

# Decontamination of Drinking Water Infrastructure

## A Literature Review and Summary



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## Abbreviations and Acronyms

BAR	Biofilm Annual Reactor
CBR	Chemical, Biological, Radiological
CDC	Centers for Disease Control
CFU	Colony Forming Unit
cPVC	Chlorinated polyvinyl chloride
Ct	Concentration of Disinfectant Multiplied by Contact Time
CWA	Chemical Warfare Agent
DCB	Dichlorobenzene
DSS	Distribution System Simulator
EC	Environment Canada
EPA	United States Environmental Protection Agency
HFO	Hydrated Ferric Oxide
ICP-AES	Inductively Coupled Plasma-Atomic Emission Spectroscopy
ICP-MS	Inductively Coupled Plasma-Mass Spectrometer
ICP-OES	Inductively Coupled Plasma-Optical Emission Spectroscopy
$K_{ow}$	Octanol-Water Coefficient
NSF	National Sanitation Foundation
p-DCB	<i>p</i> -Dichlorobenzene
PVC	Polyvinyl Chloride
SFA	Sodium Fluoroacetate
TCR	Total Coliform Rule
XRD	X-Ray Diffraction

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## Executive Summary

Chemical, biological, or radiological (CBR) contamination events or attacks on drinking water infrastructure could have significant public health, economic, and social impacts. The intentional introduction of harmful contaminants into drinking water distribution systems, for example, has the potential to contaminate the water along with pipes and pumps used to convey the water, storage tanks, service connections to buildings, and water-consuming appliances, such as water heaters. Complicating the situation is the propensity of some contaminants to adhere to corroded pipes or biofilms on the pipe walls, potentially prolonging the impact of the contamination by desorption, leaching, or otherwise detaching from the surface and into the water over time after the incident. Contamination incidents could also impact drinking water treatment plants, wastewater treatment facilities, and storm and sewer systems.

Treatment and decontamination strategy involves determining what the contaminants are, the extent and location of contamination, whether to clean or replace contaminated infrastructure, techniques for remediating the contaminants and associated hazardous degradation by-products, how long it will take, how much it will cost, and how to verify that cleanup is completed. The United States Environmental Protection Agency (EPA) and the water sector have extensive experience in treatment and cleanup, but must prepare for significant new challenges posed by intentional attacks using potent and difficult to decontaminate non-traditional or unregulated contaminants.

Factors influencing the approach and effectiveness of treatment and decontamination are often complex and multifaceted. For example, for some contaminants that could be used in an attack, fate and transport in the water environment is not well understood. Contaminants may react with or adhere to various pipe materials differently under various water flow patterns, or they may produce degradation products that are as dangerous as or more dangerous than the original substance. There is a need for knowledge and research on new or enhanced treatment and decontamination technologies and protocols. There are many data gaps that must be filled through research.

This literature review summarizes available data on the propensity of select representative target CBR contaminants to adhere to wetted drinking water infrastructure surfaces, such as pipes, and techniques for decontamination should adherence occur. Persistence and decontamination data included in this report pertain to the most common types of water pipes used in North America, including cast/ductile iron, cementitious material like cement-mortar lined ductile iron, and plastics like PVC<sup>1,2</sup>. Each section of this report includes a discussion of the current literature regarding persistence and decontamination data for a range of CBR agents on drinking water infrastructure. This is followed by an analysis of decontamination data from the literature on drinking water infrastructure, or similar materials and environments if no directly applicable data was found. Conclusions about techniques or methodologies for drinking water infrastructure

1. *Buried No Longer: Confronting America's Water Infrastructure Challenge*, 2012: American Water Works Association, Denver, CO.

2. Folkman, S., et al., *Survey of water main failures in the United States and Canada*. Journal of the American Water Works Association, 2012. 104(10): p. 70-79.

decontamination that can be drawn from the literature review are presented. Finally,

suggestions for future research are discussed.

# Chemical Agents

## Introduction

Research on chemical persistence on drinking water infrastructure has often focused on the adherence of inorganic contaminants [1-4]. There are, however, many other chemical classes of concern, including organics, pharmaceuticals, chemical warfare agents, and toxins. Historically, research into drinking water treatment of chemicals has focused on regulated inorganic contaminants such as heavy metals, disinfection byproducts, and organics such as pesticides, herbicides, and chemical discharges from industrial processes. Many of these chemical agents make their way into water distribution systems through accidental releases, discharge or agricultural runoff into ground or surface water. Drinking water treatment plants are typically designed to remove many of these contaminants from water before they enter the distribution system. However, data on persistence of many chemical agents that could potentially be used to contaminate a drinking water distribution system are very limited. This section summarizes available adherence and decontamination data for specific target chemical contaminants in regard to water distribution system infrastructure, identifies contaminant classes for which little data currently exists, and discusses which of these may be suitable for future research.

## Inorganics

### *Arsenic*

In drinking water, arsenic will be found in its oxidized forms as arsenic(III) (arsenite) or arsenic(V) (arsenate). In a drinking water contamination scenario, arsenate would be more pertinent since arsenite will be

oxidized in drinking water when it reacts with disinfectant residuals (free chlorine or chloramines or oxidants in the drinking water process). Common arsenate and arsenite compounds are soluble in water, but solubility depends on the compound. Sodium arsenate, for example, has a solubility of 61g/100 ml water @ 15 °C [5].

Association of arsenic with infrastructure surfaces, especially in drinking water, is well documented. Numerous studies have directly collected corrosion/scale samples from drinking water distribution systems, analyzed those samples for sorbed arsenic through inductively coupled plasma mass-spectrometer (ICP-MS), and included inductively coupled plasma-atomic emission spectroscopy (ICP-AES) and X-ray diffraction (XRD). All have shown the presence of sorbed arsenic [4, 6-8]. In general, arsenite (As(III)) is considered more soluble and mobile, and arsenate (As(V)) is considered more likely to associate with solid surfaces. One study found that arsenic can incorporate into calcite [9]. Calcite can be present on iron or cement-mortar pipe interiors, depending on water quality conditions.

Decontamination information is limited, but useful information has been presented by two studies. One study showed that sodium arsenite injected into chlorinated water does adhere and persist on cement-lined iron coupons cut from pipe sections [10]. The coupons were conditioned in Cincinnati, Ohio tap water and had an established biofilm. Persistence of arsenic was not influenced by flow rate. Decontamination with flushing did not consistently remove As from the coupons, and flushing with low pH (pH=4) did not increase the removal of sorbed As. Maximum removal observed with flushing (using normal tap water pH [8.5] and low pH water) was 51%. Flushing

with a phosphate buffer did not result in the removal of any additional arsenic. Acidified potassium permanganate consistently removed 54-61% of adhered As.

Implementation of NSF Standard 60 Pipe Cleaning Aid Products Flushing (NW-310/NW-400 flushing, Floran Biogrowth Remover™/Floran Catalyst™ liquid activator, and Floran Top Ultra™/Catalyst, Blue Earth Labs, Lenexa, KS) resulted in 46-67% removal. Copeland et al. (2007) show that As desorbs from iron scale over time with increasing pH (testing between pH 6-9) [11]. Increasing pH above 9 may be an effective way of removing adsorbed As [12].

### **Mercury**

Mercuric compounds (Hg(II)) are soluble in ambient and drinking water matrices and are better studied than mercurous compounds. Oxidized mercury compounds of interest are mercuric chloride, mercuric sulfate, and mercuric nitrate. Mercuric chloride (HgCl<sub>2</sub>) is more soluble than other compounds, with a solubility of 7.4 g/100 ml at 20 °C (3.6 g/100 ml at 0 °C) [5]. It is often assumed that mercury in drinking water would effectively be in an oxidized, soluble compound due to the oxic nature of drinking water with disinfectant. In some aquatic systems, mercury can be transformed by microorganisms to methylmercury, which is highly toxic [13].

Mercury persistence on drinking water infrastructure has not been studied in detail, but persistence on biofilm has been observed. One study showed that mercury (Hg(II), introduced as HgCl<sub>2</sub>) sorbed to biofilms, but found that planktonic cells were more susceptible to the toxic effects of Hg than biofilm organisms [13]. This was explained by the fact that diffusion may prevent all of the dissolved Hg from reaching the biofilm, but also that

extracellular polymeric substances material in the biofilm may sequester Hg and prevent it from impacting the biofilm organisms. Other work observed that the biofilms grown on glass slides did sorb Hg(II) and methylmercury [14]. After an initial spike, the amount of Hg detected in the biofilms decreased. This could be due to desorption, but the authors of that reference attributed the decrease to de-methylation of the mercury followed by volatilization of elemental mercury. Finally, a study focused on wastewater showed that an Hg-resistant strain of bacteria does uptake and reduce soluble mercury in wastewater [15]. A bench-scale study found less than 1% attachment of mercury when introduced at 100 mg/L on pipes made of iron, galvanized iron, PVC, cement-mortar and polyethylene, epoxy and copper, some with biofilm [16].

Like persistence data, little information on techniques for decontamination should adherence occur was found. One study that focused on drinking water found that mercury injected as mercuric chloride did persist on cement-lined iron with an established biofilm [10]. Decontamination results were mixed. Flushing results were variable with more adhered mercury found post-flushing in some experiments and 19-51% removal in others. Low pH (pH 4) consistently showed 21-23% removal. Acidified permanganate removed 72-96% of the adhered mercury. Mercury was injected as Hg(II) which is the fully oxidized state, so oxidation was not the mechanism for removal. It was speculated that the low pH of the permanganate solution dissolved adhered mercury and removed it from the pipe surface.

## Petroleum Products

One study examined petroleum product persistence on drinking water infrastructure and techniques for decontamination should adherence to infrastructure occur. The study focused on diesel fuel and cement-mortar drinking water pipe that was extracted from a water distribution system [10]. Diesel fuel was found to be persistent on the cement-mortar coupons, with 21-23 mg of diesel adhering to each coupon (19 in<sup>2</sup>). Flushing removed 36-38% of the adhered diesel, but Surfonic® TDA-6 surfactant (Huntsman Corporation, The Woodlands, TX) removed more than 96%. Surfonic TDA-6 is an ethoxylate dispersant that is also effective for chlordane and may be an effective decontaminant for other petroleum products. However, a case study of an accidental diesel fuel contamination event in drinking water showed that flushing and continual use of water for household sanitation over two days was enough to bring the amount of fuel below the taste and odor threshold [16].

## Toxins

No data were found in the open literature on plant, bacterial, fungal or algal toxin persistence on drinking water infrastructure material or techniques for decontamination should adherence to infrastructure occur. However, it is known that botulinum toxin concentration is reduced by 99.90% to 99.99% in the presence of 0.5 mg/L free chlorine at 25° C [17]. Ricin is also sensitive to free chlorine, with 99.7+ % destroyed within 20 min [18]. Free chlorine destroys cyanotoxin below pH 8, but free chlorine is ineffective against anatoxin-a. Ozone can destroy anatoxin-a as well as microcystin and cylindrospermopsin [19]. Unless the water was dechlorinated or toxins

were introduced in large enough quantities to eliminate the disinfectant residual, these toxins would likely be destroyed by free chlorine upon introduction into a drinking water distribution system. Sensitivity to free chlorine may limit persistence in drinking water systems.

## Chemical Warfare Agents

No data were found in the open literature on CWA (chemical warfare agent) persistence on drinking water infrastructure material or techniques for decontamination should adherence occur. However, since many CWA are organophosphates, data on the persistence and decontamination of the organophosphate class may be used to inform CWA class information (see the Organic Chemicals section below).

## Pharmaceuticals

No data were found in the open literature on pharmaceutical persistence on drinking water infrastructure material or on techniques for decontamination should adherence occur.

## Organic Chemicals

### *Chlordane*

A pilot-scale pipe loop study showed that flushing with Surfonic TDA-6 effectively removed chlordane from both cement-lined ductile iron and clear PVC pipe surfaces. The decontamination efficiency ranged between 89% and 91% for the cement-lined ductile iron pipe material and was 99% for the clear PVC pipe material. Chlordane showed strong adherence to both cement-lined ductile iron pipe and clear PVC pipe surfaces [10]. Another study also tested the adhesion of chlordane applied to surfaces that simulate cement-lined and PVC pipes.

Chlordane was shown to adsorb onto the pipe material coupons. Flushing evaluations suggested effective decontamination with flowing water for chlordane on concrete and PVC. Hyperchlorination with no flow was not effective at decontaminating chlordane on concrete or PVC [20].

Static bench-scale experiments using sections of various types of iron, galvanized iron, plastic and cementitious pipe showed that chlordane adsorbed to all pipe surfaces, although cement-lined ductile iron and polyethylene results were inconclusive. The insecticide demonstrated strong attachment and a relationship between duration of contaminant exposure and mass of material attaching to the pipe wall. Chlordane was selected as a high log  $K_{ow}$  chemical surrogate with a log  $K_{ow}$  value of 6.2. Surfactants were found to be effective in removing organic contaminants from pipe surfaces. The recovery of chlordane was relatively consistent by pipe material, ranging from 68% for used galvanized pipe to 120% for epoxy-coated steel pipe [16].

Chlordane was used by an unknown perpetrator to contaminate a drinking water system in Pittsburgh in 1980. A detailed firsthand account of the response to the incident is documented by an author of the report titled "Guidance for Decontamination of Water System Infrastructure" [16]. Investigations by the water utility and law enforcement confirmed that the contamination was intentional, but the perpetrator was never found. Contaminated water was contained in the distribution system and a systematic campaign of flushing was undertaken as a decontamination method. Flushing was an effective way to reduce the amount of contamination in the water over the course of days, but chlordane was still detected up to nine months after the incident. Hot water

tanks in consumers' homes were particularly difficult to decontaminate. The research studies cited in this section and this case study show that more effective decontaminants should be identified for chlordane since it has been used as a drinking water system contaminant.

### *P-dichlorobenzene*

Static bench-scale experiments using sections of various types of pipe showed that p-dichlorobenzene (p-DCB) attached at rates greater than 5% to iron pipe with biofilm; new galvanized, used galvanized, cement-lined ductile iron (with seal coat); and epoxy-coated steel pipe under non-flow conditions. There was relatively low adsorption of p-DCB to chlorinated polyvinyl chloride (cPVC) pipe surfaces. Surfactants were found to be moderately effective in removing p-DCB from pipe surfaces, with a recovery efficiency that ranged from 13% for cPVC to 60% for epoxy-coated steel pipe [16].

### *Parathion*

There is only limited data on persistence on soils and river sediments, and no data on interactions with pipes or techniques for decontamination of infrastructure should adherence occur. In surface water, parathion will usually disappear within a week, mainly by adsorption to suspended particles and bottom sediments. Adsorbed parathion is subject to degradation by microorganisms and chemical hydrolysis. For typical drinking water treatment techniques, one article reported the following degradation results: chlorine, 100%; chlorine plus aluminum sulfate, 100%; ozone, 80%; ozone plus aluminum sulfate, 80-90%; and chlorine plus activated carbon plus aluminum sulfate, 100% [21].

The chemical transformation process involves the substitution of the sulfur atom in the P-S bond of the pesticide with an oxygen atom into its oxon (oxygen analogues). This transformation is a concern because chlorination is the most commonly used disinfection technique in many US drinking water treatment plants and the product oxon of parathion is generally considered to be more toxic than the parent compound. Based on research on the rates of sorption and partitioning of contaminants in river biofilm, sorption to pipe walls is possible [22-25].

### *Chloropyrifos*

This review found no adequate information on the adhesion of chloropyrifos on drinking water system infrastructure or on techniques for decontamination should adhesion occur. Studies pertaining to water treatment or persistence in the environment indicate that chloropyrifos adsorbs strongly to soil particles, adheres to sediments and suspended organic matter in water sources, and is persistent [26]. As with most pesticides, chloropyrifos is not well removed by typical physical treatment processes. Removal from water improves with oxidation using free chlorine or ozone followed by conventional treatment [21, 27, 28]. However, rapid reaction with chlorine in water treatment yields its oxon as a primary by-product that is more stable in chlorine and more toxic than the parent compound [22, 25, 29]. Reaction with monochloramine is substantially slower [30]. There is no information about the interaction with biofilm.

### *Sodium Fluoroacetate*

Sodium fluoroacetate (SFA) is a long-lasting, toxic, and miscible-to-highly-soluble rodenticide. One bench-scale study tested the adhesion of sodium fluoroacetate applied

to simulated cement-lined pipe and decontamination by flushing and hyperchlorination. SFA was shown to adsorb onto the pipe material coupons. Persistence and flushing evaluation suggested that decontamination with flowing water (1.5-2.0 fps) for SFA on concrete was not effective. Hyperchlorination (25-50 mg/L) with no flow was not effective in decontaminating SFA on concrete [20].

## **Chemical Research Discussion and Conclusions**

Arsenic adsorption to pipe material, particularly corroded iron, is well characterized. Decontamination options have been explored, and some data suggests that higher pH may be a decontamination option for arsenic. NSF-60 pipe cleaning agents removed arsenic, but results were similar to simple flushing. Mercury was shown to adhere to cement-mortar infrastructure, but little data on iron persistence were found. Decontamination of mercury with acidified potassium permanganate was effective and future research should further examine decontamination on other common infrastructure materials such as unlined iron.

Diesel fuel has been studied in a model drinking water system and an effective decontaminant was used (Surfonic TDA-6). Gasoline is similar in nature to diesel in that they are both composed of hydrocarbons, except that the hydrocarbon chains in gasoline are shorter than in diesel. Since gasoline is at least as prevalent as diesel, determining its persistence on cement-mortar, iron and PVC may be worthwhile.

No data was found on CWA or pharmaceutical persistence on drinking water infrastructure, nor was data found on

toxin persistence. However, it is known that ricin and botulinum toxins are oxidized by free chlorine, which may limit their ability to persist on infrastructure in a drinking water distribution system.

Little direct information on organic contaminant persistence exists beyond sodium fluoroacetate, p-dichlorobenzene and chlordane. However, these three contaminants represent a wide range of octanol water partition coefficient with log  $K_{ow}$  values of -0.061, 3.4 and 6.2 for sodium fluoroacetate, p-dichlorobenzene and chlordane, respectively. As  $K_{ow}$  increases, the propensity for a chemical to associate with octanol (and not with water) increases, which can be used as a proxy for the tendency of a chemical to associate with a solid surface, like drinking water infrastructure, and not dissolve in water. Data are available on chlordane and p-dichlorobenzene on cement-mortar and iron surfaces, and with cement-mortar for sodium fluoroacetate. Future research could focus on these three chemicals, and the resulting data could be extrapolated to other chemicals with similar  $K_{ow}$  values.

The existing data show that surfactants are effective at removing chemical agents from pipe materials. Surfonic TDA-6 removed 90% of the chlordane attached to cement mortar infrastructure, and it was also effective against p-dichlorobenzene. Surfonic N-60 also achieved 90% removal of p-dichlorobenzene adhered to cement-mortar. Flushing and chlorination were not effective at decontamination of sodium fluoroacetate.

### **Future Chemical Research**

*Inorganics:* Decontamination of adhered arsenic with high pH flushing should be explored on iron and cement-mortar

infrastructure. Mercury persistence on iron infrastructure should be explored, and decontamination with acidified potassium permanganate should be attempted.

*Petroleum Products:* Diesel and/or gasoline persistence on corroded iron should be studied, and if they are persistent, the effectiveness of Surfonic TDA-6 should be assessed. Gasoline and diesel are both widely used and available hydrocarbons and are good candidates for future study.

*Toxins:* Further characterizing toxin degradation in chlorinated and chloraminated water may help determine if toxins will persist in water long enough to become attached to infrastructure surfaces. Clearly, experiments directly examining persistence on infrastructure materials should also be conducted.

*Organics:* Further examination of persistence on infrastructure materials and techniques for decontamination of infrastructure materials could be collected with sodium fluoroacetate, p-dichlorobenzene, and chlordane since these chemicals represent a wide range of  $K_{ow}$ . Specifically, sodium fluoroacetate should be examined on corroded iron, and decontamination with surfactants (like Surfonic TDA-6) should be attempted. Alternatively, a representative contaminant from classes such as organophosphorous and carbamate pesticides could be chosen and its persistence on infrastructure materials could be examined. No data on persistence of pharmaceuticals or CWA were found, so these may be priority areas of future research. Organic contaminants often react with chlorine or chloramines, so a summary of their chlorine reactivity may indicate how likely they are to persist in disinfected water and reach drinking water infrastructure. Chlordane decontaminants should be

researched further since this chemical has a history of contamination in drinking water systems.

*General:* A future research goal that crosses all chemical classes would be the development of methods for sampling and analysis. Validated methods for extraction of contaminant from coupons (iron, cement-mortar, PVC) would be helpful in any future research endeavor and lend increased credibility to the data generated from the study.

# Biological Agents

## Introduction

Compared to chemical and radiological contaminants, substantial data for persistence of biological agents (mainly bacteria) are readily available. This is primarily due to the fact that coliform persistence and regrowth in drinking water biofilm has been researched in conjunction with the promulgation of the Total Coliform Rule by the United States Environmental Protection Agency. Much of this research was conducted in the 1980s, prior to the publication of the first Total Coliform Rule in 1989, and the 1990s (<http://water.epa.gov/lawsregs/rulesregs/sdw/tcr/basicinformation.cfm>) [31-40]. This data set is a rich source of information on persistence and, in some papers, decontamination.

While vegetative coliform bacteria has been the focus of a significant amount of research, additional studies are needed on spore-forming bacteria, such as *Bacillus anthracis* (the causative agent of anthrax), which are hardier and more persistent than vegetative cells. If decontamination of spore-forming bacteria from drinking water infrastructure is successful, the same method should also be effective for vegetative cells. This chapter is divided into analyses of literature about spore-forming bacteria and vegetative bacteria persistence on drinking water infrastructure and decontamination. Data on virus such as bacteriophage are more limited, but it is discussed where applicable.

### *Spore-Forming Bacteria*

Research on *Bacillus* spore adhesion in drinking water systems has occurred on copper, PVC, and galvanized iron coupons,

which are representative of new and old household plumbing materials [41]. Results showed that 1 mg/L free chlorine has little effect on the number of spores in the bulk water phase after 1 hour of contact, but 5 and 10 mg/L achieved 97% and 99.99% removal, respectively. These inactivation data are similar to results from other studies in the literature [42-45]. Between 20 to 95% of the spores inoculated into the model system adhered to the pipe with the fewest on PVC and the most on galvanized iron. Although experimental conditions such as temperature, pH, and flow were not reported [41], the data confirm that spores injected into a drinking water distribution system can survive typical disinfectant residuals and adhere to and persist on household plumbing materials.

*Bacillus* spore persistence was examined in a study funded by the Water Research Foundation, formerly known as the American Water Works Association Research Foundation [16]. In this study, pipe sections of various infrastructure materials (iron, cement mortar, galvanized iron, copper, PVC) were filled with water, contaminated with a target contaminant, and allowed to incubate under stagnant conditions for seven days. Stagnant incubation was used to mimic a scenario where contaminated water is isolated in a drinking water system, allowed to sit stagnant until its disposition is decided, and then flushed out.

Exposing spores that had been adhered to galvanized iron pipe to 50 mg/L free chlorine for 6, 60, and 1,200 min (Ct [concentration × contact time] of 300, 3,000, and 30,000 mg/L-min, respectively) resulted in 65-84% removal when the initial number of attached spores was  $7 \times 10^4$  cfu. One hundred percent removal was reported with heavily tuberculated iron under the same

chlorination conditions. However, the authors point out that few spores were initially recovered from the iron surface (500 cfu) and the results may not be reliable. Poor recovery may have been due to inadequate sampling. Initial chlorine residual quickly decreased from 50 mg/L in galvanized and tuberculated iron pipes to nearly zero at 60 minutes and was undetectable thereafter.

A series of experiments were conducted using EPA bench-scale systems, which include biofilm annular reactors (BAR), and reactors developed by the US Center for Disease Control and Prevention (CDC). In one study, *B. cereus* spores were added to a BAR that had biofilm of *Pseudomonas fluorescens* grown on polycarbonate coupons [46]. The authors estimated that up to 3% of the  $10^5$  cfu/ml spores introduced were adhered to the biofilm. Chlorination with 2 to 3 mg/L for 7 to 8 days resulted in 2 to 3-log removal. Numerous spores still remained in the single species biofilm. Other BAR studies used iron, cement-mortar, and PVC coupons that are representative of water distribution mains currently in service [47]. Free chlorine concentration up to 70 mg/L resulted in 3-log removal of *B. globigii* spores, but persistence was still observed [48]. Application of chlorine dioxide to the same reactor/coupon set-up resulted in 4-log inactivation of adhered *B. globigii* over the course of 4 days [49]. Application of 5 mg/L free chlorine resulted in 2 to 4-log removal of *B. globigii* attached to cement-mortar coupons, with no detectable spores remaining [50].

CDC reactors were operated with PVC and copper coupons, which simulate household plumbing materials. Spores persisted on the coupons and contact with 103 mg/L free chlorine and with 49 mg/L monochloramine

produced 2-log removal in 60 minutes of contact [51]. An extension of this study examined germination of spores with L-alanine and inosine before disinfection [52]. Application of germinant reduced the number of attached spores by 1.5 to 3.0-log, with up to 4.6-log reduction after application of free chlorine at 100 mg/L. Log reductions of 0.4 to 0.6 were observed with chlorination alone. The results show that germination can enhance the decontamination potential of free chlorine.

The idea of germinant-enhanced decontamination of spores was further extended to a pilot-scale drinking water distribution system simulator (DSS) [53]. Cement-mortar and corroded iron coupons were inserted into a six-inch diameter pipe with tap water flowing through it. After a one-month conditioning period, the DSS was contaminated with *B. globigii* spores. Adhered spores were exposed to germinant, then free chlorine at 5 and 25 mg/L, followed by exposure to flowing water at a mean velocity of 1 ft/sec. Germination enhanced the effect of chlorination of cement-mortar, while flushing was enhanced on spores adhered to corroded iron. Chlorination and flushing left viable spores on each coupon type when used alone (except cement-mortar at 25 mg/L free chlorine), but adding a germinant enhanced decontamination to the extent that no viable spores could be detected on the coupons.

Other pilot-scale studies have supported the previously mentioned research. *B. globigii* spores were shown to persist on a 15 year old pipe that carried drinking water in a pilot-scale experimental system [54]. In another pilot-scale study, sections of aged cement-mortar lined pipe removed from a drinking water distribution system were placed into the DSS mentioned in the germination study and contaminated with

spores at a concentration of  $10^4$  cfu/ml [10]. Flushing at a mean velocity of 2.5 ft/sec resulted in no removal of adhered spores. Disinfection with 200 mg/L free chlorine and flushing (2.5 ft/sec) for two hours decreased adhered spores by 95% (1.3-log), with many viable spores remaining on the coupons.

### ***Vegetative Bacteria***

Much of the research related to vegetative bacteria persistence in biofilm has focused on coliforms such as *Escherichia coli* and *Klebsiella pneumoniae* [32-34, 39, 55-58]. The general trend of these studies is that coliforms can adhere to biofilm and persist or grow when exposed to simulated drinking water conditions. The authors cite the protection from disinfectants that biofilms offer, but many of these studies used dechlorinated conditions during injections of coliforms. Beyond PVC, many of these studies use glass or carbonaceous materials that are not representative of water distribution system infrastructure. One study did examine coliform persistence on mild steel with biofilm and found that 10 times more coliforms accumulated than on PVC with biofilm [32].

Another study examined *E. coli* persistence on PVC coupons colonized by drinking water biofilm in a 4-inch diameter pipe [35]. Injected *E. coli* was detectable on the PVC coupons up to 18 days in unchlorinated water in the presence of flow. Another study used corroded iron as a coupon material and assessed *Klebsiella pneumoniae* persistence in the presence of flow [59]. Experiments were conducted in chlorinated and unchlorinated water. Persistence on iron was observed up to 17 days in unchlorinated water and up to 12 days with chlorine residual less than 1 mg/L. Interestingly, *K. pneumoniae* cultured under

low nutrient conditions persisted longer than those cultured under rich conditions, indicating that environmental isolates may persist longer in drinking water systems. However, both were not detectable on the coupons after 17 days with constant flow of 1 ft/sec.

*Legionella pneumophila* persistence on biofilm has been studied by multiple researchers. Like much of the research into coliforms, substratum materials used to grow biofilm are not representative of drinking water distribution systems infrastructure. However, persistence was observed for limited periods of time, usually in unchlorinated conditions. One researcher grew a drinking water biofilm on glass and PVC slides and observed persistence between 20 to 40 days [60]. Persistence was observed for 15 days under similar conditions using stainless steel slides [61]. A study on the persistence of *L. pneumophila* on biofilm found that chlorine and monochloramine were effective at decontamination. However, monochloramine worked faster than chlorine, likely due to its lower reactivity with various biofilm components [62]. Finally, two complementary studies found that *L. pneumophila* persisted on coupons up to 12 days in the presence of a free chlorine residual of 0.2 mg/L [63]. However, fluorescent *in-situ* hybridization found DNA signatures on the coupons up to 38 days after contamination, although it was not determined if viable cells were present [64].

Data has been published on contamination of drinking water supplies with vegetative bacterial pathogens. *E. coli* 0157:H7 was the cause of the contamination of the Cabool, Missouri water supply, which likely originated from a series of water main breaks and meter replacements [65, 66]. The water supply was unchlorinated at the

time of the outbreak, which allowed the *E. coli* to persist in the water column. A program of chlorination (up to 3 mg/L) and flushing to adequately spread the disinfectant was used to decontaminate the system. After decontamination (and a “Boil Water” advisory) the number of cases of *E. coli* infection dropped precipitously.

*E. coli* 0157:H7 was also the cause of a large water supply outbreak in Walkerton, Ontario, Canada [67, 68]. The cause of the outbreak was attributed to infiltration of runoff contaminated with animal waste, and the susceptibility of the old corroded cast iron pipes to harboring *E. coli*. Remediation included distribution system flushing, disinfection of building plumbing with chlorine, as well as implementation of a program to replace the old cast iron water mains. These actions ended the outbreak, indicating that flushing and chlorination can be effective against *E. coli*.

*Salmonella typhimurium* was attributed to an outbreak of illness that originated in the water supply in Gideon, Missouri [69, 70]. Past research has shown the potential for *Salmonella* spp. persistence on biofilm [60]. Contamination occurred from bird droppings and feathers entering a water storage tank. The water system did not contain a disinfectant residual, and a flushing event before the outbreak helped spread contamination through the distribution system. In addition to a boil water advisory, the water tank was chlorinated, which stopped the outbreak. However, a systematic program of chlorination in the distribution system was not undertaken. In 2008, *Salmonella* contamination occurred in Alamosa, Colorado [71]. Prior to the outbreak, the water supply was not chlorinated. Chlorine was added to the distribution system at 25 mg/L, with a goal of maintaining at least 10 mg/L for 24 hours.

An extensive flushing program was also implemented. This strategy was successful, and the water supply was chlorinated thereafter.

### **Viruses**

Data on virus persistence in drinking water biofilm is more limited than for other microorganisms, but some studies exist. Virus persistence was studied in a pilot-scale unit using biofilm formed on PVC coupons. Poliovirus 1 persisted for less than 6 hours on the coupons, and a chlorine residual of 0.04 mg/L inhibits live virus persistence in the bulk water and biofilm [72]. Limited poliovirus persistence (6 days) was observed in another study on polycarbonate with drinking water biofilm [73]. However, the viral RNA was observed up to 35 days with no chlorine present.

Alternatively, calicivirus persisted on PVC coupons with drinking water biofilm in the presence of 0.15 mg/L free chlorine for 3 weeks [74]. *Bacteroides fragilis* phage B40-8 and coliphage MS-2 persisted up to 30 days at low levels on PVC and stainless steel [75, 76]. Interestingly, a consistent 0.5 mg/L free chlorine residual was present, indicating that some viruses were unaffected by the residual. A recent review article on viral persistence in biofilms cites numerous conference proceedings that support the idea that viruses persist in drinking water biofilms in the presence of free chlorine, although the proceedings may not be readily available [77].

## **Biological Agent Research** **Discussion and Conclusions**

A common theme that occurs in many of the cited studies is that decontamination with free chlorine leaves viable *Bacillus* spores adhered to the coupons of drinking water infrastructure. This is an important finding since chlorine is commonly used in drinking water treatment and is known to be a strong oxidant. Numerous studies have shown that *Bacillus* spores suspended in water with free chlorine are resistant to inactivation, but given enough contact time they are undetectable by culture [42-45].

Data on spore inactivation in water would support the conclusion that extending decontaminant contact time should also adequately decontaminate spores attached to pipe material. This may be true for some pipe materials, but common distribution system materials such as corroded iron or cement-mortar have been shown to be difficult to decontaminate with free chlorine. However, enhancing flushing or chlorination with germination has proven to be more effective than flushing or chlorination alone. Alternative disinfectants such as chlorine dioxide have also proven to be effective at decontaminating adhered spores without germination enhancement. Therefore, germination with a chemical suited for the *Bacillus* species followed by chlorination at least 25 mg/L and introduction of clean water moving at a velocity of at least 1 ft/sec is more effective than chlorination or flushing alone. Alternatively, application of at least 25 mg/L chlorine dioxide is more effective than chlorination at that level in bench scale applications.

The amount of literature discussing decontamination of vegetative bacteria adhered to infrastructure surfaces in experimental drinking water systems is

limited. Much of the literature focuses on initial attachment or persistence, often under conditions somewhat relevant to drinking water, but in dechlorinated water. The literature related to decontamination of vegetative bacteria includes numerous studies examining real-world decontamination incidents. Specific data on the number of pathogens associated with an infrastructure material before and after decontamination was not collected in these studies. However, the large drops in infection after decontamination measures were taken indicate that the measures were successful. In the studies that examined decontamination of adhered vegetative bacteria, free chlorine with flushing was commonly used.

Ultimately, chlorination may be adequate for decontamination of drinking water distribution systems contaminated with vegetative bacteria, with the addition of flushing to ensure adequate transport of the disinfectant. In the studies cited in this report, free chlorine levels used for decontamination ranged from undefined to 3 to 25 mg/L. However, since vegetative bacteria are more sensitive to disinfection than spore formers, methods that are effective for spore formers should also decontaminate vegetative bacteria. However, if pathogenic vegetative bacteria are harbored by amoebae in drinking water biofilm, they may be more difficult to flush or disinfect [78].

It is well documented in the literature that viruses in water are sensitive to various disinfectants such as chlorine, chlorine dioxide, and ferrate [79-83]. Although the literature presents a mixed picture on virus persistence, there is some evidence that they do persist, even in the presence of a disinfectant. The highest chlorine residual used in the cited studies was 0.5 mg/L, and

high levels used for decontamination of spores have not been studied. It is notable that virus persistence was observed on glass, PVC, and stainless steel coupons, which should be easier to decontaminate relative to iron or cement-mortar. In the absence of any firm data on virus decontamination, the techniques used for spore-forming bacteria should be applied as suggested by the Spaulding hierarchy [84].

### **Future Biological Agent Research**

*Spore-Forming Bacteria:* Given the effectiveness of chlorine dioxide on the bench scale, this disinfectant and other alternative disinfectants should be explored. Ozone and peracetic acid are good candidates for study as are peroxide, acidified nitrite, monochloramine and mixed oxidants.

*Vegetative Bacteria:* Additional data on the adequacy of specific levels of free chlorine to decontaminate infrastructure surfaces (i.e., iron and cement-mortar) could confirm or disprove what can be extrapolated from the literature on disease outbreaks in water supplies. It can also be assumed that if a technique is effective at decontaminating drinking water infrastructure surfaces with adhered spore formers, then that technique would also be effective against vegetative bacteria. Research to confirm this assumption would be of interest.

*Viruses:* Examine model virus persistence on common drinking water infrastructure materials such as iron and cement-mortar. If persistence is observed, attempt decontamination with disinfectants such as chlorine (25+ mg/L), chlorine dioxide, and monochloramine.

## **Radiological Agents**

### **Introduction**

Historically, research related to radionuclides in drinking water has focused on two areas. The first is evaluating treatment techniques to remove radionuclides from source water [85-88]. The primary goal of treatment in the U.S. is ensuring that alpha and beta emitters are below regulatory limits. The second area of research came from an observation that in some communities, the amount of a particular radionuclide (or alpha/beta emitter) was sometimes observed to be higher at a consumer's tap than it was in the water leaving a treatment plant [89-91]. Results from these papers show that low concentrations of radium originating in groundwater was accumulating on pipe surfaces over the course of years.

Desorption due to sloughing of pipe material or treatment changes led to observed concentrations above regulatory limits at the consumer's tap. This research complements other work focused on inorganics, such as lead and arsenic, which accumulate on pipe scales and later desorb [4, 92].

This section discusses similar research, but in the context of contamination of drinking water pipes with radionuclides of concern: cesium, strontium, and cobalt. As previously mentioned, radium and uranium are found in groundwater and have a track record of research in the peer-reviewed literature. Cesium, cobalt, and strontium have not previously been examined for their persistence in drinking water systems. Therefore, the literature review in this section focuses on work from nuclear cleanup sites where groundwater was contaminated with these radioisotopes. Information will be extrapolated to common

drinking water infrastructure materials (iron and cement-mortar lined iron).

### ***Cobalt***

Of the three radionuclides discussed in this section, the fewest number of papers were related to cobalt. When dissolved in disinfectant free water, cobalt forms a soluble salt that adsorbs to magnetite, which, along with hematite and goethite, is a common corrosion by-product in iron drinking water pipes [93]. Nearly 100% desorption was observed when pH was decreased below 3. Magnetite adsorption has been seen in another study, but hematite adsorption was also observed [94]. Parallel experiments with cesium and strontium did not show adsorption on hematite. The authors of that study speculate that cobalt may form a Co-Fe matrix, or precipitate on hematite surface. Cobalt adsorption to clay has also been observed, which may share some chemical species with cement-mortar (cement and sand), but are generally far more complex [95]

Cobalt was observed to adsorb to corroded iron that had been conditioned in a drinking water distribution system simulator for one month [96]. Cobalt was introduced as water-soluble cobalt, which is in the +2 oxidation state. Cobalt reacted with free chlorine in the water and oxidized to Co(III), which is insoluble in water. The Co(III) precipitated on the iron surface and persisted for 42 days. It likely would have persisted longer, but 42 days was the duration of the persistence experiment. Co(III) is only soluble in acid, and contact with sulfuric acid (0.36 M) removed over 90% of precipitated cobalt. This treatment also removed some of the corrosion layer, so, while successful, it is uncertain whether this decontamination method would be useful in a real drinking water distribution system.

### ***Strontium***

One study has specifically focused on strontium [97] adsorption to drinking water infrastructure, but numerous other studies have examined aqueous strontium adsorption to various iron oxides, clays, and silica compounds. The research study that focused on drinking water used a model distribution system and examined strontium persistence on iron oxides that form on unlined iron conditioned in potable water [98]. The iron oxides contained background strontium since stable strontium was present in the tap water used in the study. Strontium was spiked into the model system at 100 mg/L and allowed to contact the coupons for 1 hour before being flushed out. The amount of strontium adhered to the coupons increased six-fold immediately after contamination, but returned to the baseline concentration one day after contamination. This indicates that the mechanism of strontium adherence to the iron oxides was weak and reversible when the exposure was transient, even at a relatively high concentration of 100 mg/L. The baseline strontium adhered to the coupons came from long-term exposure (likely years) to low levels (0.2 to 0.3 mg/L) of strontium. This suggests that the longer strontium is in contact with iron oxides, the greater the chance it will become irreversibly adhered or incorporated into the iron oxide complex.

Strontium adsorption to iron oxides has been observed in numerous studies, although not all of them directly examine desorption. In one paper, strontium saturated the hydrated ferric oxide (HFO) adsorption sites within minutes of contact, and adsorption was successfully modeled with the Donnan diffusion model [99]. In another, strontium adsorbed to HFO after 4 hours of contact [100]. It was concluded that strontium physically adsorbed to HFO (not

chemically), meaning that desorption was possible. Later research showed that strontium adsorbed to HFO but substantially more was adhered when bacteria were present [101]. This may indicate that biofilm accumulation on iron oxides may promote adhesion. A follow up to this study showed that the mechanism of strontium adsorption to HFO was inner sphere complexation, or adsorption of strontium to HFO with no intervening water molecules [102]. Complexing of this nature means that strontium could form a persistent bond with HFO.

Research on strontium adsorption to iron oxides does expand beyond HFO. Common iron oxide/oxyhydroxide species in corrosion by-products formed in drinking water are  $\text{Fe}_3\text{O}_4$  (magnetite),  $\alpha\text{-FeOOH}$  (goethite), and  $\gamma\text{-FeOOH}$  (lepidocrocite). Studies have reported strontium adsorption to goethite [95, 103, 104] and magnetite [94], but adsorption or bonding directly to the goethite may not be the primary mechanism at play. Data from one manuscript is used to argue that strontium adsorbs to goethite as a  $\text{SrOH}^+$  complex when carbonate is not present [105]. However when carbonate is present, strontium forms complexes with carbonate that then adsorb to goethite. Dissolved carbonate ions are present in distributed tap water, and, depending on the water quality, precipitated calcium carbonate can also be present on iron scale. Strontium and calcium are neighbors in the alkaline earth metals column of the periodic table and share many similar properties. Strontium exchange with calcium in carbonate compounds might also promote strontium adhesion on goethite and other iron oxides.

Strontium-carbonate complexation and exchange with calcium are also observed in adsorption to cement or concrete-mortar.

Drinking water infrastructure is commonly lined with cement-mortar, which is a mixture of Portland cement and sand. Portland cement is largely calcium oxide and sand is silicon dioxide, which is a component of many clays. Cement-mortar can also have precipitated calcium carbonate (calcite). As with iron oxides, data shows that strontium combining with carbonate will precipitate onto the surface of cement, silica, and kaolinite clays, and adsorption can be reversible [104, 106, 107]. However, strontium carbonate precipitation was observed to be dominant below pH 8.6, while strontium complexation with the silica or kaolinite surface occurred above pH 8.6. Other researchers have observed that at strontium concentrations above 0.3 mmol, strontium precipitates onto the surface of clay. At concentrations below 0.3 mmol, strontium substitutes with calcium in calcite [108], which has also been observed in other studies [109].

Data from other studies show that while strontium incorporates into calcite structures, it can be removed. In one, strontium was put into contact with calcite containing Portland cement for three days and desorption of adsorbed strontium was observed when clean water replaced the strontium solution. Contact of clean water with the strontium-contaminated cement occurred for two days [110]. Desorption with clean water took place after six separate but identical sequential desorption steps. In two other studies, strontium in contact with kaolinite was found to irreversibly incorporate into the clay structure over weeks to months of contact [111, 112]. These studies show that strontium contact time with drinking water infrastructure may influence how it persists. Strontium adsorption has also been examined under a range of pH. When adsorption to goethite was studied under pH

ranging from 6 to 10, no strontium adsorption was observed at pH 6, while nearly all strontium spiked into a goethite suspension adsorbed at pH 10 [105]. Similar results were observed with magnetite [93]. This study was reinforced by another paper that showed 95% of dissolved strontium adhered to iron oxides above pH 7.6, but none adsorbed at pH less than 4.5 [113]. Increasing strontium adsorption (uncomplexed) at higher pH is a general trend shown in the peer-reviewed literature [85, 114, 115]. When Sr sorbed to iron oxide coated sand at alkaline pH was washed at pH 3, nearly all adsorbed Sr was released [116]. One review article discusses how the high pH in the cement/cement-mortar matrix influences strontium adsorption and precipitation, which is enhanced at high pH [117]. These studies point to low pH flushing as a potentially effective decontamination option for adhered strontium.

### *Cesium*

Two studies specifically focused on the adsorption of cesium to drinking water infrastructure, but other studies may offer insight into how cesium interacts with similar materials. In the first study, cesium was spiked into a model drinking water system at 100 mg/L and allowed to contact the iron coupons for two hours [96]. After this two-hour contact period, the cesium solution was flushed out and clean tap water was allowed to flow past the coupons for 42 days to assess persistence. The coupons were then dissolved through microwave-assisted digestions and analyzed through ICP-OES. No cesium was detected on the coupons one day after spiking or during the 42-day monitoring period. No change in the 100 mg/L bulk water cesium concentration was observed during contamination. Unlike similar experiments performed with

strontium, samples from the cesium-drinking water infrastructure study were not taken immediately after the cesium was flushed from the reactor. Samples were only taken one day after cesium was flushed from the reactor. So, if short-term persistence on the iron occurred, it was not captured in this study.

In the other study focused on drinking water, iron (with and without biofilm), galvanized iron (new and used), PVC (with and without biofilm), cement-lined iron, copper, polyethylene, PVC and epoxy pipes were exposed to a stagnant solutions of stable cesium chloride [16]. Adsorption of cesium to the pipe interior surfaces was observed with cement lined ductile iron, iron with biofilm, and used galvanize iron with percent attachments of 12.4%, 4.6% and 1.3%, respectively. Decontamination was undertaken with a free chlorine solution (25 mg/L with 20 hours of contact time) and 1% and 10% solutions of Simple Green® cleaner (Sunshine Makers, Inc., Huntington Beach, California). Free chlorine removed 23% and 26% of the adhered cesium from the used galvanized and cement-lined iron pipes, respectively. Simple Green at 10% solution removed 45% and 47% of the cesium adhered to cement-lined and galvanized pipe, but a 1% solution was significantly less effective. These results show that common cleaning agents and disinfectants can remove sorbed cesium when it is delivered to a bench-scale experimental pipe surface under stagnant conditions.

Other experiments designed to provide adsorption data for cesium-magnetite isotherms over pH 6 to 9 can be used to assess persistence. Isotherms were generated by equilibrating cesium with magnetite for one hour [93]. Cesium initially adsorbed, but desorbed from the

magnetite as soon as pH was lowered to below three. Another study found that cesium equilibrium with magnetite and hematite occurred after two hours of contact, although very little cesium adhered to hematite [94]. Taken together, these studies point toward cesium adsorbing to iron oxides, but not persisting if clean water flows after the contamination slug. Furthermore, low pH flushing may also be effective at removing an adhered cesium.

Cesium interaction has been studied with cement and cement-mortar matrices, mostly in the context of nuclear waste storage in concrete vessels. Data has shown that mixtures of calcium oxide and silicon (the main components of cement-mortar) adsorb cesium, with the most adsorption occurring at neutral pH [85]. It was also observed that cesium retention was poor when ionic strength was increased by adding salts. This suggests that other ions out-compete cesium for adsorption sites and that adsorption is reversible. Data supporting this point shows that the presence of competing ions such as sodium, magnesium, and aluminum decreases the adsorption of cesium on magnetite and ferrite [118].

Although cesium has been shown to adsorb to and persist on cement-mortar, some authors have found that the process is reversible. In one study, cesium was in contact with cement for 147 days [106]. Equilibrium was achieved in this time, but over 90% removal of cesium was observed when the cesium-loaded cement was placed in clean water. Like iron oxides, when ionic strength is high (competing ions are present), cesium adsorption on cement is inhibited [107] and competing ions could outcompete cesium for adsorption sites. An observation from multiple studies is that cesium uptake onto cement and kaolinite is slow compared to radionuclides such as

strontium. As previously mentioned, equilibration with cement took 147 days in one study [106]. Another study saw cesium equilibration with kaolinite take 1 year (compared to strontium, which equilibrated in 1 day) [112]. An interesting observation from this study is that not only did more cesium adsorb to kaolinite with time, but cesium also became more tightly bound to the clay with time. This may be explained by the fact that diffusion into the cement pore space is concurrent with adsorption [119].

### **Radiological Contaminant** **Research Discussion and** **Conclusions**

The available results indicate that cobalt interaction with drinking water infrastructure is straightforward. Soluble cobalt(II) salt introduced into drinking water with a disinfectant residual will oxidize to cobalt(III) and precipitate. Precipitation occurs as soon as the cobalt is mixed with the water, and it is known that cobalt(III) is only soluble in acidic solution. Low pH flushing with compatible acids is the only known method of removing precipitated cobalt in situ. The available literature included data on drinking water with free chlorine as a disinfectant, corroded iron as an infrastructure material, and acid as a decontaminant. It may be beneficial to study cobalt persistence with other disinfectants and infrastructure materials to determine if the published results with free chlorine and iron translate.

Strontium has been shown to adhere to both iron and cement mortar infrastructure materials, but time and pH influence its persistence. Transient exposure to iron does not appear to lead to long-term persistence, but persistence increases the longer strontium is in contact with infrastructure

materials. Strontium does associate with cement/cement-mortar. Strontium can precipitate and substitute for calcium in calcium carbonate, so the presence of carbonate greatly influences strontium persistence. Alkaline pH leads to greater strontium persistence, and low pH removes strontium from iron and cement materials. This means that adsorbed strontium may be removed with low pH flushing, but this should be tested.

Cesium interaction with iron has been studied, and although adsorption can occur, cesium does not appear to persist on iron when flow is present. Persistence was observed under completely stagnant conditions and decontamination with free chlorine and Simple Green® was moderately effective. Cesium adsorption to iron oxides is also inhibited by competing ions. Cesium adheres to cement, cement-mortar, and clays, but adsorption is slow (compared to strontium), taking months or up to a year. Some data suggests that longer cesium contact time with cement will lead to stronger binding, which could make decontamination difficult. Little data on cesium adsorption to cement-mortar was found in a drinking water environment, and this should be researched further. However, the best option for decontamination appears to be flushing at low pH and with competing ions such as sodium or magnesium present.

## **Future Radiological Contaminant Research**

*Cobalt:* Precipitation on cement mortar in the presence of free chlorine should be examined. Soluble cobalt should also be introduced into chloraminated water to assess whether precipitation is similar to chlorinated water. Chelating agents (like EDTA) could be effective at sequestering cobalt.

*Strontium:* Persistence on cement-mortar drinking water infrastructures should be assessed under alkaline conditions since this may lead to the most persistence. Low pH flushing should be tested as a decontamination method if persistence is observed.

*Cesium:* Persistence on cement mortar under drinking water conditions should be examined. If persistence is observed, decontamination with flushing, low pH flushing, and removal with competing ions should be studied.

## **Overall Concluding Remarks**

The analysis in this report shows that decontamination options for drinking water infrastructure have been explored, but important data gaps remain. In general, microbial persistence in drinking water has been studied more extensively than chemical agents and radionuclides. Several studies have been published that focus specifically on decontamination of spore-forming bacteria from drinking water infrastructure. The data show that increasing chlorine levels in a drinking water distribution system may not be effective at decontaminating spores. Techniques such as germinating spores before chlorination or using an alternative disinfectant such as chlorine dioxide have shown promise.

However, the feasibility of a technique such as germination will depend on whether sufficient quantities of an effective germinant can be realistically added to a distribution system. Decontamination of vegetative bacteria has been successful using free chlorine and flushing. However, it is possible that methods successful at decontamination of spores would also be effective for vegetative bacteria. Little information is available on virus persistence and techniques for decontamination of water infrastructure should adherence occur. Future studies focused on virus persistence in both disinfected water and on drinking water infrastructure would be beneficial.

Past research on radionuclide persistence on drinking water infrastructure has focused on radium and uranium, which are found in ground water. This study reviewed literature on cesium, strontium, and cobalt, which are fission products or are found in commercially available items and medical devices. Cobalt persistence was straightforward since soluble cobalt chloride was found to react with free chlorine and precipitate onto iron coupons. The precipitated cobalt solids were only soluble in acids. Future research should confirm precipitated cobalt persistence on other common infrastructure materials like cement-mortar, and examine decontamination solutions beyond acidification. Cesium was not found to persist on iron infrastructure when flow was present, but it did persist when in contact with iron pipes under stagnant conditions. Free chlorine and a common household cleaner removed some adhered cesium, but more effective decontamination methods should be studied. Cesium persistence is possible on cement-mortar, and this should be studied further. Strontium persisted for less than one day on iron coupons, indicating that strontium adhesion to the iron

matrix is weak and reversible. Further research on strontium persistence on cement-mortar should be undertaken.

Available data on chemical agent persistence is piecemeal. This analysis found that persistence and decontamination data exists for chlordane, p-dichlorobenzene and sodium fluoroacetate, which could be characterized as having low, medium and high affinity water based on their octanol-water coefficient. Decontamination data showed that chlordane, p-dichlorobenzene, and diesel fuel were effectively removed (greater than 90%) from drinking water with a commercially available ethoxylate dispersant (Surfonic TDA-6). This dispersant can be used as a decontaminant for other chemicals as well. Sodium fluoroacetate was not as persistent as the other compounds, but flushing and chlorination were not effective decontamination techniques. Future study of organic agents could use one representative contaminant from a contaminant class (e.g., organophosphorous pesticide, carbamate pesticides) or use a measure of a chemical's affinity for water like octanol-water coefficient. Data on inorganics (arsenic and mercury) demonstrates persistence, and decontamination was observed with flushing and application of commercially available decontamination solutions. However, most studies showed removal between 40-70%, so other decontamination techniques should be studied. No data on persistence, or techniques for decontamination should adherence to infrastructure occur, was found for pharmaceuticals, CWAs, or toxins.

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