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Characterization of elemental and structural composition of corrosion scales and deposits formed in drinking water distribution systems

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ABSTRACT

Corrosion scales and deposits formed within drinking water distribution systems (DWDSs) have the potential to retain inorganic contaminants. The objective of this study was to characterize the elemental and structural composition of extracted pipe solids and hydraulically-mobile deposits originating from representative DWDSs. Goethite (α-FeOOH), magnetite (Fe_3O_4) and siderite ($FeCO_3$) were the primary crystalline phases identified in most of the selected samples. Among the major constituent elements of the deposits, iron was most prevalent followed, in the order of decreasing prevalence, by sulfur, organic carbon, calcium, inorganic carbon, phosphorus, manganese, magnesium, aluminum and zinc. The cumulative occurrence profiles of iron, sulfur, calcium and phosphorus for pipe specimens and flushed solids were similar. Comparison of relative occurrences of these elements indicates that hydraulic disturbances may have relatively less impact on the release of manganese, aluminum and zinc, but more impact on the release of organic carbon, inorganic carbon, and magnesium.

1. Introduction

Corrosion scales affect water quality in drinking water distribution systems (DWDSs) in many important ways. The rates at which such scales are generated and chemical composition, structures, morphologies and solubilities of predominant mineralogical phases constituting them are all affected by both the pipe material (cast iron, steel or PVC) on which they are deposited and water chemistry parameters that include pH, temperature, DIC and alkalinity, concentrations of sulfate, chloride and natural organic matter (NOM), dissolved oxygen (DO) and disinfectant type and residual, presence of corrosion inhibitors (e.g., phosphate), overall conductivity of water, and hydraulic patterns (Sarin et al., 2001; Korshin et al., 1996; Vazquez et al., 2006).

Over past several years, the issue of potentially significant accumulation of trace inorganic contaminants (e.g., arsenic, vanadium, lead and others) within DWDSs has gained considerable attention. Reiber and Dostal, 2000; Lytle et al., 2004; Schock et al., 2008 and Gerke et al., 2009 have demonstrated that when these inorganic contaminants are present at concentrations below their respective maximum contaminant levels (MCLs) or even at essentially non-detect levels in water sources, they are capable of accumulating to

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measurable levels on and within deposits that exist in DWDSs. Certain compounds commonly found in corrosion scales and other deposits, including individual phases of iron(III) and manganese oxides, have been shown to adsorb and concentrate trace inorganic contaminants (Sugiyama et al., 1992; Nelson et al., 1995; Fendorf et al., 1997; Larsen and Postma, 1997; Gray et al., 1999; O'Reilly et al., 2001; Trived et al., 2001; Lytle et al., 2004; Cancès et al., 2005).

While the amount of information concerning the properties of corrosion scales, deposits and colloidal particles in DWDSs is remarkable (Tuovinen et al., 1980; Benjamin et al., 1996; Sarin et al., 2001; Teng et al., 2008; Gerke et al., 2008; Borch et al., 2008; Barkatt et al., 2009), there is a need to provide a more complete characterization of their physicochemical properties. This information can provide valuable insight on factors that influence and control the accumulation and co-occurrence of regulated trace inorganic contaminants. Accordingly, the main objective of this study was to characterize corrosion scales and deposits originating from DWDSs with varying finished water chemistries and pipe materials.

2. Materials and methods

2.1. *Participating utilities*

Twenty drinking water utilities that participated in the study were located in the contiguous United States. The selection of participated utilities and detailed information on sampling and analytical approaches can be found in the report by Friedman et al. (2010) on the relevant study funded by Water Research Foundation. Most of the utilities were from the upper Midwest while others were from the Western, Southwestern, and Northeastern regions, where groundwater tends to be softer and less mineralized than in the Midwest. Table 1 provides a summary of the utility participants and certain characteristics of each, as reported by each utility.

2.2. *Sample types and processing*

Three types of samples were collected: (1) pipe specimens, either obtained from a recent "live" extraction or from a utility storage area (referred to as "boneyard" specimens); (2) hydraulically-mobile deposit material collected during hydrant flushing events; and (3) distribution system water samples. Where appropriate, distribution system water samples were collected to correspond to each solid sample (i.e., at a site near the location where the solid sample was obtained). General water quality parameters (pH, temperature, alkalinity, disinfectant residual, and turbidity) were measured at the same time and location as water sample collection.

Samples provided by utility participants were sent to the Environmental Engineering and Science Laboratory (EES) of the University of Iowa for processing, distribution and analyses for radionuclides (to be reported elsewhere). Water samples were shipped to the Department of Civil and Environmental Engineering (CEE) of the University of Washington. Upon receipt, the samples were filtered through a 33-mm Millex®-HA syringe filter (Millipore Corporation, Bedford MA) with a 0.45-μm nominal pore size to remove particulate matter. The filtrate was acidified to reach a 1% nitric acid concentration and placed in 15 mL conical polypropylene test tubes. The samples were spiked with internal standards (^{45}Sc, ^{74}Ge, and ^{103}Rh) and stored at 4 °C until analyzed.

2.3. *Solid samples collection*

72 Solid samples were collected from 20 drinking water utilities. Of those, 26 were hydrant flush solids and 46 were pipe specimens (including 34 live pipe extractions and 12 boneyard samples). The hydrant flush samples were obtained during conventional flushing. They provided an opportunity to assess the composition of hydraulically-mobile solids released due to hydraulic disturbances. In contrast with hydrant flush solids, scale from pipe specimens can be operationally considered as hydraulically-inert material. Removal and characterization of scale allow for an assessment of total accumulation of inorganic compounds and contaminants, particularly in cases of adhering scales that are not susceptible to removal by flushing. Solid samples examined in this study are summarized in Table 1.

To obtain pipe specimens, deposit material was carefully removed from the exposed pipe surface. To obtain solids mobilized during hydrant flush events, a net assembly consisting of twin hydrant nets and/or pantyhose was used to retain the particulates. All collected solid material was dried at 103 °C for 24 h and weighed to determine its dry mass. In the case of pipe specimens, a portion of mixed dried sample was crushed using a mortar and pestle, passed through a number 50 sieve (300-μm mesh) and homogenized. The crushed/sieved material was digested (as described below) and analyzed to determine its elemental composition. For selected samples, subsets of both crushed and uncrushed material were used examined using X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) to determine their mineralogy and morphology.

Determination of the elemental composition was possible for 35 of 46 pipe specimens and 23 of 26 of hydrant flush solids. The determination of elemental sulfur and carbon content was carried out for the subsets of 48 and 36 samples, respectively.

2.4. *Analytical procedures*

All digestions of solid samples were performed at the EES using USEPA Method 3050B (Acid Digestion of Sediments, Sludges and Soils) (U.S.EPA method 3050B). For 58 samples that had adequate mass for processing, the fraction of sample mass that was digested by the above procedure ranged from 24 to 96%, with an average ± standard deviation of 78% ± 15% (Supplementary Information Table S1). Aliquots of the digests were sent to the CEE to determine their elemental composition.

Ten of the solid samples were chosen for morphological and surface elemental composition using SEM and energy dispersive spectroscopy (EDS) technique. SEM/EDS measurements were performed with a JEOL-7000F high-resolution SEM instrument (JEOL Corporation, Japan). EDS data were acquired in two modes. The first mode allowed examining the entire surface of the sample, while the second mode