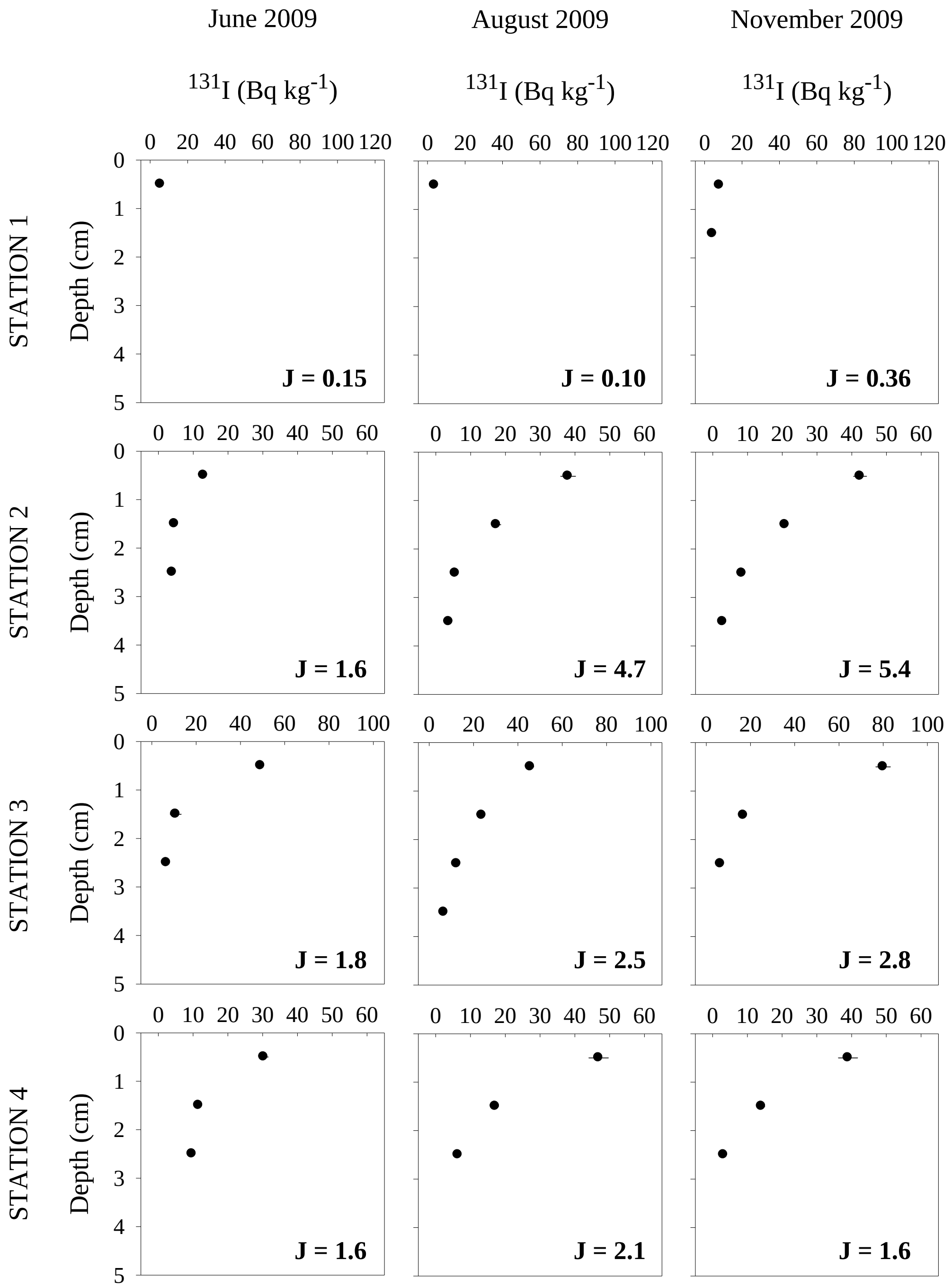

Figure 4.3. Depth profiles of ^{131}I in sediments collected from stations 1 through 4 in the Potomac River. Fluxes (J) are shown for each profile in units of 10^{-3} Bq cm^{-2} d^{-1}.

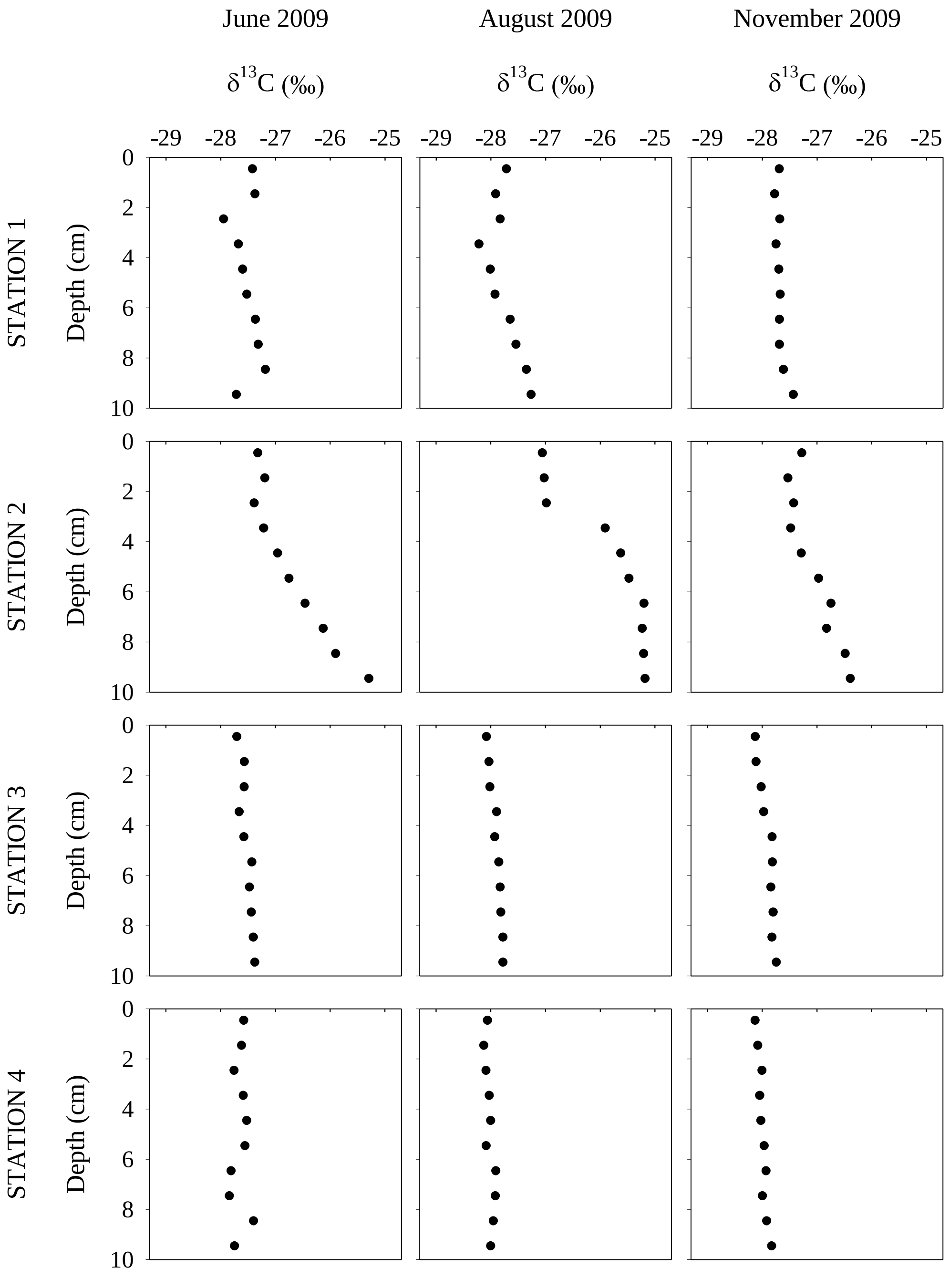

Figure 4.4. Depth profiles of $\delta^{13}C$ of organic carbon in Potomac River sediments collected from stations 1 through 4.

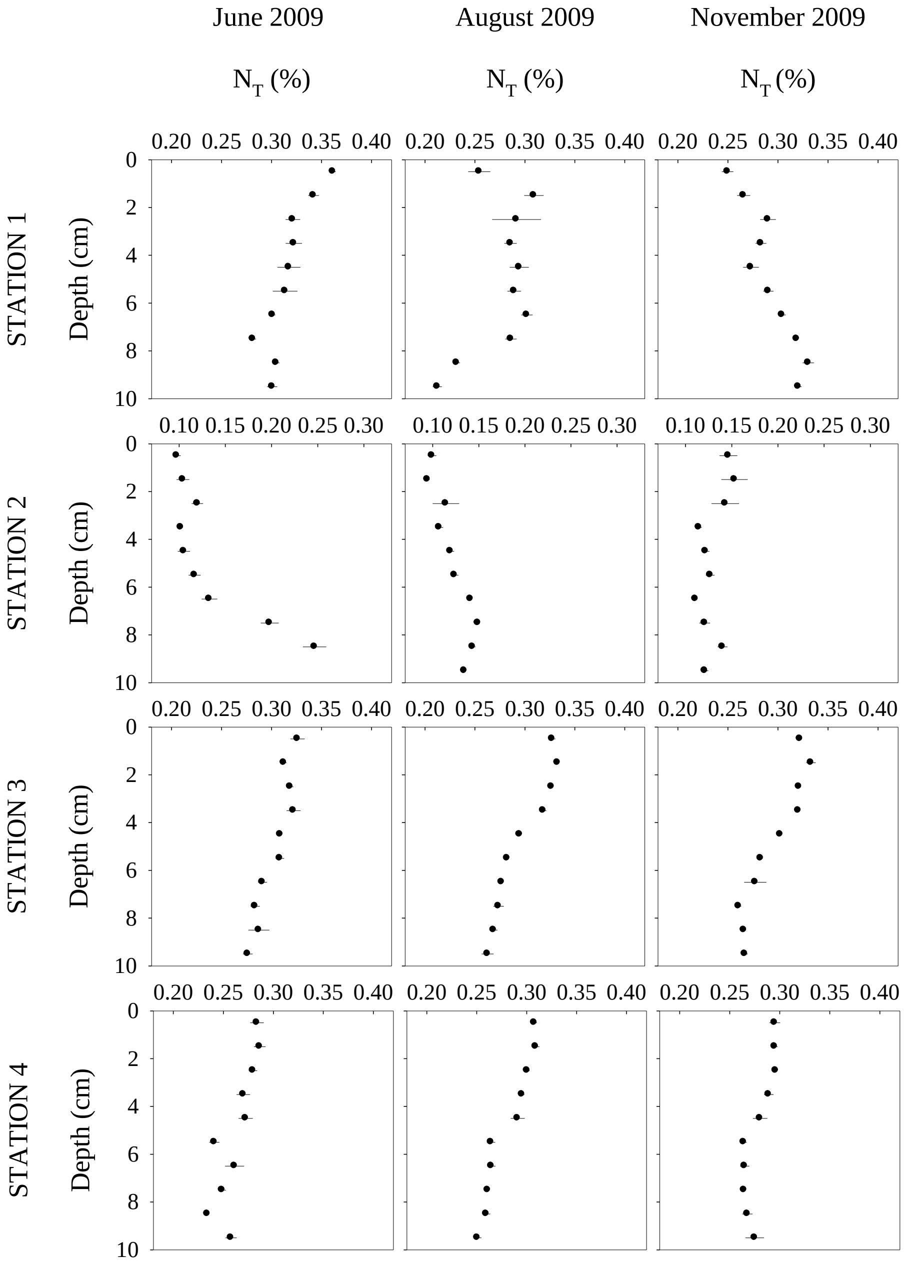

Figure 4.5. Depth profiles of percent total nitrogen (N_T) in Potomac River sediments collected from stations 1 through 4.

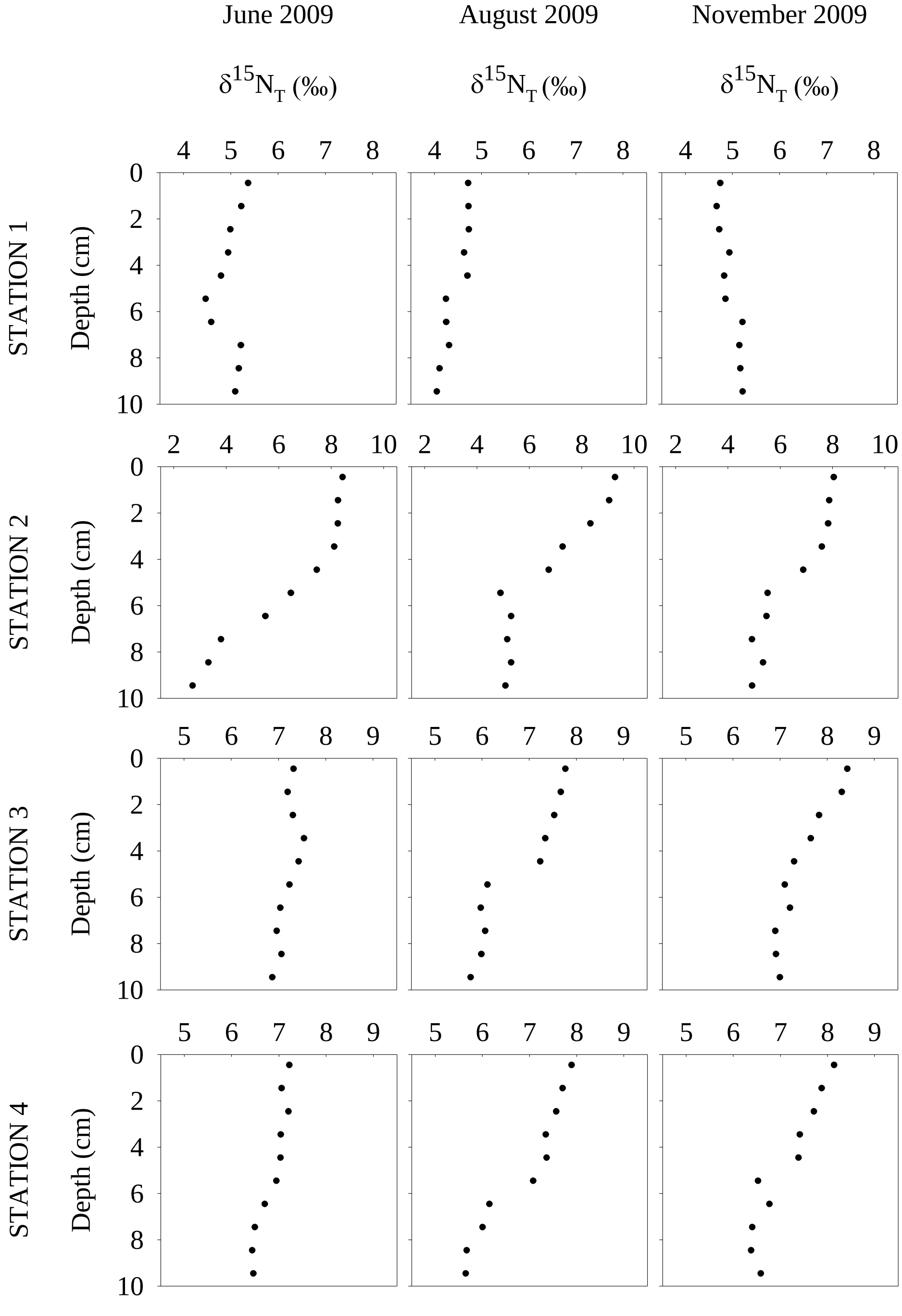

Figure 4.6. Depth profiles of $\delta^{15}N_T$ in Potomac River sediments collected from stations 1 through 4.

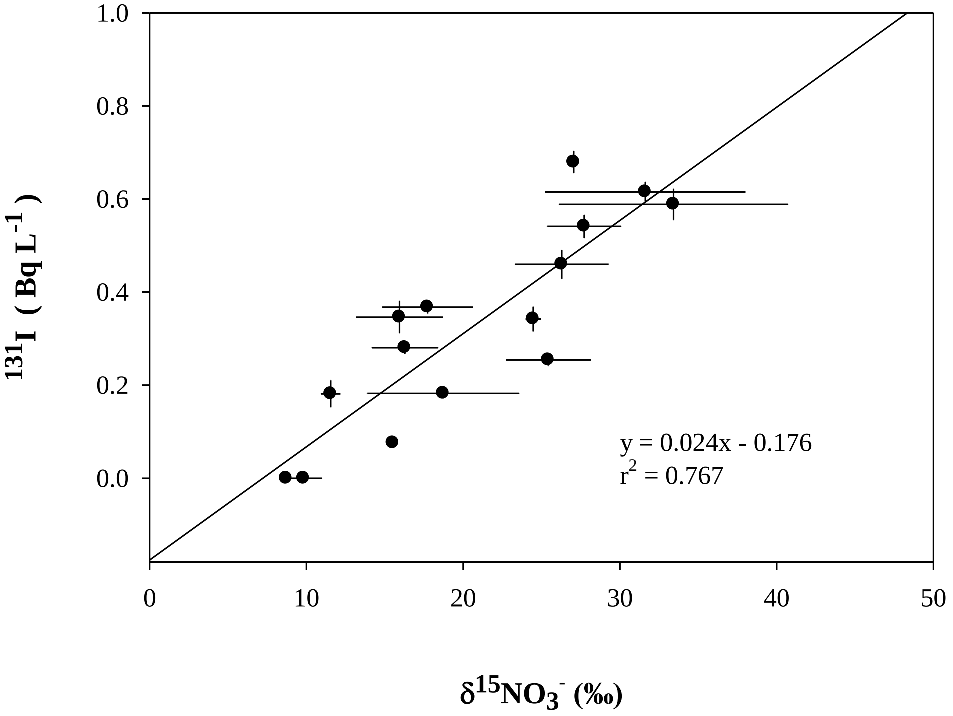

Figure 4.7. Iodine-131 versus $\delta^{15}NO_3^-$ in surface water collected from stations 1 through 4 and the Blue Plains outfall 002 in the Potomac River in June, August and November 2009.

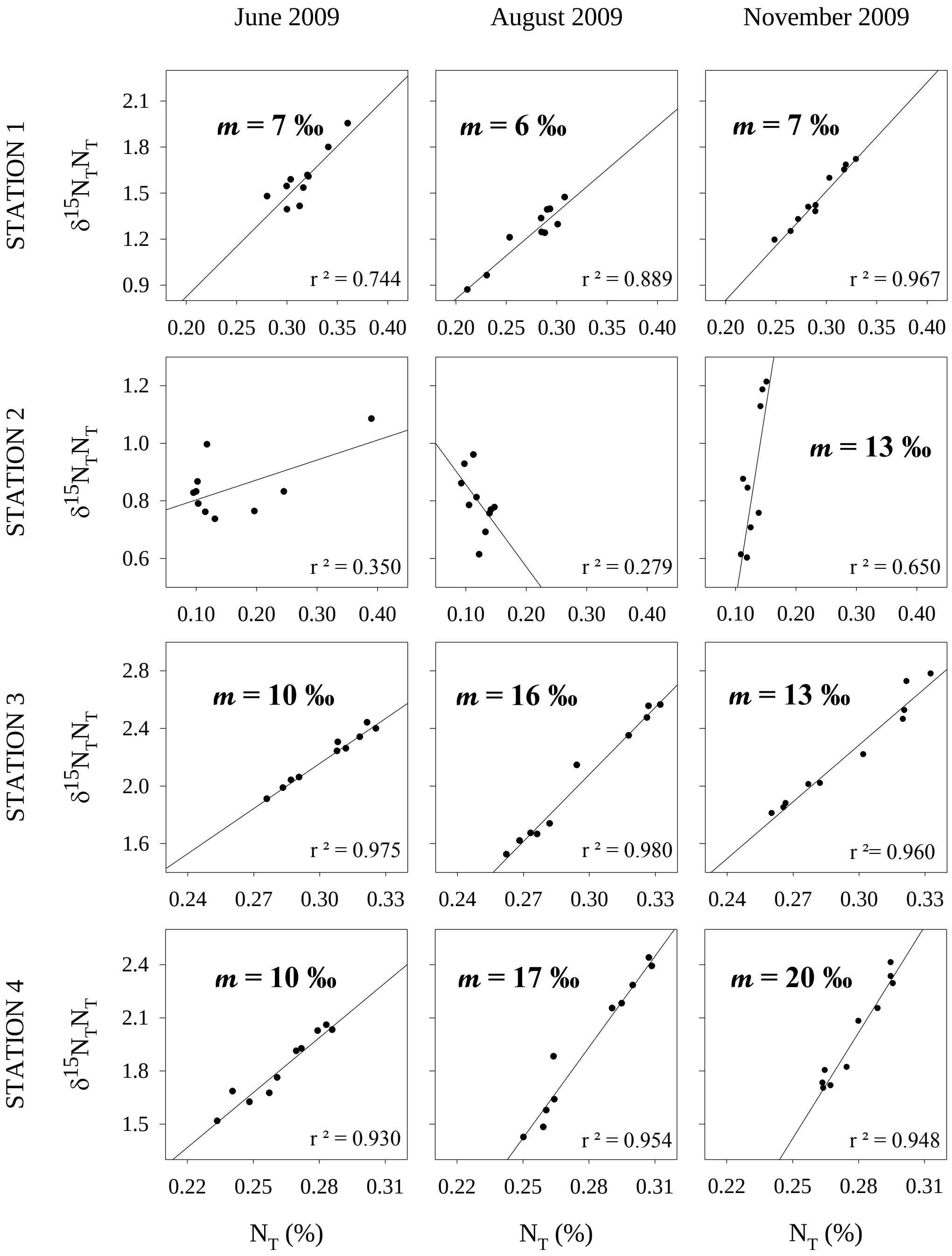

Figure 4.8. $\delta^{15}N_TN_T$ versus N_T in Potomac River sediments collected from stations 1 through 4. The linear slopes (*m*) represent the net isotopic value of nitrogen lost (or gained) from the sediments during organic matter decomposition or transport.

Chapter 5

Summary

Summary of the Dissertation

Medically-derived ^{131}I has been proposed as a tracer of biogeochemical processes in aquatic environments receiving sewage effluent discharges, yet few published data exist for the radioisotope in sewage effluent; most work has focused on sewage sludge. This work presents ^{131}I concentrations detected in sewage effluent from a small water pollution control plant (WPCP) serving a regional thyroid cancer treatment facility in Stony Brook, NY, USA. The concentrations detected in the Stony Brook water pollution control plant (SBWPCP) ranged from 1.8 ± 0.3 to 227 ± 2 Bq L^{-1} in sewage effluent. The primary source of ^{131}I is excreta from thyroid cancer inpatients treated at the Stony Brook University Medical Center (SBUMC). Time series measurements following known inputs indicated that ^{131}I is discharged for many days following an inpatient treatment. The long retention time of ^{131}I in the plant is attributed to flow recycling, or activated sludge returns, used to maintain bacterial populations required to achieve sewage treatment. The estimated sewage half-life, analogous to a radioactive half-life, of iodine in this plant is 2.9 d. This is longer than the hydraulic retention time (HRT) of this plant, which is 24 h. Continuous discharges of ^{131}I in sewage effluent are not limited to the SBWPCP.

Concentrations of ^{131}I measured in sewage effluent from the Blue Plains WPCP in Washington, DC and in the Potomac River suggest a continuous discharge there. The behavior of ^{131}I in the Potomac River agrees well with the known behavior of naturally occurring iodine in aquatic environments. Greater than 45% of ^{131}I in sewage effluent and Potomac River water is associated with colloidal material. Furthermore, ^{131}I is incorporated into particulate phases and deposited in sediments. Iodine-131 concentrations in sediments ranged from 1.3 ± 0.8 to 117 ± 2 Bq kg^{-1}. Elevated concentrations of ^{131}I in sediment pore water, relative to the overlying water column, demonstrated that ^{131}I is remineralized during diagenesis.

The nutrient-like cycling of ^{131}I in aquatic systems and its source specificity also makes it useful for investigating the fate of wastewater-derived nutrients such as nitrogen. In the sampling area, the solid phase sediment profiles of ^{210}Pb and ^{7}Be indicated that surface sediments (≤ 10 cm) are buried or removed on seasonal time scales and were not useful in determining mass fluxes of particulate matter to the sediments. Despite the dynamic sedimentary environment, solid phase sediment profiles of ^{131}I suggested steady state conditions for this isotope and provided a means to estimate a flux of iodine to the sediments. Subsequently, the flux of reactive nitrogen to the sediments was determined using the ^{131}I/N ratios in surface sediments. The estimates were reasonable but would be better constrained with ^{131}I/N ratios in suspended particulate matter.

Consistent with the presence of ^{131}I, the solid phase sediment profiles showed heavy nitrogen in the Potomac River sediments, indicative of a wastewater influence. A strong positive correlation between $\delta^{15}NO_3^-$ and ^{131}I in surface waters in the vicinity of Blue Plains identified the isotopic source signature of sewage effluent (29‰). Mass and isotopic balances for nitrogen indicated that organic matter, influenced by a wastewater source, is being deposited and remineralized in these sediments and can constitute > 50% of the total sediment nitrogen pool. This work introduces medically-derived ^{131}I as a potentially valuable tracer to study the short-term fate of wastewater nitrogen in this system. However, the utility of ^{131}I as a tracer is not limited to the Potomac River. Other studies have documented the presence of ^{131}I in aquatic systems and continuous discharges of this radioisotope in sewage effluent are likely to be widespread in urban environments.

Directions for Future Research

Medically-derived ^{131}I can be used further to estimate the rates of interconversions among the primary species of iodine in aquatic environments. The rates of transformations among the primary iodine species can be estimated using a simple box model approach and measured concentrations of the stable and radioactive species (IO_3^-, I^-, DOI, $^{131}IO_3^-$, $^{131}I^-$ and $DO^{131}I$) in both sewage effluent and receiving water. Assuming steady state conditions, measured concentrations of the individual species can be used to solve a system of equations, derived from the box model to determine rate constants. Concentration ratios for the same species in sewage effluent and receiving water (e.g., $\left[\frac{^{131}I}{I}\right]_{DOI}$) could be used to determine the relative significance of the reactions on short time scales.

Distributions of ^{131}I in receiving waters can be used to determine the dispersion pattern of sewage effluent in a receiving water body on short time scales. The relative influence of sewage effluent from the Blue Plains WPCP at sites in the tidal Potomac River was determined using surface water and sediment concentrations of ^{131}I in this study and confirmed the pattern determined previously by other investigators (Hearn, 1985). A similar study was done using *Fucus* sp. collected in Port Jefferson Harbor, NY, USA (Rose, 2003; Appendix H, this work).

Monitoring of water column concentrations of ^{131}I and in aquatic organisms, such as seaweeds in the vicinity of a sewage outfall could lead to a better understanding of uptake of stable and radioactive iodine by the organisms. More specifically, rates of uptake and concentration factors could be estimated using the ^{131}I concentrations.

Although ^{131}I has a relatively short half-life and is diluted considerably in WPCPs and receiving waters, medically-derived ^{131}I is easily measureable in the environment. This work demonstrates that sewage effluent discharges of this isotope can be used to study the biogeochemistry of iodine as well as investigate the fate of wastewater constituents.

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Appendix A. Detector counting efficiencies for ^{131}I at 364.5 keV.

	Geometry			
Detector	**Large Jar (150 mL)**	**Large Jar (Full)**	**Small Jar (Full)**	**0.7 µm GFF**
2K	0.0075	0.0071	0.0108	-
Well	0.0122	0.0105	0.0179	-
NF	0.0192	0.0167	0.0264	0.048
3KA	0.0224	0.0203	0.0319	0.058
3KB	0.0220	0.0201	0.0325	0.058
3KC	0.0226	0.0200	0.0320	0.058

Table A.1. Detector counting efficiencies for ^{131}I at 364.5 keV. Efficiencies were determined using a certified ^{131}I standard solution. Canberra intrinsic germanium detectors: 2K = 2000 mm^2 LEGe; Well = well detector used in planar mode; NF = 3000 mm^2 LEGe; 3KA, 3KB and 3KC = 3800 mm^2 LEGe. Large jar = straight-side polypropylene jar (64 mm height; 64 mm inner diameter). Small jar = straight-side polypropylene jar (41 mm height; 31 mm inner diameter). 0.7 µm GFF = 0.7 µm glass fiber filter (47 mm diameter).

Appendix B. Sample information and γ-ray spectrometry data for sewage effluent collected from the Stony Brook water pollution control plant.

Sample #	**Sample Collection Date & Time**	**Counting Start Date & Time**	**Peak Area**	**Counting Error (%)**	**Peak Integral**	**μ_B**	**L_D**	**Counting Time (s)**	**^{131}I (Bq L^{-1})**
061206E	6/12/06 1:59 PM	6/12/06 3:09 PM	3788	1.74	4068	284	58	83629.948	48.4 ± 0.8
061306E	6/13/06 1:55 PM	6/13/06 2:36 PM	1594	2.75	1758	265	56	77826.253	21.8 ± 0.6
061406E	6/14/06 11:28 AM	6/14/06 12:37 PM	8261	1.15	8671	323	62	95052.104	93 ± 1
061506E	6/15/06 2:20 PM	6/15/06 3:26 PM	4207	1.64	4474	235	53	69056.134	65 ± 1
061606E	6/16/06 10:03 AM	6/19/06 4:41 PM	3695	1.86	4167	294	59	86462.046	60 ± 1
061906E	6/19/06 2:25 PM	6/20/06 4:50 PM	1527	2.9	1758	284	58	83507.594	21.4 ± 0.6
062006E	6/20/06 2:34 PM	6/21/06 4:52 PM	1298	3.11	1470	293	59	86157.628	17.6 ± 0.5
062106E	6/21/06 10:47 AM	6/22/06 4:58 PM	1202	3.37	1434	301	60	88563.525	16.1 ± 0.5
062206E	6/22/06 2:35 PM	6/23/06 5:51 PM	1788	3.09	2448	800	96	235148.650	9.6 ± 0.3
062306E	6/23/06 11:50 AM	6/26/06 11:35 AM	1051	4.18	1505	567	81	166907.488	9.0 ± 0.4
062606E	6/26/06 12:15 PM	6/28/06 11:48 AM	1021	3.94	1305	374	66	109871.612	11.9 ± 0.5
062706E	6/27/06 2:22 PM	6/29/06 6:07 PM	1465	3.61	2099	781	95	229590.484	8.8 ± 0.3
062806E	6/28/06 2:05 PM	7/2/06 12:09 PM	612	6.93	1162	588	82	172805.528	5.5 ± 0.4
062906E	6/29/06 2:46 PM	7/4/06 12:19 PM	306	14.42	1177	937	103	275514.310	2.0 ± 0.3
063006E	6/30/06 10:48 AM	7/7/06 5:32 PM	500	8.42	1188	940	104	276446.232	4.0 ± 0.3
071706E	7/17/06 2:08 PM	7/17/06 4:53 PM	0	0	121	265	56	77815.294	$< L_D$
071806E	7/18/06 1:21 PM	7/18/06 2:37 PM	0	0	110	251	55	73913.386	$< L_D$

Table B.1. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in **unfiltered** sewage effluent collected from the Stony Brook water pollution control plant. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 2. $< L_D$ = less than detection limit. Sample volume = 0.17 L.

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
071906E-2	7/19/06 2:41 PM	7/19/06 3:28 PM	3905	1.7	4141	217	51	63706.521	65 ± 1
072006E-2	7/20/06 2:32 PM	7/20/06 4:07 PM	4056	1.69	4375	294	59	86485.371	50.2 ± 0.8
072106E	7/21/06 1:13 PM	7/21/06 4:47 PM	5215	1.59	6023	859	99	252721.313	24.1 ± 0.4
072406E	7/24/06 1:21 PM	7/25/06 3:25 PM	518	5.79	721	285	58	83866.360	7.2 ± 0.4
072506E	7/25/06 12:45 PM	7/27/06 4:10 PM	467	6.64	710	271	57	79586.329	7.5 ± 0.5
072606E	7/26/06 11:18 AM	7/26/06 2:49 PM	1085	3.28	1170	78	32	22996.222	49 ± 2
072706E	7/27/06 9:44 AM	7/27/06 10:20 AM	1198	3.1	1290	70	30	20683.308	60 ± 2
072806E	7/28/06 7:01 AM	8/1/06 12:24 PM	1424	3.34	1804	337	63	99108.088	22.2 ± 0.7
073106E	7/31/06 1:53 PM	8/2/06 4:03 PM	474	7.07	780	321	62	94316.073	6.4 ± 0.5
080106E	8/1/06 1:37 PM	8/3/06 6:23 PM	325	9.37	622	261	56	76874.556	5.4 ± 0.5
080206E	8/2/06 1:34 PM	8/4/06 3:52 PM	759	5.26	1181	558	80	164208.129	6.1 ± 0.3
080406E	8/4/06 2:13 PM	8/6/06 1:47 PM	359	8.36	634	316	61	92830.086	4.9 ± 0.4
080706E	8/7/06 1:54 PM	8/7/06 3:40 PM	160	16	412	325	62	95669.976	1.8 ± 0.3
080806E-2	8/8/06 1:05 PM	8/9/06 10:37 AM	1087	3.35	1202	96	35	28317.188	43 ± 1

Table B.1. Continued

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
080906E	8/9/06 12:47 PM	8/9/06 7:26 PM	2722	2.11	2993	218	51	63973.407	46 ± 1
081006E-2	8/10/06 2:30 PM	8/10/06 6:47 PM	6036	1.37	6400	229	53	67405.174	96 ± 1
081106E-2	8/11/06 2:26 PM	8/11/06 4:18 PM	6656	1.3	7041	225	52	66199.897	107 ± 1
081206E	8/12/06 9:13 AM	8/12/06 10:46 AM	37846	0.54	39282	671	88	197441.381	217 ± 1
081706E	8/17/06 1:35 PM	8/22/06 12:58 PM	9026	1.1	9411	264	56	77536.636	190 ± 2
081806E	8/18/06 12:29 PM	8/24/06 6:45 PM	5516	1.46	5952	284	58	83419.212	121 ± 2
082506E	8/25/06 11:44 AM	8/25/06 6:00 PM	2436	2.79	3406	887	101	260998.211	11.1 ± 0.3
082906E	8/29/06 1:35 PM	8/29/06 5:37 PM	6133	1.4	6633	295	59	86629.090	77 ± 1
083006E	8/30/06 1:46 PM	8/30/06 5:47 PM	3803	1.75	4100	299	60	87923.177	46.8 ± 0.8
083106E	8/31/06 2:35 PM	8/31/06 6:41 PM	7124	1.36	8139	940	104	276446.232	30.5 ± 0.4
091406E	9/14/06 2:22 PM	9/14/06 3:46 PM	1951	2.57	2292	326	62	83706.536	8.8 ± 0.2
091506E-A	9/15/06 2:34 PM	9/15/06 3:48 PM	1277	2.84	1302	15	15	4265.889	108 ± 3
091506E-C	9/15/06 2:35 PM	9/15/06 5:03 PM	68263	0.39	69946	874	100	253955.917	109.6 ± 0.4
091806E-A	9/18/06 2:25 PM	9/18/06 3:53 PM	10063	1.04	10605	583	82	169594.803	23.1 ± 0.2
091806E-C	9/18/06 2:26 PM	9/20/06 4:59 PM	4782	1.52	5112	316	61	91745.970	23.3 ± 0.4
092006E-A	9/20/06 1:44 PM	9/25/06 3:57 PM	1823	2.54	2030	257	55	74782.731	14.0 ± 0.4
092006E-C	9/20/06 1:45 PM	9/21/06 7:22 PM	2384	2.18	2582	238	53	69042.113	14.2 ± 0.3
092006E-E	9/20/06 1:46 PM	9/22/06 5:29 PM	7215	1.27	7995	861	99	250294.310	14.0 ± 0.2
092206E-A	9/22/06 12:41 PM	9/26/06 1:06 PM	1304	3.24	1610	329	62	95781.679	7.2 ± 0.2
092206E-B	9/22/06 12:41 PM	9/27/06 4:02 PM	1188	3.39	1478	326	62	94897.538	7.3 ± 0.2
092206E-E	9/22/06 12:43 PM	10/2/06 2:59 PM	773	4.33	1003	307	60	89297.638	7.7 ± 0.3

Table B.1. Continued

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
092906E-C	9/29/06 1:30 PM	10/5/06 2:32 PM	2411	2.07	2463	26	19	7538.196	193 ± 4
092906E-D	9/29/06 1:30 PM	10/4/06 11:33 AM	31027	0.58	31722	333	63	96937.536	183 ± 1
092906E-E	9/29/06 1:31 PM	10/5/06 4:41 PM	22930	0.67	23442	265	56	76978.922	187 ± 1
101906E-A	10/19/06 2:05 PM	10/19/06 3:08 PM	18618	0.75	19062	287	58	83533.449	83.2 ± 0.6
101906E-D	10/19/06 2:06 PM	10/23/06 5:39 PM	14866	0.84	15351	326	62	94639.080	83.7 ± 0.7
101906E-E	10/19/06 2:07 PM	11/2/06 2:57 PM	16380	0.83	17535	951	104	276446.232	81.1 ± 0.7
120606E	12/6/06 2:08 PM	12/6/06 3:23 PM	2507	2.15	2798	355	65	88827.225	10.6 ± 0.2
120706E	12/7/06 1:37 PM	12/7/06 4:12 PM	1861	2.53	2119	329	62	82363.415	8.5 ± 0.2
120806E	12/8/06 12:30 PM	12/8/06 3:15 PM	4165	1.79	5131	1014	107	253592.828	6.7 ± 0.1
121106E	12/11/06 12:43 PM	12/11/06 2:57 PM	2663	2.08	2966	367	66	91695.070	10.9 ± 0.2
121206E	12/12/06 2:22 PM	12/12/06 4:34 PM	1765	2.61	2027	335	63	83792.864	7.9 ± 0.2
121306E	12/13/06 1:00 PM	12/13/06 3:57 PM	1473	2.91	1734	356	65	88903.836	6.2 ± 0.2
121406E	12/14/06 1:47 PM	12/14/06 5:12 PM	990	3.82	1288	302	60	75550.970	4.9 ± 0.2
121506E	12/15/06 12:29 PM	12/15/06 2:17 PM	2746	2.29	3610	1055	110	263824.957	4.3 ± 0.1
121906E	12/19/06 2:05 PM	12/19/06 2:57 PM	1878	2.57	2205	398	68	99464.439	7.1 ± 0.2
122006E	12/20/06 11:22 AM	12/20/06 6:43 PM	2969	2.08	3557	646	86	161415.081	7.3 ± 0.2
010307E	1/3/07 1:48 PM	1/4/07 5:25 PM	82	19.68	227	305	60	76161.475	< L_D
010507E	1/5/07 12:42 PM	1/5/07 2:43 PM	271	10.92	764	1014	107	253409.386	< L_D
010807E	1/8/07 12:00 PM	1/8/07 1:25 PM	21	77.91	213	389	68	97188.630	< L_D
010907E	1/9/07 1:35 PM	1/9/07 4:33 PM	56	39.97	412	698	90	174505.010	< L_D
011107E	1/11/07 12:00 PM	1/11/07 5:13 PM	49	31.38	199	306	60	76567.300	< L_D

Table B.1. Continued

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
011509E	1/15/07 2:18 PM	1/15/07 6:03 PM	52	41.61	385	668	88	166958.134	< L_D
011607E	1/16/07 1:00 PM	1/18/07 11:37 AM	117	26.6	798	1404	126	351035.336	< L_D
012307E	1/23/07 12:53 PM	1/23/07 5:38 PM	855	4.11	1114	261	56	65218.425	4.9 ± 0.2
012407E	1/24/07 12:35 PM	1/24/07 4:22 PM	1135	3.52	1446	323	62	80627.649	5.3 ± 0.2
012507E	1/25/07 1:25 PM	1/25/07 3:53 PM	12723	0.91	13187	367	66	91719.830	52.2 ± 0.5
012607E	1/26/07 10:05 AM	1/26/07 4:33 PM	13752	0.94	15017	940	104	276446.232	59.4 ± 0.6
012809E[1]	1/28/09 12:00 PM	1/28/09 3:57 PM	1159	5.2	2198	483	75	120816.207	3.9 ± 0.2
031409E[1]	3/14/09 1:07 PM	3/14/09 3:45 PM	5321	1.49	5717	311	61	91470.307	67 ± 1
031509E[1]	3/15/09 1:31 PM	3/15/09 5:16 PM	1532	3.15	1893	296	59	87133.401	20.4 ± 0.6
031609E[1]	3/16/09 12:36 PM	3/17/09 6:12 PM	955	4.67	1471	611	84	179757.859	7.1 ± 0.3
031709E[1]	3/17/09 1:30 PM	3/19/09 8:00 PM	1258	4.02	1690	519	78	150994.408	4.0 ± 0.2
031809E[1]	3/18/09 12:30 PM	3/24/09 3:27 PM	382	7.91	565	284	58	82495.814	3.0 ± 0.2
031909E[1]	3/19/09 12:34 PM	3/21/09 2:34 PM	377	9.89	863	638	86	187668.420	2.9 ± 0.3
032009E[1]	3/20/09 1:00 PM	3/24/09 3:28 PM	751	6.52	1614	1456	128	428364.138	3.4 ± 0.2

[1] Sample volume = 0.15 L

Table B.1. Continued

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
013107E[1]	1/31/07 12:27 PM	1/31/07 1:26 PM	841	4.79	1236	572	81	168171.491	5.6 ± 0.3
020107E	2/1/07 12:30 PM	2/2/07 12:12 PM	6846	1.39	7820	931	103	273753.462	34.1 ± 0.5
020207E	2/2/07 12:40 PM	2/5/07 4:20 PM	1131	3.52	1354	230	53	67722.757	24.8 ± 0.9
020507E	2/5/07 12:29 PM	2/6/07 11:11 AM	734	4.99	1082	648	86	190613.270	5.0 ± 0.3
020707E	2/7/07 10:52 AM	2/10/07 8:05 PM	411	7.91	762	558	80	164063.844	4.0 ± 0.3
020807E	2/8/07 12:47 PM	2/8/07 4:34 PM	2981	1.94	3166	272	57	80065.635	43.0 ± 0.8
020907E	2/9/07 12:26 PM	2/9/07 5:03 PM	1900	2.44	2033	175	46	51501.904	42 ± 1
021207E	2/12/07 1:25 PM	2/12/07 5:43 PM	608	4.90	773	285	58	83813.138	8.4 ± 0.4
030707E	3/7/07 12:00 PM	3/7/07 3:13 PM	0	0.00	181	313	61	92149.358	< L_D
030807E	3/8/07 12:38 PM	3/8/07 4:53 PM	26	64.73	173	278	58	81619.997	< L_D
030907E	3/9/07 11:33 AM	3/9/07 4:15 PM	0	0.00	139	252	55	74144.235	< L_D
031007E	3/10/07 11:15 AM	3/10/07 12:53 PM	3518	1.80	3769	265	56	78042.174	51.6 ± 0.9
031107E	3/11/07 10:42 AM	3/11/07 11:55 AM	3175	1.92	3445	332	63	97717.538	37.5 ± 0.7
031207E	3/12/07 1:07 PM	3/12/07 3:06 PM	2005	2.51	2284	326	62	95847.100	24.2 ± 0.6
031307E	3/13/07 12:48 PM	3/13/07 5:54 PM	1892	2.53	2090	251	55	73789.703	29.7 ± 0.8
031407E	3/14/07 12:43 PM	3/14/07 2:27 PM	1944	2.54	2192	328	62	96520.702	23.3 ± 0.6
031507E	3/15/07 12:46 PM	3/15/07 5:25 PM	15918	0.82	16431	277	57	81451.546	227 ± 2
031607E	3/16/07 12:41 PM	3/19/07 12:00 PM	11004	0.99	11374	317	61	93326.514	175 ± 2

[1] *Sample volume = 0.17L*

Table B.2 Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in **filtered** sewage effluent collected from the Stony Brook water pollution control plant. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 2. < L_D = less than detection limit. Sample volume = 0.15 L.

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
032007E	3/20/07 11:12 AM	3/20/07 1:57 PM	650	4.89	848	296	59	87137.234	8.6 ± 0.4
032107E	3/21/07 1:12 PM	3/21/07 2:54 PM	701	5.77	1178	618	84	181643.728	4.6 ± 0.3
032307E	3/23/07 12:42 PM	3/23/07 5:25 PM	6193	1.38	6793	801	96	235523.787	32.9 ± 0.5
032707E	3/27/07 12:43 PM	3/27/07 6:33 PM	329	8.00	500	256	55	75306.431	5.1 ± 0.4
032807E	3/28/07 1:12 PM	3/28/07 3:35 PM	5879	1.35	6093	245	54	72205.264	93 ± 1
032907E	3/29/07 11:35 AM	3/29/07 5:20 PM	7811	1.16	8043	226	52	66530.652	136 ± 2
052307E-A	5/23/07 11:46 AM	5/23/07 2:42 PM	6855	1.27	7182	263	56	77336.082	102 ± 1
052407E-A	5/24/07 10:33 AM	5/24/07 1:23 PM	7326	1.24	7751	339	63	99786.583	85 ± 1
052507E-A	5/25/07 1:31 PM	5/25/07 5:13 PM	9252	1.21	10514	940	104	276446.232	42.5 ± 0.5
053007E	5/30/07 12:12 PM	5/30/07 1:13 PM	1419	3.35	1821	308	60	90660.545	18.0 ± 0.6
053107E	5/31/07 1:00 PM	5/31/07 4:03 PM	2637	2.74	3856	940	104	276446.232	12.1 ± 0.3
060407E	6/4/07 11:39 AM	6/4/07 3:06 PM	660	6.19	1113	586	82	172486.847	4.6 ± 0.3
060507E	6/5/07 2:09 PM	6/8/07 5:40 PM	381	12.47	1224	842	98	247750.850	2.5 ± 0.3
060607E	6/6/07 12:59 PM	6/6/07 3:11 PM	2634	2.43	3244	609	84	178970.958	17.7±0.4
060707E	6/7/07 2:18 PM	6/8/07 5:13 PM	9180	1.10	9808	672	88	168085.623	24.4 ± 0.3
060807E	6/8/07 2:25 PM	6/15/07 6:22 PM	1458	4.09	2338	836	98	245880.060	13.6 ± 0.6
061107E	6/11/07 1:33 PM	6/11/07 2:50 PM	257	10.83	513	325	62	95702.544	3.1 ± 0.3
061207E	6/12/07 2:33 PM	6/12/07 5:34 PM	353	10.08	809	582	82	171256.885	2.5 ± 0.3

Table B.2. Continued

Sample #	**Sample Collection Date & Time**	**Counting Start Date & Time**	**Peak Area**	**Counting Error (%)**	**Peak Integral**	**μ_B**	**L_D**	**Counting Time (s)**	**^{131}I ($Bq\ L^{-1}$)**
061307E	6/13/07 1:00 PM	6/14/07 5:18 PM	1971	2.75	2378	305	60	89701.102	27.8 ± 0.8
061407E	6/14/07 2:30 PM	6/18/07 4:02 PM	876	4.33	1141	294	59	86593.042	16.4 ± 0.7
061507E	6/15/07 2:30 PM	6/19/07 4:14 PM	5640	1.54	6406	587	82	172563.680	55.3 ± 0.9
061907E	6/19/07 2:31 PM	6/21/07 4:23 PM	843	5.72	1608	940	104	276446.232	4.6 ± 0.3
062007E	6/20/07 12:58 PM	6/25/07 2:32 PM	151	17.60	423	310	61	91307.854	2.9 ± 0.5
062107E	6/21/07 2:15 PM	6/26/07 4:04 PM	718	5.49	1067	299	60	87986.303	14.5 ± 0.8
062207E	6/22/07 1:13 PM	6/27/07 4:50 PM	1054	4.75	1699	568	81	167009.731	11.7 ± 0.6
062807E	6/28/07 1:55 PM	6/29/07 3:26 PM	678	8.01	1635	879	100	258584.859	3.6 ± 0.3
071207E	7/12/07 1:35 PM	7/12/07 3:12 PM	7424	1.27	7974	301	60	88438.881	97 ± 1
071407E	7/14/07 1:00 PM	7/20/07 6:58 PM	13110	0.98	14335	821	97	241472.501	115 ± 1

Table B.2. Continued

Sample #	**Sample Collection Date & Time**	**Counting Start Date & Time**	**Peak Area**	**Counting Error (%)**	**Peak Integral**	**μ_B**	**L_D**	**Counting Time (s)**	**Volume Filtered (mL)**	**Mass (g)**	**^{131}I (Bq g^{-1})**
030707E	3/7/07 12:00 PM	3/8/07 5:33 PM	0	0.00	301	315	61	80807.180	620	0.00173	< L_D
030807E	3/8/07 12:38 PM	3/9/07 4:08 PM	0	0.00	279	295	59	75590.520	518	0.00080	< L_D
030907E	3/9/07 11:33 AM	3/10/07 1:16 PM	0	0.00	278	306	60	78371.140	698	0.00151	< L_D
031007E	3/10/07 11:15 AM	3/11/07 12:10 PM	1939	2.59	2324	383	67	98202.000	691	0.00134	651 ± 17
031107E	3/11/07 10:42 AM	3/12/07 3:36 PM	1208	3.41	1542	370	66	94813.060	639	0.00167	341 ± 12
031207E	3/12/07 1:07 PM	3/13/07 6:05 PM	1438	3.45	2109	668	88	171189.020	612	0.00177	221 ± 8
031307E	3/13/07 12:48 PM	3/15/07 5:56 PM	913	3.89	1154	313	61	80194.780	740	0.00187	295 ± 11
031407E	3/14/07 12:43 PM	3/16/07 4:31 PM	2322	2.70	3324	947	104	242829.280	661	0.00160	312 ± 8
031507E	3/15/07 12:46 PM	3/19/07 12:17 PM	8420	1.14	8890	419	70	107482.730	678	0.00160	2801 ± 32
031607E	3/16/07 12:41 PM	3/20/07 6:18 PM	5027	1.48	5348	291	59	74521.190	652	0.00301	1289 ± 19
032007E	3/20/07 11:12 AM	3/21/07 3:08 PM	695	5.60	1265	691	89	177207.130	612	0.00201	91 ± 5
032107E	3/21/07 1:12 PM	3/23/07 4:45 PM	802	5.27	1486	925	103	237156.900	675	0.00219	80 ± 4
032307E	3/23/07 12:42 PM	3/26/07 10:54 AM	2073	2.70	2781	743	92	190416.000	812	0.00261	227 ± 6
032707E	3/27/07 12:43 PM	3/28/07 4:06 PM	411	8.01	868	604	84	154784.660	686	0.00161	76 ± 6
032807E	3/28/07 1:12 PM	3/30/07 11:25 AM	6662	1.44	8382	1700	138	435881.300	585	0.00110	779 ± 11
052307E-A	5/23/07 11:46 AM	5/24/07 1:44 PM	3286	1.89	3647	360	65	92210.870	748	0.00088	1797 ± 34
052407E-A	5/24/07 10:33 AM	5/25/07 5:27 PM	6939	1.41	8514	1305	122	334557.410	675	0.00297	353 ± 5
052507E-A	5/25/07 1:35 PM	5/29/07 2:56 PM	3117	2.09	3837	693	89	177814.130	700	0.00114	916 ± 19

Table B.3. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in suspended solids > 0.7 µm in sewage effluent collected from the Stony Brook water pollution control plant. Background values (μ_B) and detection limits (L_D) were calculated according to Currie (1968) as described in Chapter 2. < L_D = less than detection limit.

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Volume Filtered (mL)	Mass (g)	^{131}I (Bq g^{-1})
053007E	5/30/07 12:12 PM	5/31/07 4:33 PM	3817	2.07	5323	1352	124	346574.520	701	0.00161	345 ± 7
053107E	5/31/07 1:00 PM	6/4/07 5:12 PM	2160	2.51	2689	629	85	161197.680	798	0.00209	383 ± 10
060407E	6/4/07 11:39 AM	6/6/07 2:16 PM	1055	4.17	1655	705	90	180704.000	706	0.00193	153 ± 6
060507E	6/5/07 2:09 PM	6/10/07 4:24 PM	122	18.95	409	316	61	81081.320	759	0.00153	61 ± 12
060607E	6/6/07 12:59 PM	6/8/07 4:59 PM	461	7.68	1001	663	87	169989.990	732	0.00109	126 ± 10
060707E	6/7/07 2:18 PM	6/12/07 5:47 PM	610	6.95	1432	1003	107	257115.030	740	0.00062	260 ± 18
060807E	6/8/07 2:25 PM	6/18/07 4:17 PM	376	10.08	1132	1010	107	259024.960	701	0.00092	164 ± 17
061107E	6/11/07 1:33 PM	6/15/07 5:47 PM	310	13.01	1204	985	106	252686.150	725	0.00106	72 ± 9
061207E	6/12/07 2:33 PM	6/21/07 4:32 PM	197	15.01	676	700	90	179461.550	731	0.00090	< L_D
061307E	6/13/07 1:00 PM	6/23/07 6:40 PM	411	8.17	910	616	84	157823.910	673	0.00071	369 ± 30
061407E	6/14/07 2:30 PM	6/25/07 2:44 PM	140	17.29	446	358	65	91801.820	688	0.00078	203 ± 35
061507E	6/15/07 2:30 PM	6/26/07 4:29 PM	375	7.63	677	342	64	87586.280	679	0.00062	722 ± 55
061907E	6/19/07 2:31 PM	6/27/07 4:57 PM	233	13.41	747	652	87	167076.660	719	0.00076	154 ± 21
062007E	6/20/07 12:58 PM	6/29/07 3:34 PM	308	12.03	1046	1009	107	258654.040	702	0.00041	< L_D
062207E	6/22/07 1:13 PM	7/5/07 3:13 PM	224	19.57	1385	1362	124	349265.510	625	0.00017	< L_D
062807E	6/28/07 1:55 PM	7/9/07 4:40 PM	25	144.84	874	990	106	253761.950	719	0.00026	< L_D
071207E	7/12/07 1:35 PM	7/17/07 12:46 PM	3135	2.14	4061	1095	112	280820.630	758	0.00059	1282 ± 27
071407E	7/14/07 1:00 PM	7/23/07 3:33 PM	2308	2.42	2859	617	84	158298.520	806	0.00115	1157 ± 28

Table B.3. Continued

Appendix C. Sample information and γ-ray spectrometry data for sewage effluent collected from the Blue Plains, Arlington and Alexandria water pollution control plants.

Sample #	Date & Time Collected	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
BP_EFF_012909B	1/29/09 10:15 AM	1/29/09 11:37 PM	800	4.45	981	255	55	74238.810	4.3 ± 0.2
BP050109E-7	5/1/09 8:10 AM	5/3/09 3:38 PM	1509	3.04	1715	298	59	86597.816	8.1 ± 0.2
BP071509E	7/15/09 2:30 PM	7/17/09 1:57 PM	1749	3.41	2856	1012	107	259464.320	3.9 ± 0.1
BP072209E	7/22/09 1:00 PM	7/24/09 2:30 PM	1620	3.80	2859	989	106	253658.700	3.7 ± 0.1
BP072909E	7/29/09 2:30 PM	7/31/09 10:15 AM	1466	4.05	2690	1069	110	273986.690	3.1 ± 0.1
BP080509E	8/5/09 2:55 PM	8/7/09 11:36 AM	1091	5.19	2367	1049	109	268972.830	2.3 ± 0.1
BP081209E	8/12/09 2:25 PM	8/14/09 11:34 AM	424	10.56	1452	1041	109	266838.600	0.9 ± 0.1
BP100709E	10/7/09 10:00 AM	10/9/09 2:56 PM	285	14.42	873	946	104	242516.960	$< L_D$
BP101409E	10/14/09 11:30 AM	10/16/09 12:03 PM	630	6.95	1469	982	106	251710.350	1.4 ± 0.1
BP041210E	4/12/10 4:30 PM	4/22/10 5:06 PM	338	12.97	1229	968	105	242106.401	1.4 ± 0.2
BP052610E	5/26/10 11:30 AM	5/30/10 8:54 AM	962	5.43	1484	588	82	158920.158	3.4 ± 0.2

Table C.1. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in sewage effluent collected from the **Blue Plains Water Pollution Control Plant**. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. $< L_D$ = less than detection limit. Sample volume = 0.15 L.

Sample #	Date & Time Collected	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	^{131}I (Bq L^{-1})
AR101409E	10/14/09 10:00 AM	10/16/09 11:54 AM	179	20.72	863	759	93	189763.355	0.5 ± 0.1
AR072209E	7/22/09 9:30 AM	7/24/09 2:34 PM	505	9.72	1198	976	105	250153.373	1.0 ± 0.1
AR072909E	7/29/09 10:30 AM	7/31/09 11:56 AM	22	197.36	768	996	107	255305.575	$< L_D$
AR080509E	8/5/09 9:30 AM	8/7/09 4:59 PM	1328	4.46	2075	965	105	247485.784	2.7 ± 0.1
AR081209E	8/12/09 9:30 AM	8/14/09 4:19 PM	166	28.38	834	979	106	250923.088	$< L_D$
AR071509E	7/15/09 9:15 AM	7/17/09 12:24 PM	262	15.40	808	967	105	247968.048	$< L_D$
AR100709E	10/7/09 8:00 AM	10/9/09 2:56 PM	75	52.13	693	951	104	243730.759	$< L_D$
AR052610E	5/26/10 9:30 AM	5/28/10 1:05 PM	117	33.20	581	612	84	156815.727	$< L_D$

Table C.2. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in sewage effluent collected from the **Arlington Water Pollution Control Plant**. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. $< L_D$ = less than detection limit. Sample volume = 0.15 L.

Sample #	**Date & Time Collected**	**Counting Start Date & Time**	**Peak Area**	**Counting Error (%)**	**Peak Integral**	μ_B	L_D	**Counting Time (s)**	**^{131}I (Bq L^{-1})**
AX052610E	5/26/10 10:30 AM	5/28/10 1:05 PM	0	0	525	542	79	157682.006	$< L_D$
AX061610E	6/16/10 9:30 AM	6/18/10 12:27 PM	164	30.80	856	835	98	242876.468	$< L_D$
AX062110E	6/21/10 2:30 PM	6/24/10 11:53 PM	0	0	418	519	78	133037.853	$< L_D$
AX100709E	10/7/09 2:00 PM	10/9/09 2:56 PM	297	14.44	1179	973	105	243183.983	0.61 ± 0.09
AX101509E	10/15/09 8:30 AM	10/16/09 12:22 PM	248	17.52	823	732	92	187635.180	$< L_D$

Table C.3. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in sewage effluent collected from the **Alexandria Water Pollution Control Plant**. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. $< L_D$ = less than detection limit. Sample volume = 0.15 L.

Appendix D. Sample information and γ-ray spectrometry data Potomac River surface water and suspended solids.

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	CF	^{131}I (Bq L^{-1})
PR022009W[1]	2/20/09 8:52 AM	2/21/09 7:48 PM	9962	1.19	11561	539	79	135427.141	5	6.07 ± 0.07
PR041609-1W	4/16/09 8:25 AM	4/20/09 6:50 PM	588	7.35	1088	636	86	167413.857	20	0.100 ± 0.007
PR041609-2W	4/16/09 10:00 AM	4/20/09 4:26 PM	6114	1.56	7258	347	64	87297.847	26	1.51 ± 0.02
PR041609-3W	4/16/09 12:03 PM	4/20/09 6:54 PM	946	4.30	1234	289	59	82712.214	31	0.61 ± 0.03
PR041609-4W	4/16/09 1:46 PM	4/21/09 5:59 PM	1967	2.58	2230	310	61	88803.176	26	1.52 ± 0.04
PR041609-BW[2]	4/16/09 3:06 PM	4/19/09 3:44 PM	6564	1.32	6954	340	63	97334.082	27	3.77 ± 0.05
PR062309-1W	6/23/09 8:00 AM	7/1/09 5:27 PM	0	0	309	225	52	56189.66	54	< L_D
PR062309-2W	6/23/09 9:32 AM	7/1/09 3:13 PM	623	5.81	836	243	54	65782.275	40	0.18 ± 0.01
PR062309-3W	6/23/09 1:15 PM	6/28/09 10:16 PM	368	16.06	1400	1053	109	129344.13	18	0.18 ± 0.03
PR062309-4W	6/23/09 11:40 AM	7/1/09 3:13 PM	929	4.68	1239	250	55	65879.226	38	0.28 ± 0.01
PR062309-BW[2]	6/23/09 3:27 PM	7/1/09 3:13 PM	1193	3.50	1515	262	56	65779.309	21	0.68 ± 0.02
PR082609-1W	8/26/09 8:10 AM	9/1/09 8:40 PM	450	8.49	780	371	66	100324.161	40	0.076 ± 0.006
PR082609-2W	8/26/09 10:09 AM	9/1/09 10:32 PM	615	6.77	1014	328	62	94076.469	29	0.46 ± 0.03
PR082609-3W	8/26/09 11:58 AM	8/31/09 10:12 PM	667	7.76	1306	665	88	81703.156	27	0.34 ± 0.03
PR082609-4W	8/26/09 1:58 PM	9/1/09 9:54 PM	800	4.79	1220	387	67	96345.12	25	0.25 ± 0.01
PR082609-BOIL[2]	8/26/09 9:48 AM	9/3/09 12:51 AM	924	4.55	1242	296	59	84715.586	44	0.54 ± 0.02
PR110209-1W	11/2/09 1:13 PM	11/9/09 10:25 PM	31	73.32	290	267	56	67059.948	27	< L_D
PR110209-2W	11/2/09 10:58 AM	11/9/09 10:24 PM	762	5.65	1234	547	80	156818.354	19	0.59 ± 0.03
PR110209-3W	11/2/09 7:53 AM	11/8/09 7:19 PM	313	9.96	596	324	62	92907.354	20	0.35 ± 0.03
PR110209-4W	11/2/09 9:26 AM	11/9/09 10:25 PM	1035	3.79	1205	247	54	66773.679	31	0.37 ± 0.01
PR110209-BOIL[2]	11/2/09 12:43 PM	11/9/09 10:45 PM	1221	3.36	1393	248	55	65363.069	22	0.62 ± 0.02

[1] Sample collected at station 2; [2] Sample collected in the Blue Plains sewage outfall 002

Table D.1. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in surface water collected at stations 1 through 4 and the Blue Plains sewage outfall 002 in the Potomac River. CF = concentration factor. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. < L_D = less than detection limit.

Sample #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	CF	^{131}I (Bq L^{-1})
PR062110-4A1	6/21/10 10:28 AM	6/25/10 11:41 PM	1271	3.67	1534	248	55	67070.505	27	0.40 ± 0.01
PR062110-4A2	6/21/10 10:28 AM	6/26/10 6:24 PM	553	7.24	861	268	57	72414.469	11	0.43 ± 0.03
PR062110-4A2-F	6/21/10 10:28 AM	6/27/10 2:43 PM	882	4.63	1105	360	65	97168.774	11	0.55 ± 0.03
PR062110-4B1	6/21/10 10:28 AM	6/24/10 11:52 PM	2999	2.19	3379	304	60	82085.874	46	0.415 ± 0.009
PR062110-4B2	6/21/10 10:28 AM	6/25/10 10:49 PM	1615	3.35	2182	356	65	89547.206	23	0.45 ± 0.02
PR062110-4B2-F	6/21/10 10:28 AM	6/27/10 2:43 PM	1433	3.60	2002	386	67	96972.475	23	0.43 ± 0.02
PR062110-4C1	6/21/10 10:28 AM	6/26/10 11:44 PM	587	6.25	934	213	51	53443.323	17	0.39 ± 0.02
PR062110-4C2	6/21/10 10:28 AM	6/26/10 1:05 PM	1107	4.13	1454	348	64	91537.522	16	0.45 ± 0.02
PR062110-4C2-F	6/21/10 10:28 AM	6/27/10 2:44 PM	1036	4.62	1450	367	66	96550.116	16	0.44 ± 0.02

Table D.2. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in replicate surface water samples collected at station 4 in the Potomac River. CF = concentration factor. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. See Figure D.1 for a description of the treatments.

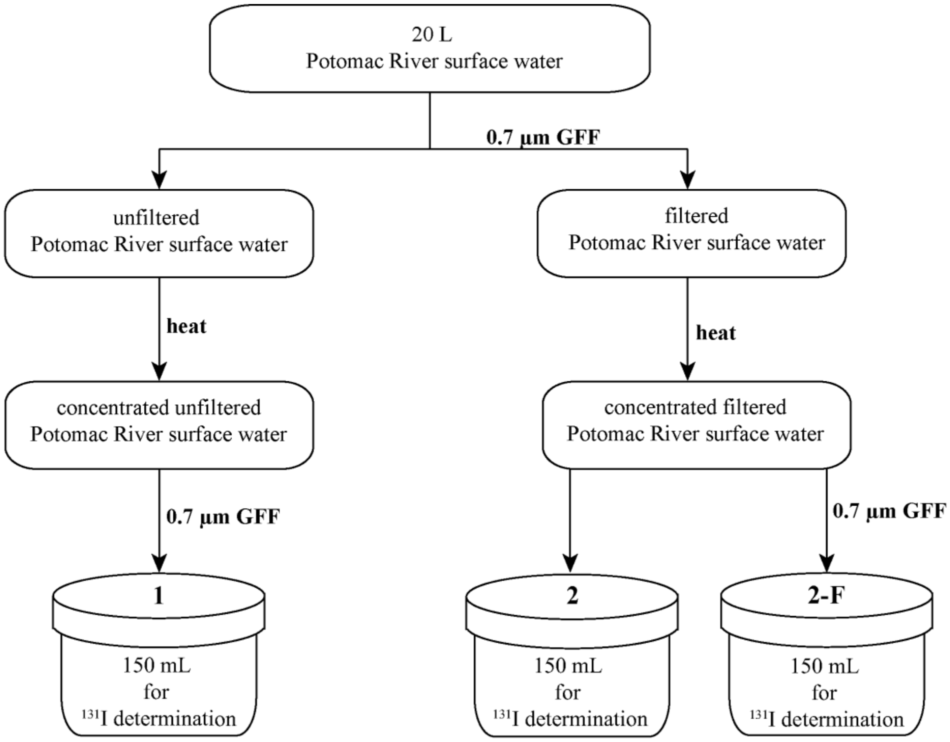

Figure D.1. Three 20 L surface water samples were collected in carboys. In the laboratory, the samples were split into two aliquots. The first aliquot was heated in an evaporating dish with no pre-treatment. After the concentration step, volume was recorded and any solids in the evaporating dish were rinsed into the container using 2.5% HNO_3. The volume was recorded to account for the addition of HNO_3 in the concentration factor. This sample was filtered through a GFF filter and 150 mL of the filtrate were used for determination of ^{131}I concentration (samples PR062110-4A1, B1 and C1). The second 10 L portion was filtered through a GFF and heated in an evaporating dish to reduce volume. After the concentration step, volume was recorded and any solids in the evaporating dish were rinsed into the container using 2.5% HNO_3. The volume was recorded to account for the addition of HNO_3 in the concentration factor. 150 mL were used for determination of ^{131}I concentration (samples PR062110-4A2, B2 and C2). A portion of this sample was filtered using a GFF filter a second time. 150 mL of this filtrate were used for the determination of ^{131}I (samples PR062110-4A2-F, 4B2-F and 4C2-F).

Sample #	**Sample Collection Date & Time**	**Counting Start Date & Time**	**Peak Area**	**Counting Error (%)**	**Peak Integral**	**μ_B**	**L_D**	**Counting Time (s)**	**Mass (g)**	**Volume filtered (L)**	**^{131}I (Bq kg^{-1})**
PR041210-1	4/12/10 9:22 AM	4/13/10 9:56 AM	0	0	1057	1953	148	283029.428	1.221	105.6	< L_D
PR041210-2	4/12/10 2:34 PM	4/13/10 9:56 AM	2235	2.58	2563	326	62	79621.740	0.408	255.0	3587 ± 93
PR041210-3	6/21/10 10:28 AM	4/14/10 8:12 AM	2510	2.62	3067	760	93	172690.485	0.811	107.3	1004 ± 26
PR041210-4	4/12/10 7:06 AM	4/14/10 8:12 AM	3807	2.14	4652	705	90	171892.309	1.155	103.5	1139 ± 24
PR041210-BOIL	6/21/10 10:28 AM	4/13/10 9:56 AM	2282	2.48	2542	352	64	79948.997	0.332	518.1	4178 ± 104
PR062110-4A	4/12/10 12:56 PM	6/23/10 8:10 PM	815	4.81	1044	437	71	99357.818	1.538	132.8	300 ± 14
PR062110-4B	4/12/10 11:35 AM	6/23/10 8:11 PM	1062	4.90	1925	1256	119	182025.059	1.576	136.1	237 ± 12
PR062110-4C	6/21/10 10:28 AM	6/23/10 8:12 PM	714	5.58	1026	406	69	98925.504	1.761	152.1	246 ± 14

Table D.3. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in suspended solids collected at stations 1 through 4 and in the Blue Plains sewage outfall 002 in the Potomac River. Mass of suspended solids on the filters was estimated by multiplying the suspended solids concentration by the volume filtered. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 2. < L_D = less than detection limit.

Appendix E. Carbon and nitrogen concentrations and stable isotope composition of Potomac River surface water and sediments.

Determination of Concentration and Stable Isotopic Composition of Dissolved Organic Carbon in Surface Water

Concentration and stable isotopic composition of DOC ($\delta^{13}C_{DOC}$) were determined using an OI Analytical 1010 TOC analyzer in-line with a Thermo Delta Plus XP isotope ratio mass spectrometer (IRMS) and a Conflo III interface using the wet chemical oxidation method described by Osburn and St-Jean (2007). All stable carbon isotope ratios are reported in units of per mil (‰) using the standard notation

$$\delta^{13}\mathrm{C} = \left[\frac{\mathrm{R_{sample}}}{\mathrm{R_{standard}}} - 1\right] \times 1000 \qquad (1)$$

where R = $^{13}C/^{12}C$ and the reference standard is Vienna Pee Dee Belemnite. DOC concentrations were normalized to a calibration curve generated for each run using potassium hydrogen phthalate (KHP) standard solutions. Stable isotopic composition of the KHP standard has been extensively calibrated against isotopic standards (IAEA C8 and USGS 40) and was used as a check standard throughout each run. Check standards were generally within 0.5 ‰ of actual values.

Station #	DOC (ppm)	$\delta^{13}C_{DOC}$ (‰)
April 2009		
1	3.0	-28.1
2	3.6	-27.0
3	3.4	-28.0
4	3.4	-27.3
June 2009		
1	4.3	-27.2
2	4.3	-27.0
3	4.1	-27.6
4	4.2	-27.6
Outfall	4.6	-26.6
August 2009		
1	3.9	-26.4
2	3.9	-26.2
3	5.1	-28.8
4	3.8	-26.7
Outfall	5.2	-28.7
November 2009		
1	4.7	-26.8
2	4.3	-26.2
3	4.5	-26.7
4	4.8	-26.6
Outfall	4.0	-26.1
April 2010		
1	2.3	-26.0
2	3.8	-25.5
3	2.8	-26.4
4	3.1	-26.3
Outfall	-	-

Table E.1. Dissolved organic carbon (DOC) concentrations and $\delta^{13}C$ values of DOC ($\delta^{13}C_{DOC}$) in surface waters collected at stations 1 though 4 and the Blue Plains sewage outfall 002 in the Potomac River. Dash indicates no data were collected.

Determination of Concentration and Stable Isotopic Composition of Carbon and Nitrogen in Sediments

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 (IRMS) and a Conflo III interface. Fifteen to 20 mg of dry, homogenized sediment were analyzed in triplicate. Total carbon and nitrogen samples were analyzed without treatment. For organic carbon, the samples were acidified with excess 10 % HCl and dried in a drying oven at 60 ˚C prior to analysis. A concentration calibration curve was generated for each run using acetanilide standard. Isotopic calibration curves were generated for δ^{15}N with isotopic standards: IAEA N2, IAEA NO3, USGS 25 and USGS 40 and for δ^{13}C with isotopic standards: IAEA C8, IAEA CH6 and USGS 40. Check standards (acetanilide, USGS 40 and IAEA C8) for both concentration and isotopic composition were analyzed throughout each run. Carbon concentrations of the check standards were no greater than 3 % difference and δ^{13}C values were within 0.4 ‰ for all analyses. Nitrogen concentrations of the check standards were no greater than 6 % of actual value. Check standards for δ^{15}N were within 0.2 ‰ for all analyses. All stable isotope ratios are reported in units of per mil (‰) using the standard notation

$$\delta^{15}\mathrm{N} \text{ or } \delta^{13}\mathrm{C} = \left[\frac{\mathrm{R_{sample}}}{\mathrm{R_{standard}}} - 1\right] \times 1000$$

where R = ^{15}N/^{14}N or ^{13}C/^{12}C the reference standards are atmospheric nitrogen and Vienna Pee Dee Belemnite, respectively. Values are reported with 1σ errors of replicate measurements.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
1	0 - 1	3.62 ± 0.03	-26.67 ± 0.07	3.47 ± 0.12	-27.66 ± 0.07	0.302 ± 0.002	7.97 ± 0.06	13.4 ± 0.5
	1 - 2	4.88 ± 0.26	-27.72 ± 0.14	4.31 ± 0.03	-27.61 ± 0.01	0.333 ± 0.011	5.44 ± 0.02	15.1 ± 0.5
	2 - 3	5.03 ± 0.09	-27.67 ± 0.04	4.64 ± 0.06	-27.62 ± 0.03	0.340 ± 0.006	5.30 ± 0.05	15.9 ± 0.5
	3 - 4	5.64 ± 0.32	-27.64 ± 0.08	5.34 ± 0.23	-28.02 ± 0.10	0.364 ± 0.009	5.28 ± 0.20	17.1 ± 0.8
	4 - 5	5.34 ± 0.04	-26.31 ± 0.01	5.30 ± 0.13	-27.67 ± 0.08	0.405 ± 0.005	5.01 ± 0.06	15.3 ± 0.4
2	0 - 1	1.54 ± 0.03	-26.81 ± 0.15	1.89 ± 0.12	-27.36 ± 0.16	0.118 ± 0.005	8.66 ± 0.06	19.0 ± 1.5
	1 - 2	1.81 ± 0.06	-26.41 ± 0.21	1.84 ± 0.04	-27.28 ± 0.17	0.116 ± 0.003	9.05 ± 0.24	18.2 ± 0.4
	2 - 3	2.07 ± 0.14	-26.35 ± 0.12	2.21 ± 0.03	-27.06 ± 0.24	0.128 ± 0.003	8.75 ± 0.19	20.2 ± 0.5
	3 - 4	2.08 ± 0.02	-26.29 ± 0.08	2.25 ± 0.17	-26.45 ± 0.14	0.170 ± 0.005	7.61 ± 0.23	15.3 ± 1.3
	4 - 5	2.42 ± 0.06	-25.74 ± 0.31	2.31 ± 0.08	-26.69 ± 0.07	0.169 ± 0.004	7.62 ± 0.09	16.0 ± 0.7
3	0 - 1	4.57 ± 0.12	-26.73 ± 0.07	4.37 ± 0.10	-27.62 ± 0.04	0.334 ± 0.005	5.01 ± 0.11	15.2 ± 0.4
	1 - 2	3.62 ± 0.06	-26.93 ± 0.03	3.43 ± 0.04	-27.61 ± 0.04	0.309 ± 0.002	8.24 ± 0.05	12.9 ± 0.2
	2 - 3	3.72 ± 0.10	-26.85 ± 0.05	3.47 ± 0.01	-27.61 ± 0.02	0.326 ± 0.008	7.57 ± 0.14	12.4 ± 0.3
	3 - 4	3.54 ± 0.10	-26.89 ± 0.03	3.39 ± 0.03	-27.56 ± 0.03	0.308 ± 0.003	7.67 ± 0.13	12.8 ± 0.1
	4 - 5	3.52 ± 0.04	-26.86 ± 0.03	3.44 ± 0.12	-27.50 ± 0.14	0.306 ± 0.002	7.42 ± 0.18	13.2 ± 0.5
4	0 - 1	3.20 ± 0.15	-26.57 ± 0.18	3.20 ± 0.13	-27.43 ± 0.11	0.223 ± 0.008	7.77 ± 0.15	16.7 ± 0.9
	1 - 2	3.26 ± 0.04	-27.38 ± 0.08	3.15 ± 0.08	-27.81 ± 0.06	0.213 ± 0.002	7.63 ± 0.11	17.2 ± 0.4
	2 - 3	3.28 ± 0.02	-27.28 ± 0.16	3.19 ± 0.03	-27.77 ± 0.10	0.215 ± 0.005	7.79 ± 0.19	17.4 ± 0.3
	3 - 4	3.36 ± 0.14	-26.82 ± 0.23	3.08 ± 0.02	-27.54 ± 0.04	0.227 ± 0.007	7.08 ± 0.18	15.8 ± 0.5
	4 - 5	3.12 ± 0.04	-26.70 ± 0.11	3.18 ± 0.06	-27.76 ± 0.04	0.214 ± 0.005	6.90 ± 0.04	17.4 ± 0.6

Table E.2. Percent total carbon (TC), $\delta^{13}C$ of total carbon ($\delta^{13}C_{TC}$), percent organic carbon (OC), $\delta^{13}C$ of organic carbon ($\delta^{13}C_{OC}$), percent total nitrogen (N), $\delta^{15}N$ of total nitrogen and molar ratios of organic carbon to nitrogen (C:N) in sediments collected at stations 1 through 4 in the Potomac River in **April 2009**. All values are reported ± 1σ errors based on replicate analyses.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
1	0 - 1	5.31 ± 0.20	-26.45 ± 0.07	4.58 ± 0.22	-27.40 ± 0.07	0.361 ± 0.003	5.39 ± 0.13	14.8 ± 0.7
	1 - 2	4.99 ± 0.20	-26.71 ± 0.09	4.47 ± 0.09	-27.36 ± 0.05	0.342 ± 0.005	5.24 ± 0.12	15.2 ± 0.4
	2 - 3	5.04 ± 0.02	-27.52 ± 0.04	4.88 ± 0.04	-27.93 ± 0.04	0.321 ± 0.007	5.01 ± 0.22	17.8 ± 0.5
	3 - 4	5.01 ± 0.21	-26.81 ± 0.10	4.77 ± 0.19	-27.66 ± 0.05	0.322 ± 0.008	4.97 ± 0.16	17.3 ± 0.8
	4 - 5	5.25 ± 0.08	-27.72 ± 0.18	4.91 ± 0.10	-27.58 ± 0.08	0.317 ± 0.011	4.81 ± 0.16	18.1 ± 0.8
	5 - 6	5.42 ± 0.09	-27.77 ± 0.09	5.22 ± 0.17	-27.50 ± 0.20	0.314 ± 0.012	4.49 ± 0.25	19.4 ± 0.9
	6 - 7	5.13 ± 0.26	-27.46 ± 0.10	5.26 ± 0.11	-27.35 ± 0.08	0.301 ± 0.003	4.61 ± 0.28	20.4 ± 0.4
	7 - 8	4.00 ± 0.10	-26.79 ± 0.12	4.12 ± 0.07	-27.30 ± 0.13	0.281 ± 0.003	5.23 ± 0.02	17.2 ± 0.4
	8 - 9	4.19 ± 0.11	-26.58 ± 0.09	4.33 ± 0.18	-27.17 ± 0.11	0.305 ± 0.003	5.19 ± 0.15	16.7 ± 0.7
	9 - 10	4.44 ± 0.04	-26.94 ± 0.04	4.23 ± 0.10	-27.70 ± 0.03	0.301 ± 0.005	5.12 ± 0.21	16.4 ± 0.4
2	0 - 1	1.49 ± 0.07	-26.90 ± 0.15	1.45 ± 0.08	-27.30 ± 0.18	0.097 ± 0.004	8.47 ± 0.08	17.6 ± 1.0
	1 - 2	1.83 ± 0.11	-26.86 ± 0.28	1.85 ± 0.20	-27.18 ± 0.08	0.104 ± 0.007	8.30 ± 0.13	20.7 ± 2.6
	2 - 3	2.07 ± 0.11	-27.11 ± 0.13	1.94 ± 0.06	-27.37 ± 0.09	0.120 ± 0.006	8.30 ± 0.10	19.2 ± 1.2
	3 - 4	1.71 ± 0.11	-26.79 ± 0.11	1.73 ± 0.08	-27.20 ± 0.11	0.102 ± 0.002	8.15 ± 0.04	19.7 ± 1.2
	4 - 5	1.77 ± 0.13	-26.89 ± 0.19	1.31 ± 0.01	-26.94 ± 0.22	0.105 ± 0.007	7.49 ± 0.06	14.7 ± 0.8
	5 - 6	1.94 ± 0.08	-26.35 ± 0.29	1.49 ± 0.0 9	-26.73 ± 0.09	0.117 ± 0.006	6.50 ± 0.27	15.1 ± 1.2
	6 - 7	1.96 ± 0.06	-25.89 ± 0.24	2.07 ± 0.08	-26.44 ± 0.14	0.133 ± 0.008	5.53 ± 0.62	18.3 ± 1.5
	7 - 8	2.88 ± 0.20	-25.65 ± 0.08	2.58 ± 0.16	-26.11 ± 0.01	0.198 ± 0.010	3.84 ± 0.46	15.2 ± 1.2
	8 - 9	3.57 ± 0.09	-25.31 ± 0.03	3.49 ± 0.14	-25.88 ± 0.16	0.247 ± 0.013	3.36 ± 0.16	16.4 ± 1.1
	9 - 10	4.25 ± 0.16	-24.69 ± 0.03	4.77 ± 0.17	-25.28 ± 0.07	0.392 ± 0.009	2.76 ± 0.14	14.2 ± 0.6

Table E.3. Percent total carbon (TC), $\delta^{13}C$ of total carbon ($\delta^{13}C_{TC}$), percent organic carbon (OC), $\delta^{13}C$ of organic carbon ($\delta^{13}C_{OC}$), percent total nitrogen (N), $\delta^{15}N$ of total nitrogen and molar ratios of organic carbon to nitrogen (C:N) in sediments collected at stations 1 through 4 in the Potomac River in **June 2009**. All values are reported ± 1σ errors based on replicate analyses.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
3	0 - 1	3.89 ± 0.07	-27.04 ± 0.04	3.59 ± 0.06	-27.69 ± 0.04	0.326 ± 0.007	7.34 ± 0.26	12.9 ± 0.3
	1 - 2	3.78 ± 0.05	-26.93 ± 0.01	3.45 ± 0.03	-27.55 ± 0.02	0.312 ± 0.003	7.22 ± 0.08	12.9 ± 0.1
	2 - 3	3.88 ± 0.06	-27.02 ± 0.06	3.50 ± 0.01	-27.55 ± 0.01	0.319 ± 0.003	7.33 ± 0.10	12.8 ± 0.2
	3 - 4	3.81 ± 0.02	-27.00 ± 0.02	3.51 ± 0.11	-27.64 ± 0.07	0.322 ± 0.007	7.56 ± 0.30	12.8 ± 0.5
	4 - 5	3.75 ± 0.02	-26.81 ± 0.02	3.57 ± 0.08	-27.56 ± 0.07	0.309 ± 0.001	7.45 ± 0.09	13.5 ± 0.3
	5 - 6	3.66 ± 0.10	-27.20 ± 0.03	3.76 ± 0.05	-27.41 ± 0.12	0.308 ± 0.004	7.25 ± 0.14	14.3 ± 0.3
	6 - 7	3.51 ± 0.11	-27.01 ± 0.04	3.39 ± 0.12	-27.45 ± 0.05	0.291 ± 0.004	7.06 ± 0.06	13.7 ± 0.5
	7 - 8	3.40 ± 0.08	-26.99 ± 0.04	3.35 ± 0.01	-27.42 ± 0.05	0.284 ± 0.005	6.98 ± 0.06	13.8 ± 0.2
	8 - 9	3.37 ± 0.12	-26.84 ± 0.23	3.43 ± 0.03	-27.39 ± 0.07	0.287 ± 0.010	7.08 ± 0.42	13.9 ± 0.6
	9 - 10	3.31 ± 0.15	-27.00 ± 0.06	3.41 ± 0.14	-27.36 ± 0.13	0.276 ± 0.005	6.89 ± 0.23	14.4 ± 0.7
4	0 - 1	3.90 ± 0.13	-26.95 ± 0.06	3.55 ± 0.07	-27.56 ± 0.04	0.284 ± 0.007	7.24 ± 0.05	14.6 ± 0.4
	1 - 2	4.02 ± 0.17	-27.03 ± 0.14	3.57 ± 0.03	-27.60 ±0.05	0.286 ± 0.006	7.08 ± 0.05	14.4 ± 0.3
	2 - 3	3.84 ± 0.05	-26.98 ± 0.05	3.73 ± 0.06	-27.74 ± 0.08	0.280 ± 0.004	7.23 ± 0.11	15.6 ± 0.4
	3 - 4	3.88 ± 0.15	-26.95 ± 0.03	3.50 ± 0.14	-27.57 ± 0.14	0.270 ± 0.007	7.06 ± 0.09	15.1 ± 0.7
	4 - 5	4.01 ± 0.18	-26.98 ± 0.01	3.52 ± 0.03	-27.51 ± 0.07	0.272 ± 0.007	7.06 ± 0.11	15.2 ± 0.5
	5 - 6	3.27 ± 0.08	-27.09 ± 0.12	3.24 ± 0.14	-27.54 ± 0.08	0.241 ± 0.005	6.97 ± 0.16	15.5 ± 0.7
	6 - 7	3.78 ± 0.15	-27.08 ± 0.12	3.38 ± 0.05	-27.79 ± 0.05	0.261 ± 0.009	6.72 ± 0.09	15.0 ± 0.6
	7 - 8	3.57 ± 0.14	-27.05 ± 0.05	3.65 ± 0.18	-27.82 ± 0.14	0.249 ± 0.004	6.52 ± 0.18	17.2 ± 0.9
	8 - 9	3.37 ± 0.08	-27.08 ± 0.18	3.41 ± 0.13	-27.38 ± 0.14	0.234 ± 0.002	6.46 ± 0.12	17.1 ± 0.7
	9 - 10	3.89 ± 0.08	-26.88 ± 0.06	3.56 ± 0.21	-27.73 ± 0.05	0.258 ± 0.006	6.48 ± 0.07	16.2 ± 1.0

Table E.3. Continued.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
1	0 - 1	4.51 ± 0.18	-26.70 ± 0.09	4.14 ± 0.41	-27.70 ± 0.10	0.254 ± 0.011	4.74 ± 0.09	19.1 ± 2.0
	1 - 2	5.66 ± 0.30	-26.72 ± 0.04	5.07 ± 0.05	-27.90 ± 0.28	0.309 ± 0.010	4.75 ± 0.07	19.1 ± 0.5
	2 - 3	4.77 ± 0.43	-26.85 ± 0.04	4.40 ± 0.15	-27.81 ± 0.16	0.292 ± 0.024	4.75 ± 0.10	17.5 ± 1.5
	3 - 4	4.94 ± 0.21	-26.78 ± 0.04	4.75 ± 0.18	-28.20 ± 0.23	0.286 ± 0.006	4.65 ± 0.15	19.4 ± 0.8
	4 - 5	5.22 ± 0.16	-26.88 ± 0.09	4.72 ± 0.22	-27.99 ± 0.27	0.294 ± 0.009	4.72 ± 0.13	18.7 ± 1.1
	5 - 6	4.37 ± 0.21	-26.82 ± 0.02	4.61 ± 0.21	-27.91 ± 0.15	0.289 ± 0.007	4.27 ± 0.18	18.6 ± 0.9
	6 - 7	4.50 ± 0.28	-26.20 ± 0.26	4.34 ± 0.13	-27.63 ± 0.08	0.302 ± 0.006	4.27 ± 0.10	16.7 ± 0.6
	7 - 8	3.92 ± 0.14	-25.81 ± 0.12	3.68 ± 0.12	-27.53 ± 0.01	0.286 ± 0.006	4.33 ± 0.15	15.0 ± 0.6
	8 - 9	3.25 ± 0.07	-25.26 ± 0.07	3.26 ± 0.12	-27.33 ± 0.10	0.232 ± 0.003	4.13 ± 0.16	16.4 ± 0.7
	9 - 10	3.24 ± 0.06	-24.90 ± 0.06	2.94 ± 0.03	-27.25 ± 0.02	0.212 ± 0.005	4.07 ± 0.12	16.2 ± 0.5
2	0 - 1	1.56 ± 0.07	-26.70 ± 0.34	1.53 ± 0.09	-27.04 ± 0.33	0.099 ± 0.005	9.31 ± 0.19	17.9 ± 1.5
	1 - 2	1.51 ± 0.09	-26.41 ± 0.13	1.44 ± 0.09	-27.01 ± 0.13	0.094 ± 0.002	9.09 ± 0.25	17.7 ± 1.3
	2 - 3	1.85 ± 0.17	-26.45 ± 0.28	1.45 ± 0.10	-26.97 ± 0.13	0.114 ± 0.014	8.37 ± 0.24	14.7 ± 2.1
	3 - 4	1.45 ± 0.11	-25.46 ± 0.27	1.62 ± 0.13	-25.89 ± 0.12	0.107 ± 0.004	7.31 ± 0.15	17.5 ± 1.6
	4 - 5	1.32 ± 0.08	-24.93 ± 0.10	1.30 ± 0.04	-25.61 ± 0.07	0.119 ± 0.004	6.78 ± 0.06	12.6 ± 0.6
	5 - 6	1.75 ± 0.23	-24.06 ± 0.45	1.32 ± 0.09	-25.46 ± 0.05	0.124 ± 0.004	4.94 ± 0.11	12.5 ± 0.9
	6 - 7	1.71 ± 0.08	-23.86 ± 0.08	1.70 ± 0.02	-25.18 ± 0.08	0.141 ± 0.001	5.34 ± 0.20	14.3 ± 0.2
	7 - 8	1.68 ± 0.03	-23.66 ± 0.05	1.56 ± 0.03	-25.21 ± 0.08	0.149 ± 0.001	5.20 ± 0.13	12.3 ± 0.3
	8 - 9	1.52 ± 0.08	-23.65 ± 0.34	1.35 ± 0.05	-25.19 ± 0.07	0.143 ± 0.003	5.34 ± 0.30	11.0 ± 0.5
	9 - 10	1.41 ± 0.03	-23.68 ± 0.06	1.36 ± 0.06	-25.16 ± 0.02	0.134 ± 0.001	5.12 ± 0.04	12.0 ± 0.5

Table E.4. Percent total carbon (TC), $\delta^{13}C$ of total carbon ($\delta^{13}C_{TC}$), percent organic carbon (OC), $\delta^{13}C$ of organic carbon ($\delta^{13}C_{OC}$), percent total nitrogen (N), $\delta^{15}N$ of total nitrogen and molar ratios of organic carbon to nitrogen (C:N) in sediments collected at stations 1 through 4 in the Potomac River in **August 2009**. All values are reported ± 1σ errors based on replicate analyses.

Station #	**Depth Interval (cm)**	**TC (%)**	**$\delta^{13}C_{TC}$ (‰)**	**OC (%)**	**$\delta^{13}C_{OC}$ (‰)**	**N (%)**	**$\delta^{15}N$ (‰)**	**C:N (mol mol^{-1})**
3	0 - 1	3.81 ± 0.01	-27.09 ± 0.01	3.77 ± 0.09	-28.06 ± 0.02	0.327 ± 0.003	7.79 ± 0.12	13.4 ± 0.4
	1 - 2	3.85 ± 0.01	-27.08 ± 0.06	3.74 ± 0.02	-28.01 ± 0.02	0.333 ± 0.001	7.69 ± 0.12	13.2 ± 0.1
	2 - 3	3.82 ± 0.04	-27.05 ± 0.02	3.68 ± 0.04	-28.00 ± 0.03	0.327 ± 0.002	7.56 ± 0.05	13.1 ± 0.2
	3 - 4	3.67 ± 0.02	-27.02 ± 0.08	3.53 ± 0.03	-27.88 ± 0.02	0.318 ± 0.003	7.36 ± 0.09	13.0 ± 0.2
	4 - 5	3.55 ± 0.02	-27.01 ± 0.01	3.56 ± 0.03	-27.91 ± 0.06	0.295 ± 0.002	7.26 ± 0.10	14.1 ± 0.2
	5 - 6	3.46 ± 0.05	-27.07 ± 0.07	3.47 ± 0.09	-27.84 ± 0.05	0.282 ± 0.001	6.14 ± 0.10	14.3 ± 0.4
	6 - 7	3.45 ± 0.06	-26.83 ± 0.36	3.42 ± 0.10	-27.81 ± 0.07	0.277 ± 0.001	5.99 ± 0.17	14.5 ± 0.4
	7 - 8	3.38 ± 0.02	-26.97 ± 0.15	3.36 ± 0.01	-27.80 ± 0.01	0.274 ± 0.005	6.09 ± 0.19	14.4 ± 0.2
	8 - 9	3.31 ± 0.05	-26.96 ± 0.04	3.36 ± 0.03	-27.76 ± 0.06	0.269 ± 0.004	6.01 ± 0.08	14.6 ± 0.3
	9 - 10	3.30 ± 0.03	-26.99 ± 0.07	3.34 ± 0.12	-27.76 ± 0.05	0.263 ± 0.006	5.78 ± 0.29	14.9 ± 0.6
4	0 - 1	4.04 ± 0.17	-26.93 ± 0.38	3.83 ± 0.09	-28.04 ± 0.12	0.308 ± 0.003	7.91 ± 0.12	14.5 ± 0.3
	1 - 2	3.99 ± 0.04	-27.25 ± 0.04	3.91 ± 0.08	-28.11 ± 0.04	0.309 ± 0.004	7.73 ± 0.20	14.7 ± 0.3
	2 - 3	4.02 ± 0.09	-27.05 ± 0.05	3.84 ± 0.03	-28.07 ± 0.11	0.300 ± 0.003	7.59 ± 0.11	14.9 ± 0.2
	3 - 4	4.03 ± 0.14	-27.10 ± 0.06	3.88 ± 0.10	-28.01 ± 0.05	0.295 ± 0.002	7.37 ± 0.16	15.3 ± 0.4
	4 - 5	4.13 ± 0.23	-27.07 ± 0.04	3.90 ± 0.02	-27.98 ± 0.13	0.291 ± 0.007	7.38 ± 0.14	15.5 ± 0.4
	5 - 6	3.95 ± 0.04	-27.83 ± 0.11	3.87 ± 0.09	-28.07 ± 0.06	0.264 ± 0.004	7.10 ± 0.28	17.1 ± 0.5
	6 - 7	3.74 ± 0.04	-27.22 ± 0.04	3.73 ± 0.02	-27.89 ± 0.07	0.265 ± 0.004	6.17 ± 0.17	16.5 ± 0.2
	7 - 8	3.82 ± 0.04	-27.18 ± 0.06	3.72 ± 0.12	-27.90 ± 0.06	0.261 ± 0.001	6.02 ± 0.08	16.6 ± 0.6
	8 - 9	3.96 ± 0.08	-27.22 ± 0.05	3.83 ± 0.20	-27.94 ± 0.09	0.260 ± 0.004	5.69 ± 0.09	17.3 ± 0.9
	9 - 10	3.84 ± 0.08	-27.18 ± 0.09	3.88 ± 0.14	-27.98 ± 0.07	0.251 ± 0.004	5.67 ± 0.05	18.0 ± 0.8

Table E.4. Continued.

Station #	**Depth Interval (cm)**	**TC (%)**	**$\delta^{13}C_{TC}$ (‰)**	**OC (%)**	**$\delta^{13}C_{OC}$ (‰)**	**N (%)**	**$\delta^{15}N$ (‰)**	**C:N (mol mol^{-1})**
1	0 - 1	4.63 ± 0.43	-27.41 ± 0.18	4.14 ± 0.04	-27.67 ± 0.11	0.250 ± 0.006	4.76 ± 0.10	19.3 ± 0.5
	1 - 2	4.59 ± 0.19	-27.64 ± 0.26	4.37 ± 0.29	-27.76 ± 0.09	0.266 ± 0.006	4.69 ± 0.13	19.2 ± 1.4
	2 - 3	5.20 ± 0.48	-27.71 ± 0.04	4.71 ± 0.11	-27.66 ± 0.24	0.290 ± 0.008	4.74 ± 0.23	19.0 ± 0.7
	3 - 4	4.66 ± 0.16	-27.50 ± 0.14	4.81 ± 0.29	-27.73 ± 0.03	0.283 ± 0.005	4.96 ± 0.03	19.8 ± 1.3
	4 - 5	4.29 ± 0.12	-27.12 ± 0.18	4.01 ± 0.05	-27.68 ± 0.09	0.273 ± 0.008	4.85 ± 0.17	17.2 ± 0.5
	5 - 6	4.26 ± 0.12	-26.84 ± 0.05	3.97 ± 0.10	-27.65 ± 0.04	0.290 ± 0.005	4.87 ± 0.29	16.0 ± 0.5
	6 - 7	4.11 ± 0.05	-26.66 ± 0.08	4.02 ± 0.09	-27.67 ± 0.10	0.304 ± 0.004	5.24 ± 0.11	15.3 ± 0.4
	7 - 8	4.26 ± 0.03	-26.54 ± 0.07	4.01 ± 0.06	-27.67 ± 0.06	0.319 ± 0.002	5.17 ± 0.14	14.7 ± 0.2
	8 - 9	4.30 ± 0.09	-26.34 ± 0.03	4.00 ± 0.02	-27.60 ± 0.11	0.330 ± 0.006	5.19 ± 0.12	14.2 ± 0.2
	9 - 10	4.09 ± 0.02	-25.98 ± 0.02	3.75 ± 0.12	-27.42 ± 0.09	0.320 ± 0.003	5.24 ± 0.02	13.7 ± 0.5
2	0 - 1	2.37 ± 0.26	-26.56 ± 0.42	2.26 ± 0.13	-27.26 ± 0.41	0.146 ± 0.010	8.09 ± 0.27	18.1 ± 1.6
	1 - 2	2.48 ± 0.37	-27.18 ± 0.29	2.10 ± 0.12	-27.51 ± 0.17	0.153 ± 0.014	7.91 ± 0.49	16.2 ± 1.9
	2 - 3	2.10 ± 0.32	-27.15 ± 0.07	2.01 ± 0.21	-27.41 ± 0.09	0.143 ± 0.015	7.87 ± 0.21	16.6 ± 2.4
	3 - 4	1.73 ± 0.15	-26.92 ± 0.15	1.71 ± 0.18	-27.46 ± 0.16	0.114 ± 0.003	7.63 ± 0.37	17.4 ± 1.8
	4 - 5	1.91 ± 0.20	-26.76 ± 0.15	1.93 ± 0.22	-27.27 ± 0.39	0.122 ± 0.004	6.92 ± 0.09	18.5 ± 2.1
	5 - 6	2.09 ± 0.14	-26.39 ± 0.18	1.87 ± 0.17	-26.95 ± 0.21	0.127 ± 0.005	5.56 ± 0.18	17.3 ± 1.7
	6 - 7	1.97 ± 0.21	-26.47 ± 0.65	1.64 ± 0.12	-26.73 ± 0.05	0.111 ± 0.002	5.52 ± 0.39	17.2 ± 1.4
	7 - 8	1.91 ± 0.19	-26.04 ± 0.17	1.74 ± 0.18	-26.80 ± 0.06	0.121 ± 0.006	4.96 ± 0.11	16.8 ± 2.1
	8 - 9	2.00 ± 0.05	-25.87 ± 0.07	1.93 ± 0.06	-26.47 ± 0.09	0.140 ± 0.005	5.39 ± 0.24	16.0 ± 0.6
	9 - 10	1.72 ± 0.12	-25.75 ± 0.04	1.70 ± 0.17	-26.37 ± 0.10	0.121 ± 0.004	4.97 ± 0.24	16.4 ± 1.7

Table E.5. Percent total carbon (TC), $\delta^{13}C$ of total carbon ($\delta^{13}C_{TC}$), percent organic carbon (OC), $\delta^{13}C$ of organic carbon ($\delta^{13}C_{OC}$), percent total nitrogen (N), $\delta^{15}N$ of total nitrogen and molar ratios of organic carbon to nitrogen (C:N) in sediments collected at stations 1 through 4 in the Potomac River in **November 2009**. All values are reported ± 1σ errors based on replicate analyses.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
3	0 - 1	3.84 ± 0.04	-27.66 ± 0.21	3.69 ± 0.04	-28.11 ± 0.04	0.322 ± 0.002	8.45 ± 0.14	13.4 ± 0.2
	1 - 2	3.79 ± 0.04	-27.91 ± 0.08	3.74 ± 0.06	-28.09 ± 0.16	0.333 ± 0.005	8.33 ± 0.15	13.1 ± 0.3
	2 - 3	3.66 ± 0.04	-27.79 ± 0.01	3.61 ± 0.01	-28.00 ± 0.03	0.321 ± 0.002	7.85 ± 0.06	13.2 ± 0.1
	3 - 4	3.68 ± 0.03	-27.73 ± 0.04	3.63 ± 0.04	-27.95 ± 0.07	0.320 ± 0.001	7.68 ± 0.12	13.3 ± 0.2
	4 - 5	3.60 ± 0.03	-27.56 ± 0.07	3.50 ± 0.04	-27.80 ± 0.05	0.302 ± 0.001	7.32 ± 0.12	13.5 ± 0.2
	5 - 6	3.48 ± 0.05	-27.46 ± 0.04	3.38 ± 0.02	-27.79 ± 0.03	0.283 ± 0.002	7.12 ± 0.22	14.0 ± 0.2
	6 - 7	3.38 ± 0.05	-27.41 ± 0.09	3.27 ± 0.03	-27.82 ± 0.02	0.277 ± 0.011	7.23 ± 0.40	13.7 ± 0.5
	7 - 8	3.25 ± 0.03	-27.38 ± 0.03	3.10 ± 0.04	-27.78 ± 0.01	0.261 ± 0.003	6.92 ± 0.19	13.8 ± 0.2
	8 - 9	3.33 ± 0.02	-27.38 ± 0.01	3.32 ± 0.15	-27.80 ± 0.09	0.266 ± 0.002	6.94 ± 0.24	14.6 ± 0.7
	9 - 10	3.27 ± 0.04	-27.30 ± 0.01	3.32 ± 0.25	-27.72 ± 0.02	0.267 ± 0.003	7.02 ± 0.05	14.5 ± 1.1
4	0 - 1	3.90 ± 0.13	-27.82 ± 0.05	3.79 ± 0.04	-28.11 ± 0.04	0.295 ± 0.005	8.16 ± 0.11	15.0 ± 0.2
	1 - 2	3.87 ± 0.07	-27.87 ± 0.05	3.92 ±0.03	-28.06 ± 0.05	0.295 ± 0.003	7.90 ± 0.15	15.5 ± 0.2
	2 - 3	3.88 ± 0.05	-27.85 ± 0.04	3.80 ± 0.02	-27.98 ± 0.03	0.296 ± 0.002	7.73 ± 0.11	14.9 ± 0.2
	3 - 4	3.86 ± 0.05	-27.78 ± 0.10	3.79 ± 0.12	-28.03 ± 0.03	0.289 ± 0.005	7.44 ± 0.19	15.3 ± 0.5
	4 - 5	3.79 ± 0.07	-27.72 ± 0.12	3.84 ± 0.16	-28.01 ± 0.05	0.280 ± 0.007	7.41 ± 0.16	15.9 ± 0.8
	5 - 6	3.79 ± 0.04	-27.60 ± 0.08	3.68 ± 0.07	-27.94 ± 0.05	0.264 ± 0.003	6.55 ± 0.07	16.3 ± 0.3
	6 - 7	3.78 ± 0.09	-27.52 ± 0.08	3.70 ± 0.12	-27.91 ± 0.07	0.265 ± 0.005	6.79 ± 0.32	16.4 ± 0.6
	7 - 8	4.01 ± 0.13	-27.59 ± 0.08	3.79 ± 0.12	-27.98 ± 0.13	0.264 ± 0.001	6.43 ± 0.27	16.7 ± 0.6
	8 - 9	4.02 ± 0.34	-27.55 ± 0.03	3.59 ± 0.04	-27.90 ± 0.03	0.268 ± 0.005	6.40 ± 0.08	15.7 ± 0.4
	9 - 10	4.05 ± 0.29	-27.37 ± 0.05	3.73 ± 0.05	-27.81 ± 0.04	0.275 ± 0.009	6.61 ± 0.13	15.8 ± 0.6

Table E.5. Continued.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
1	0 - 2	6.06 ± 0.07	-27.21 ± 0.05	5.09 ± 0.02	-28.21 ± 0.18	0.322 ± 0.008	5.33 ± 0.07	18.5 ± 0.4
	2 - 3	5.76 ± 0.14	-27.21 ± 0.15	5.13 ± 0.20	-28.23 ± 0.21	0.317 ± 0.004	5.10 ± 0.14	18.9 ± 0.8
	3 - 4	5.37 ± 0.19	-26.91 ± 0.11	4.69 ± 0.14	-27.95 ± 0.14	0.307 ± 0.007	5.26 ± 0.34	17.7 ± 0.6
	4 - 5	4.16 ± 0.22	-26.35 ± 0.21	3.68 ± 0.09	-27.76 ± 0.08	0.261 ± 0.005	5.75 ± 0.19	16.4 ± 0.5
	5 - 6	3.80 ± 0.12	-25.91 ± 0.24	3.11 ± 0.03	-27.51 ± 0.05	0.242 ± 0.009	5.83 ± 0.33	15.0 ± 0.6
	6 - 7	4.26 ± 0.08	-26.13 ± 0.07	3.65 ± 0.14	-27.57 ± 0.16	0.279 ± 0.004	5.63 ± 0.44	15.2 ± 0.7
	7 - 8	4.11 ± 0.08	-25.98 ± 0.07	3.53 ± 0.12	-27.58 ± 0.13	0.268 ± 0.006	5.72 ± 0.44	15.4 ± 0.6
	8 - 9	4.70 ± 0.13	-26.20 ± 0.02	4.07 ± 0.08	-27.72 ± 0.02	0.308 ± 0.008	5.58 ± 0.39	15.4 ± 0.4
	9 - 10	4.00 ± 0.05	-25.70 ± 0.01	3.32 ± 0.07	-27.57 ± 0.03	0.260 ± 0.011	5.65 ± 0.48	14.9 ± 0.6
2	0 - 2	2.33 ± 0.26	-26.78 ± 0.37	2.02 ± 0.11	-27.30 ± 0.18	0.134 ± 0.002	8.85 ± 0.29	17.5 ± 1.0
	2 - 4	2.28 ± 0.20	-27.11 ± 0.03	2.29 ± 0.13	-27.29 ± 0.17	0.147 ± 0.006	8.45 ± 0.25	18.2 ± 1.4
	4 - 6	2.12 ± 0.02	-26.87 ± 0.12	2.31 ± 0.08	-27.13 ± 0.09	0.125 ± 0.001	7.43 ± 0.15	21.5 ± 1.0
	6 - 8	1.50 ± 0.09	-25.95 ± 0.26	1.44 ± 0.05	-26.99 ± 0.37	0.086 ± 0.003	6.13 ± 0.12	19.5 ± 1.1
	8 - 10	2.06 ± 0.05	-25.47 ± 0.25	2.03 ± 0.20	-26.57 ± 0.07	0.147 ± 0.006	4.30 ± 0.60	16.1 ± 1.6

Table E.6. Percent total carbon (TC), $\delta^{13}C$ of total carbon ($\delta^{13}C_{TC}$), percent organic carbon (OC), $\delta^{13}C$ of organic carbon ($\delta^{13}C_{OC}$), percent total nitrogen (N), $\delta^{15}N$ of total nitrogen and molar ratios of organic carbon to nitrogen (C:N) in sediments collected at stations 1 through 4 in the Potomac River in **April 2010**. All values are reported ± 1σ errors based on replicate analyses.

Station #	Depth Interval (cm)	TC (%)	$\delta^{13}C_{TC}$ (‰)	OC (%)	$\delta^{13}C_{OC}$ (‰)	N (%)	$\delta^{15}N$ (‰)	C:N (mol mol^{-1})
3	0 - 2	3.92 ± 0.02	-27.63 ± 0.03	3.85 ± 0.03	-27.85 ± 0.06	0.320 ± 0.004	7.34 ± 0.08	14.1 ± 0.2
	2 - 3	3.93 ± 0.01	-27.69 ± 0.04	3.95 ± 0.01	-27.91 ± 0.07	0.325 ± 0.002	7.25 ± 0.10	14.2 ± 0.1
	3 - 4	3.92 ± 0.04	-27.88 ± 0.02	3.95 ± 0.01	-28.03 ± 0.06	0.324 ± 0.008	7.73 ± 0.19	14.2 ± 0.3
	4 - 5	3.82 ± 0.01	-27.96 ± 0.01	4.00 ± 0.07	-28.08 ± 0.03	0.315 ± 0.005	7.87 ± 0.04	14.9 ± 0.3
	5 - 6	3.78 ± 0.03	-27.80 ± 0.01	3.80 ± 0.06	-28.07 ± 0.03	0.308 ± 0.002	7.71 ± 0.20	14.4 ± 0.3
	6 - 7	3.71 ± 0.03	-27.81 ± 0.03	3.72 ± 0.04	-28.07 ± 0.07	0.309 ± 0.004	7.65 ± 0.08	14.1 ± 0.2
	7 - 8	3.71 ± 0.08	-27.80 ± 0.02	3.70 ± 0.04	-27.97 ± 0.05	0.299 ± 0.015	7.07 ± 0.71	14.5 ± 0.7
	8 - 9	3.66 ± 0.02	-27.70 ± 0.05	3.69 ± 0.04	-27.98 ± 0.02	0.299 ± 0.005	7.43 ± 0.18	14.3 ± 0.3
	9 - 10	3.53 ± 0.01	-27.64 ± 0.04	3.52 ± 0.05	-27.91 ± 0.07	0.285 ± 0.003	7.40 ± 0.05	14.4 ± 0.3
4	0 - 2	4.00 ± 0.02	-27.81 ± 0.05	3.98 ± 0.03	-27.96 ± 0.05	0.298 ± 0.005	7.53 ± 0.18	15.6 ± 0.4
	2 - 4	3.80 ± 0.01	-27.83 ± 0.10	3.72 ± 0.05	-27.99 ± 0.07	0.284 ± 0.013	7.39 ± 0.36	15.3 ± 0.7
	4 - 6	3.79 ± 0.10	-27.73 ± 0.14	3.85 ± 0.05	-28.04 ± 0.09	0.273 ± 0.003	7.39 ± 0.10	16.4 ± 0.2
	6 - 8	3.76 ± 0.05	-27.72 ± 0.19	3.74 ± 0.05	-27.93 ± 0.05	0.258 ± 0.011	6.42 ± 0.78	16.9 ± 0.8
	8 - 10	3.65 ± 0.03	-27.63 ± 0.09	3.84 ± 0.13	-27.76 ± 0.37	0.246 ± 0.005	6.25 ± 0.20	18.1 ± 0.8

Table E.6. Continued.

Appendix F. Sample information and γ-ray spectrometry data for Potomac River sediments.

Station #	Sampling Interval (cm)	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
1	0 - 1	4/18/09 8:07 PM	7054	1.28	7453	309	61	75315.537	34.93	117 ± 2
1	1 - 2	4/19/09 4:36 PM	21	173.51	435	379	67	86074.339	43.19	< L_D
1	2 - 3	4/21/09 4:52 PM	20	160.21	374	386	67	87735.369	45.36	< L_D
1	3 - 4	4/23/09 7:55 PM	0	0	911	915	102	89715.715	47.43	< L_D
1	4 - 5	4/24/09 8:54 PM	0	0	2343	2319	161	227381.235	39.02	< L_D
2	0 - 1	4/18/09 8:07 PM	2457	2.30	2787	330	62	70159.746	105.98	49 ± 1
2	1 - 2	4/18/09 8:38 PM	2114	4.23	4015	1658	137	162566.478	124.87	12.8 ± 0.5
2	2 - 3	4/22/09 5:37 PM	594	7.30	1128	386	67	94125.162	147.59	3.4 ± 0.2
2	3 - 4	4/22/09 6:46 PM	70	58.20	706	421	70	89671.437	146.17	1.3 ± 0.8
2	4 - 5	4/23/09 8:02 PM	631	11.93	2695	1287	121	314014.840	152.50	1.4 ± 0.2
3	0 - 1	4/18/09 8:18 PM	139	39.24	1211	511	77	74115.422	26.87	3 ± 1
3	1 - 2	4/19/09 5:15 PM	4910	1.62	5458	374	66	91165.171	54.05	46.7 ± 0.8
3	2 - 3	4/21/09 4:46 PM	1243	6.11	3055	647	86	93748.105	60.30	12.5 ± 0.8
3	3 - 4	4/22/09 6:54 PM	1177	12.00	8683	2810	177	407273.579	65.64	3.2 ± 0.4
3	4 - 5	4/23/09 7:58 PM	0	0	541	389	68	88381.209	78.39	< L_D
4	0 - 1	4/18/09 8:24 PM	4927	1.61	5367	302	60	68660.325	64.14	51.0 ± 0.8
4	1 - 2	4/19/09 4:57 PM	2658	3.07	4277	574	82	83228.546	79.34	23.3 ± 0.7
4	2 - 3	4/22/09 5:42 PM	981	4.78	1409	409	69	92963.566	88.96	7.8 ± 0.4
4	3 - 4	4/23/09 7:50 PM	136	26.39	571	338	63	72014.383	96.44	4 ± 1
4	4 - 5	4/24/09 8:57 PM	524	12.39	1743	985	106	223840.990	97.72	2.2 ± 0.3

Table F.1. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in sediments collected in **April 2009** at stations 1 through 4 in the Potomac River. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. < L_D = less than detection limit.

Station #	Sampling Interval (cm)	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
1	0 - 1	6/27/09 1:18 PM	230	14.69	567	412	70	93736.932	25.99	5.5 ± 0.8
1	1 - 2	6/28/09 3:23 PM	109	23.42	301	324	62	73551.772	23.66	$< L_D$
1	2 - 3	6/29/09 11:57 AM	74	39.33	347	353	65	80253.468	27.69	$< L_D$
1	3 - 4	6/30/09 10:51 AM	0	0	706	663	87	108735.920	28.36	$< L_D$
1	4 - 5	6/30/09 10:31 AM	10	424.96	528	422	70	102952.836	28.12	$< L_D$
2	0 - 1	6/27/09 2:17 PM	638	5.94	1104	496	76	71829.721	38.51	13.0 ± 0.8
2	1 - 2	6/28/09 10:31 AM	269	14.83	696	371	66	90499.285	39.79	4.6 ± 0.7
2	2 - 3	6/26/09 1:15 PM	264	12.59	744	619	85	89750.742	38.00	4.0 ± 0.5
2	3 - 4	6/29/09 11:58 AM	91	33.21	565	554	80	80220.780	38.68	$< L_D$
2	4 - 5	6/26/09 3:08 PM	31	85.85	462	522	78	85548.860	38.21	$< L_D$
3	0 - 1	6/26/09 1:21 PM	1851	2.78	2131	345	64	84228.813	23.27	49 ± 1
3	1 - 2	6/27/09 1:34 PM	240	23.89	1190	942	104	92387.041	24.55	11 ± 3
3	2 - 3	6/26/09 10:04 AM	325	11.66	688	430	71	97813.818	26.14	6.6 ± 0.8
3	3 - 4	6/27/09 3:03 PM	34	77.04	440	455	73	74587.000	25.73	$< L_D$
3	4 - 5	6/29/09 1:13 PM	45	66.81	548	472	74	77417.260	25.75	$< L_D$
4	0 - 1	6/28/09 11:55 AM	1034	4.35	1620	552	80	90563.300	28.31	30 ± 1
4	1 - 2	6/28/09 10:19 AM	460	7.61	914	628	85	91059.818	26.37	11.6 ± 0.9
4	2 - 3	6/29/09 11:58 AM	309	9.96	513	330	62	80366.475	26.69	10 ± 1
4	3 - 4	6/30/09 10:30 AM	79	42.30	440	453	73	102941.903	28.92	$< L_D$
4	4 - 5	6/30/09 10:31 AM	0	0	609	709	90	102793.333	30.55	$< L_D$

Table F.2. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in sediments collected in **June 2009** at stations 1 through 4 in the Potomac River. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. $< L_D$ = less than detection limit.

Station #	Sampling Interval (cm)	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
1	0 - 1	8/29/09 9:03 PM	352	16.48	1221	748	93	169979.828	31.96	3.7 ± 0.6
1	1 - 2	8/30/09 9:06 PM	79	42.42	449	365	66	89063.186	31.94	< L_D
1	2 - 3	8/28/09 9:34 PM	149	19.53	398	364	65	88760.747	32.27	< L_D
1	3 - 4	9/1/09 8:51 PM	38	92.33	458	407	69	99371.323	33.44	< L_D
1	4 - 5	9/1/09 8:45 PM	74	47.32	655	690	89	99982.787	30.60	< L_D
2	0 - 1	8/29/09 8:59 PM	782	5.77	1237	402	69	85494.147	38.55	38 ± 2
2	1 - 2	8/30/09 8:53 PM	652	7.73	1338	698	90	148539.639	45.46	17 ± 1
2	2 - 3	8/31/09 2:21 PM	449	9.44	1135	751	93	108894.245	46.25	5.6 ± 0.5
2	3 - 4	9/4/09 1:08 AM	264	15.96	805	627	85	152870.751	40.55	3.7 ± 0.6
2	4 - 5	8/31/09 10:25 PM	50	60.51	356	328	62	79996.296	34.35	< L_D
3	0 - 1	8/29/09 10:38 PM	1339	3.40	1794	468	74	76754.900	24.93	46 ± 2
3	1 - 2	8/29/09 10:26 PM	917	4.86	1302	326	62	79549.562	26.17	24 ± 1
3	2 - 3	8/30/09 8:06 PM	406	8.85	951	575	82	94302.950	24.84	12 ± 1
3	3 - 4	8/28/09 10:22 PM	228	15.83	823	530	78	86942.240	24.07	7 ± 1
3	4 - 5	9/4/09 1:07 AM	103	44.11	1107	1058	110	153355.802	26.26	< L_D
4	0 - 1	9/3/09 12:51 AM	1453	6.09	3305	1569	133	153852.942	28.46	47 ± 3
4	1 - 2	9/3/09 12:48 AM	780	5.72	1556	939	104	153982.110	28.31	17 ± 1
4	2 - 3	9/3/09 12:51 AM	365	13.37	1003	678	88	154173.044	29.08	6.4 ± 0.9
4	3 - 4	9/3/09 12:52 AM	26	132.23	619	588	83	85248.784	28.68	< L_D
4	4 - 5	9/3/09 12:52 AM	108	28.45	407	347	64	84725.519	29.11	< L_D

Table F.3. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in sediments collected in **August 2009** at stations 1 through 4 in the Potomac River. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. < L_D = less than detection limit.

Station #	Sampling Interval (cm)	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
1	0 - 1	11/6/09 8:11 PM	390	9.49	738	360	65	87827.217	32.91	7.9 ± 0.7
1	1 - 2	11/5/09 8:28 PM	220	16.94	594	431	71	98008.778	29.14	4.1 ± 0.7
1	2 - 3	11/8/09 7:19 PM	87	38.06	410	410	69	93075.315	28.28	$< L_D$
1	3 - 4	11/8/09 7:19 PM	94	32.05	528	643	86	93232.472	28.96	$< L_D$
1	4 - 5	11/8/09 7:19 PM	0	0	415	380	67	92768.226	29.97	$< L_D$
2	0 - 1	11/5/09 8:29 AM	928	4.55	3569	399	68	84873.408	40.20	42 ± 2
2	1 - 2	11/4/09 10:57 PM	1316	3.73	1860	531	78	76898.914	40.22	20.8 ± 0.8
2	2 - 3	11/6/09 11:05 AM	438	8.78	1032	544	79	89148.780	40.45	8.4 ± 0.7
2	3 - 4	11/7/09 11:59 AM	269	15.92	1247	1101	112	180546.970	41.81	2.8 ±0.4
2	4 - 5	11/4/09 10:58 PM	68	40.38	329	314	61	76490.290	44.23	$< L_D$
3	0 - 1	11/6/09 8:12 PM	916	4.26	3955	416	70	88491.321	22.75	80 ± 3
3	1 - 2	11/4/09 10:56 PM	650	6.34	950	338	63	76775.423	25.36	17 ± 1
3	2 - 3	11/6/09 11:47 PM	195	15.26	416	330	62	74896.001	24.42	6 ± 1
3	3 - 4	11/7/09 8:59 PM	114	24.66	353	321	62	78253.804	24.96	$< L_D$
3	4 - 5	11/7/09 8:59 PM	0	0	423	548	80	79365.541	23.72	$< L_D$
4	0 - 1	11/6/09 8:24 PM	918	7.24	1925	897	101	87921.569	28.26	39 ± 3
4	1 - 2	11/6/09 8:11 PM	522	7.20	998	609	84	88224.973	24.47	14 ± 1
4	2 - 3	11/5/09 8:29 PM	124	24.43	430	345	64	84125.105	25.46	3.1 ± 0.8
4	3 - 4	11/5/09 8:29 PM	27	121.89	555	565	81	81854.132	28.53	$< L_D$
4	4 - 5	11/7/09 8:58 PM	114	26.19	360	349	64	79382.569	28.18	$< L_D$

Table F.4. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in sediments collected in **November 2009** at stations 1 through 4 in the Potomac River. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. $< L_D$ = less than detection limit.

Station #	Sample Collection Date & Time	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
2	2/20/09 9:15 AM	2/21/09 7:43 PM	4429	1.92	5299	588	82	133587.969	236.05	9.4 ± 0.2
2	2/20/09 9:15 AM	3/3/09 11:40 AM*	1680	3.43	2311	525	78	119329.829	236.05	9.1 ± 0.3
1	4/12/10 7:06 AM	4/18/10 9:15 PM	117	26.08	409	382	67	93085.866	29.56	< L_D
2	4/12/10 12:56 PM	4/19/10 11:22 PM	579	6.69	868	335	63	76211.756	45.39	12.7 ± 0.9
3	4/12/10 9:22 AM	4/19/10 11:22 PM	775	5.92	1365	527	78	76305.848	22.96	33 ± 2
4	4/12/10 11:35 AM	4/19/10 11:22 AM	1655	3.21	2047	377	67	91925.587	25.56	53 ± 2

*Sediment sample collected on February 22, 2009 was counted twice

Table F.5. Sample information, γ-ray spectrometry data (364.5 keV) and ^{131}I concentrations ± 1σ counting error in sediments (top 2 cm) collected in **February 2009** and **April 2010** at stations 1 through 4 in the Potomac River. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 3. < L_D = less than detection limit.

Excess and Supported ^{210}Pb

The distribution of ^{210}Pb ($t_{½}$ = 22 y) in sediments is comprised of two components commonly referred to as supported ^{210}Pb and excess ^{210}Pb. The ultimate source of the two components is natural ^{238}U present in soils and sediments, which decays to ^{210}Pb through a series of isotopes with varying half-lives. The noble gas, ^{222}Rn, is produced in the decay series as a precursor to ^{210}Pb. A portion of the ^{222}Rn is lost to the atmosphere, decays to ^{210}Pb through a series of short-lived isotopes, and is removed from the atmosphere via precipitation. This atmospherically-derived component is scavenged by suspended particles in aquatic environments and subsequently deposited in the sediments. The ^{210}Pb that is deposited in sediments associated with particles is the excess ^{210}Pb component (Appleby and Oldfield, 1983; Cutshall et al., 1983; Koide et al., 1972).

Station #	**Depth Interval (cm)**	**Mass (g)**	**Total ^{210}Pb (Bq kg^{-1})**	**Supported ^{210}Pb (Bq kg^{-1})**	**Excess ^{210}Pb (Bq kg^{-1})**	**^{7}Be (Bq kg^{-1})**
1	0 - 1	24.18	152 ± 14	26 ± 2	128 ± 14	-
	1 - 2	22.90	214 ± 19	32 ± 2	185 ± 19	-
	2 - 3	25.21	179 ± 13	29 ± 2	152 ± 14	-
	3 - 4	25.46	230 ± 15	29 ± 2	204 ± 15	-
	4 - 5	24.35	244 ± 19	39 ± 3	206 ± 19	-
2	0 - 1	40.88	80 ± 9	15 ± 1	65 ± 9	-
	1 - 2		-	-	-	-
	2 - 3	39.26	74 ± 10	19 ± 1	55 ± 11	-
	3 - 4	35.46	68 ± 9	25 ± 1	43 ± 10	-
	4 - 5		-	-	-	-
3	0 - 1	24.68	255 ± 20	32 ± 2	223 ± 20	-
	1 - 2	26.16	151 ± 14	31 ± 2	122 ± 15	-
	2 - 3	25.63	159 ± 14	26 ± 2	135 ± 17	-
	3 - 4		-	-	-	-
	4 - 5		-	-	-	-
4	0 - 1	29.65	90 ± 12	16 ± 2	75 ± 13	-
	1 - 2	31.23	136 ± 14	29 ± 2	108 ± 14	-
	2 - 3	30.35	187 ± 15	27 ± 2	161 ± 15	-
	3 - 4	29.78	126 ± 9	28 ± 2	99 ± 10	-
	4 - 5	31.88	125 ± 9	27 ± 2	100 ± 13	-

Table F.6. Sample information, total ^{210}Pb (46.5keV), supported ^{210}Pb (352 keV), excess ^{210}Pb and ^{7}Be (477 keV) concentrations ± 1σ counting error in sediments collected in **April 2009** at stations 1 through 4 in the Potomac River. Dash indicates no data were collected.

Station #	**Depth Interval (cm)**	**Mass (g)**	**Total ^{210}Pb (Bq kg^{-1})**	**Supported ^{210}Pb (Bq kg^{-1})**	**Excess ^{210}Pb (Bq kg^{-1})**	**^{7}Be (Bq kg^{-1})**
1	0 - 1	25.99	196 ± 14	49 ± 2	150 ± 14	107 ± 6
	1 - 2	23.66	177 ± 3	44 ± 2	136 ± 11	73 ± 7
	2 - 3	27.69	211 ± 12	42 ± 2	172 ± 12	36 ± 6
	3 - 4	28.36	183± 10	41 ± 1	145 ± 10	$< L_D$
	4 - 5	28.12	187 ± 2	45 ± 1	145 ± 9	$< L_D$
	5 - 6	26.76	166 ± 11	39 ± 2	130 ± 12	$< L_D$
	6 - 7	28.03	183 ± 2	42 ± 2	144 ± 11	$< L_D$
	7 - 8	25.55	155 ± 2	33 ± 2	125 ± 15	$< L_D$
	8 - 9	26.43	179 ± 3	43 ± 2	139 ± 13	$< L_D$
	9 - 10	26.61	169 ± 9	49 ± 1	122 ± 9	$< L_D$
2	0 - 1	38.51	102 ± 7	28 ± 1	75 ± 8	12 ± 3
	1 - 2	39.79	78 ± 2	25 ± 1	53 ± 6	$< L_D$
	2 - 3	38.00	78 ± 6	26 ± 1	53 ± 6	$< L_D$
	3 - 4	38.68	80 ± 8	23 ± 1	57 ± 8	$< L_D$
	4 - 5	38.21	65 ± 3	24 ± 1	41 ± 6	$< L_D$
	5 - 6	43.16	50 ± 5	24 ± 1	26 ± 5	$< L_D$
	6 - 7	41.54	75 ± 8	27 ± 1	49 ± 8	$< L_D$
	7 - 8	37.57	84 ± 2	36 ± 1	49 ± 8	$< L_D$
	8 - 9	32.05	96 ± 9	42 ± 1	55 ± 9	$< L_D$
	9 - 10	27.51	108 ± 13	58 ± 2	50 ± 13	$< L_D$

Table F.7. Sample information, total ^{210}Pb (46.5keV), supported ^{210}Pb (352 keV), excess ^{210}Pb and ^{7}Be (477 keV) concentrations ± 1σ counting error in sediments collected in **June 2009** at stations 1 through 4 in the Potomac River. $< L_D$ = less than detection limit. Dash indicates no data were collected.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
3	0 - 1	23.27	187 ± 3	48 ±2	140 ± 13	88 ± 6
	1 - 2	24.55	138 ± 12	49 ± 2	89 ± 12	101 ± 15
	2 - 3	26.14	188 ± 14	52 ± 2	137 ± 14	41 ± 6
	3 - 4	25.73	201 ± 2	49 ± 2	154 ± 11	29 ± 5
	4 - 5	25.75	185 ± 13	49 ± 2	137 ± 14	$< L_D$
	5 - 6	25.30	192 ± 9	47 ± 1	146 ± 9	$< L_D$
	6 - 7	26.80	178 ± 2	48 ± 1	131 ± 8	$< L_D$
	7 - 8	27.54	158 ± 8	43 ± 1	116 ± 8	$< L_D$
	8 - 9	26.50	180 ± 12	50 ± 2	130 ± 12	$< L_D$
	9 - 10	27.65	156 ± 2	50 ± 2	106 ± 10	$< L_D$
4	0 - 1	28.31	152 ± 5	47 ± 1	106 ± 5	64 ± 6
	1 - 2	26.37	143 ± 8	49 ± 2	95 ± 8	46 ± 5
	2 - 3	26.69	128 ± 6	53 ± 1	75 ± 6	20 ± 4
	3 - 4	28.92	140 ± 7	51 ± 1	89 ± 7	$< L_D$
	4 - 5	30.55	132 ± 6	47 ± 1	86 ± 6	$< L_D$
	5 - 6	25.73	113 ± 5	41 ± 1	73 ± 5	$< L_D$
	6 - 7	28.66	123 ± 10	45 ± 2	80 ± 10	$< L_D$
	7 - 8	28.57	123 ± 4	44 ± 1	80 ± 4	$< L_D$
	8 - 9	29.23	100 ± 6	42 ± 1	58 ± 6	$< L_D$
	9 - 10	31.02	129 ± 8	43 ± 2	87 ± 8	$< L_D$

Table F.7. Continued.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
1	0 - 1	31.96	135 ± 2	36 ± 1	100 ± 10	23 ± 3
	1 - 2	31.94	159 ± 10	36 ± 2	124 ± 10	19 ± 4
	2 - 3	32.27	174 ± 7	37 ± 1	138 ± 7	$< L_D$
	3 - 4	33.44	152 ± 1	37 ± 1	117 ± 6	$< L_D$
	4 - 5	30.60	150 ± 8	40 ± 1	111 ± 8	$< L_D$
	5 - 6	27.99	181 ± 1	39 ± 1	144 ± 8	$< L_D$
	6 - 7	27.30	182 ± 2	52 ± 1	133 ± 9	$< L_D$
	7 - 8	27.27	166 ± 2	47 ± 1	121 ± 9	$< L_D$
	8 - 9	27.31	141 ± 1	45 ± 1	97 ± 6	$< L_D$
	9 - 10	30.20	162 ± 2	44 ± 2	120 ± 12	$< L_D$
2	0 - 1	38.55	84 ± 8	21 ± 1	64 ± 9	-
	1 - 2	45.46	55 ± 6	22 ± 1	33 ± 6	-
	2 - 3	46.25	57 ± 2	23 ± 1	35 ± 5	-
	3 - 4	40.55	66 ± 8	33 ± 1	33 ± 8	-
	4 - 5	34.35	90 ± 3	42 ± 1	48 ± 8	-
	5 - 6	33.91	93 ± 10	44 ± 1	50 ± 11	-
	6 - 7	31.59	77 ± 10	46 ± 2	30 ± 10	-
	7 - 8	28.70	97 ± 3	48 ± 1	50 ± 8	-
	8 - 9	29.94	87 ± 12	54 ± 2	34 ± 12	-
	9 - 10	28.58	109 ± 3	54 ± 2	55 ± 10	-

Table F.8. Sample information, total ^{210}Pb (46.5keV), supported ^{210}Pb (352 keV), excess ^{210}Pb and ^{7}Be (477 keV) concentrations ± 1σ counting error in sediments collected in **August 2009** at stations 1 through 4 in the Potomac River. $< L_D$ = less than detection limit. Dash indicates no data were collected.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
3	0 - 1	24.93	203 ± 13	52 ± 2	151 ± 13	67 ± 8
	1 - 2	26.17	195 ± 2	51 ± 2	145 ± 11	44 ± 5
	2 - 3	24.84	221 ± 15	56 ± 2	165 ± 15	48 ± 5
	3 - 4	24.07	174 ± 3	51 ± 2	124 ± 13	28 ± 5
	4 - 5	26.26	133 ± 13	58 ± 2	75 ± 13	< L_D
	5 - 6	24.82	165 ± 2	56 ± 2	110 ± 10	< L_D
	6 - 7	25.15	157 ± 12	49 ± 2	108 ± 12	< L_D
	7 - 8	25.45	171 ± 2	47 ± 2	125 ± 11	< L_D
	8 - 9	26.20	134 ± 11	48 ± 2	87 ± 11	< L_D
	9 - 10	25.68	178 ± 13	51 ± 2	128 ± 13	< L_D
4	0 - 1	28.46	146 ± 5	54 ± 1	93 ± 5	75 ± 5
	1 - 2	28.31	140 ± 6	49 ± 1	92 ± 6	44 ± 4
	2 - 3	29.08	135 ± 10	48 ± 2	87 ± 10	40 ± 4
	3 - 4	28.68	155 ± 10	49 ± 2	107 ± 10	24 ± 4
	4 - 5	29.11	146 ± 7	52 ± 1	95 ± 7	< L_D
	5 - 6	29.54	131 ± 7	47 ± 1	85 ± 7	< L_D
	6 - 7	29.62	128 ± 5	49 ± 1	79 ± 5	< L_D
	7 - 8	28.35	146 ± 11	46 ± 2	100 ± 11	< L_D
	8 - 9	29.60	149 ± 7	49 ± 1	100 ± 7	< L_D
	9 - 10	30.01	122 ± 9	46 ± 2	77 ± 9	< L_D

Table F.8. Continued.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
1	0 - 1	32.91	146 ± 9	34 ± 1	112 ± 9	24 ± 3
	1 - 2	29.14	154 ± 2	35 ± 1	120 ± 8	14 ± 4
	2 - 3	28.28	144 ± 7	35 ± 1	109 ± 7	< L$_D$
	3 - 4	28.96	168 ± 8	41 ± 1	128 ± 9	< L$_D$
	4 - 5	29.97	177 ± 2	44 ± 1	133 ± 8	< L$_D$
	5 - 6	27.37	168 ± 10	47 ± 2	121 ± 10	< L$_D$
	6 - 7	26.12	169 ± 9	49 ± 1	120 ± 9	< L$_D$
	7 - 8	25.01	191 ± 2	53 ± 1	139 ± 9	< L$_D$
	8 - 9	23.69	205 ± 10	51 ± 2	155 ± 10	< L$_D$
	9 - 10	21.60	186 ± 2	56 ± 2	131 ± 9	< L$_D$
2	0 - 1	40.20	104 ± 7	28 ± 1	76 ± 7	19 ± 2
	1 - 2	40.22	83 ± 7	26 ± 1	58 ± 7	17 ± 2
	2 - 3	40.45	89 ± 7	23 ± 1	66 ± 7	< L$_D$
	3 - 4	41.81	81 ± 2	24 ± 1	57 ± 6	< L$_D$
	4 - 5	44.23	82 ± 5	25 ± 1	57 ± 6	< L$_D$
	5 - 6	42.33	79 ± 4	27 ± 1	53 ± 4	< L$_D$
	6 - 7	45.53	85 ± 1	23 ± 1	62 ± 4	< L$_D$
	7 - 8	44.30	73 ± 6	26 ± 1	47 ± 6	< L$_D$
	8 - 9	43.00	78 ± 2	31 ± 1	48 ± 7	< L$_D$
	9 - 10	43.40	68 ± 2	26 ± 1	43 ± 5	< L$_D$

Table F.9. Sample information, total ^{210}Pb (46.5keV), supported ^{210}Pb (352 keV), excess ^{210}Pb and ^{7}Be (477 keV) concentrations ± 1σ counting error in sediments collected in **November 2009** at stations 1 through 4 in the Potomac River. < L$_D$ = less than detection limit.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
3	0 - 1	22.75	189 ± 2	54 ± 1	135 ± 9	61 ± 5
	1 - 2	25.36	188 ± 2	51 ± 2	138 ± 9	39 ± 4
	2 - 3	24.42	200 ± 2	49 ± 1	151 ± 9	29 ± 5
	3 - 4	24.96	171 ± 8	54 ± 1	117 ± 8	19 ± 4
	4 - 5	23.72	174 ± 10	53 ± 1	121 ± 10	$< L_D$
	5 - 6	24.36	185 ± 2	53 ± 1	134 ± 9	$< L_D$
	6 - 7	24.95	174 ± 1	53 ± 1	122 ± 7	$< L_D$
	7 - 8	25.13	134 ± 10	50 ± 1	85 ± 10	$< L_D$
	8 - 9	25.87	169 ± 2	49 ± 1	121 ± 10	$< L_D$
	9 - 10	24.75	182 ± 8	49 ± 1	134 ± 8	$< L_D$
4	0 - 1	28.26	219 ± 1	52 ± 1	168 ± 9	56 ± 4
	1 - 2	24.47	187 ± 2	45 ± 1	142 ± 8	38 ± 4
	2 - 3	25.46	195 ± 8	47 ± 1	148 ± 8	32 ± 4
	3 - 4	28.53	177 ± 10	46 ± 1	131 ± 10	$< L_D$
	4 - 5	28.18	204 ± 2	52 ± 1	152 ± 9	$< L_D$
	5 - 6	27.02	174 ± 1	49 ± 1	126 ± 7	$< L_D$
	6 - 7	27.96	193 ± 11	49 ± 1	145 ± 11	$< L_D$
	7 - 8	27.45	174 ± 2	50 ± 1	125 ± 10	$< L_D$
	8 - 9	27.79	163 ± 9	46 ± 1	117 ± 9	$< L_D$
	9 - 10	27.62	183 ± 2	47 ± 1	137 ± 8	$< L_D$

Table F.9. Continued.

Station #	**Depth Interval (cm)**	**Mass (g)**	**Total ^{210}Pb (Bq kg^{-1})**	**Supported ^{210}Pb (Bq kg^{-1})**	**Excess ^{210}Pb (Bq kg^{-1})**	**^{7}Be (Bq kg^{-1})**
1	0 - 2	29.56	124 ± 9	38 ± 1	87 ± 9	46 ± 4
	2 - 3	27.49	135 ± 9	41 ± 1	95 ± 9	24 ± 4
	3 - 4	27.16	143 ± 2	41 ± 1	103 ± 7	18 ± 4
	4 - 5	29.73	115 ± 7	42 ± 1	73 ± 8	20 ± 3
	5 - 6	30.03	141 ± 7	40 ± 1	101 ± 7	11 ± 3
	6 - 7	30.84	138 ± 8	40 ± 1	98 ± 8	20 ± 4
	7 - 8	31.16	131 ± 8	37 ± 1	94 ± 8	18 ± 3
	8 - 9	29.47	140 ± 7	42 ± 1	98 ± 7	19 ± 3
	9 - 10	30.19	151 ± 1	40 ± 1	111 ± 7	$< L_D$
2	0 - 2	45.39	92 ± 1	23 ± 1	69 ± 4	7 ± 1
	2 - 4	45.08	90 ± 2	22 ± 1	68 ± 6	$< L_D$
	4 - 6	43.27	84 ± 1	23 ± 1	61 ± 5	$< L_D$
	6 - 8	47.37	62 ± 4	17 ± 1	45 ± 4	$< L_D$
	8 - 10	35.99	87 ± 6	32 ± 1	55 ± 6	$< L_D$

Table F.10. Sample information, total ^{210}Pb (46.5keV), supported ^{210}Pb (352 keV), excess ^{210}Pb and ^{7}Be (477 keV) concentrations ± 1σ counting error in sediments collected in **April 2010** at stations 1 through 4 in the Potomac River. $< L_D$ = less than detection limit.

Station #	Depth Interval (cm)	Mass (g)	Total ^{210}Pb (Bq kg^{-1})	Supported ^{210}Pb (Bq kg^{-1})	Excess ^{210}Pb (Bq kg^{-1})	^{7}Be (Bq kg^{-1})
3	0 - 2	22.96	168 ± 7	49 ± 1	119 ± 7	39 ± 4
	2 - 3	21.73	154 ± 10	50 ± 2	104 ± 10	31 ± 5
	3 - 4	22.00	154 ± 8	49 ± 2	105 ± 9	16 ± 4
	4 - 5	23.11	199 ± 10	51 ± 2	148 ± 10	< L_D
	5 - 6	22.50	217 ± 2	49 ± 2	169 ± 9	< L_D
	6 - 7	25.22	185 ± 2	47 ± 1	138 ± 8	< L_D
	7 - 8	26.87	166 ± 7	45 ± 1	121 ± 8	< L_D
	8 - 9	26.00	125 ± 2	48 ± 1	77 ± 6	< L_D
	9 - 10	26.33	154 ± 7	48 ± 1	106 ± 7	< L_D
4	0 - 2	25.56	160 ± 9	44 ± 1	116 ± 9	44 ± 4
	2 - 4	24.39	193 ± 9	51 ± 1	142 ± 9	< L_D
	4 - 6	25.07	158 ± 8	45 ± 1	113 ± 8	< L_D
	6 - 8	29.79	181 ± 1	43 ± 1	139 ± 7	< L_D
	8 - 10	20.23	211 ± 10	67 ± 1	144 ± 10	< L_D

Table F.10. Continued.

Appendix G. Metals concentrations in Potomac River sediments.

Determination of Metals in Sediments

Metals concentrations in sediments were determined using polarized Energy Dispersive X-Ray Fluorescence (ED-XRF) on a Spectro X-Ray Extended Polarization Optical System (XEPOS). Three to 5 g of sediment were ground for five minutes in a tungsten carbide mill. XRF sample cups (31 mm diameter) were covered with a 4 µm Prolene® window. Sediment was placed in the cup and filled with polyester pillow stuffing, previously determined not to interfere with analysis (Smith, 2007). Each sample was run in triplicate under He purge. Replicate measurements of NIST SRM 2709 (San Joaquin soil) were determined on each run and used as check standards. Metals concentrations were generated using the factory installed global calibration method. The values reported varied from the standard by no more than 7%. Post-analysis, the data were normalized to the reference standard concentrations.

	Station #	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)	Ni (µg g^{-1})
April 2009	**1**	7.42 ± 0.17	26.1 ± 1.0	1.96 ± 0.03	0.98 ± 0.12	4.33 ± 0.09	48 ± 3
	2	5.39 ± 1.43	32.6 ± 1.7	1.55 ± 0.25	0.66 ± 0.03	2.90 ± 0.67	26 ±7
	3	7.52 ± 0.10	27.0 ± 0.3	1.96 ± 0.01	0.78 ± 0.11	4.47 ± 0.04	46 ± 2
	4	6.66 ± 0.05	29.0 ± 0.5	1.65 ± 0.02	0.70 ± 0.01	3.87 ± 0.05	35 ± 2
June 2009	**1**	7.16 ± 0.23	26.2 ± 0.8	1.91 ± 0.06	1.18 ± 0.08	4.12 ± 0.11	48 ± 4
	2	4.57 ± 1.10	32.6 ± 3.7	1.42 ± 0.14	0.66 ± 0.10	2.98 ± 1.19	26 ± 10
	3	7.62 ± 0.09	27.7 ± 0.4	1.97 ± 0.01	0.77 ± 0.03	4.40 ± 0.04	47 ± 3
	4	6.91 ± 0.19	27.9 ± 0.4	1.72 ± 0.03	0.70 ± 0.02	4.03 ± 0.08	43 ± 2
August 2009	**1**	7.18 ± 0.68	27.4 ± 1.0	2.01 ± 0.17	1.25 ± 0.05	4.10 ± 0.34	51 ± 7
	2	3.65 ± 0.40	35.8 ± 0.6	1.28 ± 0.07	0.59 ± 0.04	2.43 ± 0.23	24 ± 2
	3	7.59 ± 0.19	27.4 ± 0.7	1.96 ± 0.03	0.71 ± 0.05	4.45 ± 0.06	51 ± 2
	4	7.29 ± 0.09	28.1 ± 0.5	1.78 ± 0.02	0.72 ± 0.02	4.22 ± 0.06	47 ± 1
November 2009	**1**	6.95 ± 0.42	26.8 ± 0.9	1.91 ± 0.10	1.34 ± 0.24	3.89 ± 0.20	45 ± 4
	2	6.84 ± 1.92	31.9 ± 3.1	1.84 ± 0.37	0.52 ± 0.04	3.47 ± 1.05	34 ± 11
	3	7.70 ± 0.08	27.6 ± 0.5	1.99 ± 0.02	0.72 ± 0.03	4.48 ± 0.02	48 ± 2
	4	7.13 ± 0.08	28.0 ± 0.4	1.75 ± 0.02	0.73 ± 0.01	4.15 ± 0.05	45 ±1

Table G.1. Depth averaged metals concentrations ± 1σ in sediments collected at stations 1 through 4 in the Potomac River.

	Station #	Cu (μg g^{-1})	Zn (μg g^{-1})	As (μg g^{-1})	Rb (μg g^{-1})	Sr (μg g^{-1})	Pb (μg g^{-1})
April 2009	**1**	58 ± 5	233 ± 19	6 ± 1	100 ± 3	93 ± 2	44 ± 3
	2	35 ± 3	146 ± 6	3 ± 1	60 ±18	66 ± 8	33 ± 5
	3	54 ± 5	224 ± 20	6 ± 1	100 ± 2	92 ± 1	45 ± 1
	4	46 ± 1	207 ± 2	6 ± 1	73 ± 2	78 ± 1	45 ± 1
June 2009	**1**	51 ± 3	211 ± 5	6 ± 1	95 ± 5	99 ± 1	40 ± 3
	2	48 ± 28	200 ± 58	3 ± 2	52 ± 14	65 ± 15	44 ± 26
	3	51 ± 1	215 ± 2	7 ± 1	99 ± 1	89 ± 1	43 ± 1
	4	50 ± 2	226 ± 3	7 ± 1	82 ± 3	80 ± 2	50 ± 2
August 2009	**1**	46 ± 2	200 ± 4	7 ± 1	100 ± 15	100 ± 2	39 ± 2
	2	36 ± 5	155 ± 9	3 ± 1	48 ± 4	60 ± 6	33 ± 8
	3	51 ± 1	213 ± 4	7 ± 1	105 ± 1	90 ± 3	44 ± 1
	4	54 ± 1	231 ± 9	7 ± 1	89 ± 2	86 ± 3	52 ± 4
November 2009	**1**	47 ± 2	192 ± 15	5 ± 1	90 ± 9	98 ± 3	37 ± 4
	2	34 ± 8	124 ± 20	4 ± 1	85 ± 31	66 ± 10	47 ± 16
	3	52 ± 1	220 ± 2	7 ± 1	102 ± 2	89 ± 1	44 ± 1
	4	52 ± 1	229 ± 5	7 ± 1	86 ± 2	84 ± 2	50 ± 1

Table G.1. Continued.

Station #	**Depth Interval (cm)**	**Al (%)**	**Si (%)**	**K (%)**	**Ca (%)**	**Fe (%)**
1	0 - 1	7.69 ± 0.07	27.7 ± 0.1	1.97 ± 0.02	0.785 ± 0.007	4.46 ± 0.02
	1 - 2	7.31 ± 0.07	25.3 ± 0.1	1.92 ± 0.02	0.966 ± 0.010	4.31 ± 0.02
	2 - 3	7.40 ± 0.07	26.0 ± 0.1	1.94 ± 0.02	1.004 ± 0.011	4.37 ± 0.02
	3 - 4	7.44 ± 0.07	26.0 ± 0.1	1.98 ± 0.02	1.092 ± 0.011	4.28 ± 0.02
	4 - 5	7.25 ± 0.08	25.4 ± 0.1	2.01 ± 0.02	1.077 ± 0.011	4.22 ± 0.02
2	0 - 1	4.38 ± 0.04	33.7 ± 0.2	1.38 ± 0.01	0.708 ± 0.007	2.41 ± 0.01
	1 - 2	4.19 ± 0.07	33.9 ± 0.2	1.35 ± 0.02	0.650 ± 0.007	2.35 ± 0.01
	2 - 3	4.55 ± 0.07	33.8 ± 0.2	1.39 ± 0.01	0.647 ± 0.006	2.50 ± 0.01
	3 - 4	6.46 ± 0.06	30.9 ± 0.2	1.70 ± 0.02	0.647 ± 0.008	3.45 ± 0.01
	4 - 5	7.35 ± 0.05	30.7 ± 0.2	1.91 ± 0.02	0.638 ± 0.004	3.78 ± 0.01
3	0 - 1	7.40 ± 0.05	26.5 ± 0.2	1.94 ± 0.02	0.975 ± 0.006	4.41 ± 0.02
	1 - 2	7.63 ± 0.09	27.2 ± 0.2	1.95 ± 0.02	0.793 ± 0.005	4.47 ± 0.02
	2 - 3	7.56 ± 0.09	27.2 ± 0.1	1.97 ± 0.02	0.733 ± 0.004	4.50 ± 0.02
	3 - 4	7.58 ± 0.06	27.1 ± 0.1	1.97 ± 0.02	0.709 ± 0.005	4.50 ± 0.01
	4 - 5	7.45 ± 0.07	27.1 ± 0.1	1.96 ± 0.02	0.707 ± 0.005	4.46 ± 0.01
4	0 - 1	6.67 ± 0.05	29.1 ± 0.2	1.62 ± 0.02	0.714 ± 0.005	3.88 ± 0.02
	1 - 2	6.71 ± 0.05	28.4 ± 0.1	1.67 ± 0.02	0.706 ± 0.005	3.94 ± 0.01
	2 - 3	6.69 ± 0.05	28.8 ± 0.1	1.67 ± 0.02	0.691 ± 0.005	3.88 ± 0.01
	3 - 4	6.66 ± 0.06	29.5 ± 0.2	1.66 ± 0.02	0.689 ± 0.006	3.86 ± 0.02
	4 - 5	6.58 ± 0.08	29.4 ± 0.2	1.64 ± 0.02	0.695 ± 0.005	3.81 ± 0.02

Table G.2. Percent composition of Al, Si, K, Ca and Fe in Potomac River sediments collected at stations 1 through 4 in **April 2009**.

Station #	**Depth Interval (cm)**	**Ni ($\mu g\ g^{-1}$)**	**Cu ($\mu g\ g^{-1}$)**	**Zn ($\mu g\ g^{-1}$)**	**Pb ($\mu g\ g^{-1}$)**
1	0 - 1	46 ± 2	53 ± 4	210 ± 6	43 ± 3
	1 - 2	45 ± 3	58 ± 5	250 ± 8	48 ± 3
	2 - 3	52 ± 3	65 ± 6	255 ± 7	46 ± 2
	3 - 4	48 ± 6	59 ± 5	231 ± 7	44 ± 3
	4 - 5	50 ± 3	56 ± 5	222 ± 8	41 ± 2
2	0 - 1	22 ± 2	33 ± 3	142 ± 4	28 ± 1
	1 - 2	21 ± 2	31 ± 3	145 ± 4	33 ± 2
	2 - 3	22 ± 1	34 ± 3	140 ± 4	29 ± 2
	3 - 4	33 ± 5	38 ± 3	148 ± 4	36 ± 2
	4 - 5	34 ± 2	38 ± 3	156 ± 3	41 ± 2
3	0 - 1	46 ± 2	62 ± 5	260 ± 5	47 ± 2
	1 - 2	49 ± 6	52 ± 3	218 ± 5	44 ± 2
	2 - 3	44 ± 2	51 ± 3	212 ± 4	45 ± 2
	3 - 4	46 ± 2	53 ± 3	216 ± 7	45 ± 2
	4 - 5	47 ± 4	51 ± 4	214 ± 5	47 ± 3
4	0 - 1	32 ± 4	45 ± 3	203 ± 6	44 ± 2
	1 - 2	35 ± 2	46 ± 3	208 ± 7	45 ± 2
	2 - 3	37 ± 3	45 ± 3	206 ± 5	45 ± 2
	3 - 4	38 ± 4	47 ± 2	208 ± 6	45 ± 3
	4 - 5	35 ± 3	45 ± 3	208 ± 4	45 ± 3

Table G.3. Concentrations of Ni, Cu, Zn and Pb in Potomac River sediments collected at stations 1 through 4 in **April 2009**.

Station #	Depth Interval (cm)	As ($\mu g\ g^{-1}$)	Rb ($\mu g\ g^{-1}$)	Sr ($\mu g\ g^{-1}$)
1	0 - 1	7 ± 2	99 ± 1	91 ± 1
	1 - 2	5 ± 1	96 ± 2	92 ± 1
	2 - 3	6 ± 2	99 ± 2	93 ± 1
	3 - 4	6 ± 1	102 ± 1	95 ± 1
	4 - 5	6 ± 1	104 ± 3	95 ± 2
2	0 - 1	1.5 ± 0.5	47 ± 1	62 ± 1
	1 - 2	2.0 ± 0.2	46 ± 1	59 ± 1
	2 - 3	2.7 ± 0.2	49 ± 1	60 ± 1
	3 - 4	3 ± 1	73 ± 2	71 ± 1
	4 - 5	3 ± 1	85 ± 1	77 ± 1
3	0 - 1	5 ± 2	96 ± 1	94 ± 1
	1 - 2	7 ± 2	100 ± 2	92 ± 1
	2 - 3	7 ± 1	101 ± 2	92 ± 1
	3 - 4	6 ± 1	101 ± 2	92 ± 2
	4 - 5	5 ± 2	102 ± 2	90 ± 1
4	0 - 1	6 ± 1	73 ± 1	78 ± 2
	1 - 2	7 ± 1	75 ± 1	79 ± 1
	2 - 3	7 ± 1	74 ± 1	77 ± 1
	3 - 4	6 ± 1	72 ± 1	76 ± 1
	4 - 5	6 ± 2	70 ± 1	77 ± 1

Table G.4. Concentrations of As, Rb and Sr in Potomac River sediments collected at stations 1 through 4 in **April 2009**.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
1	0 - 1	7.02 ± 0.08	25.6 ± 0.3	1.90 ± 0.03	1.163 ± 0.017	4.14 ± 0.05
	1 - 2	7.29 ± 0.10	26.6 ± 0.3	1.93 ± 0.03	1.126 ± 0.015	4.14 ± 0.05
	2 - 3	6.93 ± 0.09	26.0 ± 0.3	1.85 ± 0.02	1.088 ± 0.015	4.04 ± 0.05
	3 - 4	6.88 ± 0.08	26.0 ± 0.3	1.85 ± 0.02	1.092 ± 0.015	4.06 ± 0.05
	4 - 5	7.21 ± 0.11	26.9 ± 0.4	1.90 ± 0.03	1.167 ± 0.017	4.04 ± 0.07
	5 - 6	7.13 ± 0.11	26.6 ± 0.4	1.90 ± 0.03	1.272 ± 0.017	4.02 ± 0.07
	6 - 7	7.22 ± 0.11	27.0 ± 0.4	1.92 ± 0.03	1.301 ± 0.017	4.10 ± 0.07
	7 - 8	6.92 ± 0.11	24.4 ± 0.4	1.85 ± 0.03	1.141 ± 0.015	4.01 ± 0.07
	8 - 9	7.44 ± 0.11	26.2 ± 0.4	2.00 ± 0.03	1.207 ± 0.016	4.24 ± 0.07
	9 - 10	7.58 ± 0.11	26.3 ± 0.4	2.04 ± 0.03	1.262 ± 0.020	4.37 ± 0.07
2	0 - 1	3.90 ± 0.06	35.2 ± 0.5	1.34 ± 0.02	0.609 ± 0.011	2.18 ± 0.04
	1 - 2	3.71 ± 0.06	35.6 ± 0.5	1.34 ± 0.02	0.558 ± 0.007	2.09 ± 0.04
	2 - 3	3.89 ± 0.06	34.5 ± 0.5	1.34 ± 0.02	0.584 ± 0.009	2.21 ± 0.04
	3 - 4	3.91 ± 0.03	35.3 ± 0.1	1.33 ± 0.01	0.603 ± 0.003	2.16 ± 0.01
	4 - 5	3.73 ± 0.03	35.0 ± 0.1	1.33 ± 0.01	0.570 ± 0.004	2.14 ± 0.01
	5 - 6	3.81 ± 0.04	34.7 ± 0.1	1.31 ± 0.01	0.579 ± 0.005	2.47 ± 0.01
	6 - 7	4.47 ± 0.06	32.8 ± 0.1	1.36 ± 0.01	0.673 ± 0.007	2.92 ± 0.01
	7 - 8	5.58 ± 0.05	30.1 ± 0.2	1.51 ± 0.01	0.726 ± 0.006	3.66 ± 0.01
	8 - 9	6.15 ± 0.06	27.8 ± 0.1	1.64 ± 0.02	0.804 ± 0.006	4.41 ± 0.01
	9 - 10	6.56 ± 0.07	25.2 ± 0.1	1.70 ± 0.02	0.850 ± 0.004	5.56 ± 0.02

Table G.5. Percent composition of Al, Si, K, Ca and Fe in Potomac River sediments collected at stations 1 through 4 in **June 2009**.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
3	0 - 1	7.60 ± 0.08	27.1 ± 0.1	1.98 ± 0.01	0.772 ± 0.010	4.46 ± 0.02
	1 - 2	7.68 ± 0.06	27.4 ± 0.1	1.97 ± 0.01	0.735 ± 0.006	4.45 ± 0.02
	2 - 3	7.63 ± 0.06	27.8 ± 0.1	1.98 ± 0.01	0.768 ± 0.005	4.42 ± 0.01
	3 - 4	7.79 ± 0.06	28.3 ± 0.1	1.99 ± 0.01	0.755 ± 0.008	4.45 ± 0.01
	4 - 5	7.67 ± 0.07	27.8 ± 0.2	1.97 ± 0.02	0.791 ± 0.008	4.41 ± 0.02
	5 - 6	7.67 ± 0.06	28.1 ± 0.1	1.98 ± 0.01	0.753 ± 0.006	4.40 ± 0.02
	6 - 7	7.55 ± 0.06	27.8 ± 0.1	1.96 ± 0.01	0.813 ± 0.005	4.37 ± 0.01
	7 - 8	7.54 ± 0.06	27.8 ± 0.1	1.97 ± 0.01	0.773 ± 0.006	4.36 ± 0.01
	8 - 9	7.51 ± 0.06	27.4 ± 0.1	1.95 ± 0.02	0.748 ± 0.006	4.37 ± 0.01
	9 - 10	7.53 ± 0.09	27.7 ± 0.1	1.97 ± 0.02	0.824 ± 0.007	4.35 ± 0.02
4	0 - 1	7.09 ± 0.09	28.0 ± 0.2	1.73 ± 0.02	0.736 ± 0.009	4.09 ± 0.02
	1 - 2	7.10 ± 0.04	27.4 ± 0.2	1.75 ± 0.01	0.710 ± 0.006	4.15 ± 0.01
	2 - 3	7.09 ± 0.05	27.8 ± 0.1	1.76 ± 0.02	0.698 ± 0.005	4.13 ± 0.02
	3 - 4	7.10 ± 0.04	28.4 ± 0.1	1.74 ± 0.01	0.687 ± 0.005	4.05 ± 0.01
	4 - 5	7.04 ± 0.04	28.4 ± 0.1	1.76 ± 0.02	0.703 ± 0.006	4.07 ± 0.01
	5 - 6	6.80 ± 0.03	27.2 ± 0.1	1.69 ± 0.03	0.682 ± 0.008	3.96 ± 0.01
	6 - 7	6.72 ± 0.05	27.8 ± 0.1	1.69 ± 0.01	0.681 ± 0.005	3.99 ± 0.01
	7 - 8	6.79 ± 0.04	28.0 ± 0.1	1.71 ± 0.01	0.685 ± 0.005	3.97 ± 0.02
	8 - 9	6.67 ± 0.04	27.7 ± 0.1	1.69 ± 0.01	0.684 ± 0.005	3.95 ± 0.02
	9 - 10	6.67 ± 0.03	27.9 ± 0.1	1.69 ± 0.02	0.691 ± 0.005	3.92 ± 0.01

Table G.5. Continued.

Station #	**Depth Interval (cm)**	**Ni (μg g^{-1})**	**Cu (μg g^{-1})**	**Zn (μg g^{-1})**	**Pb (μg g^{-1})**
1	0 - 1	53 ± 3	46 ± 4	204 ± 4	35 ± 3
	1 - 2	49 ± 3	49 ± 4	209 ± 5	36 ± 3
	2 - 3	46 ± 2	50 ± 5	214 ± 4	38 ± 3
	3 - 4	43 ± 2	50 ± 4	215 ± 5	41 ± 3
	4 - 5	46 ± 3	54 ± 5	210 ± 5	41 ± 2
	5 - 6	48 ± 3	55 ± 3	214 ± 5	41 ± 2
	6 - 7	49 ± 3	54 ± 3	212 ± 5	42 ± 2
	7 - 8	47 ± 3	51 ± 3	203 ± 5	40 ± 3
	8 - 9	48 ± 2	51 ± 3	207 ± 5	39 ± 2
	9 - 10	56 ± 2	51 ± 3	220 ± 5	43 ± 3
2	0 - 1	17 ± 3	28 ± 3	160 ± 4	26 ± 1
	1 - 2	18 ± 1	27 ± 2	162 ± 4	24 ± 1
	2 - 3	22 ± 2	32 ± 2	168 ± 4	26 ± 1
	3 - 4	21 ± 4	31 ± 3	163 ± 2	23 ± 1
	4 - 5	19 ± 1	29 ± 2	163 ± 2	26 ± 1
	5 - 6	20 ± 1	41 ± 5	168 ± 4	43 ± 2
	6 - 7	27 ± 2	42 ± 4	189 ± 3	40 ± 2
	7 - 8	34 ± 5	57 ± 4	225 ± 4	51 ± 2
	8 - 9	42 ± 4	82 ± 5	271 ± 3	76 ± 4
	9 - 10	45 ± 4	111 ± 7	328 ± 5	101 ± 5

Table G.6. Concentrations of Ni, Cu, Zn and Pb in Potomac River sediments collected at stations 1 through 4 in **June 2009**.

Station #	Depth Interval (cm)	Ni (µg g^{-1})	Cu (µg g^{-1})	Zn (µg g^{-1})	Pb (µg g^{-1})
3	0 - 1	51 ± 3	53 ± 5	216 ± 6	42 ± 2
	1 - 2	52 ± 2	52 ± 3	217 ± 3	42 ± 2
	2 - 3	46 ± 3	51 ± 3	213 ± 8	43 ± 2
	3 - 4	48 ± 3	52 ± 3	215 ± 8	43 ± 2
	4 - 5	46 ± 4	50 ± 5	215 ± 7	43 ± 3
	5 - 6	47 ± 2	50 ± 4	218 ± 8	43 ± 3
	6 - 7	44 ± 3	51 ± 4	215 ± 8	45 ± 3
	7 - 8	48 ± 5	51 ± 3	212 ± 7	44 ± 3
	8 - 9	46 ± 4	50 ± 3	215 ± 8	43 ± 2
	9 - 10	46 ± 3	50 ± 4	217 ± 8	45 ± 3
4	0 - 1	43 ± 5	49 ± 3	223 ± 9	48 ± 3
	1 - 2	45 ± 2	50 ± 3	222 ± 4	48 ± 3
	2 - 3	46 ± 3	51 ± 5	223 ± 5	49 ± 3
	3 - 4	44 ± 2	49 ± 5	222 ± 4	50 ± 3
	4 - 5	41 ± 1	53 ± 4	227 ± 4	50 ± 3
	5 - 6	41 ± 1	46 ± 4	226 ± 4	49 ± 3
	6 - 7	44 ± 3	51 ± 4	231 ± 5	51 ± 3
	7 - 8	41 ± 3	47 ± 5	232 ± 5	53 ± 3
	8 - 9	43 ± 1	49 ± 3	225 ± 4	53 ± 4
	9 - 10	40 ± 3	51 ± 3	224 ± 4	50 ± 3

Table G.6. Continued.

Station #	Depth Interval (cm)	As ($\mu g\ g^{-1}$)	Rb ($\mu g\ g^{-1}$)	Sr ($\mu g\ g^{-1}$)
1	0 - 1	6 ± 1	98 ± 1	101 ± 1
	1 - 2	5 ± 1	94 ± 1	99 ± 1
	2 - 3	4 ± 1	92 ± 2	99 ± 2
	3 - 4	6 ± 1	91 ± 1	100 ± 1
	4 - 5	5 ± 1	90 ± 2	99 ± 2
	5 - 6	6 ± 1	91 ± 2	99 ± 2
	6 - 7	7 ± 1	92 ± 1	100 ± 2
	7 - 8	6 ± 1	94 ± 1	96 ± 2
	8 - 9	7 ± 2	101 ± 2	98 ± 2
	9 - 10	6 ± 1	104 ± 2	100 ± 2
2	0 - 1	1.9 ± 0.2	45 ± 1	60 ± 1
	1 - 2	3 ± 1	44 ± 1	56 ± 1
	2 - 3	2 ± 1	45 ± 1	58 ± 1
	3 - 4	2 ± 1	42 ± 1	56 ± 1
	4 - 5	2.1 ± 0.3	43 ± 1	53 ± 1
	5 - 6	1.8 ± 0.8	43 ± 1	59 ± 1
	6 - 7	4.4 ± 0.4	48 ± 1	60 ± 1
	7 - 8	5 ± 1	60 ± 2	69 ± 1
	8 - 9	5 ± 2	69 ± 1	87 ± 1
	9 - 10	7 ± 2	82 ± 2	97 ± 1

Table G.7. Concentrations of As, Rb and Sr in Potomac River sediments collected at stations 1 through 4 in **June 2009**.

Station #	Depth Interval (cm)	As (μg g^{-1})	Rb (μg g^{-1})	Sr (μg g^{-1})
3	0 - 1	8 ± 1	101 ± 2	89 ± 1
	1 - 2	9 ± 1	102 ± 1	88 ± 1
	2 - 3	6 ± 1	98 ± 2	89 ± 2
	3 - 4	8 ± 1	99 ± 1	89 ± 1
	4 - 5	7 ± 1	98 ± 2	89 ± 2
	5 - 6	8 ± 1	99 ± 1	88 ± 1
	6 - 7	5 ± 1	98 ± 1	90 ± 2
	7 - 8	6 ± 1	99 ± 2	89 ± 2
	8 - 9	7 ± 2	100 ± 2	89 ± 2
	9 - 10	6 ± 2	100 ± 2	89 ± 1
4	0 - 1	6 ± 1	85 ± 1	83 ± 1
	1 - 2	7 ± 1	85 ± 1	83 ± 1
	2 - 3	6 ± 1	85 ± 1	82 ± 1
	3 - 4	7 ± 1	83 ± 1	80 ± 1
	4 - 5	7 ± 2	83 ± 1	81 ± 1
	5 - 6	7 ± 1	82 ± 1	80 ± 1
	6 - 7	8 ± 1	81 ± 1	80 ± 1
	7 - 8	5 ± 1	80 ± 1	80 ± 1
	8 - 9	7 ± 1	79 ± 1	77 ± 1
	9 - 10	7 ± 3	78 ± 1	77 ± 1

Table G.7. Continued.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
1	0 - 1	6.29 ± 0.05	29.0 ± 0.3	1.83 ± 0.02	1.183 ± 0.011	3.60 ± 0.02
	1 - 2	6.49 ± 0.08	26.4 ± 0.3	1.79 ± 0.03	1.150 ± 0.011	3.74 ± 0.02
	2 - 3	6.63 ± 0.07	26.8 ± 0.4	1.84 ± 0.02	1.167 ± 0.012	3.79 ± 0.03
	3 - 4	6.80 ± 0.06	27.3 ± 0.2	1.85 ± 0.03	1.176 ± 0.014	3.74 ± 0.01
	4 - 5	6.75 ± 0.05	26.3 ± 0.2	1.84 ± 0.03	1.162 ± 0.013	3.81 ± 0.01
	5 - 6	7.22 ± 0.05	26.2 ± 0.2	1.95 ± 0.03	1.230 ± 0.011	3.99 ± 0.01
	6 - 7	7.54 ± 0.06	26.3 ± 0.2	2.05 ± 0.03	1.431 ± 0.011	4.18 ± 0.01
	7 - 8	7.46 ± 0.05	26.2 ± 0.2	2.07 ± 0.03	1.494 ± 0.011	4.20 ± 0.02
	8 - 9	7.23 ± 0.05	26.0 ± 0.2	1.98 ± 0.03	1.607 ± 0.013	4.01 ± 0.02
	9 - 10	7.09 ± 0.04	27.7 ± 0.2	1.90 ± 0.02	1.847 ± 0.018	3.80 ± 0.02
2	0 - 1	4.14 ± 0.06	36.0 ± 0.2	1.31 ± 0.02	0.575 ± 0.005	2.11 ± 0.01
	1 - 2	3.91 ± 0.03	36.0 ± 0.2	1.30 ± 0.02	0.561 ± 0.004	2.08 ± 0.01
	2 - 3	4.76 ± 0.05	35.5 ± 0.2	1.46 ± 0.02	0.563 ± 0.003	2.30 ± 0.01
	3 - 4	6.24 ± 0.07	34.1 ± 0.1	1.64 ± 0.02	0.527 ± 0.003	2.80 ± 0.01
	4 - 5	7.68 ± 0.06	31.0 ± 0.1	2.01 ± 0.02	0.477 ± 0.005	3.61 ± 0.01
	5 - 6	7.81 ± 0.07	29.8 ± 0.1	2.02 ± 0.02	0.524 ± 0.006	3.85 ± 0.01
	6 - 7	8.11 ± 0.07	29.4 ± 0.1	2.10 ± 0.02	0.483 ± 0.007	4.19 ± 0.02
	7 - 8	8.50 ± 0.07	29.3 ± 0.2	2.17 ± 0.02	0.499 ± 0.005	4.48 ± 0.02
	8 - 9	8.61 ± 0.06	28.7 ± 0.2	2.20 ± 0.01	0.474 ± 0.004	4.66 ± 0.02
	9 - 10	8.69 ± 0.10	29.1 ± 0.2	2.20 ± 0.02	0.477 ± 0.004	4.59 ± 0.02

Table G.8. Percent composition of Al, Si, K, Ca and Fe in Potomac River sediments collected at stations 1 through 4 in **August 2009**.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
3	0 - 1	7.59 ± 0.07	27.2 ± 0.2	1.97 ± 0.02	0.792 ± 0.009	4.51 ± 0.02
	1 - 2	7.74 ± 0.06	27.3 ± 0.2	1.97 ± 0.01	0.738 ± 0.007	4.48 ± 0.02
	2 - 3	7.64 ± 0.06	27.0 ± 0.2	1.99 ± 0.02	0.710 ± 0.005	4.52 ± 0.02
	3 - 4	7.59 ± 0.06	26.9 ± 0.2	1.97 ± 0.02	0.698 ± 0.006	4.46 ± 0.02
	4 - 5	7.70 ± 0.07	27.5 ± 0.2	1.99 ± 0.01	0.698 ± 0.010	4.47 ± 0.02
	5 - 6	7.66 ± 0.06	27.5 ± 0.2	1.98 ± 0.02	0.704 ± 0.006	4.46 ± 0.02
	6 - 7	7.76 ± 0.05	28.0 ± 0.2	2.02 ± 0.01	0.712 ± 0.008	4.46 ± 0.03
	7 - 8	7.79 ± 0.07	28.1 ± 0.2	2.02 ± 0.02	0.745 ± 0.005	4.51 ± 0.03
	8 - 9	7.77 ± 0.06	28.0 ± 0.2	2.01 ± 0.02	0.699 ± 0.006	4.46 ± 0.03
	9 - 10	7.80 ± 0.05	28.1 ± 0.2	2.00 ± 0.01	0.691 ± 0.005	4.48 ± 0.03
4	0 - 1	7.16 ± 0.05	27.4 ± 0.2	1.79 ± 0.02	0.755 ± 0.006	4.25 ± 0.03
	1 - 2	7.25 ± 0.06	28.1 ± 0.2	1.79 ± 0.01	0.730 ± 0.007	4.21 ± 0.03
	2 - 3	7.06 ± 0.05	27.5 ± 0.2	1.73 ± 0.01	0.733 ± 0.005	4.17 ± 0.03
	3 - 4	7.15 ± 0.07	28.3 ± 0.2	1.74 ± 0.02	0.714 ± 0.006	4.14 ± 0.03
	4 - 5	7.00 ± 0.07	27.6 ± 0.3	1.74 ± 0.02	0.709 ± 0.009	4.14 ± 0.02
	5 - 6	7.21 ± 0.09	28.6 ± 0.3	1.76 ± 0.03	0.727 ± 0.011	4.15 ± 0.05
	6 - 7	7.15 ± 0.09	28.5 ± 0.3	1.77 ± 0.02	0.723 ± 0.014	4.11 ± 0.05
	7 - 8	7.20 ± 0.08	28.4 ± 0.3	1.76 ± 0.03	0.739 ± 0.011	4.11 ± 0.05
	8 - 9	7.02 ± 0.09	27.7 ± 0.3	1.73 ± 0.02	0.721 ± 0.010	4.09 ± 0.05
	9 - 10	7.08 ± 0.08	27.8 ± 0.3	1.72 ± 0.02	0.731 ± 0.011	4.09 ± 0.05

Table G.8. Continued.

Station #	Depth Interval (cm)	Ni ($\mu g\ g^{-1}$)	Cu ($\mu g\ g^{-1}$)	Zn ($\mu g\ g^{-1}$)	Pb ($\mu g\ g^{-1}$)
1	0 - 1	42 ± 5	46 ± 2	189 ± 5	38 ± 2
	1 - 2	41 ± 2	49 ± 3	204 ± 7	38 ± 2
	2 - 3	40 ± 3	50 ± 3	205 ± 5	40 ± 3
	3 - 4	45 ± 3	48 ± 3	201 ± 4	41 ± 4
	4 - 5	42 ± 3	47 ± 3	202 ± 6	43 ± 4
	5 - 6	48 ± 6	47 ± 3	205 ± 4	38 ± 3
	6 - 7	50 ± 3	45 ± 3	195 ± 4	37 ± 4
	7 - 8	51 ± 4	48 ± 4	188 ± 5	36 ± 4
	8 - 9	48 ± 5	45 ± 3	174 ± 4	32 ± 3
	9 - 10	45 ± 4	41 ± 4	160 ± 6	30 ± 3
2	0 - 1	18 ± 1	25 ± 2	102 ± 2	23 ± 2
	1 - 2	21 ± 2	25 ± 2	101 ± 2	22 ± 2
	2 - 3	21 ± 2	27 ± 2	98 ± 3	73 ± 3
	3 - 4	26 ± 2	28 ± 2	110 ± 3	35 ± 2
	4 - 5	36 ± 2	32 ± 3	128 ± 4	41 ± 2
	5 - 6	39 ± 3	36 ± 4	132 ± 4	51 ± 3
	6 - 7	41 ± 5	39 ± 2	140 ± 5	57 ± 2
	7 - 8	46 ± 3	43 ± 3	147 ± 5	61 ± 4
	8 - 9	44 ± 4	43 ± 4	142 ± 4	54 ± 3
	9 - 10	48 ± 4	44 ± 3	142 ± 4	49 ± 3

Table G.9. Concentrations of Ni, Cu, Zn and Pb in Potomac River sediments collected at stations 1 through 4 in **August 2009**.

Station #	Depth Interval (cm)	Ni (µg g^{-1})	Cu (µg g^{-1})	Zn (µg g^{-1})	Pb (µg g^{-1})
3	0 - 1	48 ± 5	51 ± 4	219 ± 7	43 ± 3
	1 - 2	48 ± 5	52 ± 3	220 ± 7	42 ± 2
	2 - 3	50 ± 5	53 ± 4	219 ± 6	43 ± 3
	3 - 4	48 ± 5	50 ± 4	221 ± 6	42 ± 2
	4 - 5	47 ± 3	53 ± 4	221 ± 6	44 ± 3
	5 - 6	46 ± 3	52 ± 3	221 ± 6	45 ± 3
	6 - 7	46 ± 3	51 ± 4	217 ± 5	44 ± 3
	7 - 8	54 ± 3	51 ± 5	223 ± 4	44 ± 3
	8 - 9	49 ± 4	53 ± 6	218 ± 4	45 ± 3
	9 - 10	49 ± 3	51 ± 3	220 ± 4	44 ± 3
4	0 - 1	46 ± 2	53 ± 3	219 ± 5	49 ± 3
	1 - 2	46 ± 3	54 ± 4	224 ± 4	49 ± 4
	2 - 3	47 ± 5	52 ± 3	229 ± 4	50 ± 3
	3 - 4	46 ± 3	52 ± 3	228 ± 4	50 ± 3
	4 - 5	44 ± 2	53 ± 4	227 ± 4	50 ± 3
	5 - 6	44 ± 3	51 ± 3	226 ± 4	47 ± 4
	6 - 7	45 ± 2	51 ± 3	229 ± 5	50 ± 4
	7 - 8	44 ± 5	51 ± 5	235 ± 6	51 ± 4
	8 - 9	45 ± 2	52 ± 4	235 ± 6	51 ± 4
	9 - 10	46 ± 4	50 ± 4	237 ± 4	51 ± 4

Table G.9. Continued.

Station #	**Depth Interval (cm)**	**As ($\mu g\ g^{-1}$)**	**Rb ($\mu g\ g^{-1}$)**	**Sr ($\mu g\ g^{-1}$)**
1	0 - 1	5 ± 1	79 ± 1	98 ± 1
	1 - 2	5 ± 1	83 ± 1	99 ± 1
	2 - 3	4 ± 2	83 ± 1	99 ± 1
	3 - 4	4 ± 1	83 ± 2	96 ± 1
	4 - 5	4 ± 2	84 ± 1	95 ± 1
	5 - 6	6 ± 1	94 ± 1	96 ± 2
	6 - 7	7 ± 1	103 ± 1	96 ± 1
	7 - 8	7 ± 1	104 ± 1	99 ± 1
	8 - 9	7 ± 1	98 ± 2	100 ± 2
	9 - 10	6 ± 2	89 ± 1	104 ± 1
2	0 - 1	ND	44 ± 1	54 ± 1
	1 - 2	2 ± 1	45 ± 1	55 ± 1
	2 - 3	ND	49 ± 1	55 ± 1
	3 - 4	3 ± 1	67 ± 1	58 ± 2
	4 - 5	4 ± 1	91 ± 3	68 ± 1
	5 - 6	5 ± 2	98 ± 4	70 ± 2
	6 - 7	6 ± 1	109 ± 4	75 ± 2
	7 - 8	4 ± 1	115 ± 2	77 ± 1
	8 - 9	6 ± 2	118 ± 5	75 ± 3
	9 - 10	5 ± 1	117 ± 4	76 ± 4

Table G.10. Concentrations of As, Rb and Sr in Potomac River sediments collected at stations 1 through 4 in **August 2009**.

Station #	Depth Interval (cm)	As ($\mu g\ g^{-1}$)	Rb ($\mu g\ g^{-1}$)	Sr ($\mu g\ g^{-1}$)
3	0 - 1	7 ± 1	99 ± 1	92 ± 1
	1 - 2	6 ± 1	102 ± 1	91 ± 3
	2 - 3	6 ± 1	101 ± 1	89 ± 1
	3 - 4	6 ± 1	102 ± 2	89 ± 3
	4 - 5	7 ± 2	101 ± 2	88 ± 3
	5 - 6	6 ± 2	100 ± 1	87 ± 1
	6 - 7	9 ± 2	104 ± 1	89 ± 1
	7 - 8	8 ± 1	104 ± 1	90 ± 2
	8 - 9	7 ± 2	103 ± 1	90 ± 1
	9 - 10	7 ± 2	103 ± 2	89 ± 1
4	0 - 1	8 ± 1	91 ± 1	89 ± 1
	1 - 2	7 ± 1	89 ± 1	86 ± 2
	2 - 3	6 ± 2	86 ± 2	85 ± 1
	3 - 4	7 ± 1	86 ± 1	83 ± 1
	4 - 5	7 ± 1	85 ± 1	84 ± 1
	5 - 6	9 ± 1	84 ± 1	82 ± 2
	6 - 7	6 ± 2	84 ± 1	84 ± 1
	7 - 8	7 ± 2	84 ± 1	83 ± 1
	8 - 9	7 ± 1	85 ± 1	84 ± 1
	9 - 10	8 ± 2	84 ± 2	84 ± 1

Table G.10. Continued.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
1	0 - 1	6.49 ± 0.07	29.8 ± 0.2	1.83 ± 0.02	1.221 ± 0.016	3.69 ± 0.02
	1 - 2	6.39 ± 0.08	27.7 ± 0.2	1.80 ± 0.02	1.212 ± 0.014	3.71 ± 0.02
	2 - 3	6.34 ± 0.06	27.1 ± 0.2	1.81 ± 0.02	1.224 ± 0.013	3.79 ± 0.02
	3 - 4	6.63 ± 0.06	27.3 ± 0.2	1.87 ± 0.02	1.215 ± 0.014	3.89 ± 0.02
	4 - 5	7.02 ± 0.06	28.0 ± 0.3	1.98 ± 0.02	1.219 ± 0.018	3.98 ± 0.03
	5 - 6	7.52 ± 0.07	27.8 ± 0.3	2.07 ± 0.02	1.254 ± 0.012	4.16 ± 0.02
	6 - 7	7.74 ± 0.05	27.5 ± 0.2	2.14 ± 0.02	1.245 ± 0.013	4.30 ± 0.02
	7 - 8	7.75 ± 0.09	26.5 ± 0.3	2.16 ± 0.02	1.216 ± 0.019	4.41 ± 0.05
	8 - 9	8.02 ± 0.05	26.4 ± 0.3	2.22 ± 0.02	1.280 ± 0.012	4.55 ± 0.03
	9 - 10	7.96 ± 0.06	26.3 ± 0.3	2.19 ± 0.02	1.391 ± 0.017	4.56 ± 0.03
2	0 - 1	4.16 ± 0.04	34.8 ± 0.3	1.39 ± 0.01	0.661 ± 0.007	2.47 ± 0.02
	1 - 2	4.10 ± 0.08	35.7 ± 0.4	1.33 ± 0.02	0.609 ± 0.009	2.47 ± 0.02
	2 - 3	3.84 ± 0.03	36.1 ± 0.3	1.34 ± 0.02	0.605 ± 0.007	2.33 ± 0.01
	3 - 4	3.61 ± 0.05	36.3 ± 0.4	1.28 ± 0.02	0.611 ± 0.006	2.17 ± 0.01
	4 - 5	3.56 ± 0.04	36.3 ± 0.4	1.30 ± 0.02	0.564 ± 0.006	2.28 ± 0.01
	5 - 6	3.33 ± 0.05	36.1 ± 0.4	1.23 ± 0.01	0.573 ± 0.006	2.30 ± 0.01
	6 - 7	2.89 ± 0.04	36.5 ± 0.4	1.15 ± 0.02	0.516 ± 0.006	2.17 ± 0.01
	7 - 8	3.33 ± 0.04	35.5 ± 0.4	1.22 ± 0.01	0.590 ± 0.006	2.50 ± 0.01
	8 - 9	3.99 ± 0.04	34.6 ± 0.3	1.32 ± 0.02	0.605 ± 0.009	2.88 ± 0.02
	9 - 10	3.68 ± 0.04	36.0 ± 0.3	1.23 ± 0.02	0.561 ± 0.005	2.69 ± 0.02

Table G.11. Percent composition of Al, Si, K, Ca and Fe in Potomac River sediments collected at stations 1 through 4 in **November 2009**.

Station #	Depth Interval (cm)	Al (%)	Si (%)	K (%)	Ca (%)	Fe (%)
3	0 - 1	7.75 ± 0.08	28.0 ± 0.2	1.98 ± 0.02	0.792 ± 0.007	4.53 ± 0.03
	1 - 2	7.74 ± 0.10	27.6 ± 0.2	1.99 ± 0.02	0.780 ± 0.008	4.52 ± 0.03
	2 - 3	7.61 ± 0.09	27.3 ± 0.2	1.96 ± 0.03	0.717 ± 0.008	4.48 ± 0.03
	3 - 4	7.41 ± 0.07	26.8 ± 0.2	1.92 ± 0.02	0.709 ± 0.008	4.46 ± 0.03
	4 - 5	7.51 ± 0.08	27.1 ± 0.2	1.93 ± 0.02	0.714 ± 0.006	4.43 ± 0.03
	5 - 6	7.56 ± 0.08	26.7 ± 0.2	1.95 ± 0.03	0.651 ± 0.007	4.47 ± 0.03
	6 - 7	7.34 ± 0.08	27.1 ± 0.2	1.92 ± 0.02	0.699 ± 0.010	4.38 ± 0.03
	7 - 8	7.56 ± 0.09	28.2 ± 0.2	1.98 ± 0.02	0.650 ± 0.006	4.36 ± 0.03
	8 - 9	7.46 ± 0.08	26.9 ± 0.2	1.93 ± 0.02	0.662 ± 0.007	4.37 ± 0.03
	9 - 10	7.97 ± 0.08	28.7 ± 0.2	2.02 ± 0.02	0.691 ± 0.006	4.49 ± 0.03
4	0 - 1	7.31 ± 0.07	27.7 ± 0.2	1.78 ± 0.03	0.764 ± 0.008	4.29 ± 0.03
	1 - 2	7.39 ± 0.08	27.7 ± 0.2	1.80 ± 0.02	0.733 ± 0.006	4.31 ± 0.02
	2 - 3	7.40 ± 0.07	27.4 ± 0.2	1.79 ± 0.02	0.675 ± 0.006	4.33 ± 0.03
	3 - 4	7.23 ± 0.08	27.8 ± 0.2	1.78 ± 0.02	0.696 ± 0.006	4.22 ± 0.02
	4 - 5	7.29 ± 0.08	27.9 ± 0.2	1.78 ± 0.02	0.722 ± 0.008	4.18 ± 0.02
	5 - 6	7.12 ± 0.07	27.7 ± 0.2	1.74 ± 0.02	0.694 ± 0.007	4.16 ± 0.02
	6 - 7	7.20 ± 0.07	28.3 ± 0.2	1.76 ± 0.02	0.712 ± 0.010	4.15 ± 0.03
	7 - 8	7.35 ± 0.09	28.9 ± 0.3	1.79 ± 0.02	0.734 ± 0.009	4.18 ± 0.02
	8 - 9	7.33 ± 0.07	28.8 ± 0.3	1.80 ± 0.01	0.720 ± 0.006	4.20 ± 0.02
	9 - 10	7.34 ± 0.06	28.6 ± 0.3	1.77 ± 0.01	0.719 ± 0.008	4.21 ± 0.02

Table G.11. Continued.

Station #	Depth Interval (cm)	Ni (µg g^{-1})	Cu (µg g^{-1})	Zn (µg g^{-1})	Pb (µg g^{-1})
1	0 - 1	43 ± 5	45 ± 4	191 ± 3	37 ± 2
	1 - 2	44 ± 3	47 ± 4	198 ± 4	38 ± 3
	2 - 3	43 ± 4	46 ± 5	202 ± 4	41 ± 3
	3 - 4	48 ± 3	48 ± 4	204 ± 4	42 ± 2
	4 - 5	47 ± 2	43 ± 4	195 ± 2	39 ± 2
	5 - 6	52 ± 3	49 ± 4	202 ± 4	39 ± 3
	6 - 7	58 ± 3	44 ± 4	201 ± 3	38 ± 2
	7 - 8	59 ± 3	47 ± 5	206 ± 3	38 ± 3
	8 - 9	60 ± 2	47 ± 5	202 ± 3	38 ± 2
	9 - 10	60 ± 5	46 ± 4	200 ± 2	37 ± 2
2	0 - 1	27 ± 1	39 ± 3	160 ± 5	26 ± 1
	1 - 2	27 ± 1	35 ± 4	154 ± 4	29 ± 3
	2 - 3	24 ± 3	34 ± 4	150 ± 2	27 ± 2
	3 - 4	25 ± 2	28 ± 2	145 ± 3	26 ± 2
	4 - 5	21 ± 2	33 ± 2	150 ± 3	47 ± 3
	5 - 6	22 ± 1	37 ± 4	154 ± 4	43 ± 3
	6 - 7	21 ± 1	34 ± 2	148 ± 4	29 ± 3
	7 - 8	25 ± 1	37 ± 3	155 ± 4	31 ± 2
	8 - 9	25 ± 3	41 ± 4	177 ± 4	40 ± 2
	9 - 10	25 ± 2	45 ± 4	158 ± 4	32 ± 2

Table G.12. Concentrations of Ni, Cu, Zn and Pb in Potomac River sediments collected at stations 1 through 4 in **November 2009**.

Station #	**Depth Interval (cm)**	**Ni ($\mu g\ g^{-1}$)**	**Cu ($\mu g\ g^{-1}$)**	**Zn ($\mu g\ g^{-1}$)**	**Pb ($\mu g\ g^{-1}$)**
3	0 - 1	52 ± 5	52 ± 4	216 ± 4	45 ± 2
	1 - 2	52 ± 4	53 ± 3	214 ± 5	43 ± 2
	2 - 3	51 ± 2	50 ± 4	216 ± 5	44 ± 3
	3 - 4	54 ± 3	53 ± 4	214 ± 4	46 ± 3
	4 - 5	52 ± 3	50 ± 4	213 ± 5	43 ± 3
	5 - 6	52 ± 3	51 ± 4	212 ± 5	45 ± 2
	6 - 7	50 ± 3	50 ± 3	210 ± 6	43 ± 3
	7 - 8	46 ± 3	51 ± 4	207 ± 5	43 ± 2
	8 - 9	49 ± 3	51 ± 4	212 ± 4	44 ± 3
	9 - 10	49 ± 3	52 ± 4	220 ± 6	45 ± 3
4	0 - 1	47 ± 3	54 ± 4	225 ± 5	48 ± 3
	1 - 2	46 ± 2	52 ± 3	222 ± 4	49 ± 3
	2 - 3	48 ± 5	54 ± 3	227 ± 5	49 ± 3
	3 - 4	45 ± 3	54 ± 4	229 ± 5	51 ± 3
	4 - 5	47 ± 3	55 ± 3	228 ± 5	50 ± 3
	5 - 6	46 ± 4	52 ± 4	223 ± 5	53 ± 3
	6 - 7	46 ± 3	53 ± 3	230 ± 5	54 ± 3
	7 - 8	48 ± 3	57 ± 3	240 ± 6	52 ± 3
	8 - 9	45 ± 5	55 ± 3	243 ± 6	61 ± 3
	9 - 10	47 ± 2	55 ± 3	247 ± 6	55 ± 3

Table G.12. Continued.

Station #	Depth Interval (cm)	As ($\mu g\ g^{-1}$)	Rb ($\mu g\ g^{-1}$)	Sr ($\mu g\ g^{-1}$)
1	0 - 1	5 ± 1	82 ± 1	101 ± 1
	1 - 2	5 ± 1	82 ± 1	102 ± 1
	2 - 3	7 ± 2	86 ± 1	102 ± 1
	3 - 4	5 ± 1	90 ± 1	101 ± 1
	4 - 5	6 ± 1	95 ± 1	100 ± 1
	5 - 6	7 ± 1	103 ± 1	100 ± 1
	6 - 7	7 ± 1	108 ± 1	99 ± 1
	7 - 8	8 ± 2	115 ± 1	97 ± 1
	8 - 9	9 ± 1	119 ± 1	98 ± 2
	9 - 10	8 ± 1	119 ± 2	98 ± 1
2	0 - 1	5 ± 1	53 ± 1	68 ± 1
	1 - 2	ND	54 ± 1	67 ± 1
	2 - 3	2.1 ± 0.3	51 ± 1	66 ± 1
	3 - 4	2.4 ± 0.4	47 ± 1	59 ± 1
	4 - 5	2.5 ± 0.3	47 ± 1	59 ± 1
	5 - 6	2.8 ± 0.3	45 ± 1	55 ± 1
	6 - 7	3.4 ± 0.4	41 ± 1	51 ± 1
	7 - 8	3.6 ± 0.4	45 ± 1	56 ± 1
	8 - 9	4 ± 1	51 ± 1	62 ± 1
	9 - 10	3 ± 1	46 ± 1	55 ± 1

Table G.13. Concentrations of As, Rb and Sr in Potomac River sediments collected at stations 1 through 4 in **November 2009**.

Station #	Depth Interval (cm)	As (µg g^{-1})	Rb (µg g^{-1})	Sr (µg g^{-1})
3	0 - 1	6 ± 1	104 ± 2	95 ± 1
	1 - 2	7 ± 1	105 ± 3	93 ± 1
	2 - 3	5 ± 1	107 ± 1	91 ± 1
	3 - 4	5 ± 1	108 ± 1	91 ± 1
	4 - 5	7 ± 1	106 ± 1	89 ± 1
	5 - 6	7 ± 1	106 ± 1	88 ± 2
	6 - 7	7 ± 1	106 ± 1	88 ± 1
	7 - 8	7 ± 1	102 ± 1	86 ± 1
	8 - 9	7 ± 2	106 ± 1	88 ± 1
	9 - 10	7 ± 2	104 ± 1	88 ± 1
4	0 - 1	7 ± 2	89 ± 1	91 ± 1
	1 - 2	7 ± 2	92 ± 1	89 ± 1
	2 - 3	7 ± 2	92 ± 2	87 ± 1
	3 - 4	6 ± 1	89 ± 1	87 ± 1
	4 - 5	8 ± 1	89 ± 1	85 ± 1
	5 - 6	7 ± 1	88 ± 1	84 ± 1
	6 - 7	8 ± 2	87 ± 1	84 ± 1
	7 - 8	8 ± 1	87 ± 1	84 ± 1
	8 - 9	7 ± 2	87 ± 1	84 ± 1
	9 - 10	7 ± 1	86 ± 1	84 ± 1

Table G.13. Continued.

Appendix H. Iodine-131 in *Fucus* sp. collected from Port Jefferson Harbor, NY, USA.

Previous Work

Sewage effluent from the Stony Brook Water Pollution Control Plant (WPCP) travels through a pipe to Port Jefferson, NY where it is combined with sewage effluent from the Port Jefferson WPCP (Suffolk County Sewer District #1). The combined effluent is discharged to Port Jefferson Harbor, an embayment of Long Island Sound. The shared outfall is located near the southwestern shore of the harbor (Figure H.1). Medical use is the only known source of ^{131}I to Port Jefferson Harbor. Previous work showed that sewage effluent from the SBWPCP is the primary source of ^{131}I to Port Jefferson Harbor. In the same study, ^{131}I was measured in *Fucus* sp. in and around Port Jefferson Harbor. The *Fucus* sp. was collected at the three sites shown in Figure H.1. Seaweed collected from site 1, located near the Setauket Yacht Club, had the highest concentrations of ^{131}I. The concentrations decreased from site 2 to 3. The concentrations measured in the seaweed reflected the flow pattern of sewage effluent leaving the sewage outfall in the harbor as determined by Nuzzi (1984). In that study, it was determined that sewage effluent leaving the outfall had a tendency to be carried toward the eastern shore on an ebb current in a counterclockwise manner. The concentrations measured in the seaweed at site 1 on three different dates were 224 ± 2, 254 ± 2 and 260 ± 5 Bq kg^{-1} (Rose, 2003). These values suggest steady state conditions despite differences in the number of patients treated in the month prior to collection of samples which would imply daily variations in ^{131}I concentrations in the sewage effluent.

This Work

Sample Collection and Determination of ^{131}I

In the current study, *Fucus* sp. was collected at site 1 (now known as Harborfront Park) on three different days. Samples were collected at or near low water. Whole seaweed fronds and holdfast were removed from rocks. Several fronds were collected from rocks distributed throughout the sampling area. In the lab, the samples were rinsed with deionized water to remove particulate matter. The entire sample was chopped in a blender. A portion of the composite sample was packed into a pre-weighed 150 mL straight-side polypropylene jar (64 mm height; 64 mm diameter) for counting. After counting, the sample was dried at 60 ˚C to obtain dry mass.

Iodine-131 was determined by γ-ray spectrometry using Canberra low energy germanium detectors. The activity of ^{131}I was measured using the emission at 364.5 keV. Previous work demonstrated that there is no self-absorption of γ-rays emitted at 364.5 keV by concentrated sewage sludge samples and therefore no self-absorption correction was applied to the data (Rose, 2003). Samples were counted for approximately one day, depending on activity. Due to the relatively short half-life of ^{131}I, activities were corrected to account for decay during data acquisition as described in Hoffman and Van Camerik (1967).

The counting efficiency of each detector at 364.5 keV in the 150 mL jar geometry was determined using a certified ^{131}I standard solution. Deionized water was spiked with a ^{131}I standard solution and counted three times on each detector. The mean counting efficiency of replicate analyses was used to calculate sample activities.

Multi-day continuum background counts were determined for each detector at 364.5 keV. Counting time for each sample and the mean background was used to determine limit of detection (L_D) as described by Currie (1968). The L_D for ^{131}I was ≤ 0.5 Bq kg^{-1} for seaweed. All activities are reported for time of collection ± 1σ counting error. Specific activities for solids are reported in units of dry mass.

Results and Discussion

The concentration of ^{131}I measured in *Fucus* sp. collected from site 1 ranged from 2.5 ± 8 to 516 ± 5 Bq kg^{-1}. Sample information and γ-ray spectrometry data are reported in Table H.1.

In this study, it was determined that the concentrations of ^{131}I in the sewage effluent discharged from the SBWPCP are a function of thyroid cancer inpatient treatments at the Stony Brook University Medical Center (SBUMC). Due to the frequency treatments and retention in the plant, sewage effluent discharges of ^{131}I are fairly continuous (Chapter 2).

The ^{131}I concentration in the seaweed collected on August 18, 2006 (253 ± 8 Bq kg^{-1}) is similar to the values reported previously and described above (224 ± 2, 254 ± 2 and 260 ± 5 Bq kg^{-1}). While the ^{131}I concentrations measured in sewage effluent discharged from the SBWPCP may vary by two orders of magnitude (1.8 ± 0.3 to 227 ± 2 Bq L^{-1}), the seaweed values suggest steady state conditions on monthly time scales.

The difference in concentrations of ^{131}I measured in *Fucus* sp. collected on March 10 (2.5 ± 0.8 Bq kg^{-1}) and March 16 (516 ± 8 Bq kg^{-1}) can be explained by a month-long break between inpatient treatments at the SBUMC. In general, several patients are treated at SBUMC per month (Chapter 2, Table 2.5). However, between February 8 and March 8 there were no inpatients treated. This time was sufficient to allow concentrations of ^{131}I in sewage effluent from the SBWPCP to fall below detection limits (see Chapter 2, Table 2.3). Four inpatients were treated between March 9 and 14 which resulted in more than a 200-fold increase in ^{131}I in the seaweed.

Additional measurements following another break in patient treatments at the SBUMC and its subsequent discharge of ^{131}I from the SBWPCP could help to further understand iodine uptake in seaweeds. Time series measurements of ^{131}I concentrations in the sewage effluent, the water column and in *Fucus* sp. in Port Jefferson Harbor may provide insight into the kinetics of its uptake by the macroalgae. Speciation of ^{131}I may play an important role in its uptake and concentration by the macroalgae. Future work could include ^{131}I speciation measurements in the water column and sewage effluent. Additionally, a concentration factor for ^{131}I in *Fucus* sp. in Port Jefferson Harbor can be derived from total ^{131}I measurements in the water and seaweed.

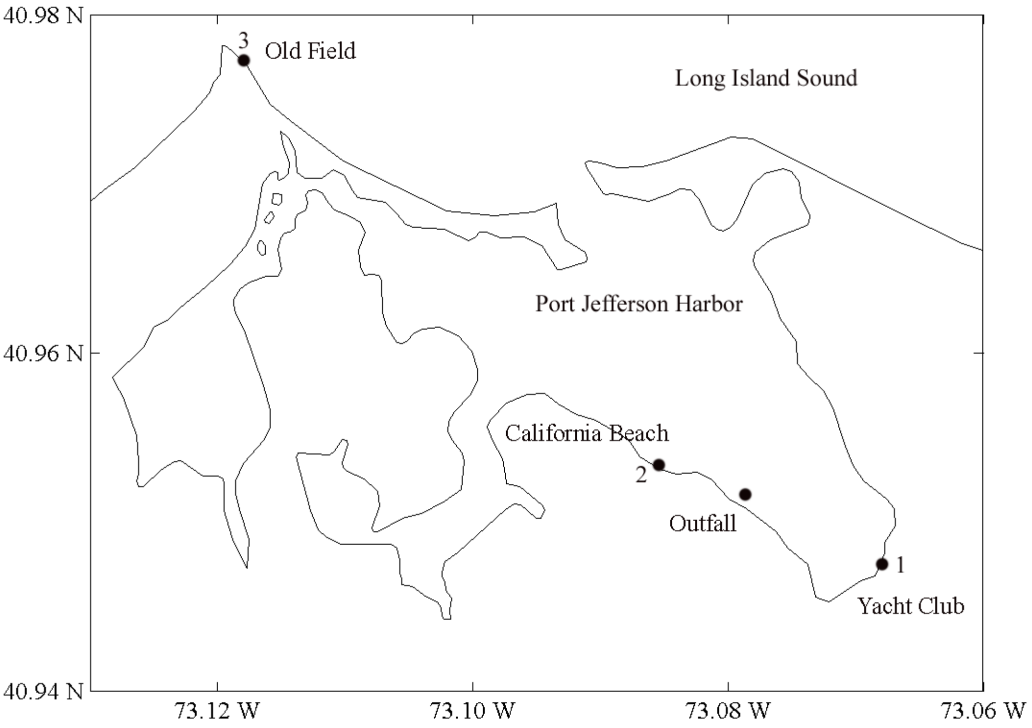

Figure H.1. Location of shared outfall of the Stony Brook and Port Jefferson WPCPs in Port Jefferson Harbor, NY. *Fucus* sp. collection sites 1 through 3 from previous work as described in Rose (2003). Samples were collected from site 1 only, in this work.

Sample #	Date & Time Collected	Counting Start Date & Time	Peak Area	Counting Error (%)	Peak Integral	μ_B	L_D	Counting Time (s)	Sample Mass (g)	^{131}I (Bq kg^{-1})
PJ081606F	8/16/06 12:00 PM	8/18/06 2:52 PM	1304	3.17	1540	190	48	54412.046	17.971	253 ± 8
PJ031007F	3/10/07 10:55 AM	3/10/07 12:59 PM	65	32.74	342	275	57	78456.146	14.366	2.5 ± 0.8
PJ031607F	3/16/07 3:20 PM	3/16/07 5:07 PM	11975	0.99	12974	839	98	239609.115	16.862	516 ± 5

Table H.1. Sample information, γ-ray spectrometry data (364.5 keV peak) and ^{131}I concentrations ± 1σ counting error in *Fucus* sp. collected from site 1 in Port Jefferson Harbor, NY. Background values (μ_B) and detection limits (L_D) were determined according to Currie (1968) as described in Chapter 2.