

Available online at www.sciencedirect.com

ScienceDirect

www.journals.elsevier.com/journal-of-environmental-sciencesJOURNAL OF
ENVIRONMENTAL
SCIENCESwww.jesc.ac.cn

A comprehensive study of treatment of arsenic in water combining oxidation, coagulation, and filtration

Terracciano Amalia, Ge Jie, Meng Xiaoguang*

Center for Environmental Engineering, Stevens Institute of Technology, Hoboken, NJ 07030, USA

Arsenic is one of the most common inorganic contaminants in groundwater worldwide, mainly due to the release of naturally occurring arsenic from aquifer sediments (Amini et al., 2008; Li and Cai, 2015; Rahman et al., 2015). Naturally occurring arsenic exists predominantly in arsenate and arsenite species in groundwater. Arsenite is more toxic and more difficult to remove from water than arsenate (Hughes et al., 2011; Pena et al., 2005; Bang et al., 2005; Du et al., 2015).

Development of effective technologies for removal of arsenic from water by coagulation and adsorption requires in-depth knowledge of arsenic chemistry, surface interactions of arsenic species and coexisting solutes with the adsorbents, and redox transformation of arsenic species. Recently, Cui et al. (2015) conducted a comprehensive study on the treatment of arsenic by ferric salts. The research spanned from field evaluation of a household coagulation filtration system to spectroscopic analysis of chemical bonding structures of adsorbed arsenic species with surface functional groups. Moreover, the leachability of arsenic in the solid residue generated during the water treatment was tested for determination of the proper disposal options of the waste material.

Several technologies have been used for removal of arsenic from water, including coagulation or coprecipitation with ferric salts, adsorption with metal oxides and hydroxides, treatment with zero valent iron, filtration with anion exchange resins, and membrane filtration (Banerjee et al., 2008; Jing et al., 2009; Bordoloi et al., 2013; Xia et al., 2014). Coagulation processes are typically used in large drinking water treatment plants and industrial wastewater treatment systems. Granular metal hydroxides and oxides, such as iron, titanium, aluminum, and zirconium, can be easily used in small water treatment systems and water filters for households, although the cost of granular adsorbents is higher than that of ferric salts.

Coagulation with ferric salts, such as ferric sulfate and chloride, is one of the most cost-effective processes for arsenic treatment, because the chemicals are readily available

at low cost and the fresh ferric hydroxide precipitate formed in the water has very high surface area and high adsorption capacity for arsenic (Lakshmanan et al., 2010). After the coagulation treatment, the fine precipitate in the water has to be removed from the water by sedimentation and sand filtration processes. The two-bucket system (Fig. 1) developed by Cui et al. (2015) integrated oxidation of arsenite to arsenate by calcium hypochlorite, coagulation to convert soluble arsenate to adsorbed arsenate forms on fresh ferric hydroxide, sand filtration removal of the precipitate, and activated carbon filtration removal of residual chlorine. The field-test results demonstrated that the system is an effective and low-cost method for households in rural areas to prepare clean drinking water.

The removal of the arsenic species by metal oxides and hydroxides, in fresh precipitate or granular forms, is affected by water pH and the chemical composition of water. Coexisting anions, such as phosphate, silicate, and bicarbonate, can compete with the arsenic species for adsorption sites on the metal oxides and hydroxides (Meng et al., 2000; Wang et al., 2014), while calcium cations improve the adsorption of arsenic (Wang and Tsang, 2013). Cui et al. (2015) investigated the effect of 12 types of cations and anions in well water on the removal of arsenic by ferric coagulation and determined a significant negative correlation between arsenic removal efficiency and silicate concentration. They also observed that arsenic was more difficult to remove in three of the ten well water samples tested in the study, even though the arsenic and silicate concentrations were similar in the water.

Adsorption of arsenic species by metal oxides and hydroxides occurs through interactions of the arsenic species with the hydroxyl groups on the solid surfaces (Waychunas et al., 1993). Currently, the most powerful technique used for *in situ* measurement of the bonding structures between the adsorbates and the surfaces is X-ray absorption spectroscopy. Since extended X-ray absorption fine structure (EXAFS) was first

* Corresponding author. E-mail: Xiaoguang.Meng@stevens.edu (Xiaoguang Meng).

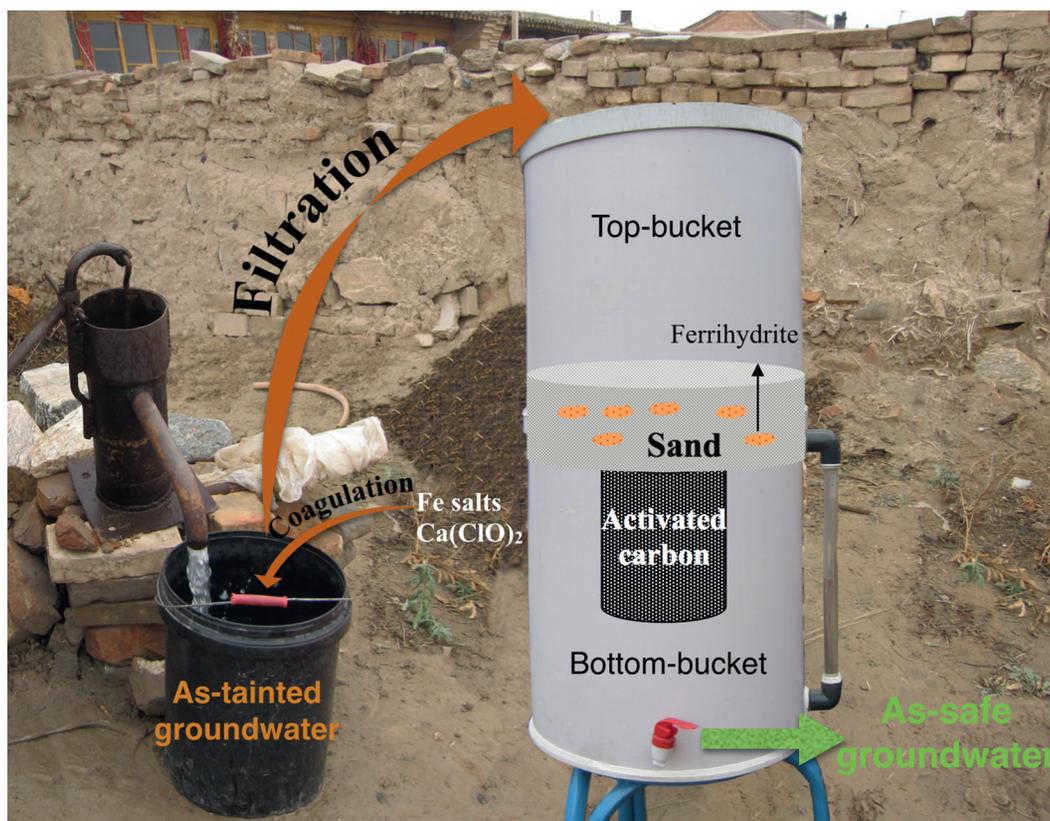


Fig. 1 – Removal of arsenic from groundwater using a two-bucket system (Cui et al., 2015). Calcium hypochlorite and ferric salts are added to arsenic-containing water. Calcium hypochlorite oxidizes arsenite to arsenate, and ferric hydroxide coagulation adsorbs arsenate. Sand filtration removes the precipitate, and activated carbon filtration removes residual chlorine. Figure provided by Dr. Chuanyong Jing, State Key Laboratory of Environmental Chemistry and Ecotoxicology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing.

used to determine structural information (inner- and outer-sphere complexes, bond distances, and coordination numbers) of adsorbed selenite and selenate at an α -FeOOH (goethite)-water interface (Hayes et al., 1987), the technique has been widely applied in adsorption studies (Waychunas et al., 1993; Manning et al., 2002; Ona-Nguema et al., 2005; Jing et al., 2005; Pena et al., 2006). Cui et al. (2015) performed EXAFS and X-ray absorption near-edge structure (XANES) analysis of the solid residue generated in the ferric coagulation treatment of well water and determined that arsenic formed a bidentate binuclear complex on ferrihydrite. The high adsorption capacity of the ferric hydroxide precipitate for arsenic and the low mobility of arsenic in the solid residue were attributed to the strong chemical bonds between the adsorbed arsenic and the surface sites.

Cui et al. (2015) successfully combined the development of an arsenic treatment technique with fundamental research. The study also showed the challenges and needs in the research and development of arsenic treatment processes. For instance, the existing techniques are not effective for the removal of arsenite without oxidizing it to arsenate. At the present, we cannot accurately predict the removal of arsenic in different waters by ferric coagulation and by filtration with granular metal oxides and hydroxides, because of the complicated effect of water chemistry and lack of knowledge about interactions of the coexisting solutes with the solids and the interactions among

adsorbed species at the solid-water interface. The current research is an important step toward filling this knowledge gap.

REFERENCES

- Amini, M., Abbaspour, K.C., Berg, M., Winkel, L., Hug, S.J., Hoehn, E., Yang, H., Johnson, C.A., 2008. Statistical modeling of global geogenic arsenic contamination in groundwater. *Environ. Sci. Technol.* 42 (10), 3669–3675.
- Banerjee, K., Amy, G.L., Prevost, M., Nour, S., Jekel, M., Gallagher, P.M., et al., 2008. Kinetic and thermodynamic aspects of adsorption of arsenic onto granular ferric hydroxide (GFH). *Water Res.* 42 (13), 3371–3378.
- Bang, S.B., Korfiatis, G.P., Meng, X.G., 2005. Removal of arsenic from water by zero-valent iron. *J. Hazard. Mater.* 121, 61–67.
- Bordoloi, S., Nath, S.K., Gogoi, S., Dutta, R.K., 2013. Arsenic and iron removal from groundwater by oxidation-coagulation at optimized pH: laboratory and field studies. *J. Hazard. Mater.* 260, 618–626.
- Cui, J.L., Jing, C.Y., Che, D.S., Zhang, J.F., Duan, S.X., 2015. Groundwater arsenic removal by coagulation using ferric(III) sulfate and polyferric sulfate: a comparative and mechanistic study. *J. Environ. Sci.* 32, 42–53.
- Du, M., Wei, D.B., Tan, Z.W., Lin, A.W., Du, Y.G., 2015. The potential risk assessment for different arsenic species in aquatic environment. *J. Environ. Sci.* 27 (8), 1–8.
- Hayes, K.F., Roe, A.L., Brown Jr., G.E., Hodgson, K.O., Leckie, J.O., Parks, G.A., 1987. *In situ* X-ray absorption study of surface

- complexes: selenium oxyanions on α -FeOOH. *Science* 238, 783–786.
- Hughes, M.F., Beck, B.D., Chen, Y., Lewis, A.S., Thomas, D.J., 2011. Arsenic exposure and toxicology: a historical perspective. *Toxicol. Sci.* 123 (2), 305–332.
- Jing, C.Y., Meng, X.G., Liu, S.Q., Baidas, S., Patraju, R., Christodoulatos, C., et al., 2005. Surface complexation of organic arsenic on nanocrystalline titanium oxide. *J. Colloid Interface Sci.* 290, 14–21.
- Jing, C.Y., Meng, X.M., Calvache, E., Jiang, G.B., 2009. Remediation of organic and inorganic arsenic contaminated groundwater using a nanocrystalline TiO₂-based adsorbent. *Environ. Pollut.* 157, 2514–2519.
- Lakshmanan, D., Clifford, D.A., Samanta, G., 2010. Comparative study of arsenic removal by iron using electrocoagulation and chemical coagulation. *Water Res.* 44 (19), 5641–5652.
- Li, Y., Cai, Y., 2015. Mobility of toxic metals in sediments: assessing methods and controlling factors. *J. Environ. Sci.* 31, 203–205.
- Manning, B.A., Hunt, M.L., Amrhein, C., Yarmoff, J.A., 2002. Arsenic(III) and arsenic(V) reactions with zero valent iron corrosion products. *Environ. Sci. Technol.* 36, 5455–5461.
- Meng, X.G., Bang, S.B., Korfiatis, G.P., 2000. Effect of silicate, sulfate and carbonate on arsenic removal by ferric hydroxide. *Water Res.* 34 (4), 1255–1261.
- Ona-Nguema, G., Morin, G., Juillot, F., Calas, G., Brown Jr., G.E., 2005. EXAFS analysis of arsenite adsorption onto two-line ferrihydrite, hematite, goethite, and lepidocrocite. *Environ. Sci. Technol.* 39, 9147–9155.
- Pena, M.E., Korfiatis, G.P., Patel, M., Lippincott, L., Meng, X.G., 2005. Adsorption of As(V) and As(III) by nanocrystalline titanium dioxide. *Water Res.* 39, 2327–2337.
- Pena, M., Meng, X.G., Korfiatis, G.P., Jing, C.Y., 2006. Adsorption mechanism of arsenic on nanocrystalline titanium dioxide. *Environ. Sci. Technol.* 40, 1257–1262.
- Rahman, M.M., Dong, Z., Naidu, R., 2015. Concentrations of arsenic and other elements in groundwater of Bangladesh and West Bengal, India: potential cancer risk. *Chemosphere* 139, 54–64.
- Wang, Y., Tsang, C.W., 2013. Effects of solution chemistry on arsenic(V) removal by low-cost adsorbents. *J. Environ. Sci.* 25 (11), 2291–2298.
- Wang, Z.F., Shi, M., Li, J.H., Zheng, Z., 2014. Influence of moderate pre-oxidation treatment on the physical, chemical and phosphate adsorption properties of iron-containing activated carbon. *J. Environ. Sci.* 26 (3), 519–528.
- Waychunas, G.A., Rea, B.A., Fuller, C.C., Davis, J.A., 1993. Surface chemistry of ferrihydrite. EXAFS studies of the geometry of coprecipitated and adsorbed arsenate. *Geochim. Cosmochim. Acta* 57 (10), 2251–2269.
- Xia, S.Q., Shen, S., Xu, X.Y., Liang, J., Zhou, L.J., 2014. Arsenic removal from groundwater by acclimated sludge under autohydrogenotrophic conditions. *J. Environ. Sci.* 26 (2), 248–255.