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Medically-derived ^{131}I in municipal sewage effluent

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ABSTRACT

This work presents ^{131}I ($t_{½}$ = 8.04 d) concentrations in sewage effluent from the Stony Brook Water Pollution Control Plant (WPCP), a small plant serving a regional thyroid cancer treatment facility in Stony Brook, NY, USA. The concentrations detected in sewage effluent ranged from 1.8 ± 0.3 to 227 ± 2 Bq L^{-1}. The primary source of ^{131}I is excreta from thyroid cancer inpatients treated at the Stony Brook University Medical Center. Based on several time series measurements following known inpatient treatments, the mean sewage half-life (T_s) of iodine is 3 d in this plant. The T_s, analogous to a radioactive half-life, describes the time it takes for half of a wastewater component to be removed from a WPCP. Flow recycling, or activated sludge, used to maintain bacterial populations necessary for sewage treatment causes iodine to remain in this plant far longer than its hydraulic retention time. The experimental results suggest that most ^{131}I entering the Stony Brook WPCP leaves in sewage effluent, not in sewage sludge. Patient treatments can result in continuous discharges of ^{131}I to surface waters where it can be used as a tracer of sewage-derived material and to understand the behavior of ^{131}I in aquatic environments.

1. 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, primarily to treat hyperthyroidism and thyroid cancer. In developed countries, the average number of hyperthyroidism treatments is 150 per million people and 38 thyroid cancer treatments per million people; however, the latter represents a greater potential source to water pollution control plants (WPCPs). 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 4000–8000 MBq compared to 100–1000 MBq of ^{131}I for hyperthyroid treatments and most of the initial dose is eliminated from the body in urine (ICRP, 2004; UNSCEAR, 2000). In the United States, patient excreta are exempt from sewer discharge regulations and are therefore released directly into sewer systems (Martin and Fenner, 1997).

Iodine-131 has been measured in aquatic environments receiving sewage effluent discharges, yet few published data exist for the radioisotope in sewage effluent (Chang et al., 2011; Erlandsson et al., 1989; Fischer et al., 2009; Kleinschmidt, 2009; Puhakainen, 1998; Rose, 2003; Smith et al., 2008; Sodd et al., 1975); most work has focused on sewage sludge. 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, 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 medically-derived

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^{131}I is readily measurable in the environment (Fischer et al., 2009; Howe and Hunt, 1984; Howe and Lloyd, 1986; 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 (Ullman and Aller, 1980, 1983, 1985; Upstill-Goddard and Elderfield, 1988). The nutrient-like behavior of iodine suggests that medically-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.

2. Methods

2.1. Study site

The Stony Brook WPCP, 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 Stony Brook University Medical Center 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 4–6 h. The oxidation ditch no longer receives mixed liquor returns 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 (Rose, 2011).

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 (Rose, 2011).

The hydraulic retention time of sewage in the Stony Brook WPCP is approximately 24–36 h, depending on the University schedule as mentioned above. The mean cell residence time, or the residence time of organic matter in the system, is about 36 d (Rose, 2011).

2.2. 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 high density polyethylene bottle. More than half of the samples were analyzed with no further treatment. The remaining samples were vacuum filtered through a 47 mm 0.7 μm glass fiber filter. Aliquots of the filtrate were retained and analyzed with no further treatment. Sample volumes analyzed were 150 mL and 170 mL. Straight-side polypropylene jars (64 mm height; 64 mm diameter) were used for counting. Pre-weighed filters 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 Stony Brook WPCP on six additional days.

2.3. 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. 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 (Eckert & Ziegler Isotope Products, Valencia, CA). 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 (3 replicates per detector). The ^{131}I standard solution was applied to three filters with a pipette, using enough liquid to wet the filter entirely. Each filter was counted three times on each detector to determine the counting efficiency of the suspended solids samples (9 replicates per detector). In each case, the mean counting efficiency was used to calculate sample activities.

Multi-day continuum background counts were determined for each detector with several ^{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 $\leq$1.7 Bq L^{-1} for sewage effluent samples and $\leq$50 Bq g^{-1} for suspended solids.

3. Results

3.1. Unfiltered effluent

Iodine-131 concentrations detected in unfiltered sewage effluent collected on 77 different days between June 2006 and March 2009