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Genesis and mobilization of fluoride in groundwater of India: Statistical evaluation, health impacts, and potential remedies

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ABSTRACT

Groundwater contamination by fluoride (F^{-} >1.5 mg/L) is pervasive and typically confined to arid and semi-arid regions. Therefore, several parts of India are contaminated with F^{-}. However, genesis, sources, and mobilization of F^{-} in groundwater are unclear or so far evaluated based often on studies conducted at a local scale. To understand the severity of F^{-} contamination at the national scale and to devise remedial strategies, we performed a statistical evaluation between F^{-} and its influencing factors, including geology, hydro-meteorology, and potential hydro-chemical parameters based on a large dataset (n = ~2000) published in the last two decades throughout India. Results revealed that (a) alkalinity plays a pivotal role in the mobilization of F^{-} into groundwater from the sediments/rocks, (b) high F^{-} in groundwater is more pronounced in the arid and semi-arid areas of alluvial plains than hard rock regions, and (c) positive correlation of elevated F^{-} with SiO_2 and K^{+} indicates the dominance of geogenic sources linked to the weathering of fluorine bearing silicates. Investigations show that one-third of the Indian drinking water wells are contaminated with F^{-}, thereby risking the health of over millions of people through the drinking water pathway. Findings from this study have addressed the most possible sources, pathways, and regional prevalence of F^{-} contamination in the groundwater of India, and suggested the suitable remedial measures based on prevailing surface and sub-surface conditions. Lastly, this review also addresses challenges and propose future research directions to tackle high F^{-} groundwater and ensure safe drinking water supply in India.

Introduction

Fluorine is one of the lightest halogen elements, electronegative, and highly mobile at high temperatures. It is one of the abundant elements in the Earth crust (625 mg/kg), and in aqueous solution; it behaves as F^{-} ions (Ali 2017). World Health Organization (WHO 2011) and the Bureau of Indian Standards (2012) have set the upper safe limit to 1.5 mg/L of F^{-} in drinking water; however, excess of this is considered to be contaminated. Intake of F^{-} via the drinking water pathway beyond the permissible/safe limit can cause dental fluorosis, especially in children, and have dangerous severe, irreversible effects in the long-term and evident in the form of skeletal fluorosis (Ali et al., 2016; Mohammadi et al., 2017; Kumar et al., 2018; Nordstrom and Smedley, 2022). However, an optimum level of F^{-} (0.5 mg/L) is needed for tooth enamel and bone mineralization (Ali et al., 2016). Various studies on F^{-} contamination were earlier documented to study its sources and pathways in different parts of the globe (Ozsvath et al. 2006; Guo et al., 2007; Kumar et al., 2015Vithanage and Bhattacharya 2015; Dehbandi et al., 2018; Ali et al., 2018; Fuge 2019; Kashyap et al., 2020; McMahon et al., 2020).

F^{-} contamination can be derived from both geogenic and anthropogenic sources, while geogenic sources are widespread (Nordstrom and Smedley, 2022). In India, groundwater from both crystalline rocks, as well as unconsolidated aquifers, are largely reported to be contaminated by F^{-} (Saha et al., 2016; 2020). Nearly 300 minerals were reported to contain F^{-}; however, the most commonly occurring minerals are fluorite (CaF_2), fluorapatite [$Ca_5(PO_4)_3F$], biotite [$K(Mg, Fe)_3(AlSi_3O_{10})(OH, F)_2$], phlogopite [$KMg_3(AlSi_3O_{10})(F, OH)_2$], apatite [$Ca_5(PO_4)_3(F, Cl, OH)$], cryolite ($Na_3AlF_6$), and topaz [$Al_2SiO_4(F, OH)_2$] (Nordstrom and

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Smedley, 2022; Kumar et al., 2023a). The pH of groundwater is one of the critical factors responsible for the release of F^- from the sediments/rocks. Fertilization in agriculture and its abundant use in industrial sectors such as aluminum smelting, fertilizers manufacturing, and coal power stations are likely anthropogenic sources (Ali et al., 2016; Rao et al., 2017; Kumar et al., 2019; Kumar et al., 2023a). Apart from drinking water, F^- can also be consumed through food, beverages, and teas, particularly F^- bearing brick tea used in China (Ali et al., 2016). Even though, the content of F^- in beverages is comparatively lower than in drinking water, the daily consumption of F^- through these beverages can significantly contribute to fluorosis (De et al., 2021).

Elevated level of F^- in the groundwater is now a global problem and are frequently reported. Typically, these regions are confined to the arid and semi-arid areas which mainly fall in the developing world. In Africa, high F^- levels were found in numerous countries and chiefly reported from the vicinity of the East African Rift Valley (Kut et al., 2016; Kimambo et al., 2019; Ijumulana et al., 2021; Sunkari et al., 2022). In Latin America, the elevated levels were largely reported from Argentina, Mexico, and Brazil. However, studies from other South American countries are limited (Gomez et al., 2009; Alarcón-Herrera et al., 2013; Martins et al., 2018). While in Asia, China, Mongolia, and Indian subcontinents have often recorded high F^- (Wu et al., 2015; Ali et al., 2016; 2019; 2022; Rahman et al., 2020; Khattak et al., 2022; Ling et al., 2022). For example, Chandrajith et al. (2020) investigated drinking wells in Sri Lanka and found that F^- contaminates nearly 50% of the wells in dry zones. Coastal aquifers of China were also found to be contaminated (Cao et al., 2023). Prevalent regions containing elevated F^- in groundwater were reported in many countries; some of the prominent studies that investigated F^- contamination across the globe are mentioned here: India (Jacks et al., 2005; Ali et al., 2016); China (Dong et al., 2022; Hao et al., 2022; Wu et al., 2015); Ethiopia (Rango et al., 2012); Mongolia (Guo et al., 2012); Kenya (Olaka et al., 2016); and Malawi (Addison et al., 2020). Ali et al. (2016) earlier documented the F^- contamination and systematically investigated the worldwide situation. Consequently, other reviews, such as by Chowdhury et al. (2019), have reported five F^- bearing belts globally linked to the prevalence of F^- contamination. Further, Yadav et al. (2019a) investigated the Asian continent on F^- contamination. Similarly, Ali et al. (2019), Mukherjee and Singh (2018), and Podgorski et al. (2018) studied F^- contamination in Indian groundwater. Nordstrom and Smedley (2022) and Podgorski and Berg (2022) are the most recent documents on worldwide F^-. Though, the occurrence and intensity of the F^- contamination were reported, the association between F^- and its influencing factors have not been dealt with in detail.

Therefore, the current study aims to identify the inter-relationship between F^- with geology, hydro-meteorology, and potential hydrochemical parameters that influence the genesis and mobilization of F^- into the groundwater. This study is based on the systematic development of a large secondary dataset with more than 2000 drinking water wells published in the past two decades in India. Thus, this work aims to (a) analyze the sources, distribution, genesis, and mobilization of high F^- concentrations in groundwater, (b) correlation between co-contamination of F^- with other potential ions, and (c) health impacts and potential remedies in India. Lastly, this review concluded with significant challenges and proposed future research to tackle high F^- groundwater and put forward the option of safe and clean drinking water supply in India.

Distribution of high fluoride groundwater in India

Groundwater with high fluoride (F^-) is now reported as a major global contaminant (Kumar et al., 2023a; Neeti et al., 2023; Podgorski and Berg 2022; Li et al., 2021; Ali et al., 2016). Nordstrom and Smedley (2022) reported that fluorosis in India was first recognized in 1937.

Kumar et al. (2019) investigated the Indo-Gangetic Alluvial plain and reported 5.8 mg/L of F^- in groundwater. In Bihar, Mridha et al. (2021) reported that groundwater samples from Gaya and Nawada districts are highly contaminated with F^-, and found mean value of 2.15 ± 1.78 mg/L and 3.2 ± 1.64 mg/L, respectively. Kumari and Mishra (2023) also reported that F^- levels in groundwater of Munger district were up to 12 mg/L. Nizam et al. (2022a) studied the Kanpur region of the Gangetic plain and observed 5.2 mg/L of F^-. High values of F^- were frequently reported in the groundwater of Haryana state. For example, Yadav et al. (2019b) reported 18 mg/L, and Ali et al. (2018) reported 18.5 mg/L of F^- from the Mahendergarh and Bhiwani districts, respectively. Malik and Kavita (2022) reported 4.5 mg/L of F^- from the Karnal district. In adjoining Punjab, Duggal and Sharma (2022) observed more than 10 mg/L of F^- from few localities in the Malwa region. Ahada and Suthar (2018) investigated southern regions of Punjab and found up to 5 mg/L of F^-. Recently, Nizam et al. (2022b) investigated the Patiala district, Punjab, and found the F^- value up to 9.2 mg/L. In Rajasthan state, 5.74 mg/L of F^- was reported from the Jhunjhunu district (Jandu et al., 2021). The highest value reported from Jaisalmer of Rajasthan was 6.6 mg/L (Singh and Mukherjee, 2015).

In East India, Mukherjee et al. (2019) and Thapa et al. (2017) reported 18.25 mg/L and 20.4 mg/L of F^- from the drinking well of West Bengal state, which is higher than ten folds of the safe limit of WHO and BIS. However, the geogenic sources from West Bengal are undocumented.

Southern Indian states are long known to be prone to F^- contamination due to bedrock. Hot spots like the Nalgonda district, Telangana (previously in Andhra Pradesh) have granitic rocks with F^- concentration reported up to 1706 mg/kg by Mondal et al. (2009) and as high as 3125 mg/kg by Reddy et al. (2010). Prevalence of endemic fluorosis in this district and adjacent areas has led to several studies on the occurrence, sources, and distribution of F^- in groundwater. Groundwaters from this district have recurrently reported elevated F^- ranging from 3 to 7.6 mg/L (Reddy et al., 2010), 0.7 to 19 mg/L (Mondal et al. (2009)), and 0.1 to 8.8 mg/L (Brindha and Elango, 2011; Brindha and Elango, 2013). Other districts in Telangana, such as the Yadadri-Bhuvanagiri district, have also recently recorded a maximum of 3.56 mg/L F^- in groundwater (More et al., 2021). The adjacent state of Andhra Pradesh has often reported high F^- in its groundwaters. F^- in Andhra Pradesh from various districts, such as the Prakasam district, ranged between 0.7 and 2.8 mg/L (Subba Rao, 2017), and from the Guntur district, ranged between 0.3 and 2.3 mg/L (Subba Rao, 2003).

Weathering of granite and granitic gneiss host rocks in the Ilkal area of Karnataka had led to F^- in groundwater from 0.3 to 6.5 mg/L (Tirumalesh et al., 2007). Studies in the granitic belt of Badami Taluk and Hunagund Taluk of Karnataka consistently recorded F^- >2 mg/L in all investigated locations (Gaonkar et al., 2019). Several other taluks and districts also have F^- at excessive levels, such as in the Indi taluk (0.26 to 3.57 mg/L) reported by Ugran et al. (2017), and Kolar district (0.36 to 3.34 mg/L) and Thumkur district (0.78 to 5.35 mg/L) reported by Mamatha and Rao (2010).

Dharmapuri district in Tamil Nadu is another F^- endemic area in southern India. Here, F^- between 0.15 and 6.48 mg/L was reported by Jagadeshan et al. (2015a) from epidote hornblende biotite gneiss and charnockite areas containing average F^- of 59.4 mg/kg and 35.8 mg/kg, respectively (Jagadeshan et al., 2015b). Maximum F^- from Tiruvannamalai region in Tamil Nadu was reported to be 3.2 mg/L from open wells and 3.8 mg/L in bore wells (Chicas et al., 2022). Groundwater analyzed from areas with charnockite rocks in Madurai district (Thivya et al., 2017) and Dindigal district (Ramachandran et al., 2012) in Tamil Nadu had F^- up to 1.8 mg/L and 2.5 mg/L, respectively. In the northern part of Tamil Nadu, namely in Kancheepuram district, F^- was observed to range from 1 to 3.24 mg/L by Dar et al. (2011).

Compared to the studies on F^- in the southern Indian states, Kerala has only a very few documented studies. However, this does not imply that groundwater has low F^- content. Reported studies from Chittur Block had a maximum of 6.3 mg/L (Shaji et al., 2018), Alleppey recorded 0.68 to 2.88 mg/L (Raj and Shaji, 2017), and Palakkad district documented 0.2 to 5.75 mg/L of F^{-} (Shaji et al., 2007).

Methodology

Systematic searches for data mining

For the present work, systematic searches were conducted on the Google Scholar and Scopus web engines to include the maximum research articles on F^{-} contamination in the groundwater of India in international peer-reviewed journals (Fig. 1).

Various key words like 1) fluoride in India, 2) fluoride in groundwater of India, 3) fluoride in alluvial plains, 4) fluoride in arid and semi-arid regions, and 5) fluorosis in India; were used to collect maximum articles. Various articles falling in the following criteria were discarded: 1) studies conducted on water other than the groundwater, and 2) articles specifying only min-max ranges of F^{-}. After deletion, all articles were thoroughly studied. At this stage, there were 64 research articles. Further, an Excel sheet (Microsoft Office) was prepared where all the parameters along with F^{-} level were compiled. For the bivariate plots, elevated F^{-} content with other physio-chemical parameters were only considered (expect some exceptions). At this stage, it was found that the articles investigated for the F^{-} contamination were not studied with all the hydro-chemical parameters. So, even if one parameter was mentioned, the article is considered for this work. Therefore, the number of variables varies during the correlation analysis. Finally, a total of more than 2000 wells (hand pumps, tube wells, and bore wells) were compiled along with potential parameters recorded in the groundwater for this study.

As mentioned, research articles in the international domain from the last two decades comprising more than 2000 drinking water wells across India were investigated. Further, statistical correlation of F^{-} with other potential hydro-chemical parameters was performed to investigate the possible genesis and mobilization of F^{-} release into the groundwater (Fig. 2). In some studies, all hydro-chemical parameters were not analyzed; however, available parameters were used for this study; and thus, differ in the number of variables during the statistical correlation. F^{-}, along with other hydro-chemical parameters, were initially considered for statistical correlation. However, during the analysis, it was observed that a significant correlation could not be drawn by considering all the data of F^{-}. Therefore, F^{-} values greater than the safe limit (>1.5 mg/L) were only considered for in-depth statistical correlation. This study suggests that nearly one-third of India's drinking wells have elevated levels of F^{-}, thus a major concern for the water supply. The results revealed significant findings and are discussed in the subsequent sections.

Results and discussion

The unconsolidated alluviums are dominantly composed of sand, silt, and clay mixed with kankars (calcareous nodules) with varying proportions found to form suitable aquifers in India. The hard crystalline rocks also form suitable aquifers wherever they were found to be jointed and fractured (Saha et al., 2016; 2020). Groundwater largely occurs in these aquifers with distinct quantity and quality. In general, the wells installed in the unconsolidated aquifers have higher discharge rates than the crystalline ones. The inhabitants primarily depend on the groundwater which is extensively used for drinking from both aquifers.

In this study, a detailed statistical analysis of F^{-} was conducted with various other potential hydro-chemical parameters for the evaluation of possible genesis, mobilization, and causes of contamination of F^{-} in the groundwater of India. The F^{-} content from both of the aquifers are included in the present work. The association of hydrochemical parameters with F^{-} are discussed below.

Bivariate plot of fluoride with hydro-chemical parameters

pH and EC

The elevated F^{-} content with pH showed an insignificant positive correlation ($n = 1155$; Fig. 2a). Box and Whisker diagram was plotted for all the F^{-} values, which suggests that the groundwater with elevated F^{-} levels has higher pH values (Fig. S1). The bivariate plot confirmed that the pH plays a pivotal role in triggering the F^{-} from fluorine-bearing sediments or rocks to the aqueous medium (Ali et al., 2016). Further, the statistical correlation between F^{-} (>1.5 mg/L) with electrical conductivity (EC) was performed ($n = 1079$; Fig. 2b). The bivariate plot between F^{-} and EC revealed that the F^{-} has weak positive correlation with EC. This is possible due to the longer water-rock/sediment interaction, which could enrich high F^{-} content in the groundwater (Ali et al., 2021).

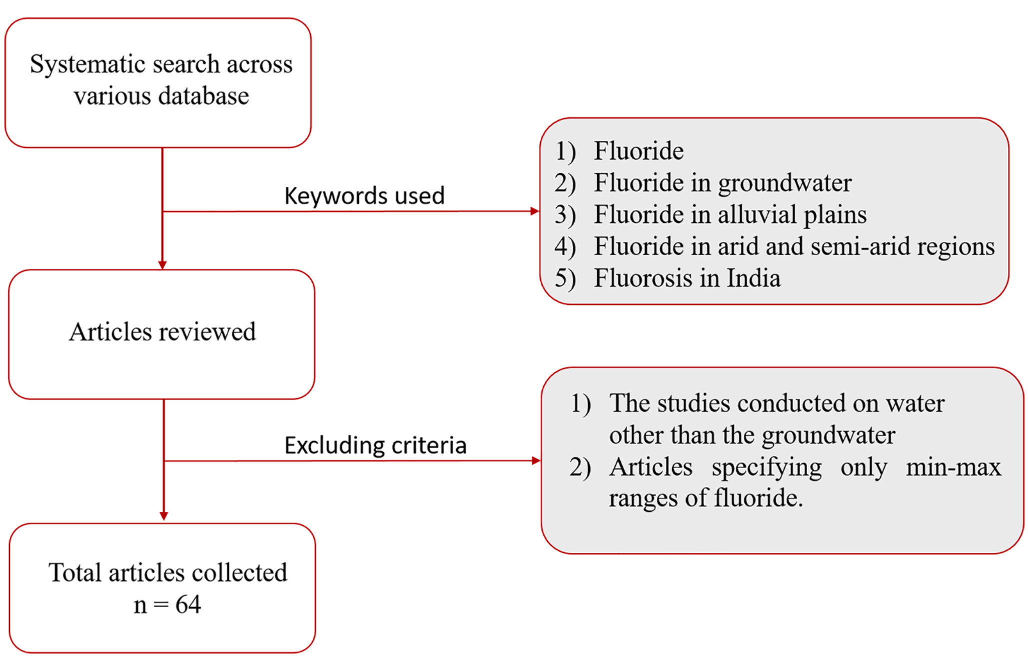

Fig. 1. Flowchart showing exclusion and inclusion criteria of articles selection.

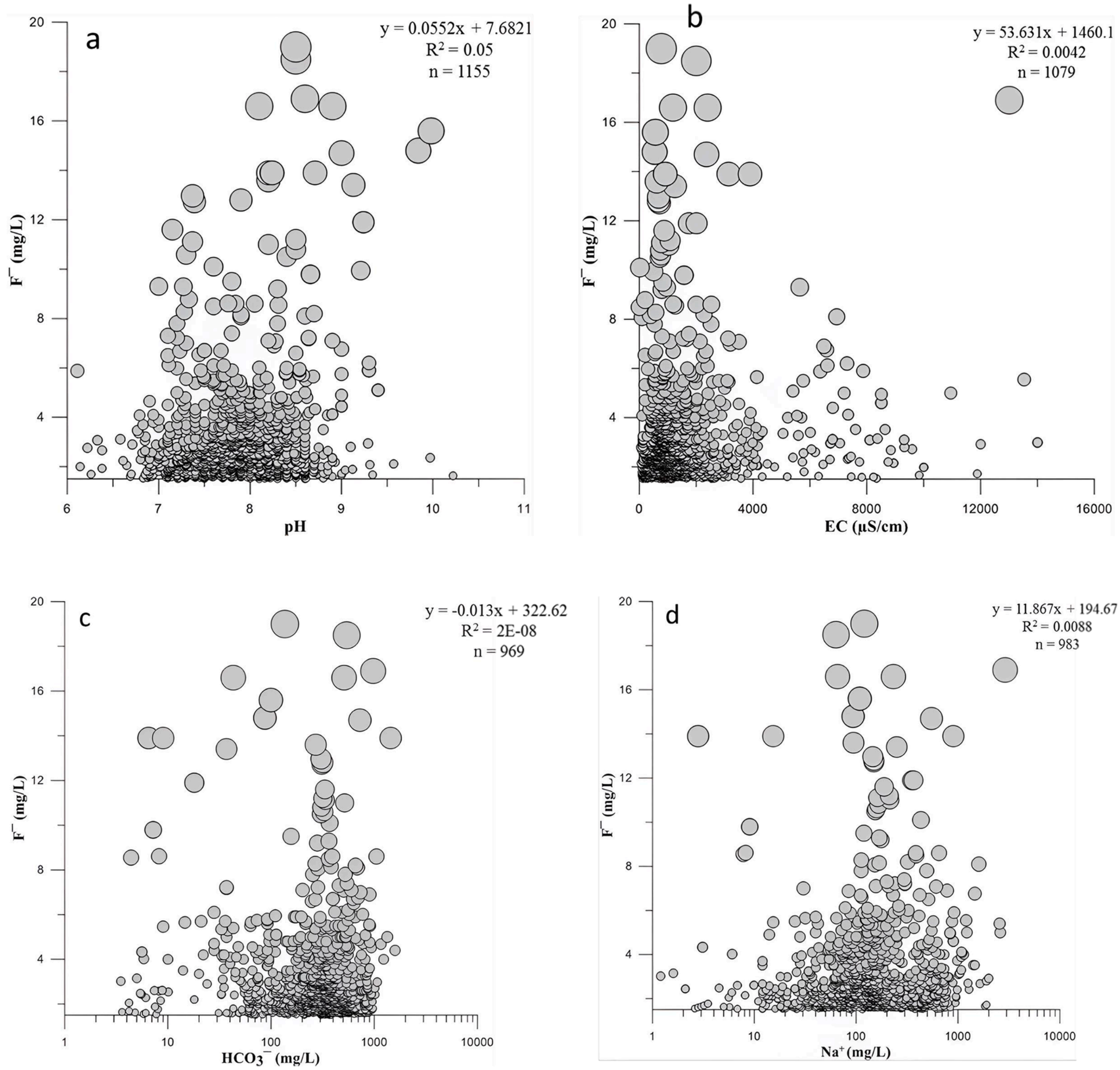

Fig. 2. Bivariate plots between F^- with (a) pH, (b) EC, (c) HCO_3^-, (d) Na^+, (e) SO_4^{2-}, (f) NO_3^-, (g) Mg^{2+}, (h) Ca^{2+}, (i) K^+, (j) Fe, and (k) SiO_2 (bubbles size is proportional to F^- content; continued, $n = 64$ and given here alphabetically).
Acharya et al. (2018); Adimalla et al. (2018); Adimalla et al. (2018a); Ahada and Suthar (2018); Ali et al. (2021); Ali et al. (2018); Arulbalaji and Gurugnanam (2017); Arveti et al. (2011); Batabyal (2017); Bhuiyan and Ray (2017); Chatterjee et al. (2008); Dar et al. (2011); Das et al. (2003); Datta et al. (2014); Datta et al. (1999); Devadas et al. (2007); Duraiswami and Patankar (2011); Gawle et al. (2021); Gupta and Misra (2018); Gupta et al. (2006); Haritash et al. (2008); Jha et al. (2010); Joshi and Seth (2011); Kantharaja et al. (2012); Karthikeyan et al. (2010); Kashyap et al. (2020); Keesari et al. (2014); Khanna (2015); Kodate et al. (2007); Kumar (2017); Kundu et al. (2001); Madhnure and Malpe (2007); Madhnure et al. (2007); Meenakshi and Malik (2004); Misra and Mishra (2007); Mondal et al. (2014); Mondal et al. (2009); Narsimha and Sudarshan (2017); Pandith et al. (2017); Prasad and Rao (2018); Raj and Shaji (2017); Raju (2017); Raju et al. (2009a); Raju et al. (2009b); Rao (2017); Rao et al. (2012); Rao (2009); Rao (2008); Sahu et al. (2017); Salve et al. (2008); Saxena and Ahmed (2003); Shaji et al. (2007); Shankar et al. (2008); Sharma et al. (2012); Singh et al. (2013); Sudarshan et al. (2018); Sujatha (2003); Sujatha and Reddy (2003); Tirumalesh et al. (2007); Tiwari et al. (2020); Usham et al. (2018); Vikas et al. (2009); Viswanathan et al. (2009); and Yadav et al. (2021).

HCO_3^-, Na^+, SO_4^{2-}, and NO_3^-

Bivariate plot between F^- and HCO_3^- revealed a statistically insignificant correlation (Fig. 2c). Further, the bivariate plot of F^- with Na^+ suggests a statistically weak positive correlation (Fig. 2d). This correlation is well supported with our earlier studies (Ali et al., 2016; 2021). In another plot between F^- and SO_4^{2-}, positive correlation was observed (Fig. 2e). The bivariate plot of F^- with NO_3^- revealed negative correlation (Fig. 2f). This supports the lower possibilities of anthropogenic involvement in F^- enrichment in Indian groundwater.

Mg^{2+}, Ca^{2+}, and K^+

Other ions, such as Mg^{2+}, Ca^{2+}, and K^+, were also evaluated for this study. The plot of F^- with Mg^{2+}also revealed negative correlation (Fig. 2 g). The bivariate plot between F^- and Ca^{2+}revealed an insignificant correlation (Fig. 2h). Further, the plot between F^- and $K^+$was also attempted, which revealed poor positive correlation between each of the ions (Fig. 2i). This correlation also supports the geogenic sources of F^- which simultaneously releases K^+ with F^- (Ali et al., 2016).

Fe and SiO_2

The plot between F^- and Fe revealed an insignificant negative

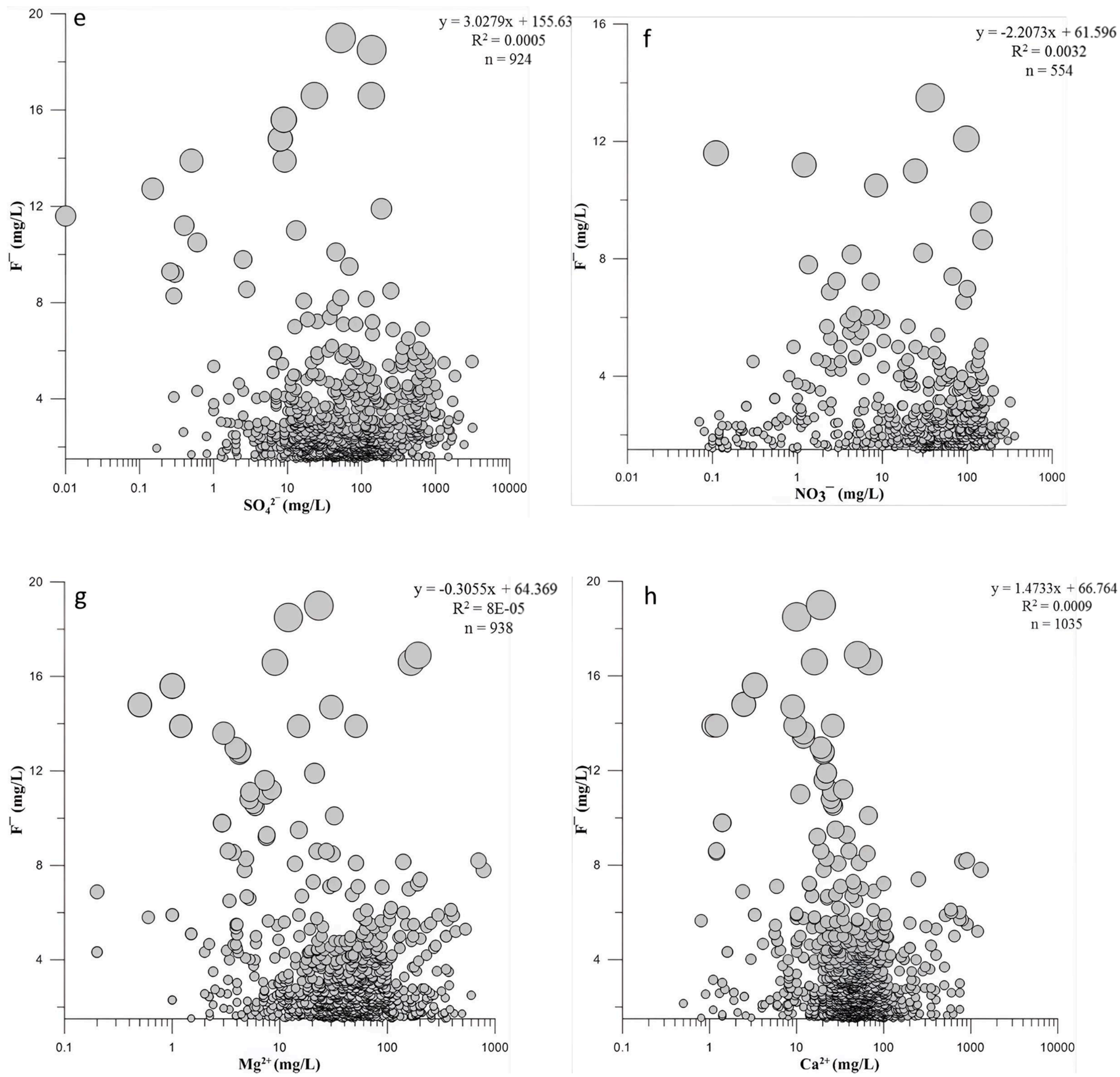

Fig. 2. (*continued*).

correlation, although the correlation is only based on the limited available Fe data (Fig. 2j). The bivariate plot between F^- and SiO_2 is crucial for revealing the geogenic contamination. The plot has also shown positive correlation between these two parameters (Fig. 2k). This suggests that the geogenic sources are most likely possible contamination sources which could be linked to the weathering of fluorine-bearing silicates.

Statistical correlation

Statistical correlation of F^- with other parameters is shown in Table 1. For this work, statistical analysis is broadly divided into two groups aiming to study the possible association with F^- contamination: 1) all F^- values and 2) F^- >1.5 mg/L.

Table 1 showed a noticeable positive correlation of pH with F^-. This revealed that alkalinity has a pivotal role in the mobilization of F^- into the groundwater (Ali et al., 2016). In addition, the table also suggests a noticeable positive correlation of F^- with SiO_2 and K^+. This is possibly linked to the weathering of the fluorine-bearing silicates. Further, Box and Whisker diagrams were plotted for all the states of India. This is created to highlight the regions with F^- concentration above the safe limit to demarcate the contaminated states based on the research conducted in the last two decades (Fig. 3).

Identification of fluoride-polluted areas

This study highlighted the states having F^- content greater than the safe limit in drinking water of India. States like, Andhra Pradesh, Chhattisgarh, Haryana, Jammu and Kashmir, Kerala, and Punjab, are highly contaminated (Fig. 3). Further, Fig. 3 also revealed that except significant parts of Jammu and Kashmir, other states falling in the arid and semi-arid regions are typically contaminated by F^-. Further, this study suggests that the F^- contamination is more predominant in states having drinking water in alluvial zones (Delhi, Gujarat, Haryana, Punjab, Rajasthan, Uttar Pradesh, and West Bengal) than the states having hard rock aquifers (including all states other than the alluvial ones; Fig. S3). This could be due to the leaching of F^- in the alluvium derived from the hard rocks, whereas F^- content in hard rocks are least mobilized. Further, this study suggests that the F^- are commonly found in the alluvial areas, mainly in arid and semi-arid regions, followed by the hard rocks regions (Fig. S4). Further, this study does not include most of the eastern part of India, as few studies were conducted in those regions.

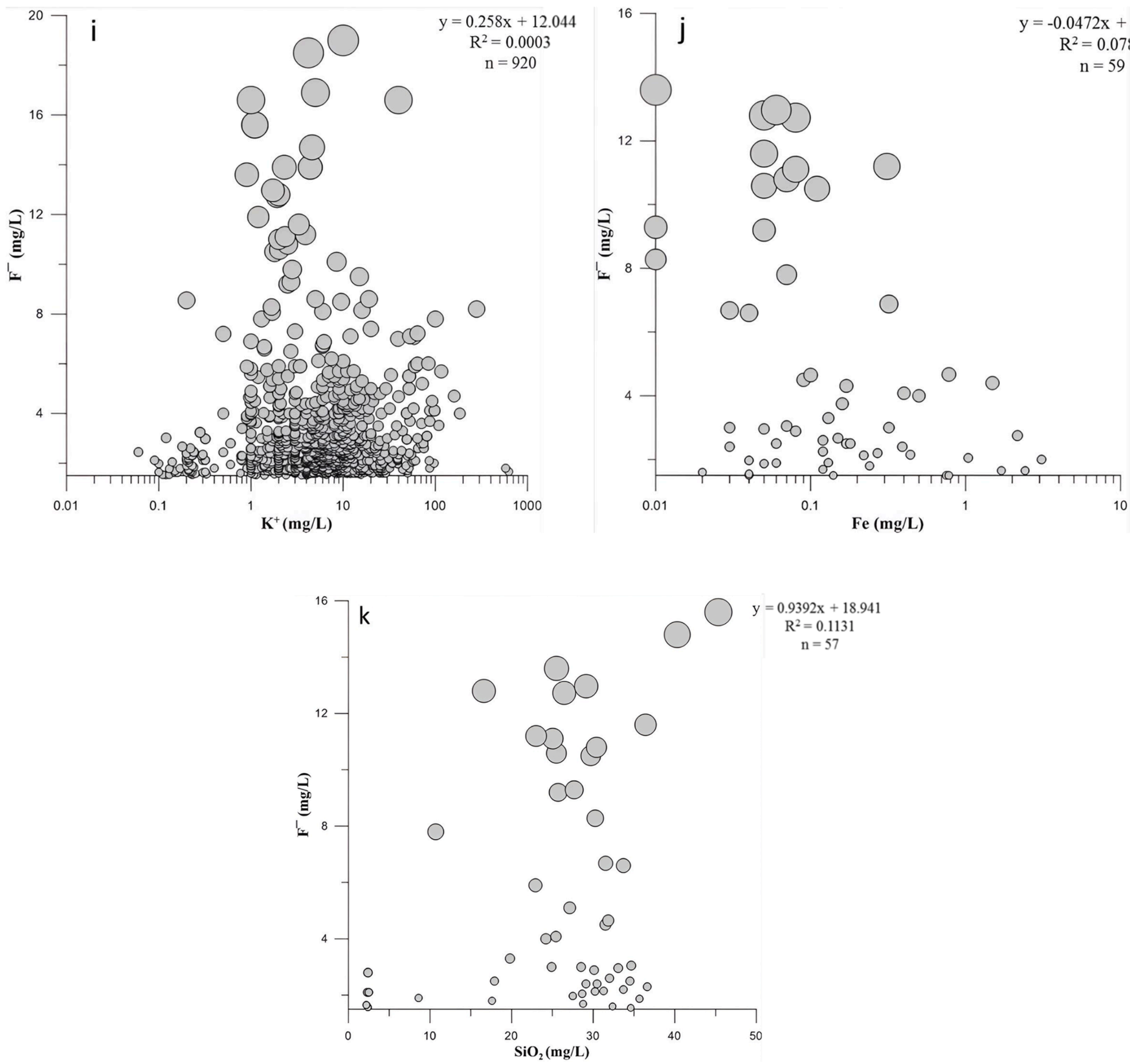

Fig. 2. (*continued*).

Finally, all F^- content were plotted on the hydrogeological map of India. For this work, each data point in the groundwater was initially classified into three groups, i.e., less than 1.5 mg/L, 1.5–3 mg/L, and more than 3 mg/L (Fig. 4). In this study, prevalent contaminated regions were shown as a single point on the hydrogeological map of India (Fig. 4; www.cgwb.gov.in). For clear distinction, five data pertaining to each point were plotted on the map. It is evident from Fig. 4 that the F^- concentration in the groundwater greater than the safe limit is typically restricted to the semi-arid and arid regions of India.

This study revealed that alkalinity plays a pivotal role in triggering F^- into the groundwater from the fluorine-bearing sediments/rocks. The contamination is more pronounced in the arid and semi-arid areas of alluvial plains than in the hard rock regions. The significant correlation of elevated F^- with SiO_2 and K^+suggests the possibility of the dominance of geogenic sources over anthropogenic and is possibly linked to the weathering of the silicates (Fig. 5). The results showed that the F^- contamination in the groundwater is primarily contaminated by geogenic sources. This study highlighted the prevalent areas and thus significant for policymakers where safe water supply must be priorities.

Genesis and mobilization of fluoride in groundwater

F^- in natural water is primarily governed by pH, anion exchange between hydroxyl groups with F^- and residence time of water in the aquifer (Apambire et al., 1997). The most accepted mobilization process is ion exchange of F^- by hydroxyl ions (OH^-; Edmunds and Smedley 2013; Ali et al., 2018). It was also reported that the aquifers with low hydraulic conductivity retain water for a longer time, resulting in elevated F^- levels. However, this association is not well documented.

High evaporation and scanty rainfall in the arid and semi-arid regions facilitate the release of F^- from the host rocks. Handa (1975) earlier observed a link between sodium bicarbonate facies in groundwater and high F^-. Documentation of worldwide studies (Ali et al., 2016) showed that F^- is significantly negatively correlated with calcium ions and positively correlated with sodium and bicarbonate ions. While limited studies are conducted on the identification of contamination sources of F^- globally (Ali et al., 2021). Ali et al. (2021) investigated the Delhi older alluviums in India and attributed mica as a possible geogenic source for F^- ions. Ali (2022) recently observed that F^- in the groundwater of Delhi is primarily controlled through water-sediment interactions, local hydro-meteorological conditions, and mean water residence time. Further, Ali (2022) investigated the possible release of F^- from sediments/rocks. The release of F^- from common F^- bearing minerals is given below:

$$\underset{\text{Biotite}}{2K(Mg,Fe)_3(AlSi_3O_{10})(F,OH)_2} + 2O_2 + 4CO_2 + 19H_2O \rightarrow \underset{\text{Kaolinite}}{Al_2Si_2O_5(OH)_4} + 2K^+ + 4HCO_3^- + 6Fe(OH)_3 + 4H_4SiO_4 + 6Mg^{2+} + 2F^- \tag{1}$$

(Chakraborty et al., 2022)

$$\underset{\text{Phlogopite}}{KMg_3(AlSi_3O_{10})F_2} + 2OH^- \rightarrow KMg(AlSi_3O_{10})(OH)_2 + 2F^- \tag{2}$$

(Guo et al., 2007)

$$\underset{\text{Muscovite}}{KAl_2(AlSi_3O_{10})F_2} + 2OH^- \rightarrow KAl_2(AlSi_3O_{10})(OH)_2 + 2F^- \tag{3}$$

(Guo et al., 2007)

Water-rock interaction is one of the most significant processes resulting in the operation of various other processes leading to the enrichment of F^- in groundwater. Ali (2022) recently reported that the enrichment of F^- is primarily governed via sediment geochemistry, water-sediment interactions, residence time, and existing local hydro-meteorological (temperature and rainfall) condition in Delhi. Kumar et al. (2019) concluded that the weathering of hard rocks in the Indo-Gangetic alluvial plains was responsible for high F^- in the region. However, limited studies only reported the sources of F^- in the groundwater of India (Ali et al., 2018; 2021).

Health impacts of drinking high fluoride groundwater

Extensive use of high F^- groundwater for drinking poses potential threat to humans. As discussed earlier, the importance of F^- studies has increased due to their adverse effects on humans. The consequence of F^- contaminated water is evident in the form of dental and skeleton fluorosis. This may lead from initial mild dental fluorosis to prolonged effects of irreversible crippling skeleton fluorosis (Nordstrom and Smedley 2022; Table 2).

F^- within the permissible limit is also a boon to humans, which helps in formation of tooth enamels and bone mineralization. Therefore, it is also said to be a sword with two edges. It was estimated that nearly 200 million people globally are potentially exposed to fluorosis (Edmunds and Smedley 2013), and significant vulnerable population lives jointly in India and China (Podgorski and Berg 2022). Predicting affected people from fluorosis is challenging; therefore, the exposed number is infect clearly more than estimated. Recently, random forest modeling was also performed for F^- prediction and demarcation of contaminated areas on a regional scale in India (Sarkar et al., 2023; Podgorski and Berg 2022).

Numerous researchers have evaluated the probabilistic non-carcinogenic human health risk assessment of F^- consumption (Ali et al., 2019; Li et al., 2019; Keramati et al., 2019; Kaur et al., 2020; Kumar et al., 2023). These studies reported that children are highly vulnerable than adults due to their lower body weight. Few studies documented the noticeable link between pregnant women and F^- exposure. On examining the F^- levels in groundwater and the potential risk through the drinking water pathway, Green et al. (2019) reported that F^- exposure during pregnancy can be associated with lower IQ in children. Studies also showed negative effects on the development of children intelligence (Karimzade et al., 2014). However, this association needs further investigation. Further, Kheradpisheh et al. (2018) investigated F^- links with human thyroid disease and found a good relation between them. Ahmad et al. (2022) recently reported that the kidney, liver, and heart are three crucial parts of the human body exposed to excess consumption of F^-, which could accelerate various diseases.

Table 1
Correlation coefficient between fluoride and other hydro-chemical parameters shown as heat map for better visualization.

	F^-	F^- >1.5 mg/L
F^-	1	1
Fe	-0.17[1]	-0.28[13]
HCO_3^-	0.05[2]	0.00[14]
SiO_2	0.23[3]	0.34[15]
SO_4^{2-}	0.10[4]	0.04[16]
NO_3^-	0.01[5]	-0.06[17]
Cl^-	0.05[6]	0.06[18]
Mg^{2+}	0.03[7]	-0.01[19]
K^+	0.00[8]	0.02[20]
Ca^{2+}	0.00[9]	0.03[21]
Na^+	0.17[10]	0.09[22]
pH	0.17[11]	0.22[23]
EC	0.10[12]	0.06[24]

n^1 = 153; n^2 = 2347; n^3 = 255; n^4 = 2268; n^5 = 1464; n^6 = 2591; n^7 = 2222; n^8 = 2226; n^9 = 2391; n^{10} = 2370; n^{11} = 2688; n^{12} =2291; n^{13} = 55; n^{14} = 968; n^{15} = 57; n^{16} = 901; n^{17} = 554; n^{18} = 1054; n^{19} = 938; n^{20} = 920; n^{21} = 1035; n^{22} = 983; n^{23} = 1155; n^{24} = 1079 (n is the number of data available for correlation).

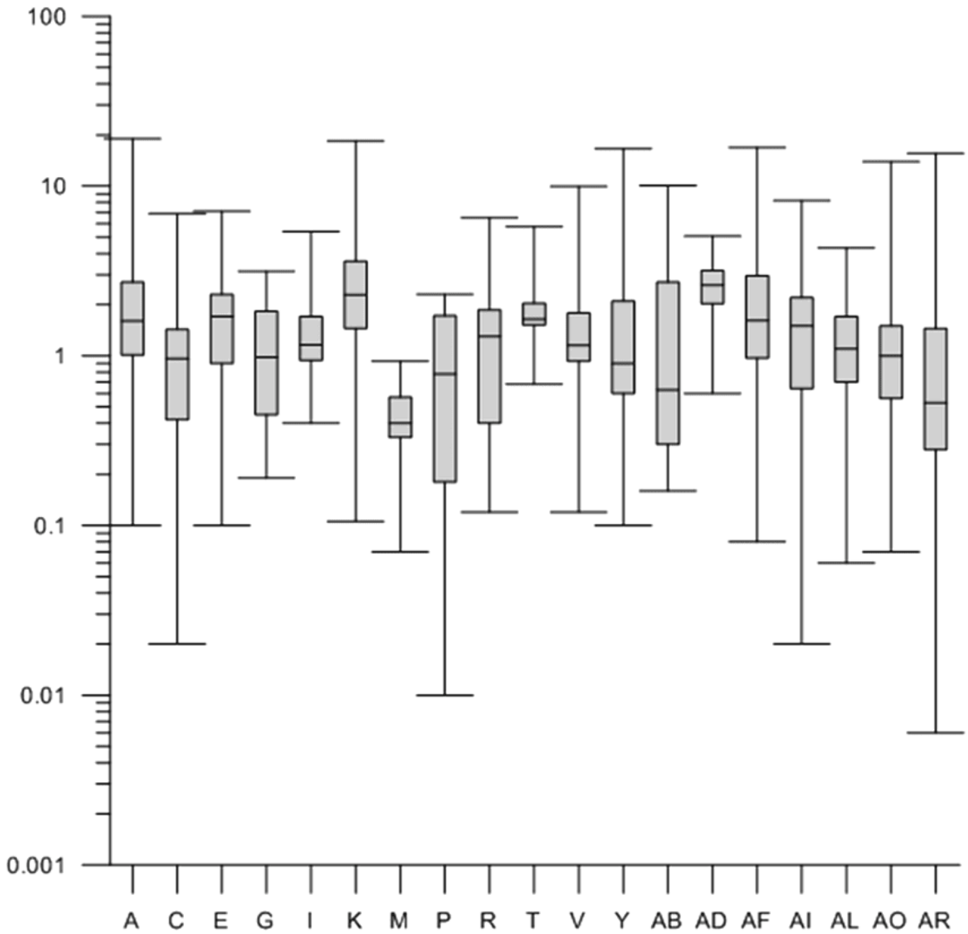

Fig. 3. Box and Whisker diagram showing the range of F^{-} concentration (mg/L) in the states of India (Fig. S2).
A: Andhra Pradesh; C: Assam; E: Chhattisgarh; G: Delhi; I: Gujarat; K: Haryana; M: Jammu and Kashmir; P: Jharkhand; R: Karnataka; T: Kerala; V: Madhya Pradesh; Y: Maharashtra; AB: Odisha; AD: Punjab; AF: Rajasthan; AI: Tamil Nadu; AL: Telangana; AO: Uttar Pradesh; AR: West Bengal. $n^{A} = 439$; $n^{C} = 64$; $n^{E} = 33$; $n^{G} = 44$; $n^{I} = 40$; $n^{K} = 122$; $n^{M} = 26$; $n^{P} = 32$; $n^{R} = 103$; $n^{T} = 31$; $n^{V} = 43$; $n^{Y} = 207$; $n^{AB} = 36$; $n^{AD} = 76$; $n^{AF} = 238$; $n^{AI} = 327$; $n^{AL} = 242$; $n^{AO} = 345$; $n^{AR} = 223$ (n is the number of data available for correlation. Indian states boundaries with name are shown in the Fig. S2).

Kadam et al. (2020) and Senthilkumar et al. (2021) investigated western India and observed that children and infants are vulnerable to fluorosis. Further, Duggal and Sharma (2022) investigated the northern region of India and also observed that children are more vulnerable to fluorosis than other age groups in the Punjab state. Ali et al. (2019) reported that children are more likely to be highly vulnerable than adults in numerous states of India. Kumar et al. (2023a) summarized the potential F^{-} impacts on humans and observed that high F^{-} consumption might affect the liver, kidney, and reproductive systems.

Karunanidhi et al. (2020) investigated the Coimbatore region of South India and found that children are more vulnerable to adult females and males. Adimalla and Qian (2022) also studied the southern region and found that infants are more vulnerable than other age groups. Thabrez et al. (2023) reported that people from the Sira region in Karnataka are more prone to dental fluorosis than skeletal fluorosis. This study also showed that children (<8 years) face moderate risk of dental caries, dental fluorosis, and skeletal fluorosis, whereas adolescents and adults do not encounter such risk. Clinical studies by Ugran et al. (2017) found F^{-} content ranging from 0.34 to 3.41 mg/L in urine samples of people who consume elevated F^{-} groundwater. This study also observed symptoms of clinical fluorosis among these sample populations, which include arthritis, joint pains, gastrointestinal discomfort, and lower limb deformities (Ugran et al., 2017). Jose et al. (2014) conducted a survey among school students and observed dental fluorosis in 36% of the sample population, more prevalent in children aged between 9 and 10 years and with boys more frequently affected by dental fluorosis than girls.

Potential remedies/techniques for high fluoride groundwater in India

Various defluoridation methods were earlier investigated and documented by numerous researchers like Mohapatra et al. (2009), Jagtap et al. (2012), Vithanage and Bhattacharya (2015), Kut et al. (2016), Yadav et al. (2019), Kashyap et al. (2021), and Kumar et al. (2023a). The defluoridation methods can efficiently remove up to 70–90% from various defluoridation methods; providing sustainable F^{-} free water to large communities is challenging. However, cost-effective defluoridation methods impose a potential challenge on the scientific community. Fluoridation issue is highly debatable even though, in the United States of America; the optimum level of F^{-} is mandated in the water supply (www.cdc.gov). Solving this F^{-} problem remains a major challenge even today, especially in developing countries. Treating the water using ion exchange or precipitation methods is technically feasible and has been tried. However, these methods have typically been met with limited success in developing countries. For example, in India, defluoridation technologies have been implemented nationwide for a safe and clean drinking water supply.

Earlier, several studies have reported that the "*Nalgonda procedure*," designed by NEERI, Nagpur, is the defluoridation technology most commonly employed for domestic use at the grassroots level (Nawlakhe et al., 1975; Bulusu et al., 1979; Nawlakhe and Rao, 1990; Nawlakhe and Paramasivam, 1993). Community-oriented "*defluoridation plant*" was set up in the Nalgonda district of Andhra Pradesh to remove F^{-} from potable water; NEERI invented the technique in 1961. In the 1930's, activated alumina was used as an effective defluoridation method for domestic water uses. Using activated alumina, the Sathya Sai University for Higher Education, Puttaparthi, created the Prasanthi technology.

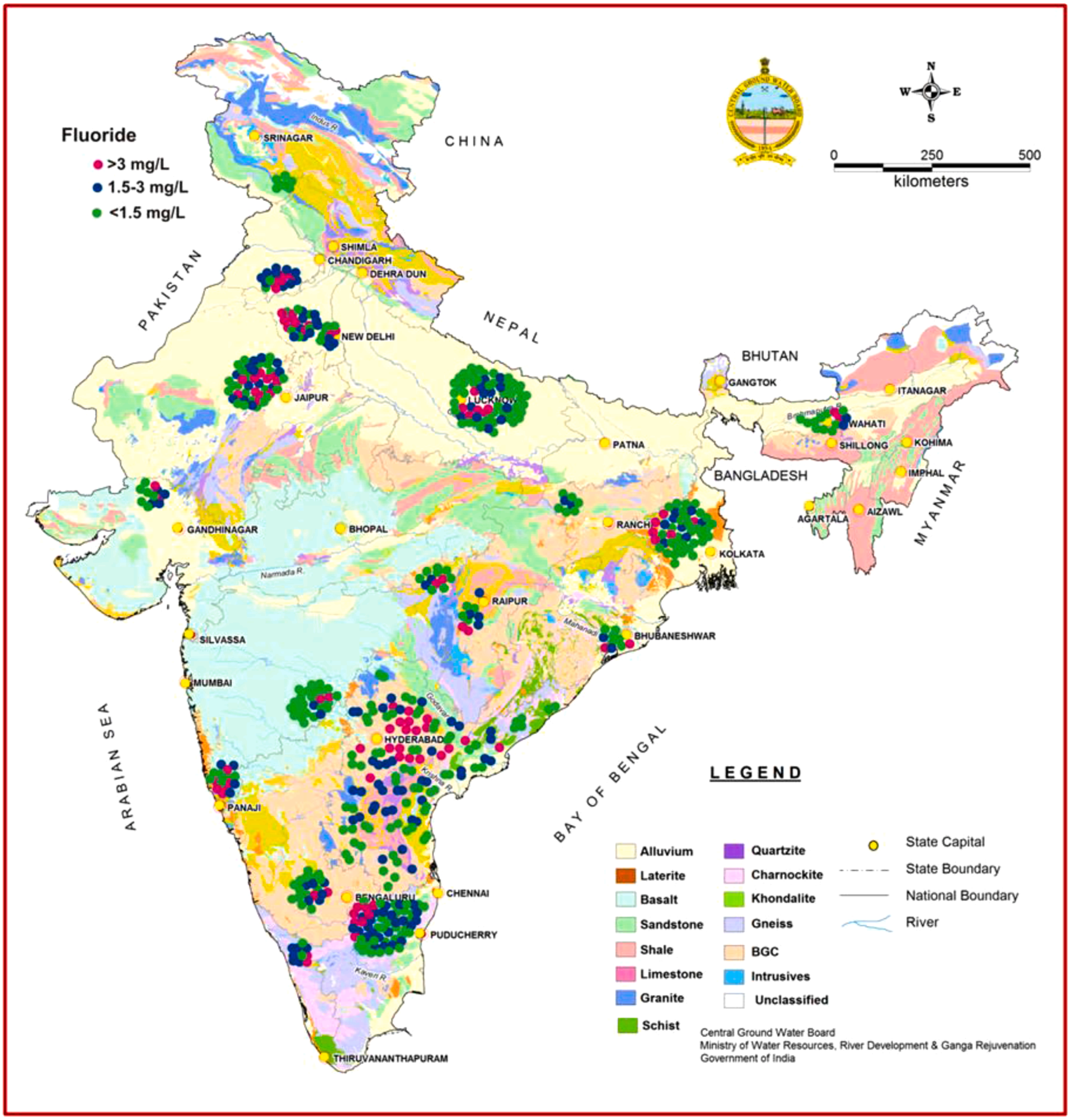

Fig. 4. Fluoride concentration in the groundwater of India (five data points pertaining to each point are shown on the hydrogeological map of India; after CGWB, India).

Drawbacks to this technology include high cost, the need for skilled workers to reprocess the filter material, and a byproduct of slightly higher concentrations of residual aluminum (Agarwal et al., 1999). Ayoob et al. (2008) reported defluoridation methods most frequently in locations where F^{-} contamination is a problem via coagulation and adsorption/ion exchange. In addition, Ayoob et al. (2008) summarized that in several developing nations, including India and Tanzania, the Nalgonda method and bone char adsorption—or integrated approaches—have been applied at both individual and communal levels. Even though membrane separation methods guarantee high-quality water, they are "*highly technical and expensive*" alternatives for most fluoride-endemic countries. Electrochemical approaches are "*energy-intensive*," which provide good F^{-} removal. During the 1990's, it has been more common for rural Indian communities to look for home-based water quality remediation solutions for their drinking water supply. In India, since 1991, the UNICEF initiative program, defluoridation units attached to hand pumps, and domestic defluoridation units were built using locally produced activated alumina (Eswar and Devaraj, 2011). Further, evidence of the usefulness and efficiency of activated alumina in removing F^{-} was provided by down-flow column tests (Srimurali and Karthikeyan, 2008). The IISc technology of F^{-} treatment at Kolar, Karnataka, comprises of blending cum sedimentation unit with geared manual-operated mechanical stirrer for blend of MgO and water (Rao and Mamatha, 2004; Eswar and Devaraj, 2011). Furthermore, adsorption has been reported as cost-effective and simple method for defluoridation (Agarwal et al., 2002; Karthikeyan et al., 2005). Afterward, Sulaiman et al. (2009) reported the application of granular alumina cement on fixed beds for F^{-} removal from groundwater and demonstrated how granules performed in rural Indian villages.

Lunge et al. (2011) have synthesized lanthanum-treated chitosan granules and applied them for groundwater monitoring through pre-packaged loose absorbents, cloth-in-pouches, and porous bamboo

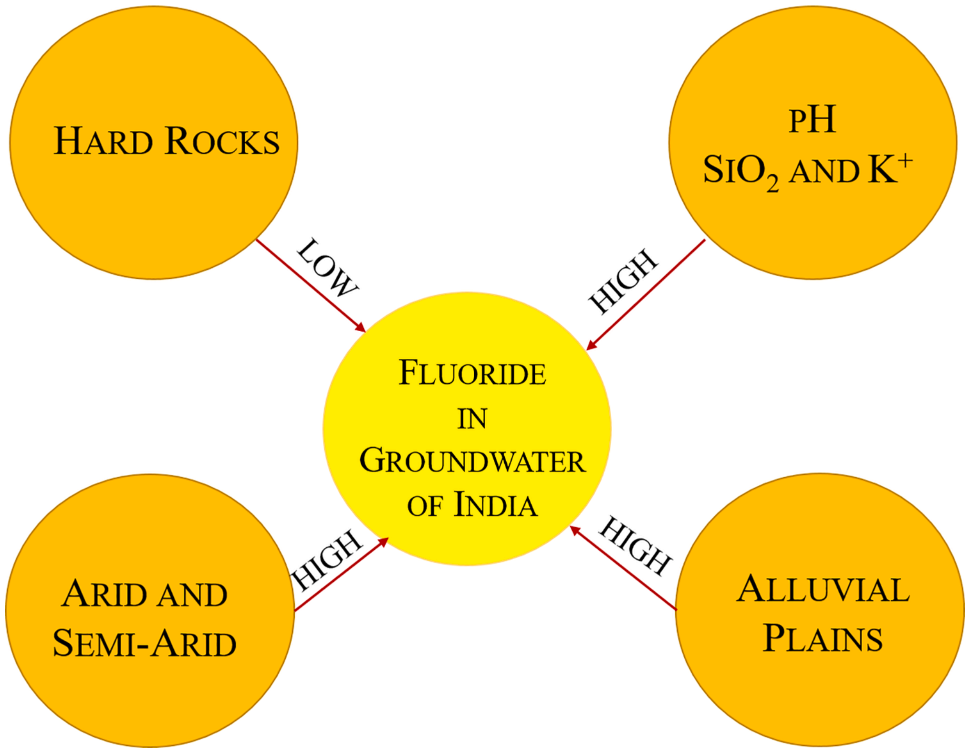

Fig. 5. Conceptual circular diagram showing major causes for high fluoride concentration in groundwater of India.

Table 2
Effects of fluoride consumption on human health (Ali 2016).

Fluoride content (mg/L)	Effect on humans
<0.5	Promotes dental caries
0.5–1.5	Required for strong bones and teeth
1.5–4.0	Dental fluorosis in children
>4.0	Dental and skeletal fluorosis
>10	Crippling skeletal fluorosis

pillars in rural areas of Madhya Pradesh. Lunge et al. (2011) reported that F^{-} levels were reduced to 1 mg/L for domestic utilities, such as drinking and cooking purposes, to avoid costly techniques and to implement in-situ applications. Feedback from end-users showed that implementing this technology was widely accepted at the community level in the Dhar district of Madhya Pradesh, India. Anjaneyulu et al. (2012) described that the F^{-} levels in rainwater collected from the upper surface were less than the desired limit/level of 1 mg/L and can be employed for cooking/drinking purposes. The current effort focuses on studies including the implications of "*solar distillation for defluoridation purposes*". In Bangalore, "*an inclined basin-type solar distillation setup*" consisting of sand-water was used for defluoridation based on Thomas's design (http://www.planetkerala.org/). Andey et al. (2013) summarized that NEERI, India, has developed electrolytic defluoridation (EDF) technology for treating excessive F^{-} in water sources. EDF technology offers a technologically sound, affordable, and dependable community drinking water defluoridation system for providing safe drinking water that complies with the WHO guideline value. Iyer et al. (2013) used state-of-the-art solar-based technologies – passive reverse osmosis units – or 77% of 100 installed units and also performed interactive surveys with residents and local officials. However, none of the Nalgonda technology or activated alumina units are currently in use due to high operational costs and skilled labour requirements.

Even though activated alumina has been found to remove F^{-}, and its long-term commercial uses are prevented due to bacterial buildup by its sluggish rate of adsorption and pH correction (Gill et al., 2014). The effectiveness of community-based defluoridation in avoiding severe fluorosis effect in Kaiwara village in the Indian state of Karnataka was evaluated by Isaac et al. (2020). After receiving ethical approval, this community interventional investigation was carried out in the hamlet of Kaiwara. This study highlights the significance of offering defluoridated water to people in villages as a potential fluorosis treatment. Agrawal et al., 2023 recently reported that defluoridation in rural regions of India could create potential employment opportunities. Sludge conversion into inexpensive construction raw low-cost materials can be a successful strategy for small-scale enterprises that uses both professional and unskilled labours. In this study, Agrawal et al. (2023) examined the methods for achieving this, which would give them much-needed job and transform it into a small-scale enterprise. Ramanarayanan et al. (2022) proposed that different adsorbents based on flowering trees have been employed to develop a defluoridation approach that is inexpensive and well-liked based on the adsorption process. The samples were subsequently filtered, and F^{-} levels were defined using a "*fluoride ion-specific electrode technique*". Kumar et al. (2023b) have recently utilized rice-husk biochar for F^{-} removal through batch and column sorption experiments, which showed efficient sorption at pH 7. Hand pumps and tube wells are frequently used in rural regions for cooking/drinking purposes, where biochar-mediated saturated fixed bed sand columns can effectively remove F^{-} from surface/groundwater at a larger scale.

Challenges and future research

Our literature review shows that high caring for teeth is not a cultural practice in the developing world. Therefore, the initial consequences are often ignored due to the unnoticeable immediate effects of F^{-}. Further, practical and feasible solutions for defluoridation methods are challenging. Membrane-based filtration is the most acceptable at present; however, the method is quite expensive and fails to provide water to large communities, mainly in India, and thus, only modern societies are benefited. Providing well locations with F^{-} level data by continuous monitoring will be a remarkable success in the reported countries. Now, there is a need to look for safe aquifers to avoid contaminated sources. The dilution of F^- content through recharge from rain, canal, or river water is evident in many studies. Therefore, the rejuvenation of water bodies can be highly helpful (Kalpana et al., 2019; Brindha et al., 2016).

Even though, there are marked contaminated wells in few regions, many wells are still installed and frequently used for drinking due to the lack of adequate water supply in India. With no other drinking water option, inhabitants are forced to drink the contaminated water. Therefore, consumption of contaminated water is frequent. Some local defluoridation methods are available, but most are undocumented and confined only to the local level. Its application on a wider scale is challenging. Therefore, providing sustainable water supply is challenging for the scientific community (Gutierrez et al., 2021). A remarkable achievement can only be achieved by involving local defluoridation methods, working with local people, and adopting practical and feasible science-based policy intervention measures over traditional methods.

Conclusions

In this study, the statistical correlation of F^- with other hydrochemical parameters was investigated based on the extensive dataset published in the last two decades for deciphering the genesis, mobilization, and contamination of F^- in the groundwater of India. The study suggests that the alkalinity of the groundwater facilitates the mobilization of F^- from sediments/rocks. It was observed that the elevated F^- levels in the groundwater of India is mainly confined to the alluvial areas falling in the arid and semi-arid regions, followed by the hard rocks regions. This study also revealed that the high F^- levels correlates well with SiO_2 and K^+, indicating F^- in groundwater is mainly derived from the weathering of fluorine-bearing silicates and, thus, possibly geogenic in nature. Further, this study highlighted the contaminated regions in India where safe water must be supplied for drinking for the inhabitants. Besides, defluoridation should be implemented at the grassroots level to treat high F^- groundwater for safe and clean drinking water and sustainable water supply. The policies intervention in contaminated areas in the developing world were often ineffective and had to be monitored more practically. To conclude, targeting uncontaminated aquifers for sustainable water supply and subsequent monitoring of drinking wells, and the willingness of the concerned authorities could be immensely helpful to mitigate the problem.

Ethical approval

All authors agreed and approved the final version of this manuscript.

Consent to participate

Not Applicable

Consent to publish

Not Applicable

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Availability of data and materials

The data can be retrieved from the mentioned studies.

A preprint server prior to submission has not been submitted elsewhere.

CRediT authorship contribution statement

Shakir Ali: Methodology, Investigation, Data curation, Formal analysis, Visualization, Writing – original draft, Writing – review & editing. **Shashank Shekhar:** Writing – review & editing. **Rakesh Kumar:** Writing – review & editing. **K. Brindha:** Writing – review & editing. **Peiyue Li:** Writing – review & editing.

Declaration of Competing Interest

The author declares that there are no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.hazadv.2023.100352.

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