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High levels of fluoride in groundwater from Northern parts of Indo-Gangetic plains reveals detrimental fluorosis health risks

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ABSTRACT

Fluoride contamination in groundwater is a worldwide phenomenon. Excess fluoride in drinking water causes serious health risks, and as a result, fluoride contamination of water resources is a global concern. In this study, an attempt has been made to provide the distribution of fluoride and related non-carcinogenic health hazards to local individual groups (males, females, and children separately) in the fluoride endemic region of Patiala, Punjab located in the Northern Indo-Gangetic Plain (IGP). The study shows that the dissolved groundwater fluoride concentration ranged between 1.5 and 9.2 mg/L with ~98% of the sampling locations having fluoride levels higher than the permissible limit. Samples collected from deeper aquifers (>284 m bgl) showed ~27% more fluoride contamination compared to those collected from <284 m bgl. Maximum incidence of elevated fluoride concentrations was observed in the eastern part of the study area in-sync with groundwater movement. The hazard quotient of fluoride ($HQ_{Fluoride}$) calculated to assess the non-carcinogenic health risk was higher than the unitary value in all individual groups suggesting a prevalence of distressful fluorosis and chronic health risk. Results show that the children are the most vulnerable to fluoride toxicity followed by males and females. Our results are consistent with the recent trends in an increase in dental, skeletal fluorosis, and liver functional damage problems reported in children and adults of the studied region. The study area, therefore, needs the urgent attention of policymakers and government agencies to implement proper water management and cost-effective fluoride remedial measures to reduce the current and future chronic health risks associated with high fluoride intake.

1. Introduction

Sustainable groundwater supply is among the most critical issues as 2.5 billion people worldwide are dependent on groundwater resources (Grönwall and Danert, 2020). However, groundwater availability and quality are constantly changing due to various natural and anthropogenic causes (Nizam et al., 2021; Rashid et al., 2021;, 2019, 2018; Shukla and Sen, 2021). Among the various water quality parameters fluoride is an important contaminant that causes serious health risks and has received significant attention (Rashid et al., 2020). It is well established that high levels of fluoride in groundwater mainly originate from the interaction of meteoric water and surface runoff with fluoride-bearing minerals (Kimambo et al., 2019; Marimon et al., 2007), hence, its origin is mostly geogenic in nature. In addition to the geogenic sources, fluoride can also be derived from anthropogenic sources such as chemical fertilizers, industrial effluents, sewage plant discharge, deposition from combustion sources, landfill leachate, and excessive groundwater pumping can also cause significant fluoride enrichment in groundwater (Iqbal et al., 2021; Podgorski et al., 2018; Rasool et al., 2017; Talpur et al., 2020).

Fluoride is classified as among the twelve most hazardous contaminants by the US Agency for Toxic Substances and Disease Registry (ATSDR, 2003) because of its high reactivity and toxicity. Ingestion of fluoride above the optimum level (>1.5 mg/L, (WHO, 2011) can cause severe dental and skeletal fluorosis in humans and animals (Dehghani et al., 2019; Sahu et al., 2017; Yousefi et al., 2019;, 2018a). Fluoride has the inherent capability of replacing the calcium content of teeth and bones making them fragile and ultimately causing osteoporosis, particularly in adult and old-aged persons (Ibrahim et al., 2011; Mohammadi et al., 2017). The health impacts of fluoride are not acute but chronic, which means that prolonged exposure to high fluoride doses is required to adversely affect human beings. Recent studies reported high levels of

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fluoride intake would cause detrimental health effects such as hypertension (Yousefi et al., 2018b), neurological defects and nervous system damage (Jiang et al., 2019), destructive gene effects (Cao et al., 2016), damage of kidney, liver, and thyroid gland (Yang et al., 2019), and may even cause carcinogenic disease in lungs, bones, bladder, and uterus (Alexander and Olsen, 2007; Yang et al., 2000). It is noteworthy to mention that an optimum amount of fluoride (0.5–1.5 mg/L) in drinking water is also essential to prevent the teeth from demineralization and plaque bacterial activity producing acid in the dental cavity. Considering the beneficial effects of fluoride, fluoridated drinking water has been supplied to the public as a natural cure for dental caries problems in several countries (Aoun et al., 2018).

Fluoride contamination of groundwater resources has been reported in more than 30 countries including India (Maliyekkal et al., 2006; Podgorski et al., 2018). In India, around 120 million people accounting for ~9% of the total population in India are at fluorosis risk due to groundwater fluoride consumption (Podgorski et al., 2018). A regional survey by Central Ground Water Board (CGWB, 2010) revealed 19 Indian states are severely contaminated with high levels of groundwater fluoride. The most affected groundwater fluoride endemic regions are the northwestern states of Rajasthan, Gujrat, Punjab, Haryana, and Delhi and southern states of Andhra Pradesh, Telangana, Karnataka, and Tamil Nadu (CGWB, 2010; Podgorski et al., 2018) where fluoride concentration is many folds higher than the maximum permissible limit (>1.5 mg/L).

This study is focused on the state of Punjab, India considered a global hotspot of groundwater fluoride contamination. The concentration ranges reported from various districts in the state of Punjab are as follows: 0.5–2.7 mg/L at Mansa (Sharma et al., 2021b), 0.05–1.38 mg/L at SBS Nagar (Mittal et al., 2020), 0.29–4.79 mg/L at Bathinda, 0.37–2.3 mg/L at Barnala and 0.08–2.75 mg/L at Ludhiana (Kumar et al., 2021), 1–3 mg/L at Bathinda and 0.3–2.5 mg/L at Faridkot (Sharma et al., 2021a), with the highest levels reported in the districts of Patiala (9.2 mg/L) and Fatehgarh Sahib (11.6 mg/L) (Virk, 2018). According to the annual water quality report from the State Government of Punjab, Patiala district is among the worst affected fluoride endemic region that documented 65% of the total fluorosis cases of Punjab due to groundwater fluoride intake (AWQR, 2021). Therefore, the aim of this study is to investigate the concentration and spatial distribution of fluoride in groundwater and its probabilistic health hazards to human individuals (male, female, and children) in the district Patiala, Punjab, India. Further, fluoride contamination and fluoride exposure risk are compared with major global hotspots to depict the current and future vulnerability of non-cacogenic and chronic health risks due to high fluoride intake in the local residents.

2. Materials and methods

2.1. Study area

The study area for the investigation is Patiala district, which is part of Northern IGP located in the Malwa region of Southern Punjab, India (Fig. 1). It has an aerial coverage of about 3218 km^2 lying between 29^o 49' and 30° 40' N latitudes and 75° 58' and 76° 48' E longitudes. The land surface has a gentle slope of 0.8 meters per kilometer in the northeast-southwest direction. Geology of the Patiala district mainly includes Indo-Gangetic alluvium comprising of sandy clay extending up to an average depth of ~4.2 m followed by 9 m thick hard clay resting on 17.9 m thick coarse grey sand body (Kumar et al., 2007). The composition of the subsurface lithology, however, is heterogeneous in character consisting of gravel, pebble, and kankar deposited by the Ghaggar River. Groundwater is mainly contained in sand and gravel under both phreatic and confined conditions up to the depth of 49–400 m below ground level (bgl) (CGWB, 2013). Depth of the water table, in general, is shallower in the vicinity of the canals and rivers varying between 4.4 and 20.6. m bgl. The subsurface water movement in Patiala follows the

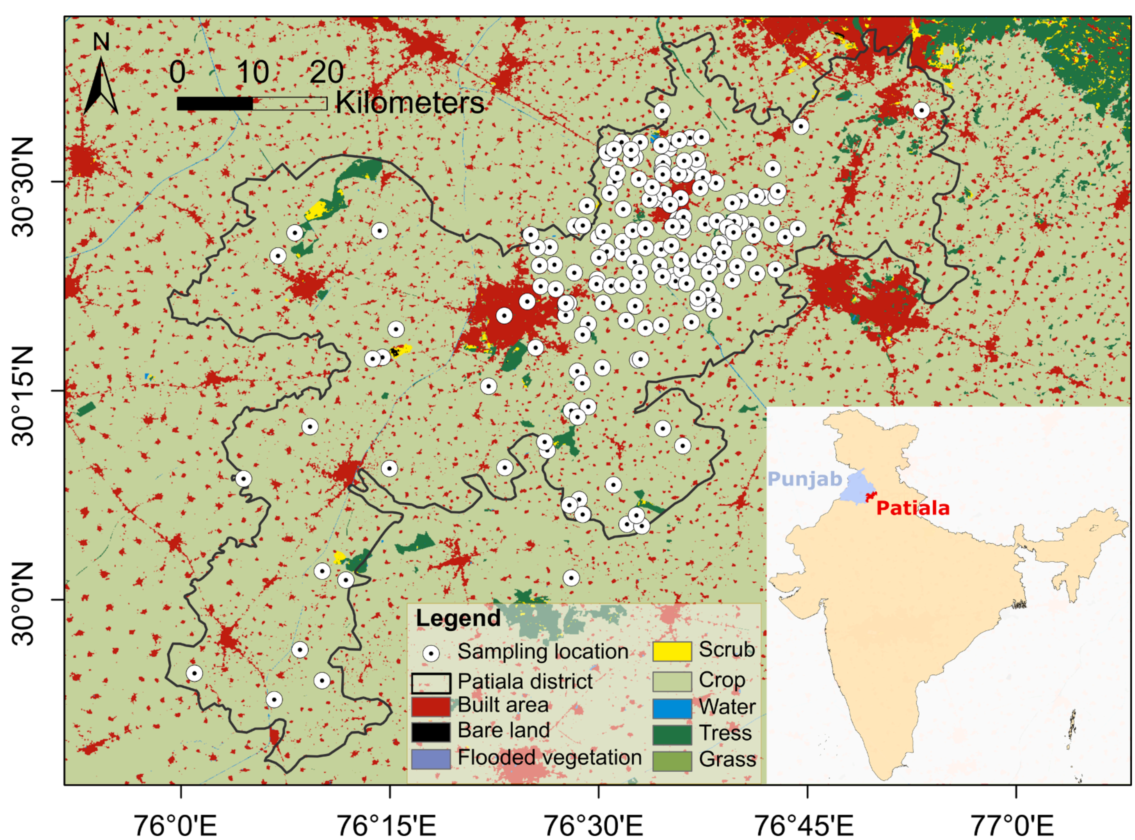

Fig. 1. Land-use and land-cover map showing groundwater sampling location in Patiala district, Punjab, India.

direction of the land surface slope (CGWB, 2013). The local climate of Patiala district is tropical steppe, semi-arid having very hot summer and cold dry winter except during monsoon. The total population of the Patiala districts is ~1.8 million (2011 Census) which is mainly dependent on agriculture. About 82% of the total area of the Patiala district is under intensive agricultural cultivation, while 4% is under forest cover, and the rest 16% is available for of non-agricultural use (CGWB, 2013). The main recharge sources of underground aquifers in the study area are meteoric water, irrigation canals and rivers (Joshi et al., 2018). Artificial recharge and rainwater harvesting programs have been also introduced in Patiala to cope with the rapid drawdown of groundwater level (CGWB, 2013; Singh and Virdi, 2015; WRED and CGWB, 2018).

2.2. Sampling details and fluoride analysis

Total 173 samples of groundwater were collected following random technique from shallow and deep aquifers from different localities in the Patiala district of Punjab, India (Fig. 1). Briefly about 200 ml of samples were collected directly in precleaned high-quality polyethylene (HDPE) bottles from tube wells and hand pumps installed at a varying depth ranging between 43 and 443 m bgl. Prior to sample collection, tube wells and handpumps were flushed out for 5–10 min to minimize the standing water interference inside the metal casing. Each sampling bottle was rinsed thrice to remove any background contamination. Samples were filtered through a 0.45 µm Millipore filter and collected in pre-cleaned 250 ml HDPE bottles and capped firmly to avoid interference with atmospheric CO_2. All the samples were stored in a dark place at 4 °C until the laboratory analysis.

Dissolved fluoride concentration in groundwater samples was analyzed using Ion Chromatograph at the Punjab Water Supply and Sanitation Department (PWSSD) Laboratory, Mohali (Punjab). The detection limit of the instrument was calibrated at 0.1 mg/L. Data quality and reproducibility of the analysis was monitored by running calibration standard (from Sigma Aldrich) of 5.0 mg/L at regular interval and the average value obtained was 5.14 ± 0.26 ($n = 8$, 1 SD, RSD = 0.18). Ten replicates were measured to monitor the reproducibility of the analysis and the results showed an average reproducibility of 101 ± 4% ($n = 10$, 1 SD). The overall accuracy of the standard analyzed was <3% with a precision better than ≤2%. The analysis results of all the samples are shown in Table S1.

2.3. Human exposure health risk assessment

Fluoride can enter the human body through three different pathways: ingestion (food, and drinking), inhalation (breathing), and dermal contact. However, drinking is the major pathway of groundwater fluoride exposer accounting for about 75–90% of the total intake (Fawell et al., 2006; WHO, 2010). Moreover, the non-carcinogenic fluoride health effect associated with inhalation and dermal exposure routes is negligible (Mukherjee et al., 2019). Therefore, health risk due to ingestion of excess fluoride through drinking water was estimated following the standard protocol (USEPA, 1993). In this method, first, Chronic Daily Intake (CDI) of fluoride in an individual is calculated using the equation given as follows:

$$CDI(mg/kg/d) = \frac{[F_c \times DID \times TEF \times ED]}{[ABW \times AET]} \quad (1)$$

where Fc: Fluoride content in groundwater samples (mg/L); DID is Daily: Ingestion Dose of drinking water (L/day); TEF: Total Exposure of drinking water; ED: Exposure Duration; ABW: Average Body weight; and AET: Average Exposure Time (calculated as the product of the number of years and number of days) and the values of the parameters are provided in Table 1. Finally, fluoride health impact due to ingestion of fluoride contaminated water which is a non-carcinogenic health risk is calculated in terms of Hazard Quotient of fluoride ($HQ_{Fluoride}$) using the following

Table 1
Numerical values of the parameters used in the calculation of the fluoride health risks.

Parameter	Physical significance	Values	Units	Reference
F_c	Fluoride concentration	1.5 - 9.2	mg/L	This study
DID	Daily ingestion dose	Males: 4 Females: 3 Children: 1	L/day	Naz et al. (2016)
TEF	Total exposure frequencies	365	day/year	Ahada and Suthar (2019), USEPA (1999)
ED	Exposure duration	Males: 64 Females: 67 Children: 12	year	WHO (2013)
ABW	Average body weight	Males: 65 Females: 55 Children: 15	kg	ICMR (2009)
AET	Average exposure time	Males: 23360 Females: 24455 Children: 4380	day	WHO (2013)

equation:

$$HQ_{Fluoride} = \frac{CDI}{RfD} \quad (2)$$

Where RfD is the reference dose for chronic oral exposure of fluoride taken as 0.06 mg/kg/d prescribed by USEPA (1993). Calculated CDI and $HQ_{Fluoride}$ results calculated for human individual are given in Table S2.

3. Results and discussion

3.1. Fluoride concentration and spatial distribution

Dissolved fluoride in the groundwater samples collected from Patiala varies between 1.5 and 9.2 mg/L with an average value of 2.8 ± 1.6 mg/L ($n = 187$, 1 SD, Fig. 2a). Surprisingly, 98% of the total samples analyzed exhibit fluoride concentration above the 1.5 mg/L drinking water specification set by the Indian standard (BIS, 2012).In general, fluoride concentration of 1.5–2 mg/L is common throughout the study region, whereas, higher groundwater fluoride levels (> 2.5 mg/L) are mainly concentrated in eastern parts of the study area with few discrete patches in central and western parts. According to the 2021 Punjab Government annual report (AWQR, 2021), the southern part (Malwa Region) of Punjab is most vulnerable to groundwater fluoride contamination. Fluoride concentration in districts of Northern Punjab (Majha and Doaba region) is below the BIS guideline for drinking water but exceeded the permissible limit in several districts (Bathinda, Faridkot, Fatehgarh Sahib, and Patiala) of Southern Punjab (Fig. 2b) with the largest incidences of fluoride contamination observed in the Patiala district (AWQR, 2021). Groundwater fluoride contamination in Patiala district has drastically increased over the past two decades. For instance, groundwater fluoride concentration in Patiala was within the permissible limit (range: 0.06–0.66 mg/L) during the year 2003 (Kumar et al., 2007), later exceeded 1.5–9.8 mg/L in 2021, and posed a serious threat to local residents' health and the ecosystem (Ahada and Suthar, 2018; Gupta et al., 2014; NAPCC, 2011), The ranges and mean concentration of fluoride observed in groundwater of the Patiala district is comparable or higher than the fluoride endemic hotspots of the South Asia (viz. Bangladesh, China, Iran, Pakistan, Vietnam including India) and Europe (Sweden and Tunisia) (Table 1). The mean fluoride concentration is highest across the IGP despite of similar subsurface geology and climatic condition (Mukherjee et al., 2020; Samal et al., 2020; Yadav et al., 2019). In contrast, groundwater from Africa (Kenya and Tanzania), North America (Mexico), and South America (Argentina) exhibit much higher fluoride content due to hot climate and distinct subsurface lithology enriched in fluoride bearing minerals. However, groundwater

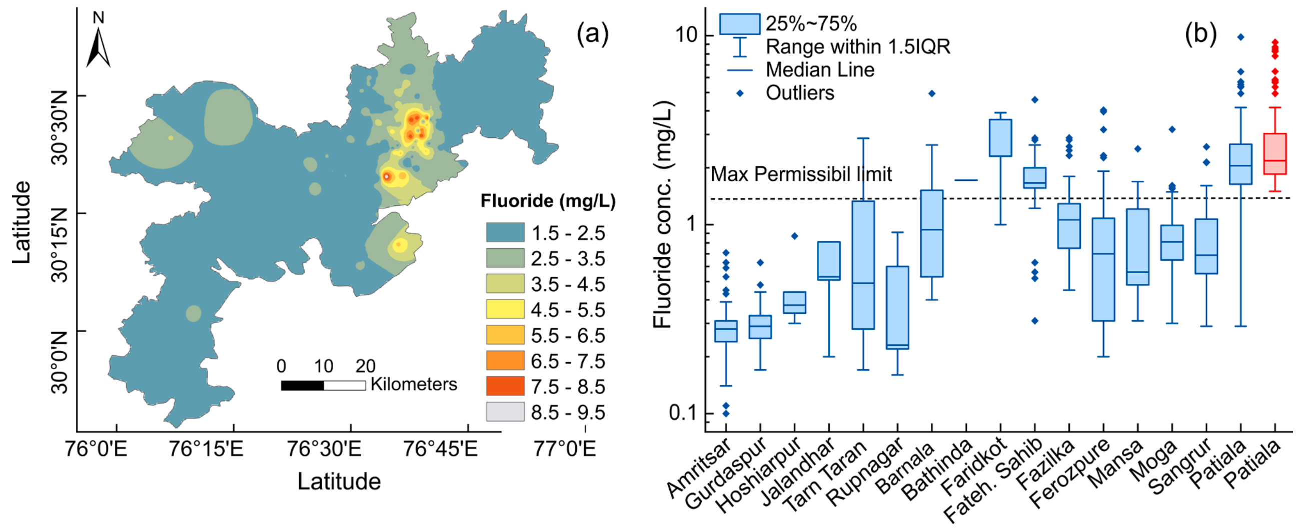

Fig. 2. Spatial distribution of (a) dissolved fluoride concentration in Patiala district and (b) comparison of average groundwater fluoride levels with other districts of Southern Punjab, India.

fluoride concentration in parts of the South Asian region viz. Faryab in Afghanistan may exceed up to 79 mg/L (4.2 ± 8.4), even higher than present study and as well as Africa and America due to arid climate and diversified geological and anthropogenic factors (Hayat and Baba, 2017). It is interesting to note that fluoride contamination is more persistent in Patiala compared to all regional and global sites as all of the groundwater samples exhibit similar or higher than the threshold value (1.5 mg/L) of the safe drinking water.

The significant spatial variation observed in groundwater fluoride concentration in the present study suggests that the heterogeneity in fluoride distribution could be due to varying source input, and different controlling factors, viz. local hydrogeology, subsurface lithology and redox condition, pH and dissolution and precipitation of F-bearing minerals as well as rain water composition (Ali et al., 2019; Nizam and Sen, 2018; Rasool et al., 2017). Groundwater sampling depth recorded during the sampling plotted against the dissolved fluoride revealed significant but limited control of depth on fluoride content of the aquifer systems of the Patiala region (Fig. 3). Several groundwater samples exhibited high fluoride concentration collected from deeper aquifers. For instance, groundwater sampled from >284 m bgl in Sahal, Chatar Nagar, Gandian, Ghagar Sarai, Salempur Jattan and Salempur Sekhan

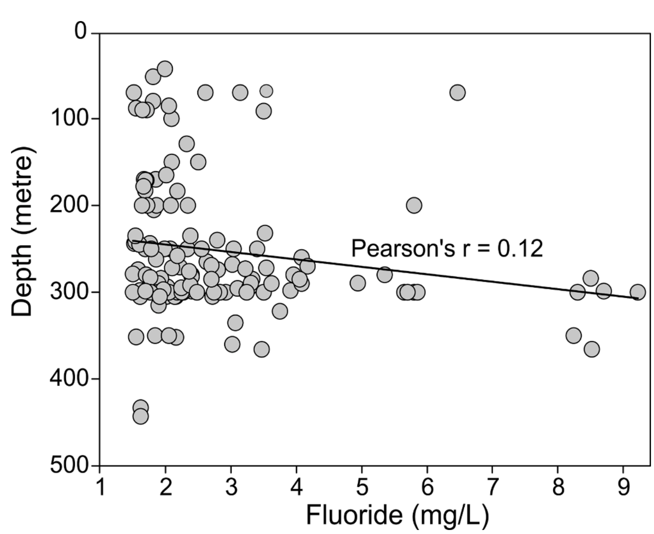

Fig. 3. Relationship between fluoride concentration and sampling depth of groundwater (aquifer) in Patiala district, Punjab, India.

localities have very high fluoride concentrations (>6.5 mg/L). This coupled with the weak positive Pearson correlation ($R = 0.18, p < 0.05$) between fluoride concentration and sampling depth suggests the fluoride vulnerability in deeper aquifers is much likely attributed to downward migration of the anthropogenic/geogenic contaminants as observed in Bist Doab region in Northwest India, and central part of the IGP (Kumar et al., 2019; Lapworth et al., 2017). Nevertheless, a significant population of groundwater samples collected from shallow aquifers (depth: 70 m bgl) also contain high fluoride concentration (2.6 mg/L at Akbarpur) apprehends the presence of additional local factors over geogenic control in groundwater fluoride enrichment. For instance, prolonged application of pesticides and phosphate fertilizers in agricultural production activates fluoride leaching from the cultivation field to the underlying groundwater system which can be distributed through groundwater movement (Kabata-Pendias and Pendias, 2000). Most incidences of elevated fluoride concentration observed in the southeastern part of Patiala are consistent with the transfer of fluoride through groundwater movement along the gradient of the land surface elevation. Therefore, excessive groundwater abstraction for irrigation and intensive fertilizer application for enhanced agricultural production, which is the main industrial venture in the Patiala region could have enhanced massive fluoride leaching to the groundwater under favorable oxygenated environment (Ahada and Suthar, 2018; Mohapatra et al., 2021).

3.2. Non-carcinogenic health risk due to excess fluoride intake

The persistence of high levels of fluoride beyond the permissible limit observed in most of the groundwater samples in the study area makes it inadequate for drinking purposes due to the negative health impact of fluoride contamination in drinking water (Agnihotri et al., 2014). Fig. 4.a summarizes the health effects on the local population in the study region depending upon different levels of fluoride exposure through groundwater ingestion. None of the samples fall in class-I suggesting that there is no risk of dental decay problems. Similarly, there is no risk of crippling fluorosis associated with Class-V in the study area. There are only 3 samples accounting for about 2% of the total samples belonging to Class-II having fluoride concentration within the optimum requirement (0.5–1.5 mg/L) for good human health. In contrast, Class -III and Class-IV together constitute the largest population (~98%) of the groundwater samples implying that prolonged ingestion of groundwater may pose detrimental health risk such as mottling of teeth, crippling of bones, calcification of ligaments, and other neural and hormonal problems to the local individual soon in near the future

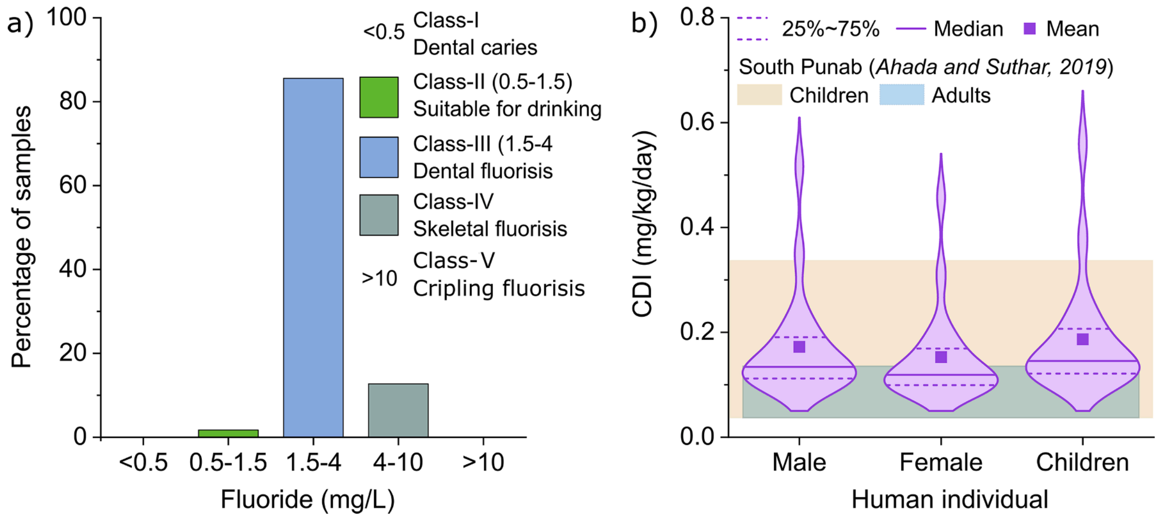

Fig. 4. (a) Effect of the fluoride toxicity due to different levels of fluoride intake. (b) Statistical summary of chronic daily intake (CDI) of fluoride compared to CDI range for adults and children estimated for groundwater samples collected from the entire Malwa Region, South Punjab, India (Ahada and Suthar, 2019).

(Fawell et al., 2006).

Chronic daily intake (CDI) of fluoride through groundwater ingestion in the Patiala region was in the ranges of 0.09–0.57, 0.08–0.50, and 0.10–0.61 mg/kg/day for males, females, and children, respectively (Fig. 4b). The maximum CDI value observed in both the adults and children at Salempur Jattan locality is consistent with the highest fluoride concentration (9.22 mg/L) recorded in groundwater samples of this region. Similarly, groundwater samples from other localities namely Bhatonia Kalan, Gandian, Chatar Nagar, Ghagar Sarai, and Salempur Sekhran contained higher fluoride concentration (>7 mg/L) also showed very high CDI value >0.4 mg/kg/day. The CDI range for human individuals recorded in this study is significantly higher than those reported from the entire Malwa region (0.04–0.14 mg/kg/day), which makes Patiala the main fluoride hotspot in Punjab (Ahada and Suthar, 2019). Significant differences in the CDI estimates of the Patiala region in the current study and previous findings could be due to differences in the sampling depth as the previous study results were based on limited samples collected from shallow aquifers ranging between 7 and 41 m bgl.

For a better assessment of non-cariogenic fluoride health risk to the human individual residing in the Patiala district, the magnitude of hazard index i.e., $HQ_{Fluoride}$ was calculated for different age groups (Fig. 5). $HQ_{Fluoride} \geq 1$ means there is a high chance of development of fluoride-induced health hazards to the individuals living in the fluoride endemic region, whereas $HQ_{Fluoride} < 1$ suggests no health risk. The range of $HQ_{Fluoride}$ values in the study region was 1.5–9.5 (mean: 2.9), 1.4–8.4 (mean: 2.5) and 1.7–10.2 (mean: 3.1) for males, females, and children, respectively. The $HQ_{Fluoride}$ range is up to 3 times higher for both children (0.67–5.63) and adult (0.29–2.41) compared to those reported in southern Punjab (Ahada and Suthar, 2019). Similarly, the groundwater fluoride health risk to human individuals in the current study is higher or similar to most of the global fluoride endemic hotspots viz. China (Chen et al., 2017), Iran (Yousefi et al., 2018a), Kenya (Mwiathi et al., 2022), Pakistan (Noor et al., 2021), Mexico, (Fernández-Macias et al., 2020), Tunisia (Guissouma et al., 2017), and Vietnam (Nguyen et al., 2021) including Northern and Sothern India (Ahada and Suthar, 2019; Kumar et al., 2019; Shukla and Saxena, 2020). However, largest magnitude of the $HQ_{Fluoride}$ observed for children followed by males and females (respectively) implying most risk vulnerability to children than the adults is consistent with all aforementioned studies. In general, individuals residing in fluoride endemic regions with excess groundwater fluoride concentrations are more prone to fluoride toxicity. Greater fluoride exposure health risks in children compared to adults are usually attributed to the lower body size of the children that accumulates more contaminants (He and Wu, 2019). The $HQ_{Fluoride}$ in the current study exceeded the unitary value in all of the groundwater samples for both the adults and children demonstrating that the local inhabitants in the district of Patiala are under distressful fluorosis health problems. However, the severity of the fluoride toxicity may vary depending on its concentration in groundwater, ingestion rate, the longevity of fluoride exposure as well as local climate (air temperature: a key driver of daily water intake). For instance, exposure to groundwater fluoride levels above the safe limit led to the development of dental fluorosis (1.5–3 mg/L), skeletal fluorosis (3–6 mg/L), and bone crippling (>6 mg/L) in human individuals (WHO, 1994). Prolonged uptake of low level of fluoridated water (0.8 mg/L) can also lead to the development of dental fluorosis (Brouwer et al., 1988).

Non-carcinogenic fluoride exposure risks to human individuals estimated in the Patiala region in the current study are consistent skeletal and non-skeletal fluorosis (collectively known as hydrofluorosis) problems reported in the fluoride endemic Malwa region of south Punjab (Ahada and Suthar, 2019). For instance, dental fluorosis survey data of school children (<18 years) and adults from fluoride endemic districts, viz. Bathinda and Patiala revealed the widespread prevalence of dental fluorosis of varying grades. Both children and adults from Bathinda showed the development of white striation and opaque-yellow patches residing in areas with normal groundwater fluoride levels whereas dark brown patches with structural damage in dental enamels were observed in individuals residing in high fluoride areas (Chahal and Chahal, 2016a, 2016b). Compared to adults, children and teenagers showed more cases of dental fluorosis than skeletal fluorosis because of developing teeth that require fluoride for dental growth, but higher concentrations disturb the tooth enamels. Similarly, 40% of the total 1600 children examined in Patiala city showed the prevalence of dental caries problems (Kaur et al., 2020). The fluorosis problem in the Patiala district is expected to worsen in the future since 80% of its total villages had groundwater fluoride concentrations higher than the permissible limit (Singh et al., 2021). This has been reflected as the largest number of hydrofluorosis cases (65% of total Punjab) reported in the year 2021 in Patiala (AWQR, 2021). Large dental fluorosis problems due to high fluoride intake in children compared to adults has been reported (~60 and 31% of the children) in China and Pakistan, respectively (Chen et al., 2017; Rashid et al., 2020). Similarly, ~9% of the total Indian population are at fluorosis risks among which, 62 million population are children (Podgorski et al. 2018, and references therein). In Tunisia, two third of the population is facing dental caries problems whereas 25% of

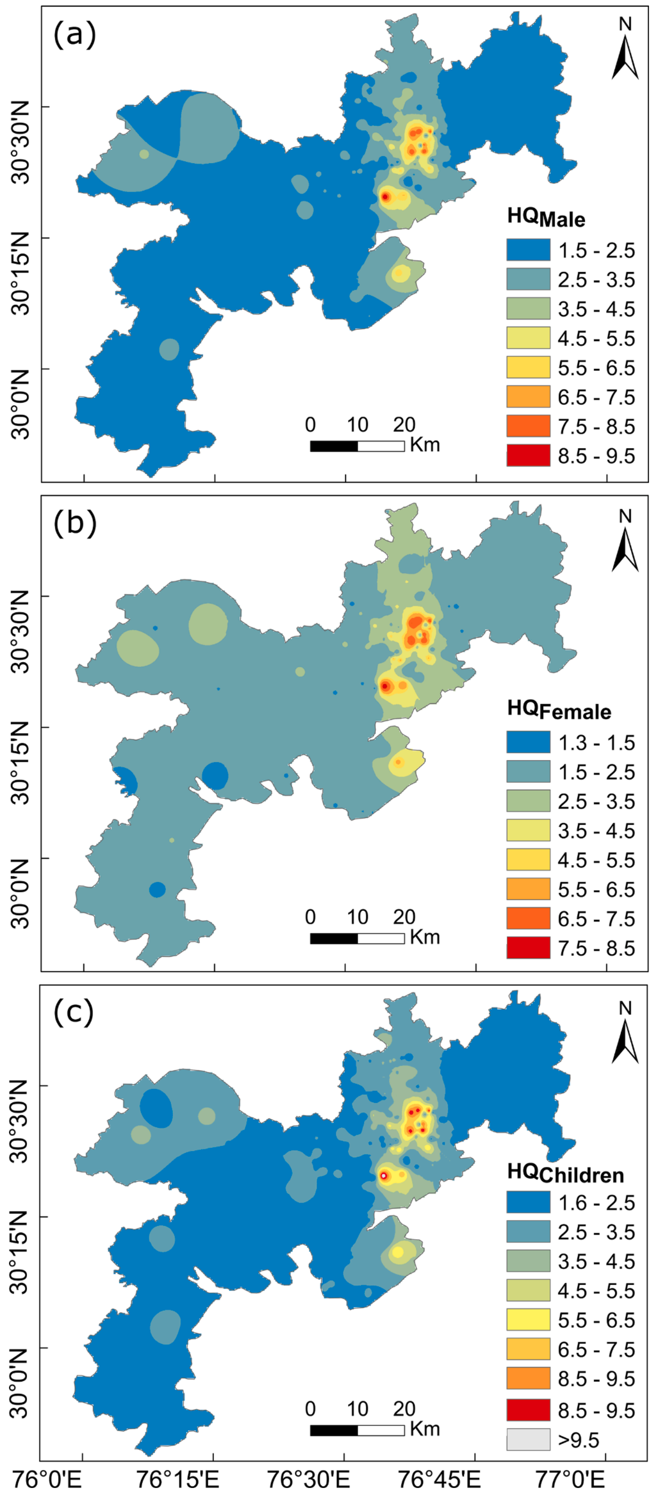

Fig. 5. Summary of fluoride exposure health risk ($HQ_{Fluoride}$) on (a) male (b) female and (c) children due to groundwater ingestion in Patiala district, Punjab, India.

the population is living under dental fluorosis risk (Guissouma et al., 2017). Prevalence of fluoride concentration in excess of 4 mg/L in 23% of the total groundwater samples in the study region further suggests skeletal risk particularly to the adults living in the study region, For instance, individuals exposed to high groundwater fluoride comparable to our region in Iran reported ~18% higher skeletal fluorosis cases than those residing in region with low fluoride concentration (Mohammadi et al., 2017). Recent findings from central India revealed about 8% of the population suffering from skeletal fluorosis due to high groundwater fluoride (4–4.5 mg/L) intake (Sonali et al., 2013). Similarly, increased skeletal fluorosis stress has been observed all over the globe and reported in fluoride endemic hotspots of the 24 countries including India (Srivastava and Flora, 2020 and references therein). Moreover, domestic animals in the fluoride endemic regions have been reported to be affected by hydrofluorosis risks in several states of India but underexplored in the case of Punjab, which is among the largest domestic animals users in agricultural sectors of India (Choubisa, 2018). Therefore, groundwater fluoride intake is one of the major potential causes of the public health deterioration in the study region.

3.3. Implication for chronic health risk

Long-term ingestion of high fluoride often gives rise to chronic health disease, viz. respiratory failure, liver damage, paralysis, blood pressure problem, and chronic fluoride poisoning that causes anaemia, cachexia, and weight loss (Ibrahim et al., 2011). High fluoride intake also adversely affects the male's fertility and reproduction (Ortiz-Pérez et al., 2003). For instance, decreased birth rate with increasing fluoride concentration reported in 30 regions of the U.S. having groundwater fluoride concentration in excess of 3 mg/L (Freni, 1994). Similarly, fluoride uptake of 2–4 mg/L negatively affects children's visuos-patial abilities and reaction times that cause lower Intelligence Quotient (IQ) scores in real-time tests (Aravind et al., 2016; Saxena et al., 2012; Wang et al., 2007). In addition, few studies showed bone cancer (osteosarcoma) and neurotoxicity mainly in children (compare to adults) due to excess ingestion of fluoride. Although such cases are not common, further research is required particularly in regions like Patiala where both shallow and deep aquifers are severely contaminated. Moreover, a recent study examined the relationship between fluoride and liver function in adults from normal and seven fluoride endemic regions of Punjab (including Patiala) found an increased alteration in liver functions due to cellular damage in fluorotic patients living in fluoride endemic regions (Shashi and Bhardwaj, 2011). Closure resemblance in fluoride concentration and hydrofluorosis problems in human indicate that the situation would become alarming in near future due to widespread occurrence of high levels of fluoride in groundwater systems of Patiala as well as other regions of Southern Punjab and need urgent attention to implement proper water management plan to cope with the current scenario of fluoride contamination.

Table 2.

4. Conclusions

This study gives an overview of the high resolution (depth wise) spatial distribution of groundwater fluoride concentration and its human health hazards in one of the endemic fluorosis regions (Patiala district) of the Northern IGP, India. The fluoride level in groundwater samples was up to six-fold higher than the maximum permissible limit prescribed by WHO and BIS. Of all the groundwater samples analyzed, about 98% of samples showed fluoride concentrations >1.5 mg/L. Aquifers at deeper depth show more contamination than at shallower depth plausibly due to downward movement of the contamination through the subsurface. Overall, fluoride contamination observed in groundwater of the Patiala district is comparable or higher than most of the fluoride endemic hotspots of the South Asia (except few local hotspots in Afghanistan) and Europe including India, but lower than African and American countries. Groundwater fluoride concentration and probabilistic health risk in the local individual groups (male, female, and children) marked by using GIS mapping revealed persistent fluoride health hazard. The $HQ_{Fluoride}$ index shows children and teenagers are most vulnerable to fluoride toxicity (and health risk) followed by males and females, which is consistent with most of the previous studies across the globe. About 23% of the samples contain >4 mg/L fluoride and revealed high probable risk of skeletal fluorosis and other chronic health

Table 2
Statistical summary of the dissolved fluoride concentration in groundwater of the study are and its comparison with some of the major fluoride endemic hotspots of India and world.

Sampling location	Year of sampling/study	Min (mg/L)	Max (mg/L)	Average ± 1 SD (mg/L)	Total samples analyze	Percentage of samples > WHO limit	References
Patiala, Northern India	2016	1.5	9.2	2.8 ± 1.6	187	98	This study
Jamui, Eastern India	2014	0.01	5.8	0.98–1.38	119	18–34	Kumar et al. (2019)
Raebareli, Central, India	2016-17	0.13	8.28	1.85–2.04	28	43	Shukla and Saxena (2020)
Siddipet, Southern India	2016	0.4	4.2	1.26–2.2	158	31–80	Narsimha and Rajitha (2018)
Dhaka, Bangladesh	2015-2017	0.01	16.1	0.53–0.80	840	4–7	Rahman et al. (2020)
Dargai, Northern Pakistan	2020	0.5	8.65	2	75	51	Rashid et al. (2020)
Poldasht, Northwest Iran	2014-2016	0.23	10.3	1.7	112	57	Yousefi et al. (2018a)
Faryab, Afghanistan	2013	0.02	79	4.2 ± 8.4	380		Hayat and Baba (2017)
Ningxia Hui, China	2021	0.17	5.1	0. 81	144	22	Liu et al. (2021)
Zhongning, China	2012	0.11	6.33	0.85 ± 1.14	50	50	Chen et al. (2017)
Xuyen Moc, Vietnam	2017	0	16.8	1.7–2.3	14	14	Nguyen et al. (2021)
Sweden	1998-2007	0.1	15	1	4800	24	Erdal and Buchanan (2005)
Tunisia	2014	0.05	2.4	0.12–2.08	100		Guissouma et al. (2017)
San Luis Potosí, Mexico	2019	0.2	3.5	0.6–1.75	35	50	Fernández-Macias et al. (2020)
La Pampa, Argentina	2011	0.5	14.2	3.1–4.2	44	78-100	Aullón Alcaine et al. (2020)
Northern Tanzania	2014-2016	0.01	74	3.36 ± 6.4	507	42	Ijumulana et al. (2020)
Nakuru, Kenya	2018-2019	0.01	23.5	3.35	32	40	Mwiathi et al. (2022)

Note: Empty cell indicate data not reported and average value in ranges refer to average reported for different season and or region.

diseases with continued consumption of the untreated groundwater intake in the Patiala region. Therefore, this study provides valuable data that will help the government and water management agencies in the formulation of better policies and remedial measures to protect the local human individuals exposed to groundwater in the studied region.

5. Limitation and future research outlook

This study provides a comprehensive overview of the fluoride distribution in shallow and deep aquifers and associated fluorosis and chronic health risk to human individuals in parts of the Northern IGP. The study however lacks information on visual health effect. Therefore, local surveys are required to be conducted to document the effect of the groundwater in local residents and animals due to varying ground fluoride ingestion. Moreover, the average groundwater fluoride concentration in the study region is comparable or higher than most of the fluoride endemic regions of India and other parts of the world despite the fact that the region is exclusively underlain by alluvium with no fluoride rich formation in the basement. It is therefore critical to constrain the mechanism and role of different factors (natural versus anthropogenic) driving such a high fluoride release in the aquifer systems of the Patiala district for better implication of preventive measures.

CRediT authorship contribution statement

Sarwar Nizam: Conceptualization, Writing – original draft, Writing – review & editing. **Hardev Singh Virk:** Data curation, Methodology, Formal analysis, Funding acquisition, Writing – review & editing. **Indra Sekhar Sen:** Data curation, Methodology, Formal analysis, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Full data and supporting information are available as supplementary material.

Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.envadv.2022.100200.

References

Agnihotri, N., Pathak, V.K., Khatoon, N., Rahman, M., 2014. Hydrochemical assessment and factor analysis of groundwater with special reference to fluoride in Kanpur Dehat, U.P., India. IOSR J. Appl. Chem. 7, 52–56. https://doi.org/10.9790/5736-07315256.
Ahada, C.P.S., Suthar, S., 2019. Assessment of human health risk associated with high groundwater fluoride intake in Southern districts of Punjab, India. Expo. Health 11, 267–275. https://doi.org/10.1007/s12403-017-0268-4.
Ahada, C.P.S., Suthar, S., 2018. Assessing groundwater hydrochemistry of Malwa Punjab, India. Arab. J. Geosci. 11 https://doi.org/10.1007/s12517-017-3355-8.
Alexander, B.H., Olsen, G.W., 2007. Bladder cancer in perfluorooctanesulfonyl fluoride manufacturing workers. Ann. Epidemiol. 17, 471–478. https://doi.org/10.1016/j.annepidem.2007.01.036.
Ali, S., Fakhri, Y., Golbini, M., Thakur, S.K., Alinejad, A., Parseh, I., Shekhar, S., Bhattacharya, P., 2019. Concentration of fluoride in groundwater of India: a systematic review, meta-analysis and risk assessment. Groundw. Sustain. Dev. 9, 100224 https://doi.org/10.1016/j.gsd.2019.100224.
Aoun, A., Darwiche, F., Hayek, S.Al, Doumit, J., 2018. The fluoride debate: the pros and cons of fluoridation. Prev. Nutr. Food Sci. 23, 171–180. https://doi.org/10.3746/pnf.2018.23.3.171.
Aravind, A., Dhanya, R.S., Narayan, A., Sam, G., Adarsh, V.J., Kiran, M., 2016. Effect of fluoridated water on intelligence in 10-12-year-old school children. J. Int. Soc. Prev. Commun. Dent. 6, S237–S242. https://doi.org/10.4103/2231-0762.197204.
ATSDR, 2003. Agency for toxic substances and disease registry Toxicological profile for fluorine, hydrogen fluoride, and fluorides. 10.1201/9781420061888_ch86.
Aullón Alcaine, A., Schulz, C., Bundschuh, J., Jacks, G., Thunvik, R., Gustafsson, J.P., Mörth, C.M., Sracek, O., Ahmad, A., Bhattacharya, P., 2020. Hydrogeochemical controls on the mobility of arsenic, fluoride and other geogenic co-contaminants in the shallow aquifers of northeastern La Pampa Province in Argentina. Sci. Total Environ. 715, 136671 https://doi.org/10.1016/j.scitotenv.2020.136671.
AWQR, 2021. Annual water quality report 2021 department of water supply and sanitation Government of Punjab. availaibale at https://dwss.punjab.gov.in/downloads/.
BIS, 2012. Indian standard drinking water specification (second revision). Bur. Indian Stand. IS 10500, 1–11.
Brouwer, I.D., De Bruin, A., Dirks, O.B., Hautvast, J.G.A.J., 1988. Unsuitability of World Health Organisation guidelines for fluoride concentrations in drinking water in Senegal. Lancet 331, 223–225. https://doi.org/10.1016/S0140-6736(88)91073-2.
Cao, J., Chen, Y., Chen, J., Yan, H., Li, M., Wang, J., 2016. Fluoride exposure changed the structure and the expressions of Y chromosome related genes in testes of mice. Chemosphere 161, 292–299. https://doi.org/10.1016/j.chemosphere.2016.06.106.
CGWB, 2013. Groundwater information booklet Patiala district, Punjab. Chandigarh.

CGWB, 2010. Ground water quality in shallow aquifers of India. Central Ground Water Board and Ministry of Water Resources Government of India.

Chahal, R.P.S., Chahal, P.K.P., 2016a. Incidentce of dental fluorosis among children of Bathinda District in the Punjab State. J. Adv. Med. Dent. Sci. Res. 4, 7–9.

Chahal, R.P.S., Chahal, P.P.K., 2016b. Effect of fluoride content of drinking water on dental fluorosis in the Punjab. J. Adv. Med. Dent. Sci. Res. 4, 4–6.

Chen, J., Wu, H., Qian, H., Gao, Y., 2017. Assessing nitrate and fluoride contaminants in drinking water and their health risk of rural residents living in a Semiarid region of Northwest China. Expo. Health 9, 183–195. https://doi.org/10.1007/s12403-016-0231-9.

Choubisa, S.L., 2018. A brief and critical review on hydrofluorosis in diverse species of domestic animals in India. Environ. Geochem. Health 40, 99–114. https://doi.org/10.1007/s10653-017-9913-x.

Dehghani, M.H., Zarei, A., Yousefi, M., Asgharia, F.B., Haghighat, G.A., 2019. Fluoride contamination in groundwater resources in the southern Iran and its related human health risks. Desalin. Water Treat. 153, 95–104. https://doi.org/10.5004/dwt.2019.23993.

Erdal, S., Buchanan, S.N., 2005. A quantitative look at fluorosis, fluoride exposure, and intake in children using a health risk assessment approach. Environ. Health Perspect. 113, 111–117. https://doi.org/10.1289/ehp.7077.

Fawell, J., Bailey, K., Chilton, J., Dahi, E., Fewtrell, L., Magara, Y., 2006. Flouride in Drinking-water. IWA Publishing.

Fernández-Macias, J.C., Ochoa-Martínez, Á.C., Orta-García, S.T., Varela-Silva, J.A., Pérez-Maldonado, I.N., 2020. Probabilistic human health risk assessment associated with fluoride and arsenic co-occurrence in drinking water from the metropolitan area of San Luis Potosí, Mexico. Environ. Monit. Assess. 192, 712. https://doi.org/10.1007/s10661-020-08675-7.

Freni, S.C., 1994. Exposure to high fluoride concentrations in drinking water is associated with decreased birth rates. J. Toxicol. Environ. Health 42, 109–121. https://doi.org/10.1080/15287399409531866.

Grönwall, J., Danert, K., 2020. Regarding groundwater and drinkingwater access through a human rights lens: self-supply as a norm. Water 12. https://doi.org/10.3390/w12020419 (Switzerland).

Guissouma, W., Hakami, O., Al-Rajab, A.J., Tarhouni, J., 2017. Risk assessment of fluoride exposure in drinking water of Tunisia. Chemosphere 177, 102–108. https://doi.org/10.1016/j.chemosphere.2017.03.011.

Gupta, I., Singh, B.P., Angurala, M.L., 2014. Central ground water board: ground water year book of Punjab state and Chandigarh (UT).

Hayat, E., Baba, A., 2017. Quality of groundwater resources in Afghanistan. Environ. Monit. Assess. 189 https://doi.org/10.1007/s10661-017-6032-1.

He, S., Wu, J., 2019. Hydrogeochemical characteristics, groundwater quality, and health risks from hexavalent chromium and nitrate in groundwater of Huanhe formation in Wuqi County, Northwest China. Expo. Health 11, 125–137. https://doi.org/10.1007/s12403-018-0289-7.

Ibrahim, M., Prabhakar, P., Sumalatha, M., Prabhakar, P., 2011. Effects of fluoride contents in ground water: a review. Int. J. Pharm. Appl. 2, 128–134.

ICMR, 2009. Nutrient requirements and recommended dietary allowances for Indians, a Report of the Expert, A Report of the Expert Group of the Indian Council of Medical ResearchExpert Group of the Indian Council of Medical Research.

Ijumulana, J., Ligate, F., Bhattacharya, P., Mtalo, F., Zhang, C., 2020. Spatial analysis and GIS mapping of regional hotspots and potential health risk of fluoride concentrations in groundwater of Northern Tanzania. Sci. Total Environ. 735, 139584 https://doi.org/10.1016/j.scitotenv.2020.139584.

Iqbal, J., Su, C., Rashid, A., Yang, N., Baloch, M.Y.J., Talpur, S.A., Ullah, Z., Rahman, G., Rahman, N.U., Earjh, E., Sajjad, M.M., 2021. Hydrogeochemical assessment of groundwater and suitability analysis for domestic and agricultural utility in Southern Punjab, Pakistan. Water 13, 3589. https://doi.org/10.3390/w13243589.

Jiang, P., Li, G., Zhou, X., Wang, C., Qiao, Y., Liao, D., Shi, D., 2019. Chronic fluoride exposure induces neuronal apoptosis and impairs neurogenesis and synaptic plasticity: role of GSK-3B/B-catenin pathway. Chemosphere 214, 430–435. https://doi.org/10.1016/j.chemosphere.2018.09.095.

Joshi, S.K., Rai, S.P., Sinha, R., Gupta, S., Densmore, A.L., Rawat, Y.S., Shekhar, S., 2018. Tracing groundwater recharge sources in the northwestern Indian alluvial aquifer using water isotopes (δ18O, δ2H and 3H). J. Hydrol. 559, 835–847. https://doi.org/10.1016/j.jhydrol.2018.02.056.

Kabata-Pendias, A., Pendias, H., 2000. Trace elements in soils and plants. Br. Med. J. https://doi.org/10.1136/bmj.2.4640.1355-a (Clin. Res. Ed). CRC Press.

Kaur, S., Kaur, A., Singh, R., Avasthi, A., Fatima, A., 2020. Prevalence of dental caries in 5- to 12-year-old school children of Patiala City, Punjab. Dent. J. Adv. Stud. 08, 01–04. https://doi.org/10.1055/s-0040-1703026.

Kimambo, V., Bhattacharya, P., Mtalo, F., Mtamba, J., Ahmad, A., 2019. Fluoride occurrence in groundwater systems at global scale and status of defluoridation – state of the art. Groundw. Sustain. Dev. 9, 100223 https://doi.org/10.1016/j.gsd.2019.100223.

Kumar, M., Kumari, K., Ramanathan, A., Saxena, R., 2007. A comparative evaluation of groundwater suitability for irrigation and drinking purposes in two intensively cultivated districts of Punjab, India. Environ. Geol. 53, 553–574. https://doi.org/10.1007/s00254-007-0672-3.

Kumar, R., Mittal, S., Sahoo, P.K., Sahoo, S.K., 2021. Source apportionment, chemometric pattern recognition and health risk assessment of groundwater from southwestern Punjab, India. Environ. Geochem. Health 43, 733–755. https://doi.org/10.1007/s10653-020-00518-1.

Kumar, S., Singh, R., Venkatesh, A.S., Udayabhanu, G., Sahoo, P.R., 2019. Medical Geological assessment of fluoride contaminated groundwater in parts of Indo-Gangetic Alluvial plains. Sci. Rep. 9, 1–16. https://doi.org/10.1038/s41598-019-52812-3.

Lapworth, D.J., Krishan, G., MacDonald, A.M., Rao, M.S., 2017. Groundwater quality in the alluvial aquifer system of northwest India: new evidence of the extent of anthropogenic and geogenic contamination. Sci. Total Environ. 599–600, 1433–1444. https://doi.org/10.1016/j.scitotenv.2017.04.223.

Liu, L., Wu, J., He, S., Wang, L., 2021. Occurrence and distribution of groundwater fluoride and manganese in the Weining Plain (China) and their probabilistic health risk quantification. Expo. Health. https://doi.org/10.1007/s12403-021-00434-4.

Maliyekkal, S.M., Sharma, A.K., Philip, L., 2006. Manganese-oxide-coated alumina: a promising sorbent for defluoridation of water. Water Res. 40, 3497–3506. https://doi.org/10.1016/j.watres.2006.08.007.

Marimon, M.P.C., Knöller, K., Roisenberg, A., 2007. Anomalous flouride concentration in groundwater - is it natural or pollution? A stable isotope approach. Isotopes Environ. Health Stud. 43, 165–175. https://doi.org/10.1080/10256010701360132.

Mittal, S., Kumar, R., Sahoo, P.K., Sahoo, S.K., 2020. Geochemical assessment of groundwater contaminants and associated health risks in the Shivalik region of Punjab, India. Toxin Rev. 0, 1–17. https://doi.org/10.1080/15569543.2020.1802597.

Mohammadi, A.A., Yousefi, M., Yaseri, M., Jalilzadeh, M., Mahvi, A.H., 2017. Skeletal fluorosis in relation to drinking water in rural areas of West Azerbaijan, Iran. Sci. Rep. 7, 4–10. https://doi.org/10.1038/s41598-017-17328-8.

Mohapatra, A.K., Sujathan, S., Ekamparam, A.S.S., Singh, A., 2021. The role of manganese carbonate precipitation in controlling fluoride and uranium mobilization in groundwater. ACS Earth Sp. Chem. https://doi.org/10.1021/acsearthspacechem.1c00133.

Mukherjee, I., Singh, U.K., Patra, P.K., 2019. Exploring a multi-exposure-pathway approach to assess human health risk associated with groundwater fluoride exposure in the semi-arid region of east India. Chemosphere 233, 164–173. https://doi.org/10.1016/j.chemosphere.2019.05.278.

Mukherjee, I., Singh, U.K., Singh, R.P., Kumari, Anshumali, D, Jha, P.K., Mehta, P., 2020. Characterization of heavy metal pollution in an anthropogenically and geologically influenced semi-arid region of east India and assessment of ecological and human health risks. Sci. Total Environ. 705, 135801 https://doi.org/10.1016/j.scitotenv.2019.135801.

Mwiathi, N.F., Gao, X., Li, C., Rashid, A., 2022. The occurrence of geogenic fluoride in shallow aquifers of Kenya Rift Valley and its implications in groundwater management. Ecotoxicol. Environ. Saf. 229, 113046 https://doi.org/10.1016/j.ecoenv.2021.113046.

NAPCC, 2011. Final report September 2011 Appendix 2 lower Sutlej Sub Basin.

Narsimha, A., Rajitha, S., 2018. Spatial distribution and seasonal variation in fluoride enrichment in groundwater and its associated human health risk assessment in Telangana State, South India. Hum. Ecol. Risk Assess. 24, 2119–2132. https://doi.org/10.1080/10807039.2018.1438176.

Naz, A., Mishra, B.K., Gupta, S.K., 2016. Human health risk assessment of chromium in drinking water: a case study of Sukinda chromite mine, Odisha, India. Expo. Health 8, 253–264. https://doi.org/10.1007/s12403-016-0199-5.

Nguyen, A.H., Nguyen, M.P.L., Pham, N.T.T., Tat, V.M.H., Luu, L.K., Vo, P.L., 2021. Health risk assessment of groundwater consumption for drinking and domestic purposes in Xuyen Moc District, Ba Ria - Vung Tau Province, Vietnam. IOP Conf. Ser. Earth Environ. Sci. 652. https://doi.org/10.1088/1755-1315/652/1/012018.

Nizam, S., Sen, I.S., 2018. Effect of Southwest monsoon withdrawal on mass loading and chemical characteristics of aerosols in an urban city over the Indo-Gangetic Basin. ACS Earth Sp. Chem. 2, 347–355. https://doi.org/10.1021/acsearthspacechem.7b00140.

Nizam, S., Sen, I.S., Shukla, T., Selby, D., 2021. Melting of the Chhota Shigri Glacier, Western Himalaya, insensitive to anthropogenic emission residues: insights from geochemical evidence. Geophys. Res. Lett. 48, 1–12. https://doi.org/10.1029/2021GL092801.

Noor, S., Rashid, A., Javed, A., Khattak, J.A., Farooqi, A., 2021. Hydrogeological properties, sources provenance, and health risk exposure of fluoride in the groundwater of Batkhela, Pakistan. Environ. Technol. Innov. 25, 102239 https://doi.org/10.1016/j.eti.2021.102239.

Ortiz-Pérez, D., Rodríguez-Martínez, M., Martínez, F., Borja-Aburto, V.H., Castelo, J., Grimaldo, J.I., De la Cruz, E., Carrizales, L., Díaz-Barriga, F., 2003. Fluoride-induced disruption of reproductive hormones in men. Environ. Res. 93, 20–30. https://doi.org/10.1016/S0013-9351(03)00059-8.

Podgorski, J.E., Labhasetwar, P., Saha, D., Berg, M., 2018. Prediction modeling and mapping of groundwater fluoride contamination throughout India. Environ. Sci. Technol. 52, 9889–9898. https://doi.org/10.1021/acs.est.8b01679.

Rahman, M.M., Bodrud-Doza, M., Siddiqua, M.T., Zahid, A., Islam, A.R.M.T., 2020. Spatiotemporal distribution of fluoride in drinking water and associated probabilistic human health risk appraisal in the coastal region, Bangladesh. Sci. Total Environ. 724 https://doi.org/10.1016/j.scitotenv.2020.138316.

Rashid, A., Ayub, M., Javed, A., Khan, S., Gao, X., Li, C., Ullah, Z., Sardar, T., Muhammad, J., Nazneen, S., 2021. Potentially harmful metals, and health risk evaluation in groundwater of Mardan, Pakistan: application of geostatistical approach and geographic information system. Geosci. Front. 12, 101128 https://doi.org/10.1016/j.gsf.2020.12.009.

Rashid, A., Farooqi, A., Gao, X., Zahir, S., Noor, S., Khattak, J.A., 2020. Geochemical modeling, source apportionment, health risk exposure and control of higher fluoride in groundwater of sub-district Dargai, Pakistan. Chemosphere 243, 125409. https://doi.org/10.1016/j.chemosphere.2019.125409.

Rashid, A., Guan, D.X., Farooqi, A., Khan, S., Zahir, S., Jehan, S., Khattak, S.A., Khan, M.S., Khan, R., 2018. Fluoride prevalence in groundwater around a fluorite mining area in the flood plain of the River Swat, Pakistan. Sci. Total Environ. 635, 203–215. https://doi.org/10.1016/j.scitotenv.2018.04.064.

Rashid, A., Khan, S., Ayub, M., Sardar, T., Jehan, S., Zahir, S., Khan, M.S., Muhammad, J., Khan, R., Ali, A., Ullah, H., 2019. Mapping human health risk from exposure to potential toxic metal contamination in groundwater of Lower Dir, Pakistan: application of multivariate and geographical information system. Chemosphere 225, 785–795. https://doi.org/10.1016/j.chemosphere.2019.03.066.

Rasool, A., Farooqi, A., Xiao, T., Ali, W., Noor, S., Abiola, O., Ali, S., Nasim, W., 2017. A review of global outlook on fluoride contamination in groundwater with prominence on the Pakistan current situation. Environ. Geochem. Health 40, 1265–1281. https://doi.org/10.1007/s10653-017-0054-z.

Sahu, B.L., Banjare, G.R., Ramteke, S., Patel, K.S., Matini, L., 2017. Fluoride contamination of groundwater and toxicities in dongargaon block, Chhattisgarh, India. Expo. Health 9, 143–156. https://doi.org/10.1007/s12403-016-0229-3.

Samal, A.K., Mishra, P.K., Biswas, A., 2020. Assessment of origin and distribution of fluoride contamination in groundwater using an isotopic signature from a part of the Indo-Gangetic Plain (IGP), India. HydroResearch 3, 75–84. https://doi.org/10.1016/j.hydres.2020.05.001.

Saxena, S., Sahay, A., Goel, P., 2012. Effect of fluoride exposure on the intelligence of school children in Madhya Pradesh, India. J. Neurosci. Rural Pract. 3, 144–149. https://doi.org/10.4103/0976-3147.98213.

Sharma, T., Bajwa, B.S., Kaur, I., 2021a. Contamination of groundwater by potentially toxic elements in groundwater and potential risk to groundwater users in the Bathinda and Faridkot districts of Punjab, India. Environ. Earth Sci. 80, 1–15. https://doi.org/10.1007/s12665-021-09560-3.

Sharma, T., Litoria, P.K., Bajwa, B.S., Kaur, I., 2021b. Appraisal of groundwater quality and associated risks in Mansa district (Punjab, India). Environ. Monit. Assess. 193 https://doi.org/10.1007/s10661-021-08892-8.

Shashi, A., Bhardwaj, M., 2011. Study on blood biochemical diagnostic indices for hepatic function biomarkers in endemic skeletal fluorosis. Biol. Trace Elem. Res. 143, 803–814. https://doi.org/10.1007/s12011-010-8944-2.

Shukla, S., Saxena, A., 2020. Groundwater quality and associated human health risk assessment in parts of Raebareli district, Uttar Pradesh, India. Groundw. Sustain. Dev. 10, 100366 https://doi.org/10.1016/j.gsd.2020.100366.

Shukla, T., Sen, I.S., 2021. Preparing for floods on the Third Pole. Science 372 (6539), 232–234. https://doi.org/10.1126/science.abh3558.

Singh, B., Kaur, S., Litoria, P.K., Das, S., 2021. Development of web enabled water resource information system using open source software for Patiala and SAS Nagar districts of Punjab, India. Water Pract. Technol. 16, 980–990. https://doi.org/10.2166/wpt.2021.050.

Singh, J., Virdi, S.S., 2015. Environment management through public private partnership : rain water harvesting model 1, 18–24.

Sonali, D., Varsha, D., Jaya, K., Rashmi, U., 2013. An epidemiological study of skeletal fluor- osis in some villages of Chandrapur district, Maharashtra, India 7.

Srivastava, S., Flora, S.J.S., 2020. Fluoride in drinking water and skeletal fluorosis: a review of the global impact. Curr. Environ. Health Rep. 7, 140–146. https://doi.org/10.1007/s40572-020-00270-9.

Talpur, S.A., Noonari, T.M., Rashid, A., Ahmed, A., Jat Baloch, M.Y., Talpur, H.A., Soomro, M.H., 2020. Hydrogeochemical signatures and suitability assessment of groundwater with elevated fluoride in unconfined aquifers Badin district, Sindh, Pakistan. SN Appl. Sci. 2, 1–15. https://doi.org/10.1007/s42452-020-2821-1.

USEPA, 1999. Guidance for Performing Aggregate Exposure and Risk Assessments. Office of Pesticide Programs, Washington DC.

USEPA, 1993. Reference Dose (RfD): Description and Use in Health Risk Assessments. USEPA. https://www.epa.gov/iris/reference-dose-rfd-description-and-use-health-risk-assessments.

Virk, H.S., 2018. Fluoride contamination of ground waters of two Punjab districts and its implications. OmniSci. Multidiscip. J. 8, 25–31. https://doi.org/10.13140/RG.2.2.21040.66566.

Wang, S.X., Wang, Z.H., Cheng, X.T., Li, J., Sang, Z.P., Zhang, X.D., Han, L.L., Qiao, X.Y., Wu, Z.M., Wang, Z.Q., 2007. Arsenic and fluoride exposure in drinking water: children's IQ and growth in Shanyin county, Shanxi Province, China. Environ. Health Perspect. 115, 643–647. https://doi.org/10.1289/ehp.9270.

WHO, 2013. World health statistics, SBN 978 92 4 156458 8.

WHO, 2011. Guidelines for Drinking-water Quality, 4th ed. World Health Organization, Geneva, Switzerland. https://doi.org/10.1007/978-1-4020-4410-6_184.

WHO, 2010. Inadequate or Excess Flouride: A major public health concern. WHO. GenevaPublic Heal. Environ.

WHO, 1994. Fluoride and Oral Health: Report on Oral Health Status and Fluoride Use. WHO, Geneva.

WRED and CGWB, 2018. Groundwater resources of Punjab state (As on 31st March, 2017). https://dswcpunjab.gov.in/contents/docs/publications/Draft%20Report%20Punjab%20Groundwater%20Resources%202017.pdf.

Yadav, K.K., Kumar, S., Pham, Q.B., Gupta, N., Rezania, S., Kamyab, H., Yadav, S., Vymazal, J., Kumar, V., Tri, D.Q., Talaiekhozani, A., Prasad, S., Reece, L.M., Singh, N., Maurya, P.K., Cho, J., 2019. Fluoride contamination, health problems and remediation methods in Asian groundwater: a comprehensive review. Ecotoxicol. Environ. Saf. 182, 109362 https://doi.org/10.1016/j.ecoenv.2019.06.045.

Yang, C.Y., Cheng, M.F., Tsai, S.S., Hung, C.F., 2000. Fluoride in drinking water and cancer mortality in Taiwan. Environ. Res. 82, 189–193. https://doi.org/10.1006/enrs.1999.4018.

Yang, K., Liang, X., Quan, C., 2019. Fluoride in drinking water: effect on liver and kidney function. Encyclopedia of Environmental Health, 2nd ed. Elsevier Inc. https://doi.org/10.1016/B978-0-12-409548-9.11083-8.

Yousefi, M., Ghalehaskar, S., Asghari, F.B., Ghaderpoury, A., Dehghani, M.H., Ghaderpoori, M., Mohammadi, A.A., 2019. Distribution of fluoride contamination in drinking water resources and health risk assessment using geographic information system, northwest Iran. Regul. Toxicol. Pharmacol. 107, 104408 https://doi.org/10.1016/j.yrtph.2019.104408.

Yousefi, M., Ghoochani, M., Hossein Mahvi, A., 2018a. Health risk assessment to fluoride in drinking water of rural residents living in the Poldasht city, Northwest of Iran. Ecotoxicol. Environ. Saf. 148, 426–430. https://doi.org/10.1016/j.ecoenv.2017.10.057.

Yousefi, M., Yaseri, M., Nabizadeh, R., Hooshmand, E., Jalilzadeh, M., Mahvi, A.H., Mohammadi, A.A., 2018b. Association of hypertension, body mass index, and waist circumference with fluoride intake; water drinking in residents of fluoride endemic areas, Iran. Biol. Trace Elem. Res. 185, 282–288. https://doi.org/10.1007/s12011-018-1269-2.