

EPA/600/R-21/006 | March 2021 www.epa.gov/emergency-response-research

Persistence of Surrogate Radionuclides on Wastewater Collection System Infrastructure

Office of Research and Development Homeland Security Research Program

Persistence of Surrogate Radionuclides on Wastewater Collection System Infrastructure

by

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Contract EP-C-14-012, Task Order 4-06

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The U.S. Environmental Protection Agency (EPA) through its Office of Research and Development funded and managed the research described herein under Contract EP-C-14-012, Work Assignment 4-06 with Aptim Federal Services, LLC. It has been subjected to the Agency's review and has been approved for publication. Note that approval does not signify that the contents necessarily reflect the views of the Agency. Any mention of trade names, products, or services does not imply an endorsement by the U.S. Government or EPA. The EPA does not endorse any commercial products, services, or enterprises.

Foreword

The U.S. Environmental Protection Agency (EPA) is charged by Congress with protecting the Nation's land, air, and water resources. Under a mandate of national environmental laws, the Agency strives to formulate and implement actions leading to a compatible balance between human activities and the ability of natural systems to support and nurture life. To meet this mandate, EPA's research program is providing data and technical support for solving environmental problems today and building a science knowledge base necessary to manage our ecological resources wisely, understand how pollutants affect our health, and prevent or reduce environmental risks in the future.

The Center for Environmental Solutions and Emergency Response (CESER) within the Office of Research and Development (ORD) conducts applied, stakeholder-driven research and provides responsive technical support to help solve the Nation's environmental challenges. The Center's research focuses on innovative approaches to address environmental challenges associated with the built environment. We develop technologies and decision-support tools to help safeguard public water systems and groundwater, guide sustainable materials management, remediate sites from traditional contamination sources and emerging environmental stressors, and address potential threats from terrorism and natural disasters. CESER collaborates with both public and private sector partners to foster technologies that improve the effectiveness and reduce the cost of compliance, while anticipating emerging problems. We provide technical support to EPA regions and programs, states, tribal nations, and federal partners, and serve as the interagency liaison for EPA in homeland security research and technology. The Center is a leader in providing scientific solutions to protect human health and the environment.

If a wide area or water system contamination event involving radionuclides occurs, large volumes of contaminated water could be generated during clean-up activities, or during rain events. Should this water enter a sewer system, radiological contamination could persist on collection system infrastructure, or travel with the sewage to the wastewater treatment plant. The extent of radionuclide persistence on sewer infrastructure materials is currently unclear. This report contains data on the persistence of non-radioactive cesium, cobalt and strontium on sewer system infrastructure. The report is the first step in collecting a data set that decision makers could use to determine response actions should radiological contamination enter a sewer system.

Gregory Sayles, Director

Center for Environmental Solutions and Emergency Response

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Executive Summary

Contamination from a radiological dispersal device, improvised nuclear device or nuclear power plan accident could be widespread. Cleanup activities or precipitation events could result in radioactive contamination entering a wastewater or stormwater collection system. In this study, collection system infrastructure materials such as brick, clay, concrete, high density polyethylene, iron, polyvinyl chloride (PVC), and rubber were conditioned in wastewater flowing through six 6-inch (15.2 cm) diameter PVC pipes for two months. Conditioning allowed biofilms to form and wastewater solids to accumulate on the coupons surface. Subsequently, non-radioactive cesium chloride, cobalt chloride and strontium chloride were injected into the wastewater flow, and persistence on the infrastructure coupons (excised sample materials) was determined over time. Flow in each channel was approximately 50 gallons per minute (189.2 liters per minute), and each non-radioactive surrogates for radionuclides would adhere to or persist on common collection system infrastructure materials.

Results for cesium and cobalt showed that metal adhesion to conditioned infrastructure materials was undetectable. Strontium was detected on concrete coupons at levels above the method detection limit. However, further analyses of the concrete suggested that strontium in the concrete matrix was being detected, not strontium from the contaminant injection. Although the data suggests that none of the surrogate radionuclides adhered to or persisted on collection system infrastructure materials, research with real radionuclides at lower concentration levels could confirm these results.

Abbreviations and Acronyms

BOD	biochemical oxygen demand
CESER	Center for Environmental Solutions and Emergency Response
cm ²	square centimeter
Со	cobalt
Cs	cesium
EPA	United States Environmental Protection Agency
FEMA	Federal Emergency Management Agency
gpm	gallon(s) per minute
HDPE	high density polyethylene
HSPD	Homeland Security Presidential Directive
IAEA	International Atomic Energy Agency
IND	improvised nuclear device
L	liter(s)
m	meter(s)
MDL	method detection limit
mg	milligram(s)
mgd	million gallons per day
min	minute(s)
μS	micro Siemens
mL	milliliter(s)
MPN	most probable number
MSDGC	Metropolitan Sewer District of Greater Cincinnati
NA	not applicable
NH3	ammonia
NIST	National Institute of Standards and Technology
PVC	polyvinyl chloride
QA	quality assurance
QAPP	quality assurance project plan
QC	quality control
RDD	radiological dispersal device
RPD	relative percent difference
SETBC	secondary effluent test bed channels
Sr	strontium
T&E	Test and Evaluation
TSS	total suspended solids
TV	true value

1.0 Introduction

During the last two decades, there has been an increased focus on the potential impacts of radiological materials that could be used in intentional attacks or resulting from accidental contamination in the United States. For example, the Federal Emergency Management Agency (FEMA) uses 15 all-hazards National Planning Scenarios as a basis for assessing national preparedness and implementing Homeland Security Presidential Directive 8: National Preparedness (HSPD-8) (FEMA 2003). The National Planning Scenarios encompass cyberthreats, natural disasters, pandemics and attacks with chemical, biological or radiological materials. The radiological scenarios focus on radiological dispersal devices (RDD) and on improvised nuclear devices (IND). In the case of an RDD, radiological material is dispersed with conventional explosives, while an IND is the detonation of a nuclear device. In both cases, the impacts of these devices could be significant if they were detonated in an urban area (USEPA 2007)

Significant research has been undertaken to understand the impacts of RDD or IND detonations in urban environments (Biancotto et al. 2020, Lee et al. 2010, Regens et al. 2007, USEPA 2007). One often overlooked aspect of this research is understanding how radionuclides could interact with sewer collection system infrastructure. Radionuclides could enter a sewer system due to remediation activities such as washing down contaminated road and building surfaces. Water runoff from cleanup activities should be collected and then treated or disposed of, but accidental releases of contained water are possible. Furthermore, if a rain event were to occur after an RDD or IND event, it might be difficult to prevent all runoff from entering a sanitary or stormwater sewer system. Furthermore, should a nuclear power plant accident occur, such as the 2011 incident in Fukushima, Japan, radionuclides could be spread over a wide area. Collection systems would be vulnerable to radiological nuclear power plant contamination from remediation activities or rain events. Should radionuclides enter a sewer system, persistence on collection system infrastructure or contamination at the treatment plant could pose a long term health risk for workers, and potentially disrupt the wastewater treatment process.

Research has been conducted to determine the fate of biological agents on water infrastructure surfaces, including drinking water pipes (LeChevallier et al. 1988, De Beer et al. 1994, Chu et al. 2003, Emtiazi et al. 2004, Szabo et al. 2007, Miller et al. 2015) and wastewater collection infrastructure (USEPA 2017). The persistence of radionuclides has also been examined on drinking water pipe surfaces (Szabo et al. 2009, USEPA 2016, USEPA 2018). The purpose of this study was to fill a research gap and to produce data on the persistence of water-soluble radionuclide surrogates on collection system infrastructure. Small sections of collection system infrastructure material (brick, clay, concrete, HDPE, iron, PVC, and rubber) were conditioned over two months in flowing wastewater, which allowed for formation of wastewater biofilms and accumulation of solids from the wastewater flow. The coupons were then exposed to non-radioactive salts of cesium (Cs), cobalt (Co) and strontium (Sr). These compounds are common homeland security radionuclides of concern (USEPA 2007). Subsequently, persistence of these radionuclides on the infrastructure coupons (excised sample materials) over time was determined.

2.0 Materials and Methods

2.1 Description of the pilot scale wastewater collection system setup

Experiments were conducted at the United States Environmental Protection Agency's (EPA's) Test and Evaluation (T&E) Facility in Cincinnati, Ohio. The pilot scale system used was the secondary effluent test bed channels (SETBC) (Figure 1). The SETBC consists of six 6-inch (15.2 cm) diameter PVC pipes arranged horizontally with the upper section of the pipe removed, which allowed access to the open channel flow in the pipes. Unchlorinated secondary treated effluent was pumped from the adjacent Metropolitan Sewer District of Greater Cincinnati's (MSDGC) 100 million gallons per day (mgd) (378.5 liters per day) Mill Creek wastewater treatment plant and into a manifold where flow was distributed to each pipe. Before reaching the SETBC, untreated wastewater had undergone primary settling, treatment with activated sludge, and secondary settling. The secondary treated effluent was diverted to the SETBC before chlorination.

Although secondary treated effluent is lower in solids and biological activity than raw sewage, it is more consistent in quality and more amenable to pumping through the SETBC channels. Flow rates in each of the six SETBC pipes was adjusted to approximately 50 gallons per minute (gpm) (189.2 liters per minute). Each of the six PVC pipes has its own flow control valve, two sections of fabricated horizontal open grids to mount test material coupons, an injection port, and a flow monitoring sensor and a data logger. In addition, the system also consists of two sets of pH, conductivity, and temperature monitoring sensors. One-inch (2.54 cm) diameter coupons were cut from seven unused collection system infrastructure materials (brick, clay, concrete, HDPE, iron, PVC, and rubber) were secured to metal bars which spanned the section cut from the top of pipe (Figure 2). Using this setup, the coupons were mounted to the horizontal grids and inserted into the unchlorinated secondary effluent flow.



Figure 1: Schematic of the secondary effluent test bed channels (SETBC) (left) including the overall orientation of the pipes, source of secondary effluent, injection ports and location of flow sensor. The picture (left) shows two of the pipes and their layout.



Figure 2: Coupons of collection system infrastructure. From top to bottom: high-density polyethylene, brick, rubber, concrete, iron (uncorroded), clay, PVC.

2.2 Conditioning of infrastructure coupons

Collection system infrastructure coupons were conditioned using unchlorinated secondary effluent pumped from an adjacent 100 million gallons per day (mgd) wastewater treatment plant. In order to disinfect the SETBC between experiments, the system was drained and sprayed with undiluted bleach between the tests and then flushed with unchlorinated secondary effluent. Similarly, coupons were wiped with undiluted bleach prior to conditioning. Coupons were randomly placed amongst the six channels, but each material was represented in equal numbers in each channel. Each coupon was mounted so that the face of the coupon was parallel to the direction of the water flow. Coupons were conditioned in unchlorinated secondary treated effluent for two months with flow at approximately 50 gpm in each channel.

2.3 Contamination and sampling of SETBC

After conditioning, the second phase of the experiment focused on the adhesion of Cs, Co and Sr to the conditioned infrastructure material coupons. Contamination experiments with Cs, Co and Sr were conducted separately, and each experiment was conducted in duplicate for a total of six injection experiments. Before contamination, two coupons (conditioned uncontaminated controls) of each material were removed from the SETBC and sampled for the metal being used in each test. Injection solutions (1000 mg/L) of cesium, cobalt, and strontium were prepared separately by dissolving chloride salts of Cs, Co, and Sr in deionized water. Metal salt solutions were injected through the injection ports in each pipe (Figure 1) for one minute at 1,000 mL/min using a pre-calibrated peristaltic pump into each pipe to achieve a target metal concentration of 5 mg/L in the flow. Coupons of each material were harvested in pairs after injection in order to determine persistence. Sampling timeframes varied and are further discussed in the results.

2.4 Determination of metals adhered to coupon surfaces

After the coupons were removed from the SETBC, their surfaces were sampled to determine the concentration of adhered Cs, Co and Sr. Each coupon was washed with 5 mL of concentrated trace metal grade nitric acid. Each acid extract was diluted in deionized water if needed, filtered through a 0.45 μ m filter and stored 4.0 \pm 0.5 °C prior to analysis. Samples were analyzed separately to determine the metal concentration either by a 240Z Graphite Furnace Atomic Absorption (Agilent, Santa Clara, CA) for cesium or an Optima 2100 DV Inductively Coupled Plasma-Optical Emission Spectrometer (PerkinElmer, Waltham, MA) for cobalt and strontium using EPA Methods 200.9 (USEPA 1994) and 6010B (USEPA 1996), respectively.

Method detection limits (MDL) for Co and Sr were 100 μ g/L and 5 μ g/L in the acidified extract solution, respectively. The MDL for Cs using atomic adsorption was not determined, and the lowest reportable value was 20 μ g/L, which was the lowest point on the calibration curve. Data reported here is in the form of the mass of surrogate radionuclide recovered from the surface of the coupon (μ g/cm²). Coupon surface area exposed to water flow was 10.7 cm² (1.7 in²), which includes the front and back of the coupon. Once the metal concentration in solution was known, the concentration was multiplied by the solution volume so the total mass was known. The total mass was then divided by 10.7 cm² to get the amount of adhered metal recovered from the coupon. This resulted in the lowest reportable values of 9.1 μ g/cm², 45.6 μ g/cm², and 2.3 μ g/cm² for Cs, Co and Sr, respectively.

2.5 Other unchlorinated secondary treated effluent parameters

Coliform/*Escherichia coli* levels were measured via Colilert-18 Quanti-tray 2000 (Idexx Laboratories, Westbrook, ME). Briefly, unchlorinated secondary treated effluent samples were serially diluted in pH 7.2 Butterfields phosphate buffer (Fisher Scientific, Waltham, MA), plated and incubated according to the manufacturer's instructions (35° C). The pH, temperature and specific conductance were measured by online sensors. Conductivity, temperature and pH were measured with a GF Signet 2820 series sensor (George Fisher/GF Piping Systems, Schaffhausen, Switzerland). Water quality parameters were measured continuously over the course of approximately one year, which was the time frame in which the experiments took place. Sensors were maintained and calibrated according to the manufacturer's instructions. Data was reported at 2-minute intervals. MSDGC provided weekly data on 5-day biochemical oxygen demand

(BOD), total suspended solids (TSS) and ammonia (NH₃). BOD, TSS, and NH₃ were analyzed according to methods 5210, 2540, and 4500 in Standard Methods for the Examination of Water and Wastewater (Baird et al. 2017). The weekly MSDGC data spanned a one year period when the experiments were taking place.

2.6 Quality control and data quality

Quality control (QC) samples for the contaminant reference method included continuing duplicate samples, controls and laboratory blanks. The data quality objectives for each of these quality control samples are provided in Table 1. The acceptable ranges limit the error introduced into the experimental work. All analytical methods operated within the QC requirements for controls and laboratory blanks, and unless otherwise noted in the Deviations (Section 2.6.2), all data quality objectives in Table 1 were met.

Measurement	Matrix	QA/QC Check	Frequency	Acceptance Criteria	Corrective Action	
Temperature	Water	NIST certificate expiration date	Before each use	Within expiration data	Replace/recalibrate thermometer	
BOD	Water	Calibration Check	Every batch	Within 30% true value	Rerun samples	
		Blank	Every batch	Sample is compared against blank	NA	
		Duplicate	Every 10 samples (bench)	± 30% RPD	Discard data point, repeat experiment if insufficient data points	
pН	Water	Initial calibration	Before each use	±0.1 pH units	Check standard buffers for	
		Calibration check	Every 10 samples	±0.1 pH units	contamination, check electrode for electrolyte, replace probe if required	
Ammonia	Water	Calibration Check	Every batch	Within 30% true value	Rerun samples	
		Blank	Every batch	Sample is compared against blank	NA	
		Duplicate	Every 10 samples (bench)	± 30% RPD	Discard data point, repeat experiment if insufficient data points	

Table 1: Quality Control Data Objectives

				Acceptance	
Measurement Matrix		QA/QC Check	Frequency	Criteria	Corrective Action
TSS	Water	Calibration Check	Prior to analysis, every 10 samples, and at the end of the batch	±10% TV	Recalibrate and/or reanalyze affected samples.
		Duplicates	Once per batch or every 10 samples	RPD<20%	Repeat analysis on the same sample; if sample volume does not allow, choose another sample and document accordingly
E. coli	Sterile buffer / water	Positive control using stock	Once per batch	± 10 fold of the stock	Investigate laboratory technique. Re-analyze the stock and change if necessary
E. coli	Sterile buffer / water	Negative control using sterile buffer / sterile water	Once per batch	0 MPN / plate	Investigate laboratory technique. Re-analyze the sterile buffer / sterile water and change if necessary
		Equipment Calibration	Initially	$R^2 \ge 0.995$	Re-calibrate
	10% nitric water	Initial calibration verification	Once immediately after calibration	±10 % of the actual concentration	Re-calibrate
Metals (Cs, Co,		Laboratory check blanks	One per batch	≤ instrument detection limit	Re-run blank
Sr)		Matric Spike	One per batch of 20 Samples	± 20 % recovery	Re-run spike, Re-prepare Spike
		Lab Duplicate	One per batch of 20 samples	$\leq 10\%$ RPD	Re-run duplicate Re prepare duplicate
		Continuing Calibration Verification	Every 10 sample	± 15 % recovery	Re-prepare QCs Re run affected samples

BOD, biochemical oxygen demand; MPN, Most Probable Number; NA, Not Applicable; NIST, National Institute of Standards and Technology; QA, quality assurance; QC, quality control; RPD, relative percent difference; TSS, total suspended solids; TV, true value.

2.6.1 Data quality

At least 10% of the data acquired during the evaluation were audited. The data were traced from the initial acquisition, through analysis, to final reporting, to ensure the integrity of the reported results. All calculations performed on the data undergoing the audit were checked. No significant adverse findings were noted in this audit.

2.6.2 Deviations

Coliforms/E. coli measurements were not in the original quality assurance project plan (QAPP) developed for this study. The QC criteria for coliforms/*E.coli* measurement is described in Table 1 and the methodology in section 2.5.

3.0 Results and Discussion

3.1 Unchlorinated secondary effluent water quality

Unchlorinated secondary effluent temperature varied from 13-27 °C depending on the season in which the experiment took place, but averaged 22 °C. The pH varied from 6.7 to 7.4, with an average value of 7.0. Conductivity ranged from 760 to 2000 μ S, with an average value of 1,500 μ S across all experiments. Coliform and *E. coli* levels were measured on four occasions and ranged from 20,000 to 150,000 most probable number (MPN)/100 ml and 1,300 to 7,500 MPN/100 mL, respectively. MSDGC provided data on BOD, TSS, and NH₃, which is summarized in Table 2.

	BOD	TSS	NH₃
	(mg/L)	(mg/L)	(mg/L)
Average	3.35	6.02	0.75
Standard Deviation	1.78	2.52	0.67
Maximum			
Observed	10.0	14.0	2.86
Minimum Observed	2.00	2.50	0.10
Number of Samples	48	61	31

Table 2: Unchlorinated Secondary Treated Effluent 5-day, BOD, TSS and NH₃ Data

BOD, biochemical oxygen demand; TSS, total suspended solids

3.2 Evaluation of cesium and cobalt adhesion to infrastructure materials

Contamination experiments with cesium and cobalt were conducted in duplicate. Contamination injections were conducted the same across experiments with a one-minute injection that achieved 5 mg/L of the metal in the flow. For cesium, sampling of coupons occurred for 28 days after injection in the first test. Across all infrastructure materials, cesium was not detected in the coupon extract over the course of the 28 days of sampling. Because of this result, the duplicate test was conducted for 5 days. No cesium was detected in the coupon extract during the 5-day sampling period. Since no cesium was detected on the any infrastructure material, experiments with cobalt were limited to 5 days of post-injection sampling. Like cesium, no cobalt was detected in any coupon extract across all infrastructure materials.

3.3 Evaluation of strontium adhesion to infrastructure materials

Like cesium and cobalt, strontium tests were conducted in duplicate. The first strontium test (Test 1) was conducted for 28 days (Figure 3, top). In the first test, Sr was detected above the MDL during one sampling event on concrete coupons (day 21). On concrete, the Sr concentration varied from 0.45 to 2.32 μ g/cm², with the 2.32 μ g/cm² data point being the only one above the MDL. The second strontium sampling experiment (Test 2) was conducted for 5 days (Figure 3, bottom). Similar to the first test, strontium was detected in all coupon types, but only samples from concrete coupons were above the MDL. These concentrations ranged from 3.7 to 6.3 μ g/cm² (Figure 3). Note that concentration values were determined for sample points below the MDL and are shown in Figure 3, but it cannot be stated that these values are greater than zero with 99% confidence.



Figure 3: Strontium recovered from collection system infrastructure materials. One experiment lasted for 28 days (top) and one lasted 5 days (bottom).

Table 3 summarizes two sets of uncontaminated conditioned control data. Data points in bold are above the MDL. The sections labeled uncontaminated conditioned control show the coupons of each material that were conditioned in secondary effluent but were extracted and analyzed at time point zero before strontium was introduced into the test bed channels. The rows labeled "Test 1" and "Test 2" show the mean of duplicate time zero samples harvested during the respective test along with the range between them. The rows labeled "All Tests" average the duplicate time zero controls across all four experiments and show the standard deviation of those samples.

	Material	Brick	Clay	Concrete	HDPE	Iron	PVC	Rubber	
Uncontaminated conditioned control (μg/cm ²), n=2									
Test 1	Mean	0.26	0.18	1.03	0.37	0.33	0.21	0.18	
(28 day)	Range	0.14	0.02	0.30	0.39	0.02	0.00	0.01	
Test 2	Mean	0.10	0.04	5.88	0.22	0.16	0.14	0.11	
(5 day)	Range	0.13	0.00	1.90	0.20	0.00	0.07	0.03	
	Unc	ontaminat	ed conditio	oned contro	l (μg/cm²),	n=4			
	Mean	0.18	0.11	3.45	0.30	0.24	0.18	0.15	
All Tests	Standard								
	Deviation	0.12	0.08	2.91	0.20	0.09	0.05	0.04	
Uncontaminated coupons soaked in acid (μ g/cm2), n=3									
	Mean	4.65	6.43	97.02	2.17	10.54	21.16	3.40	
	Standard								
	Deviation	0.30	0.27	3.08	0.01	0.33	1.19	0.05	

 Table 3: Strontium Extracted from Uncontaminated Conditioned Control Coupons (Top Rows) and Those Soaked in Acid to Extract More Cesium (Bottom Rows)

HDPE, high density polyethylene. Note: Values in bold are above the MDL. Values in normal font are below the MDL

In Test 1, all control samples are below the MDL. However, during the 28 day collection period, three concrete samples were above the range of the control (days 3, 4 and 21) and three were below (days 7, 10 and 28). In Test 2, only strontium extracted from concrete was above the MDL. In this case, all samples harvested after introduction of strontium into the test bed channels fall within the range of the control, except for the last sample on day 5 that is below (Figure 3). All concrete samples analyzed for strontium during both duplicate tests (four total) fall within the range of the standard deviation of the "All Test" pre-injection conditioned control samples, except the sample from Test 1, day 10, which was less.

Table 3 also includes a section titled "uncontaminated coupons soaked in acid". Data for this section was collected by either grinding or shredding one gram of the clean (not conditioned in secondary effluent), uncontaminated collection system material into the smallest pieces possible. These pieces were then microwave digested using EPA method 3051A (USEPA 2007a) using a Mars Xpress microwave digester (CEM Corporation, Matthew, NC), and the liquid extract analyzed for strontium. This process was performed in triplicate. For each material, more strontium was extracted compared to the rinsing process used during contamination experiments. However, the rinsing method was designed to remove adhered strontium, not strontium in the material itself. The most strontium was associated with concrete, which suggests that there is strontium in the cement or mortar material that make up the concrete coupon that could leach into the rinse solution. Note that data from this section of Table 3 was first normalized by the mass of material samples, but then extrapolated to the area of the coupon for comparability to the control samples.

Taken together, the data suggests that strontium detected on the concrete coupons came from the concrete itself, and was not due to adhesion of the strontium to the coupons. It cannot be ruled out that some strontium adhered to the coupons, but any amount was small enough that it could not be distinguished from the inherent strontium extracted from the coupon itself. It should be noted that all concrete coupons were cut from concrete sewer pipe, but samples used in Test 1 and Test 2 came from two different concrete pipes. Assuming strontium detected during sampling came from the pipe itself, it is possible that the different pipes contained a different mix of cement and mortar, which affected the inherent strontium levels. This would explain the overall difference in the amount of strontium detected in the coupon between tests. However, it should be noted that most strontium detected in Test 1 was below the MDL.

3.4 Discussion, implications and future research

The data presented in the previous sections indicate that cesium and cobalt were not detectable on the collection system infrastructure coupons used in this study. In particular, no results were recorded for cesium on the coupons, even below the method detection limit. Similar results were found with strontium, but results were often recorded even though they were below the detection limit. However, data from the pre-contamination control samples and the ground coupon materials digested in nitric acid suggest that the strontium found on the coupons was not from the injected strontium, but from the material itself. It is possible that some cesium, cobalt or strontium was present on the coupons, but the instrumentation used in this study was not sensitive enough to detect them. It should also be noted that the data in this study is applicable to straight runs of pipe. The coupons used in this study are not designed to replicate pipe bends or elbows.

Two similar studies have been carried out on the bench scale using iron, concrete, copper and PVC surfaces in chlorinated drinking water and non-radioactive salts of cesium, strontium and cobalt (Szabo et al. 2009, USEPA 2016) In both studies, cesium did not persist on any material, or was undetectable. Strontium and cobalt were found to persist on concrete surfaces. Cobalt was found to persist on iron (Szabo et al. 2009) and concrete (USEPA 2016), but this was due to cobalt forming an insoluble precipitate when in contact with chlorinated tap water, which deposited on the coupon surfaces. Strontium was found to be persistent on concrete in USEPA, 2016. However, in that study, strontium was deposited directly onto the coupon surface and allowed to soak in in the absence of flow, which likely lead to greater persistence. In the study reported here, wastewater flow was continuously present in the pilot scale SETBC, which is closer to resembling flow in a real wastewater system and makes adhesion more difficult. Finally, drinking water is a different matrix than wastewater, which may account for differences in the results.

When considering the implications of the results presented above, the typical uses of the radioactive forms of the metals used this study should be considered. Radioactive cobalt-60 is commonly used in solid metallic form in instruments ranging from medical devices (e.g., blood/tissue irradiators, brachytherapy, teletherapy) to level gauges and food sterilization devices. Strontium-90 is used in medical devices, thickness gauges, level measurements, and automatic control processes. However, one of its most common historical uses was in radioisotope thermal electric generators, which are used to generate electricity and can contain tens of thousands of curies of activity. However, strontium is often used in devices as strontium

titanate, in a ceramic form, or as a metal foil (EPA 2007, IAEA 2000).

Should cobalt-60 or strontium-90 be used in an RDD, it would likely be in a form that has low solubility in water. If pieces of radioactive cobalt or strontium were dispersed and then washed into a collection system, it would likely be in insoluble pieces that may settle out or be carried in the wastewater flow (USEPA 2007). Any dissolved fraction would be small, although it is possible to solubilize cobalt and strontium into a soluble salt. The insolubility of radioactive cobalt and strontium source material coupled with the fact that neither was detected on collection system infrastructure surfaces in this study when in their soluble form indicate that it is unlikely that either would persist on collection system infrastructure.

Unlike radioactive colbalt and strontium, cesium-137 is commonly used as a soluble chloride salt in medical devices, sterilizers and industrial gauges (IAEA 2000). Should it be used in a RDD, the soluble salt would be dispersed, and could dissolve in wastewater or stormwater flowing through a collection system (USEPA 2007). However, it was not detected on any of the infrastructure surfaces when introduced into flowing wastewater at 5 mg/L. For cesium chloride, 5 mg/L translates to activity in the water of 0.44 Ci/L (4.4×10^{11} pCi/L), with the equivalent of 1 Ci injected over the course of 1 minute using the injection conditions in this study. The isotope and amount of activity in a RDD are impossible to know and could take many different forms. However, past scoping studies have shown that the metal concentrations used in this study are feasible (USEPA 2007).

Finally, as noted earlier, it is possible that some contaminant adhered to the coupons, but, in this study, the analytical instrumentation was not sensitive enough or else the extraction methods was not efficient enough to detect it. This could be resolved by introducing the actual radionuclide into the SETBC and measuring residual activity on the surface of the coupon. One recent study did use cesium-137 as the contaminant in a bench scale drinking water system with concrete, copper and PVC surfaces (USEPA 2018). Cesium-137 was introduced at 10 μ Ci/L (concrete) and 100 μ Ci/L (PVC and copper). This translates to 1.14×10^{-7} mg/L (10 μ Ci/L) and 1.14×10^{-6} mg/L (100 μ Ci/L), which is less than the levels used in this study. Activity on the coupons was measured using a sodium iodide spectroscopy system and a liquid scintillation counter. The results showed that after 24 hours of stagnant exposure to dissolved cesium, there was transient adhesion of cesium to PVC and concrete, but 91% and 93%, respectively, were removed after the coupons were flushed with clean water. Further flushing and use of decontamination agents removed over 99% of the adhered cesium.

A key difference between these studies was that 24 hours of stagnant contact between the cesium and infrastructure coupons was used in USEPA, 2018, whereas coupon to exposure cesium was only one minute with constant flow in the SETBC wastewater system. The fact that residual cesium activity was detected on the coupons was likely due to this extended initial contact time. However, even after flushing and decontamination with chemical cleaning agents, some residual activity was still detected on the coupons. In the future, it would be beneficial to repeat the experiments described in this report with radioactive cesium, cobalt, and/or strontium. Although large scale persistence is unlikely, research with real radionuclide would determine if a small fraction does adhere and persist on the coupons. Still, the results of this study suggest that any cesium, cobalt or strontium washed into a wastewater collection system would not persist on the infrastructure, with dissolved material traveling with the flow to a combined sewer outfall or wastewater treatment plant. Also, as noted above, not all radionuclides could enter a sewer system in soluble form. The fate of insoluble radioactive particles in sewer collection system is a topic that deserves further study.

4.0 Conclusions

The primary conclusions from this study are as follows:

- Cesium, cobalt and strontium salts injected into a pilot scale wastewater collection system using secondary treated effluent were not detectable on common collection system infrastructure materials. It should be noted that non-radioactive salts were used and were analyzed by atomic adsorption and inductively coupled plasma.
- Strontium was the only metal detected above the MDL. However, analyses of the strontium content of the control (pre-injection) coupons suggested that detectable strontium came from the coupon itself and did not adhere after injection of strontium into the wastewater flow.
- Past studies examining the persistence of radioactive cesium on infrastructure materials in a drinking water environment did find some residual activity after flushing the surfaces with clean water and other decontamination techniques. Repeating the experiments presented in this report with real radionuclides would help determine if a small fraction of cesium, strontium or cobalt do adhere to the infrastructure coupons, but could not be detected with the analytical techniques used in this study.

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