

Practical evaluation of inorganic contaminant presence in a drinking water distribution system after hydraulic disturbance

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ABSTRACT

The accumulation of loose deposits in a drinking water distribution system (DWDS) is a potential risk for water quality. In this work, hydraulic disturbance was simulated in different areas of a large DWDS. The water quality features before and after the disturbance were compared, and the solid deposits captured from different experimental sites were characterized. The results showed that the hydraulic disturbance increased the water turbidity and color, which can be attributed to the re-suspension of iron and manganese oxide particulates. The concentrations of arsenic and lead increased after the disturbance, and a significant positive correlation was observed between the iron and arsenic concentrations. The solid deposits exhibited distinct characteristics in different sites based on the normal hydraulic conditions and pipe material. The main crystalline iron substances were goethite and pyrosiderite. The iron and arsenic contents in the deposits had a positive correlation with their concentrations in the disturbed water. The combination status of manganese, chromium, and lead in the deposits was different from that of arsenic.

Key words | drinking water distribution system, hydraulic disturbance, inorganic contaminants, loose deposits, water discoloration

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INTRODUCTION

Loosely attached deposits on pipe walls in a drinking water distribution system (DWDS) are ubiquitous and can greatly influence the water quality. The most common and appreciable water quality problems in a DWDS usually result from the re-suspension of loose deposits, such as an occasional or intermittent turbidity increase or water discoloration at consumer taps (Husband *et al.* 2008).

The origins of loose deposits are complicated, and they can come from residual particles in raw water, precipitates of residual metallic elements in raw water or from treatment chemicals (e.g., iron, aluminum, and manganese), corrosion products of pipe materials, and biofilms that have detached from pipe walls (Kanakoudis & Tolikas 2002; Kanakoudis 2004; Liu *et al.* 2017). Iron is considered to be one of the most common constituents of loose deposits (Hill *et al.* 2010; Peng *et al.* 2010). In addition to residual iron in treated

water, another main source of iron is iron pipe corrosion product release. Iron-based distribution pipes, such as unlined cast iron pipes and steel pipes, still widely exist in current DWDSs worldwide (McNeill & Edwards 2001; Peng *et al.* 2010). Cast iron pipe walls are usually covered with corrosion scales, and a thin layer of loosely attached iron oxides exists on top of the scales (Sarin *et al.* 2004). Manganese is another element that is frequently found in loose deposits. Even low levels (less than 0.05 mg/L) of residual, dissolved Mn(II) species in finished water can be chemically or microbially oxidized and accumulated in a DWDS (Sly *et al.* 1990; Prasad & Danso-Amoako 2014). In addition, cement-lined iron pipes are popular in the water supply industry and have been reported to have a high potential for releasing aluminum containing particulates as they age (Fujita *et al.* 2014).

Although loose deposits mainly contain fine mineral particles, organic matter is often also present (Gauthier *et al.* 1999; Lehtola *et al.* 2004; Peng *et al.* 2010; Pocas *et al.* 2013). Gauthier *et al.* (1999) investigated loose deposit samples from a DWDS and water storage tanks, and they discovered that the loose deposits contained variable amounts of organic matter (less than 11% as organic carbon). In addition to a turbidity increase and water discoloration, the loose deposits can provide a habitat for microorganisms to avoid disinfectant exposure and damage (LeChevallier & Gibbon 2010). Moreover, the extracellular polymeric substances produced by microorganisms help the loose deposits attach by occluding the release of fine particles and strengthening their adherence to pipe walls (Pocas *et al.* 2013, 2015). Toxic inorganic contaminants, such as lead, nickel, chromium, and antimony, can also accumulate in loose deposits (Hill *et al.* 2010; Peng *et al.* 2012; Yang *et al.* 2012). Lytle *et al.* (2004) discovered that iron oxides in pipe solids have a high tendency to accumulate arsenic, and 10 to 13,650 µg of As/g of solid (as high as 1.37 wt %) was reported. Strontium, which is a recently recognized potential contaminant, can be released from iron corrosion products (Gerke *et al.* 2013).

Under normal conditions (no significant disturbances), the heavy metals accumulate and remain in the deposits due to the deposited compounds adsorbing the heavy metals. However, when the water chemistry parameters, such as the pH, alkalinity, and oxidation/reduction potential, change, desorption of these heavy metals from the deposits into the bulk water can occur (Copeland *et al.* 2007; Hill *et al.* 2010). Most of the fine, light particles will move into the bulk water when a hydraulic disturbance occurs. It is well known that particle-associated bacteria are the main type of bacteria in drinking water, and their effects on microbial safety are attracting more and more attention (Camper *et al.* 1986; Gauthier *et al.* 1999; Vreeburg *et al.* 2008; Winkelmann & Harder 2009; Liu *et al.* 2013a, 2013b). Consequently, the microorganisms and heavy metals attached to the loose deposits could simultaneously enter the water and cause a potential health risk.

Events such as seasonal or daily water demand variations, pipe breaks, and fighting fires can cause sudden increases in the shear strength on the pipe wall and re-suspend deposits (Lehtola *et al.* 2004). Due to the irregular

occurrences and short durations of re-suspension events, it is hard to monitor and evaluate such incidences (Vreeburg & Boxall 2007). Currently, detailed documents and analyses on deposit re-suspension-related cases are relatively rare. Meanwhile, the complexity of loose deposit-related problems and the necessity for control strategies have not received enough attention in the water supply industry (Hill *et al.* 2010).

The objective of this work was to evaluate the effects of re-suspension of deposits on water quality variations in a large DWDS. A hydraulic disturbance was simulated and implemented by opening hydrants. Water quality variations (mainly esthetic parameters and heavy metal element concentrations before and after the hydraulic disturbance) were analyzed and evaluated. Solid deposits were captured from hydrants during the disturbance experiments and were characterized by X-ray diffraction (XRD) and elemental analysis. The relationship between the deposit composition and the water quality after the disturbance was discussed.

MATERIALS AND METHODS

Operation of the hydraulic disturbance and sampling

To simulate a hydraulic disturbance in a DWDS, a high water flow was achieved by fully opening the hydrants installed on the distribution pipes. In total, 21 sites were selected for the disturbance tests, and all the sites were close to community water supply pipes. The pipes in the tested areas were mainly old, unlined cast iron pipes (around 20 years), but cement-lined iron pipes also existed in some places (which was determined based on the small cement mortar blocks observed in the obtained solid samples).

Before opening the hydrants, water samples were collected from a drinking tap near the experimental sites (denoted 'normal tap water'), and pre-flushing was conducted to discharge the stagnant water in the plumbing system. Experimental water samples (denoted 'disturbed water') were collected after opening the hydrants for approximately 10 seconds (to discharge the stagnant water in the short standpipes connecting the distribution pipes and the hydrants).

After collecting the disturbed water samples, the hydrant was shut off, and a tailor-made nylon net assembly was

either mounted on the outlet of the hydrant or connected by a fire-fighting hose. Then, the hydrant was opened again for approximately 5 minutes. Solid deposit particles greater than the pore size of the net were captured as the disturbed water flowed through the net assembly. The pore size of the net in this work was 50 μm . (For nets with smaller sizes, a sufficient flow was hard to achieve.) Figure S1 (available with the online version of this paper) is the working scene picture for the solid deposit sample collection. The flow velocities were not measured, but they were high enough to flush out the loose deposits in pipes, which was evidenced by the substances with a relatively high density (like gravels, cement mortar blocks, and sands, see Figure S2 (available online)) that were caught by the sampling net assembly. However, pipe corrosion scales were scarcely observed in the collected samples, which indicated that the flow velocities were not high enough to remove the pipe corrosion scales. After the net assembly was removed from the hydrant, it was placed in a sealed plastic bag and transported to the laboratory for characterization.

Water quality analysis

The turbidity, residual chlorine, and pH of all the water samples were measured immediately after the field sampling using a turbidimeter (2100Q, Hach, USA), portable spectrophotometer (DR1900, Hach, USA) and pH-meter (FE20 K, Mettler-Telodo, Switzerland), respectively. The alkalinity was measured by titration following standard methods. The sulfate, chloride, and nitrate concentrations were determined by ion chromatography (IC1500, Dionex, USA).

For the metal element analysis, the samples were first digested by adjusting their pH to less than 2.0 with concentrated nitric acid. The calcium, magnesium, and iron concentrations were analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, OPTIMA 2000, Perkin Elmer, USA). The metal elements present in low levels, such as manganese, arsenic, chromium, copper, and lead, were analyzed using inductively coupled plasma mass spectrometry (ICP-MS, NexION 300X, Perkin Elmer, USA).

Characterization of solid deposits

The solid deposit samples were first vacuum-freeze-dried at $-50\text{ }^{\circ}\text{C}$ for 18 hours. The appearances of the dried solids

were not the same, and some typical pictures of the dried solid samples are provided in Figure S2. Some samples contained relatively large gravel and/or cement mortar fragments (Figures S2(a) and S2(b)), which came from the pipe cement lining breaking. (The brownish color of the gravel in Figure S2(a) is due to a thin surface layer of iron oxide.) Most of the solid samples exhibited good homogeneity in their color and particle size although the color varied for different samples (Figures 2(c) and 2(d)). Before the instrumental characterization, the large gravel and cement fragments were removed, and the samples were ground with an agate mortar and pestle under anaerobic conditions. Small gravel, cement mortar fragments, and other non-mineral impurities were sieved out using a 160 μm mesh sieve. X-ray diffraction (XRD, PANalytical X'Pert PRO MPD) was performed to characterize the crystalline matter in the samples. The XRD operation parameters were: Ni_0 -filtered $\text{Cu-K}\alpha$ radiation (λ of 1.5406 Å), 2θ values from 3° – 70° with a 0.02° step size. The crystalline phase was identified using the Jade XRD software, and the crystalline phase composition was determined quantitatively using the contrasted parameters of the intensity method (Chung 1975).

After the aqua regia digestion, the elemental composition of the solid samples was determined using ICP-OES and ICP-MS for regular and trace level metals, respectively.

RESULTS AND DISCUSSION

Esthetic characteristics of disturbed water

Figure 1(a) shows the cumulative percentage profiles of the turbidity for normal tap water and the disturbed water samples from the 21 DWDS sites. Obviously, the disturbed water exhibited a much higher turbidity than normal tap water. (The data are given in Table S1, available with the online version of this paper.) Only one sample out of all the normal tap water samples had a turbidity value greater than 1.0 NTU, which is the regulation limit for turbidity in China. In contrast, for the disturbed water, 16 out of the 21 samples (76%) exceeded the turbidity limit of 1.0 NTU, seven samples (33%) exceeded 10 NTU, four samples (19%) exceeded 30 NTU, and two samples (9%) had turbidities greater than 50 NTU. The appearances of the disturbed water samples with different turbidities

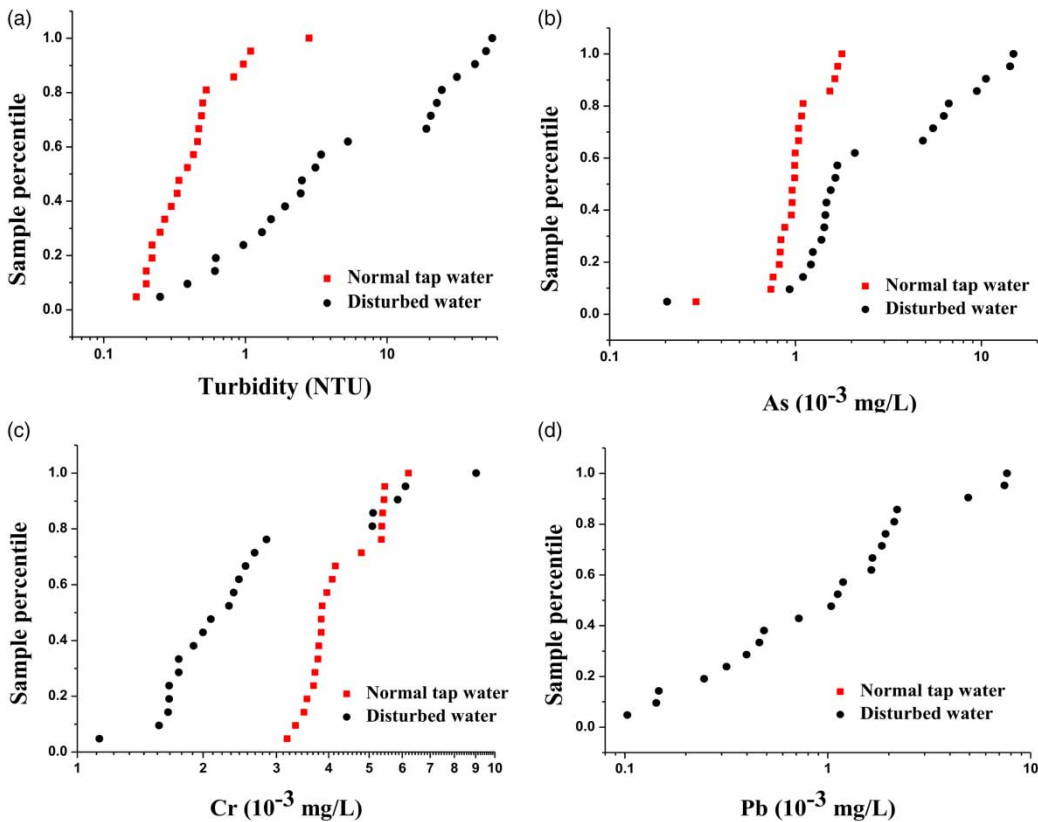


Figure 1 | Cumulative percentile profiles for the turbidity (a), arsenic (b), chromium (c), and lead (d) levels for normal tap water and the disturbed water samples.

are shown in Figure S3 (available online). As the turbidity increased, the yellowish color increased accordingly. When the turbidity was greater than 3.0 NTU, the color was easily perceptible by the naked eye. Therefore, the extent of the hydraulic disturbance caused by opening a hydrant could result in consumer complaints.

To investigate the occurrence scenarios of inorganic contaminants, the metallic elemental composition of the disturbed water and normal tap samples was analyzed. (The data are given in Table S2, available online.) Iron is the most common element that causes color and turbidity in a DWDS because of the large percentage of iron material pipes. The iron concentrations in normal tap water were rather low (all less than 0.03 mg/L), but the iron concentrations in the disturbed water samples were all greater than 0.3 mg/L, which is the regulation limit for iron in China. Iron concentrations up to 18.0 mg/L were detected, and corresponded to the samples with the highest turbidities. The relationship between the iron concentration and the turbidity in the disturbed water samples is presented in

Figure 2(a). Clearly, a positive linear correlation exists between the two parameters, which agrees with the previous finding that iron oxide particulates (mainly from iron pipe corrosion products) are the main constituents of turbidity and color-forming substances (Imran *et al.* 2005).

Another common turbidity and color-forming element is manganese. Even low levels of residual Mn(II) in finished water can accumulate on pipe walls via chemically or microbially influenced oxidation (Sly *et al.* 1990). The manganese concentrations in the disturbed water samples were a magnitude lower than the iron concentrations. Sixteen samples (76%) had manganese concentrations lower than 0.1 mg/L, which is the regulation limit for manganese in China. The manganese concentration of the remaining five samples (24%) was in the range of 0.1–0.2 mg/L. Furthermore, it was found that the relative turbidities of the five samples with the highest manganese concentrations were also high. Figure 2 (b) shows that a linear correlation exists between the turbidity and the manganese concentration. Meanwhile, a good linear correlation also existed between manganese and iron

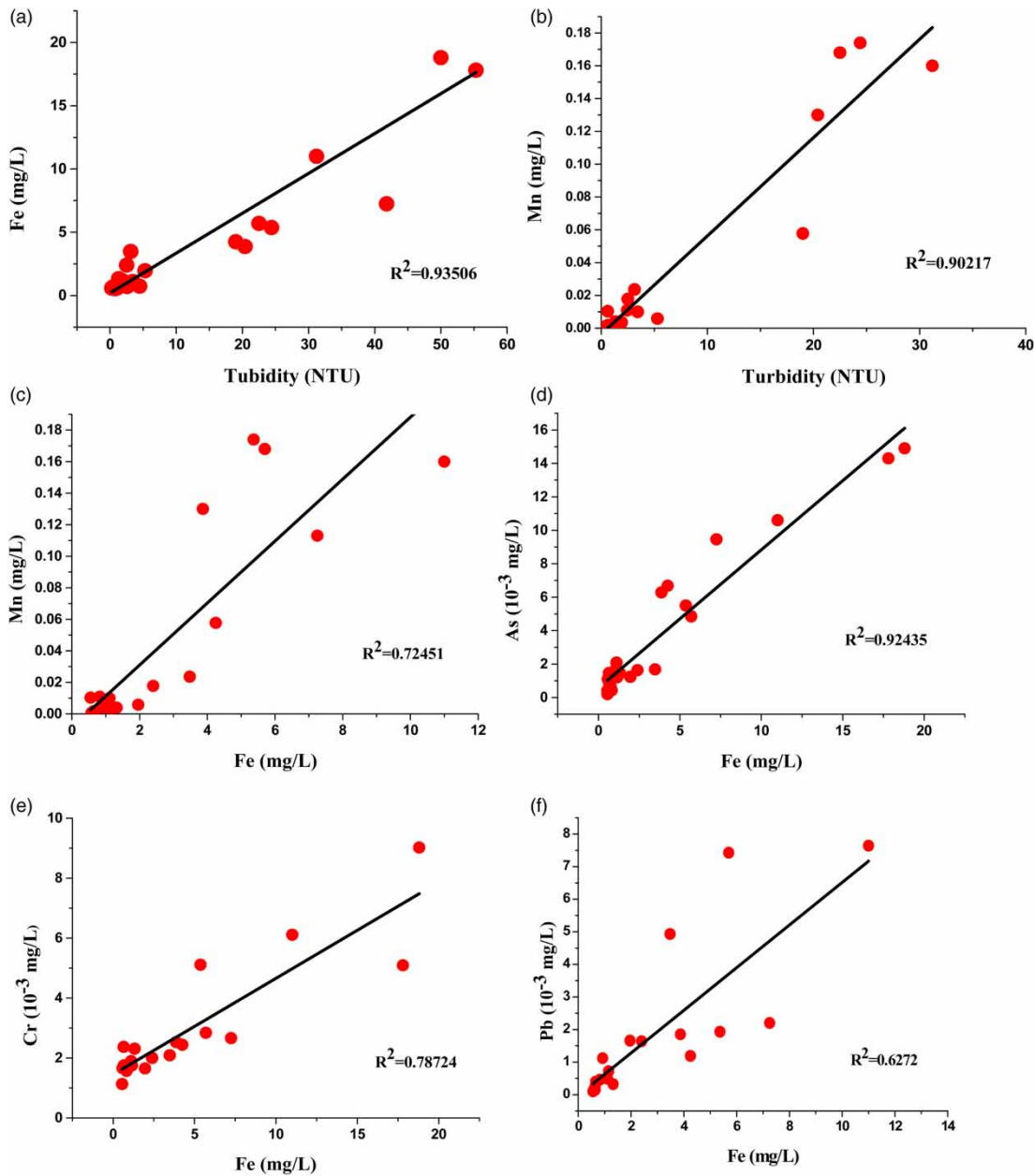


Figure 2 | Correlation analyses for some parameters/elements in disturbed water: (a) turbidity vs. Fe; (b) turbidity vs. Mn; (c) Fe vs. Mn; (d) Fe vs. As; (e) Fe vs. Cr; and (f) Fe vs. Pb.

(Figure 2(c)). These correlations indicate that manganese coexists with iron oxides in the disturbed water. However, the lower correlation coefficient between manganese and iron implies that manganese might also exist with particulate matter other than iron oxides. It should be noted that the manganese concentrations in the normal tap water samples were extremely low. More than half of the normal tap water samples had manganese concentrations lower than

0.001 mg/L, and the maximum detected value was only 0.006 mg/L. The manganese accumulation in the pipe deposits can be attributed to the oxidation of low levels of dissolved Mn (II) species into insoluble manganese oxides (Sly *et al.* 1990; Cerrato *et al.* 2006; Gerke *et al.* 2016) or adsorption of Mn(II) by other pipe solids (Wang *et al.* 2012).

An unusual color-forming element is copper. Copper-related problems are often associated with the use of

copper tubing in households. In this investigation, the maximum copper concentration detected in all the samples (including normal tap water and the disturbed water) was 0.009 mg/L, which is far below the regulation limit of 1.0 mg/L. Thus, copper is not a problem in the DWDS areas of this investigation.

Toxic inorganic contaminants in disturbed water

The regulation limits for iron, manganese, and copper for drinking water are generally based on esthetic water quality control. In addition to these three metals, some regulated toxic metals were also measured, including barium, beryllium, cadmium, nickel, arsenic, chromium, and lead. Among these elements, only arsenic, chromium, and lead were detected in the disturbed water samples, and only arsenic and chromium were detected in both the disturbed and normal tap water samples. (The data are shown in Table S2.) Figures 1(b)–1(d) show the cumulative percentage profiles for the three metals. First, the concentrations of these metals for all the normal tap water samples were far below their respective regulation limits (0.01 mg/L, 0.05 mg/L, and 0.01 mg/L for arsenic, chromium, and lead, respectively). The detected maximum concentrations of arsenic and chromium for the normal tap water samples were 0.002 mg/L and 0.006 mg/L, respectively. For the disturbed water samples, the detected maximum concentrations of arsenic, chromium, and lead were 0.015 mg/L, 0.009 mg/L, and 0.008 mg/L, respectively. Although these levels did not violate the regulation limits for these toxic metals, it should be noted that the arsenic concentrations in the disturbed water were significantly higher than those in normal tap water. The lead in normal tap water was undetectable, but lead was detected in the disturbed water. For chromium, it is interesting that the concentrations in the disturbed water were lower than those in normal tap water. This indicated that the chromium source in normal tap water might come from the household tubing or water taps instead of the re-suspended deposits.

Further analyses determined that the arsenic, chromium, and lead concentrations in disturbed water had positive correlations with the iron concentration (Figures 2(d)–2(f)), which meant that these heavy metals were mainly adsorbed on the particulate iron oxides. The adsorption capability of

iron oxides for trace inorganic contaminants has been reported by other researchers (Lytle *et al.* 2004; Peng *et al.* 2012). In addition, the adsorption of arsenic by iron oxides can be affected by its speciation, i.e., arsenite or arsenate (Raven *et al.* 1998; Bose & Sharma 2002). Therefore, control of corrosion and corrosion product release in a DWDS is helpful to avoid discolored water and to reduce the contents of toxic elements related to health risks.

Crystalline substance composition of the solid deposits

As mentioned in the experimental section, the appearances and amounts of solid deposits collected from the hydrants were obviously different at each site. The factors influencing the amounts and compositions of the captured deposits can include the pipe material type, water quality characteristics, and, more importantly, the hydraulic shearing strength. If the experimental site area has a historically high shearing strength, fine particulate deposits might not be able to accumulate, and only gravel or cement mortar fragments were captured. For these areas, the turbidities of the corresponding disturbed water samples were much lower. In this investigation, there were nine site areas where fine particulates were not obtained, and XRD and elemental characterizations could not be performed. Therefore, only 12 deposit samples were characterized.

After analyzing the XRD patterns of the 12 samples, 14 crystalline substances were identified in total. The XRD pattern of each crystalline substance is provided in Figure S4, and the mass percentage of the content is given in Table S3. (Figure S4 and Table S3 are available online.) Seven main substances were selected for the content comparison (Figure 3), and the selection rules were a high detection frequency (more than three out of 12 samples) and high mass percentage (more than 10% in at least one sample). The remaining crystalline substances were added up and marked as 'rest' in Figure 3. Quartz existed in all 12 samples and at a relatively larger percentage. Quartz can come from the residual particles in treated water or the erosion of the cement linings in iron pipes. Although the quartz content was high in all these samples, it cannot increase the turbidity or be a color-forming material due to its relatively large density and particle size. In contrast, a weak negative correlation was observed between the

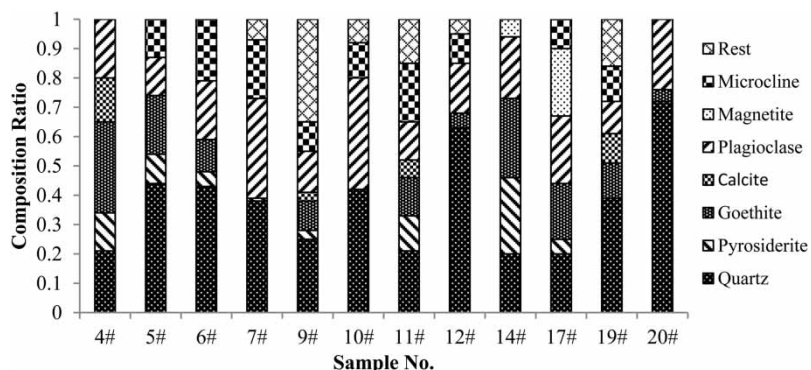


Figure 3 | Compositions of the crystalline substances in the deposit samples.

quartz content in the deposits and the turbidity of the disturbed water (Figure S5, available online). Plagioclase was another substance that was detected in all 12 samples. Plagioclase and microcline are two types of aluminum silicates, and they can either come from residual mineral particles that originate in the source waters or be formed by the reaction between residual coagulant aluminum and silica in natural water in a DWDS (Snoeyink *et al.* 2003). Calcite was only detected in three samples at a relatively lower content, and it is mainly derived from the precipitation of calcium in bulk water under some circumstances. The main iron oxides that were detected were goethite and pyrosiderite. Magnetite was only found in two samples (14# and 17#). It should be noted that in three samples (10#, 19#, 20#), no iron oxides were detected, and sample 7# only had 1% of pyrosiderite. The remaining crystalline substances were primarily non-iron oxides.

Elemental compositions of the deposit samples

The inorganic element contents in the deposit samples are listed in Table S4 (available online). The unit 'mg/kg' indicates '(mg element)/(kg dry solids)'. Iron had the highest content with a range of 31,700 to 94,000 mg/kg or mass percentage of 3.17% to 9.40%. Since the main iron oxides detected by XRD were goethite and pyrosiderite, the calculated iron oxide (in the form of FeOOH) content range was 5.03% to 14.94%. It was also determined that the samples without crystalline iron oxides have a certain iron element content (31,700 mg/kg, 48,800 mg/kg, and 53,500 mg/kg for samples 10#, 19#, and 20#, respectively). This indicated that iron can exist in an amorphous form in the deposit samples.

Similar to the element contents in the disturbed water, the manganese content in the deposits was a magnitude lower than that of iron. Heavy metals, such as arsenic, chromium, copper, and lead, were also detected in the deposits. A significant positive correlation between arsenic and iron was observed (Figure 4). However, a significant relationship was not observed for manganese, lead, and copper with iron. Extremely low levels of cadmium were detected in half of the deposit samples, but it was not detected in all the disturbed water samples.

Since the particulates in the disturbed water were from the re-suspension of the deposits, the relationships of the iron, arsenic, chromium, and lead contents in the deposits to their concentrations in the corresponding disturbed water were explored. Positive correlations were observed for the iron and arsenic contents in the deposit samples and their concentrations in the corresponding disturbed

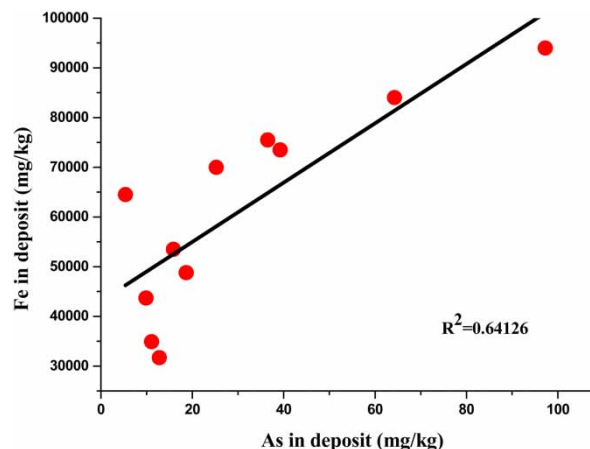


Figure 4 | Relationship between arsenic and iron in the solid deposits.

water samples (Figure 5). However, significant relationships were not observed for the manganese, chromium, and lead contents in the deposits and their concentrations in the disturbed water. This result confirmed that the iron oxides can be re-suspended in a more homogenous manner, resulting in an increase in the turbidity and color formation, and arsenic was closely associated with the iron oxides. Manganese and other heavy metals may exist in different forms or be combined with particulates other than iron oxides. As mentioned previously, the particle size, density, and constituents of the deposits were highly heterogeneous, and thus, the particulates in the disturbed water were only those fractions with a finer size and lighter density in the deposits. Further research is needed to illustrate the combination status and re-suspension behaviors of such metals.

Further discussions and implications

The most important function of a DWDS is to transport treated water from the treatment plant to consumers. Unfortunately, during the transportation process, the water quality will unavoidably change to some extent. Pursuing good chemical and biological stabilities for the treated water is critical for maintaining the water quality in a DWDS. Water quality with a good chemical stability should have a low corrosivity for pipe materials and a low residual turbidity and/or color-forming metals such as iron and manganese. However, it is very difficult to establish well accepted guidelines for a chemical stability evaluation due to the current lack of understanding on this issue.

One important factor that complicates water quality problems in a DWDS is the hydraulic conditions, which can vary greatly with time and space, particularly for a large-scale DWDS in metropolitan cities. Based on the current knowledge and technology, biofilm growth and loose deposit accumulation in a DWDS cannot be avoided. Unstable hydraulic conditions are the main cause of loose deposit re-suspension and the biofilm peeling off.

In this investigation, the effect of a hydraulic disturbance on the water quality deterioration was demonstrated to be significantly different based on the specific conditions in each DWDS area. Heavily turbid and discolored water was observed in areas with more iron corrosion product accumulation. Unlined cast iron pipes are the primary source of the iron oxides in the deposits. The iron corrosion product release in a DWDS can be affected by the water quality characteristics, such as the chloride and sulfate content, alkalinity, and Larson ratio (Imran et al. 2005; Shi & Taylor 2007). In the city used for this investigation, different source waters and different treatment trains are used, and the water quality parameters of the treated water vary greatly (Table S1). Generally, the corrosivity of all the waters for iron pipes was relatively low because the alkalinity concentrations were all relatively high (greater than 130 mg/L as CaCO₃), and the Larson ratio, a common index for iron corrosivity evaluation, was relatively low (less than 0.6). No significant relationship was found between the disturbed water turbidity and the water corrosivity. Thus, the heavily turbid and discolored water could not be attributed to the corrosivity of the transported water. In contrast, the captured solids mainly

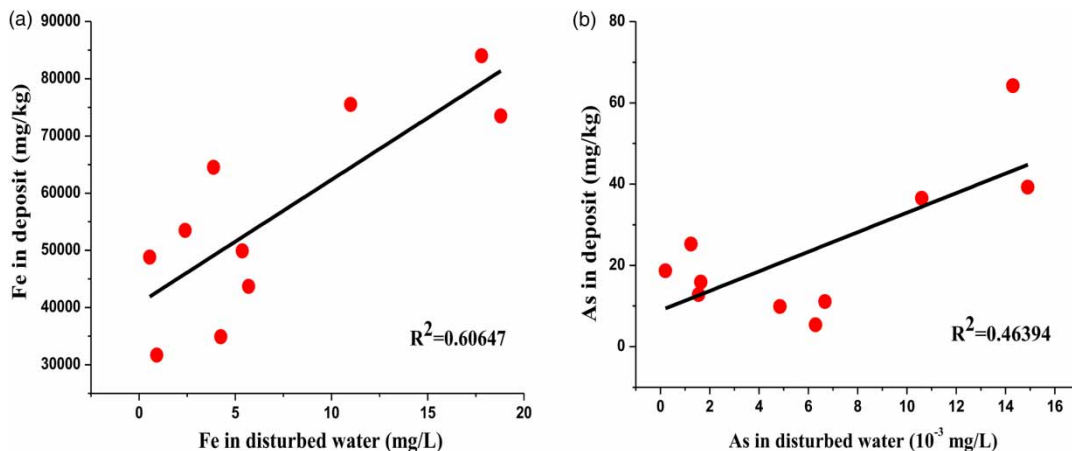


Figure 5 | Relationships between the contents in the deposits and the concentrations in the disturbed water for iron (a) and arsenic (b).

consisted of gravel and/or cement mortar fragments, and iron oxides and other fine particulates were hardly captured, which corresponded to lower disturbed water turbidity. This indicated that the normal hydraulic shear strength in those areas was high enough to prohibit the accumulation of fine particulates.

This investigation also revealed that toxic inorganic contaminants can accumulate in loose deposits, and the re-suspension of the loose deposits might increase the risk of consumer exposure to these contaminants. However, the treated water met the required regulations. The combination of toxic contaminants with loose deposits could vary based on the elemental speciation characteristics, water chemistry, and composition of the deposits.

Some implications for the practical operation of a DWDS can be drawn from this investigation. First, utilities should evaluate the possible deposit accumulation status in their DWDS. The evaluation could be conducted via hydraulic modeling to identify locations with undesirable flow conditions. Second, a detailed geographical information system of the DWDS should be established to help locate pipes with a high risk, such as old, unlined cast iron pipes, and a replacement strategy for the pipes should be created. Third, routine pipe cleaning measures should be planned based on a systematic evaluation of the DWDS status. Planned cleaning could efficiently reduce the water contamination risk during the transportation process.

CONCLUSIONS

Hydraulic disturbance simulation experiments were conducted in different areas of a large city DWDS. The main findings obtained from this investigation are as follows:

- Hydraulic disturbances can significantly increase the water turbidity and apparent color, and iron oxide particulates are the main turbidity and color-forming substances.
- The manganese, arsenic, and lead concentrations in disturbed water were higher than those in normal tap water, and a significant positive correlation existed between the arsenic and iron concentrations.
- Goethite and pyrosiderite were the main crystalline iron oxides in the deposits, and quartz, plagioclase, and microcline were the main non-iron particulates.

- The iron and arsenic concentrations in the deposits had a significant positive correlation with those in the disturbed water, but no such correlations were observed for manganese, chromium, and lead.

Planned, routine pipe cleaning with high flow flushing could effectively reduce the risk of water quality deterioration in DWDSs. The replacement of old, unlined iron pipes is necessary to maintain the water quality in the distribution process.

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