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Editorial

Seven 21st century challenges of arsenic-fluoride contamination and remediation

ABSTRACT

Arsenic (As) and Fluoride (F) are two commonly occurring geogenic contaminants in groundwater environment, causing a range of carcinogenic and non-carcinogenic adverse health effects worldwide. Several studies have been conducted in past and many are ongoing to address As and F contamination issue of natural water. This special issue is conferring in recent times one of the emerging fields of science regarding co-occurrences of multi-contaminants within a given system and associated health risks. This special issue is divided into three sections. Section I deals with the occurrence and co-existence of As, F, and trace elements (TE) in the environment. As and F occurrence [including trace elements (TEs)] in groundwater at a global scale (example: India, Sri Lanka, Bangladesh, Ghana, and Iran, etc.) has been highlighted (Section I). The geological, and anthropogenic factors affecting As and F contamination have been observed. The state-of-art, removal techniques for As and F have been discussed. Section II and Section III incorporate all the advanced removal methods for As and F, respectively. Arsenic and F removal comprises assessing natural remediation potential (phytoremediation) including different advanced absorbents. The new findings published here, bring together a wide range of new insights on As and F behavior in the groundwater environment.

1. Introduction

The co-occurrence of multiple-contaminants in drinking water and associated health risks is a concern. Organic and inorganic forms of arsenic (As) and fluoride [F^-: Fluoride is the simplest fluorine anion) are widely present in the environment. Living organisms can be exposed to inorganic As and F through food and water. Arsenic and/or F occurrence in groundwater (GW) and surface water (SW) is principally caused by geogenic processes associated with the dissolution of As and/or F-containing minerals present in rocks and soils (Fig. 1) (Maity et al, 2011a, 2011b, 2011c, 2017). The deposition of industrial airborne pollutants, ash (e.g. fly ash), and application of fertilizers (agricultural sector) are the primary anthropogenic sources of As and F (Bhattacharya et al., 1997, 2002, 2004, 2006; 2007; Brindha and Elango, 2011; Bundschuh et al., 2004, 2011; Coomar et al., 2019; Kimambo et al., 2019; Litter et al., 2019a, 2019b; Aullon Alcaine et al., 2020; Jacks et al., 2005; Jha et al., 2011; Kumar et al., 2017; Mukherjee et al., 2014, 2008; Nriagu et al., 2007; Vithanage and Bhattacharya, 2015). The As and F contamination in groundwater is a worldwide phenomenon (Ali et al., 2019; Brindha et al., 2011; Jha et al., 2011; Kumar et al., 2017; Lacson et al., 2021). Worldwide, > 300 million people are currently dependent on As and/or F contaminated groundwater for drinking and irrigation, which are the main source of toxicity to human health (Kumar et al., 2020; Patel et al., 2019; Singh et al., 2020a). There are several toxic effects of As in biotic health, such as arsenicosis, malignancies, cancers in the bladder, lung, liver, breast, and kidney, whereas fluorosis, dental caries is caused by F. The guidelines for As and F in drinking water are set at 10 μg/L and 0.5–1.5 mg/L, respectively (Kapaj et al., 2006; Maity et al., 2012; WHO, 2017; WHO, 2017). Fluoride also enters the human body by dental products, mouthwash and toothpaste, F supplements (tablets, chewing gums, etc.), and F gel. The principal route of human exposure to As and F is through water, soil, food; thus, those (As and F) are necessary to prevent by the treatment process.

Environmental protection agencies cover issues of As and F contamination, health effects, limits, which are cohesive for all the nations (EPA, 2002; WHO, 2017; Singh et al., 2020b; UNICEEF, 2018). The environmental protection institutions provide reports based on research and development (R&D) for the benefits as well as the protection of the ecological system. UNICEF published the 'Arsenic Primer' for guidance on arsenic monitoring, assessment, and mitigation (https://www.unicef.org/wash/files/UNICEF_WHO_Arsenic_Primer.pdf). This covers the changes associated with the Sustainable Development Goals (SDG), the framework for safe drinking water, and the experience over the last decade in the implementation of As mitigation programs (UNICEF, 2018; Bundschuh et al., 2017). In a potential short-term strategy to minimize the As-exposure, the focus must be given on rainwater harvesting and storage, cooking with As-safe water, and application of As removal technologies (Halder et al., 2014; Ahmad et al., 2017; Maity et al., 2020). For the long-term As mitigation solution, the emphasis must be given to the information related to regional conditions, habits and practices of regional communities, and management practices of water supply for agriculture, food production, and public health (Hossain et al., 2014, 2015, 2017; UNICEF, 2018). In particular, SDG 6.1 of UNICEF and UN, the major focus is given to the drinking water safety to achieve 'by 2030, achieve universal and equitable access to safe and affordable drinking water for all'. UNESCO works on As contaminated regions in South Asia including Afghanistan, Bangladesh, Bhutan, Maldives, Nepal, Pakistan, and Sri Lanka (https://en.unesco.org/sites/default/files/usr15_south_asia.pdf). UNESCO keeps an agenda for Sustainable Development Program (SDP) on the groundwater As within 2030. The World Health Organization (WHO) set the drinking water guideline of 10 μg/L and 1.5 mg/L for As and F, respectively by working

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on exposure sources, health effects, the problem magnitude, and prevention/control (WHO, 2017). EPA set the safe and allowable As limit in drinking water as 10 μg/L (https://www.epa.gov/dwreginfo/drinking-water-arsenic-rule-history#Review). The Netherlands and Denmark have focused on removing As to a level that is much lower than 10 μg/L (Ahmad et al., 2020; Ahmad and Bhattacharya, 2019). Thus, monitoring, mitigation, and planning are essential for SDP for As and F in the environment (Singh et al., 2020b).

This special issue focuses on the As and F occurrence in the environment including the co-existence of multi-contaminants within a given system, and their associated health impacts, as well as their removal technologies, to achieve As and F free water. Arsenic and F exist together within a single aquifer system. The occurrence of As and F including TEs in groundwater systems at a global scale is highlighted in this special issue. The state-of-art, As and F removal methods are included. The special issue is divided into three different sections where the global occurrence of As, F, and TEs in the environment and their co-existence are described in Section I. Section II and Section III cover all advanced and sustainable (green technology and nanotechnology) removal methods which may be implemented for As and F remediation from water.

2. Section I: Global arsenic, fluoride, and TEs occurrence

Elevated F-concentration in the drinking water supply in many regions of the world has caused the widespread prevalence of dental and skeletal fluorosis. Global F contamination of water in different geographic regions was reviewed (Kimambo et al., 2019). Fluorosis still represents a serious and widespread health problem particularly in rural communities, which depends on untreated F-rich water supplies (Kimambo et al., 2019). The defluoridation methods need to be developed for fluoride removal from potable water in the areas with no other alternative sources of water is available. Dongzagla et al. (2019) examined the F occurrence in groundwater of Jirapa and Kassena-Nankana municipalities of Ghana. Fluoride concentration in groundwater sources was in the range of 0.6–2 mg/L, whereas high F levels exist in the upper east region of Ghana. The F was >1.5 mg/L in 1.4% of boreholes in the study area, where 1.5% of the population is exposed to high F in drinking water. Badeenezhad et al. (2020) reported the affecting groundwater quality factors in Shiraz (Iran), which was used for drinking water. The hydro-chemical properties of groundwater in this area revealed that natural processes have limited influence, and the elevated F in the groundwater in Shiraz are mainly caused by anthropogenic activities. High F concentrations (0.6–18.5 mg/L) in the aquifers related to Seonath River, northern tehsils of Chhuikhadan and Khairagarh in Rajnandgaon, Chhattisgarh, India were investigated by Yadav et al. (2020). The seasonal variations displayed the F dilution process during monsoon. Fluoride concentration in GW increases mainly due to the bedrock (rhyolites) weathering and agricultural practices. An average prevalence rate of 25% skeletal fluorosis in cattle was reported in Rajnandgaon. Kumar et al. (2019) draw attention to groundwater quality and trace metals (TEs) potential sources in Saharanpur, Uttar Pradesh (India). Enrichment of Fe, Al, As, B, Cu, Mn, Pb, and Cd, where pollution index exceeded critical value at few locations due to vicinity of industrial setup was recorded. However, Fe, Mn, and Pb were associated with the mixed origin of geogenic and anthropogenic activities. B and Cu were controlled by anthropogenic activities while As was derived from the geogenic sources. Co-occurrence of F and As in groundwater of the Dharmanagar region, North Tripura (India), and associated health risk were assessed by Bhattacharya et al. (2020). A positive correlation (r = 0.6) between groundwater F (<0.005–4.8 mg/L) and As (<0.003–0.044 mg/L) was reported. The cumulative estimated daily intake (EDI) in the studied population was 0.07–0.1 and 0.13–0.18 mg/kg-day for Central Tendency Exposure (CTE) and Reasonable Maximum Exposure (RME) scenarios, respectively [children are at high risk due to 0.1 mg/kg-day EDI (fluoride)]. Consequently, the groundwater of the Dharmanagar region is alarming, and thus for reducing threats to human health, the government should immediately take necessary action to supply safe drinking water for the residents. Occurrence and identification of As sources along with Fe, Mn, Ba, Zn, and Al were reported in the groundwater which was used for drinking purposes in the Rangpur district, Bangladesh (Islam et al., 2019). Fe, Mn, and Ba concentrations exceeded the drinking water permissible limits set by the WHO, and the DoE (Department of Environment, the Environment Conservation Rules, Dhaka, Bangladesh). Fe, Mn, Ba, and As originated from geogenic sources, while Zn and Al came from anthropogenic activities in this region. The geogenic contribution is much higher than anthropogenic inputs for the reported elevated trace element concentrations. Carcinogenic risk (CR) of As in both adults and children population is a positive agreement with the potential health risk. Chandrajith et al. (2020) present the geogenic As and F contamination in Sri Lankan groundwater known since the past 30 years. This study shows that the numerous dry regions (semi-arid conditions) of Sri Lanka are affected by excessive quantities of F in the groundwater (due to low precipitation and high evaporation), where dental fluorosis, skeletal fluorosis, and chronic kidney disease are prevalent. In Sri Lanka, over 80% of the population

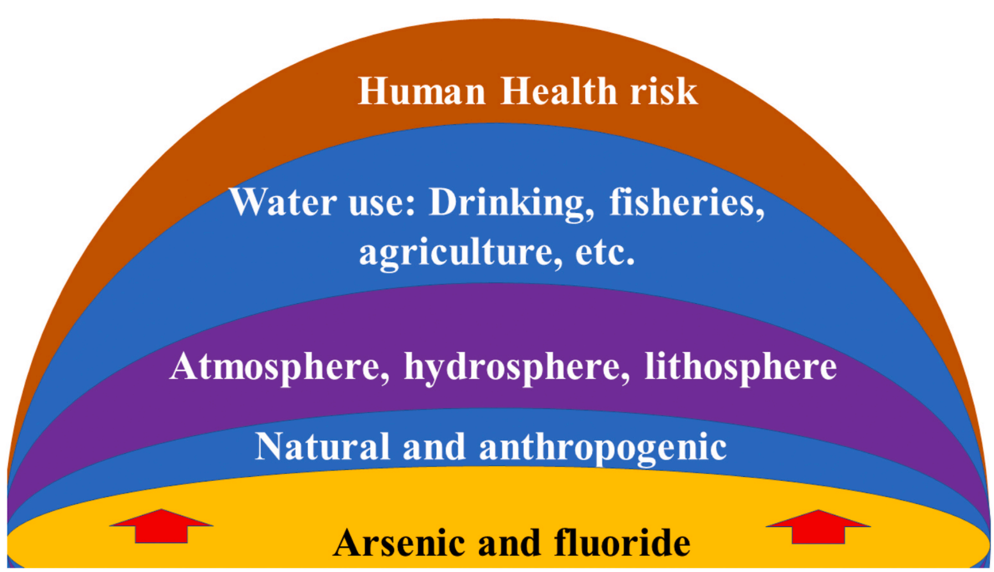

Fig. 1. Geogenic arsenic and fluoride mobilization pathways by natural and anthropogenic processes and human health rick.

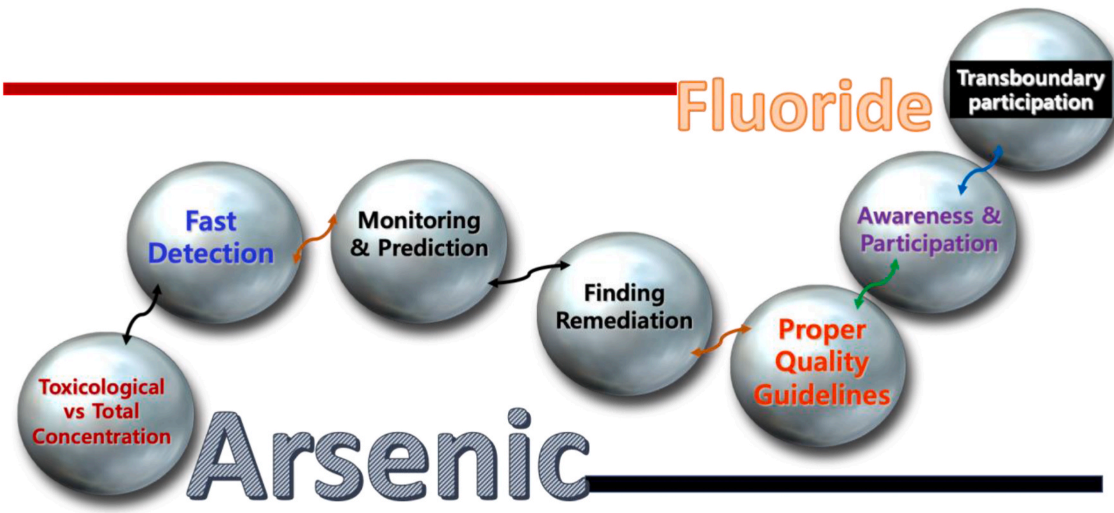

Fig. 2. Seven samurai in the field of arsenic and fluoride contamination in the groundwater that are going to remain in the focus in the 21st century of the humankind.

uses groundwater for drinking purposes, where >50% of wells in the dry zone regions have high F (>1.0 mg/L) levels. Furthermore, the F content was higher in deep wells versus shallow wells. However, the As in groundwater is not yet considered as a serious issue in Sri Lanka, particularly in aquifers in the metamorphic terrain (0.06–1.9 μg/L), however, the As levels were recorded higher (0.10–66 μg/L) in sedimentary terrains.

3. *Section II: Advanced arsenic removal methods*

Arsenic contamination of drinking water, soil, and related to As-rich food consumption has been a major threat to human health in the last decade. Thus, As removal from water and soil is a major issue for environmental safety and human health. Precipitation/co-precipitation, membrane filtration, adsorption, ion exchange, and permeable reactive barriers were most commonly used for As removal and also adopted by United States Environmental Protection Agency (USEPA) (EPA, 2002; Maity et al., 2020). Arsenic removal mainly depends on As-concentration, speciation, temperature water hardness, interfering chemical species, contaminated water volume, and sustainability. Iron oxide nanoparticles have been developed for As removal from water (Maity et al., 2020; Hao et al., 2018; Habuda-Stanić and Nujić, 2015). Gonzalez et al., 2019; Mukherjee et al., 2020; Raval et al., 2020; Raval and Kumar, 2021; Shim et al., 2019 successfully remove As(V) using nanofiltration (NF) membrane from water at 25–50°C. As(V) removal enhanced with the rise in temperature. At low-pressure (down to 1.5 bar), the NF can be applied as an alternative for rural communities. A granular activated carbon (GAC) column (carbon treated with 5% sodium hydroxide solution for 3 h and then washed with distilled water, and dried at 110 °C for 2.5 h) was successfully used for 96% As removal (initial As(III) concentration = 0.5 mg/L) by photo-oxidation process (Salehi et al., 2020). This process is based on UV-persulfate at an optimum pH 3 and persulfate concentration of 14 mM/L, where the UV-activated persulfates can significantly increase the As oxidation efficiency (Salehi et al., 2020). FeS was synthesized and used within the natural sand packed porous media *in situ* and immobilized the As(III) (>80% up to 17 PVs) in water, which can be applicable in aqueous As mitigation (Tiwari et al., 2020).

4. *Section III: Advanced fluoride removal methods*

Defluoridation methods include precipitation, ion exchange, adsorption, electrocoagulation, and membrane processes (Lacson et al., 2021; Maity et al., 2018; Darchen et al., 2016). In this SI, many articles on aqueous F removal are included. Phytoremediation can be a conceivable route to remove F from water (Weerasooriyagedara et al., 2020). Fluoride uptake depends on plant sensitivity and stress tolerance mechanism. The F-hyper-accumulator plant would be a safe, secure, and cost-effective for removing F from soil and water. In successful F remediation, the diverse groups of terrestrial and aquatic plants are essential to identify, which can accumulate the F more than their origin (from where the plant has been collected for F removal). The water hyacinth beads doped with hydrous aluminium oxide (WH-HAO), hydrous iron oxide (WH–HFO), and hydrous aluminium oxide-iron oxide mixture (WH-HAO-HFO) successfully remediated aqueous F (Murambasvina and Mahamadi, 2020). The highest adsorption capacity was achieved in WH-HAO (4.43 mg/L) followed by WH–HFO (4.25 mg/L) and WH-HAO-HFO (4.18 mg/L). More than 80% F uptake on WH-HAO was recorded in 3 sorption-desorption cycles, where immobilized adsorbent shows higher removal potential in a continuous flow system. Thus, the hydrous metal oxide-doped water hyacinth can be used as an alternative adsorbent for aqueous F immobilization.

It is essential to search for cheaper, effective, and easily available adsorbents for water defluoridation. A mud pot with a cement paste layer on its inner surface successfully reduced (0.25 mg/g) fluoride (Shyamal and Ghosh, 2019). Portland pozzolana cement (layer of cement pastes on the inner side of mud pots) reduces 86 and 54.15% F from synthetic water and groundwater, respectively (Shyamal and Ghosh, 2019). Pectin biopolymer-based binary metal oxide composite [pectin-Al-Fe (PAF)] was used for aqueous F adsorption. This composite adsorbent (surface area of 122.78 m^2/g) adsorbs 333 mg/g F from water (Raghav and Kumar, 2019). Iron-impregnated activated carbons derived from waste *Citrus limetta* peels (AC-CLPs) were used for F uptake (Siddique et al., 2020). Fluoride adsorption decreased with a rise in F initial concentration from 5 to 30 mg/L (95–5.0% by AC-CLP_{250} and 94.8%–33.3% by AC-CLP_{500}). Thus, AC-CLPs can effectively be utilized for F decontamination. Entele and Lee (2020) estimated the household willingness to pay (WTP) for F-free 'water connection' in the Rift Valley region of Ethiopia, where the income shares for WTP ranges at 8–16% for F-free water service. A cost-benefit analysis shows that the F-free water supply is economically feasible. The F-free 'water service connection' at home is more valued versus the F-free water supply at the public tap. However, a public-private partnership model is essential to fulfill the safe water demand. The distribution of safe water is increasing in developed counties but it is still challenging in developing countries due to the socioeconomic structure. Implementation of the existing F removal technologies requires local research as communities differ in socioeconomic status. Further research is required to establish the suitability and sustainability of the existing research results in the pilot and full-scale operations.

5. The integration

Arsenic and fluoride contamination of water is a huge problem on a global scale. Researches are being actively pursued to address the As and F contamination, health risk, and mitigation. Arsenic and F poisoning represent a serious and widespread health problem particularly in rural communities, which depend on untreated As and F-rich water. Significantly, the co-occurrence of As and F in groundwater and associated health risk was observed in the Dharmanagar region, North Tripura (India). Arsenic and F contamination has been mainly dependent on region-specific geological and anthropogenic factors. For example, elevated F in the groundwater in Shiraz (Iran) is mainly caused by anthropogenic activities (natural processes have limited influence), whereas, in Chhattisgarh, India, F concentration in GW increases mainly due to the bedrock (rhyolites) weathering and agricultural practices. Enrichment of elements (Fe, Al, As, B, Cu, Mn, Pb, and Cd) are reported to be in a mixed origin of geogenic and anthropogenic activities. The geogenic contribution is much higher than anthropogenic inputs for elevated trace element concentrations in Bangladesh. Arsenic concentrations were noted higher particularly in aquifers in sedimentary terrains compare to metamorphic terrain in Sri Lanka.

Remediation of As and F contaminated water is essential. The absorbent GAC or FeS can be applicable in aqueous As removal, due to high efficiency (80–96%). Nanofiltration (NF) membrane is effective for the removal of As at 50 °C temperature, which can be used for As [as As (V)] contaminated geothermal waters (temperature limit 50 °C). The absorbent, pectin biopolymer-based binary metal oxide composite [pectin-Al-Fe (PAF)] or Iron-impregnated activated carbons derived from waste *Citrus limetta* peels (AC-CLPs) are potentially effective to remove aqueous F. The mud pot with cement paste (layer on its inner surface) has also been shown to be effective in removing F from water. Phytoremediation is one potentially cost-effective means for removing As and F from water. The hydrous metal oxide-doped water hyacinth can be used as an alternative adsorbent for aqueous F immobilization. To solve the As and F contamination problem will require a variety of approaches from different fields of research. Arsenic-tolerant phenotype transgenics plants, the seed priming technique can be used for As and F removal from water, soil, and sediment. The combination of biological and chemical treatment is often efficient for As and F removal. Furthermore, the rainwater harvesting, use of As and F safe aquifer and surface water are the alternative ecofriendly options for As and F water mitigation. Proper monitoring is essential. The governmental and non-governmental organizations must work together with the researcher for proper mitigation of As and F with Sustainable Development Goals (SDG) considering the framework for safe drinking water. Like UNESCO, every governmental and non-governmental organization should keep an agenda for Sustainable Development Program (SDP) for safe drinking water.

6. Future perspectives: the editor's take

Our experience with this special issue has been summarized in form of seven samurai in the field of arsenic and fluoride contamination to the groundwater in Fig. 2, where each challenge of this domain has been represented as a bead that is often used for removal techniques as well. The key to the sustainable tackling of the problem lies in its effective integrations for which a common interface has to be there for the proper interactions among interdisciplinary scientists, industrialists, academicians, stakeholders, and policymakers.

While, monitoring and prediction will remain in the focus for the field earth scientists, environmental engineers are likely to focus on finding the remediation technique, chemical engineers and industries are likely to focus on the advancement of fast analytical methods as well as in-situ removal where lab to field conversion efficacy will be significantly tested. Biologists and medical practitioners are needed to further decode the toxicological aspects of the cumulative effect of the As–F co-occurrences so that proper guidelines may be put forward. Social scientists have to come forward to sketch out effective and curated region-specific mechanisms for enhancing awareness and participation among various stakeholders. Above all, the transboundary cooperation among the administrative boundaries, national or international, has to be firmly in place, not only to tackle the issue but even to make a slight change in the present scenarios where co-occurrences of geogenic contaminants have emerged as a new threat to the welfare of the humankind.

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