CHAPTER

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Non-carcinogenic health risk assessment of fluoride in groundwater of the River Yamuna flood plain, Delhi, India

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1 Introduction

Rivers provide water to humans for sustenance and are a vital component of livelihood. In India, rivers are an essential source of aquifer recharge and water supply. The Yamuna River is one of the largest tributaries of the Ganga and traverses around 1376 km in north India before it meets the Ganga River.[1] Various scientific communities working on the Yamuna water quality reported that the water is highly polluted in the plains,[2–9] and the existing sewage treatment plants (STPs) in Delhi have sub-optimal treatment capacity in comparison to the sewage load.[1] In addition, antibiotics have also been reported in the effluents of STPs.[10–12]

Delhi contributes approximately 79% of the pollution load to the Yamuna, leaving behind Uttar Pradesh (16%) and Haryana (5%) states of India.[2,9] However, the monsoon flood of the river recharges floodplain aquifers on an annual basis and can sustain its exploitation for desired uses.[13–15] Furthermore, the bank filtrate of the river can also provide safe drinking water.[16] The study area is entirely residential, except for a small area under agricultural activity in the northern part of Delhi.

The high concentration of fluoride (F^-) in groundwater in parts of India and its consumption by humans can have serious short (dental fluorosis) and long (skeletal fluorosis) consequences.[17–26]

Thus, this study was conducted to evaluate the groundwater quality with respect to F^- in the Yamuna flood plains (YFP) from a geochemical perspective. Furthermore, in this study, the health risk assessment of humans due to the consumption of F^--contaminated groundwater was evaluated. In general, it was observed that the groundwater is safe with respect to F^- in YFP and has the potential to serve as a source of sustainable water supply in the future, provided that other organic and inorganic pollutants are within safe limits. The hazard quotient (HQ_{ORAL}) in the area shows that children consuming groundwater in Kherakalan and Narela localities are likely to have adverse health effects (Fig. 1). This study is the first integrated attempt to gain insight into F^- contamination and the major ion chemistry of the groundwater of YFP from the perspective of local sediment geochemistry. It also incorporates health hazard assessment in the context of the prevailing F^- concentration in the groundwater of the study area.

2 Study area

The aquifer system of Delhi is a part of the highly stressed regional aquifer system of NW India, where the solution to the groundwater crisis is an hour.[27,28] In the case of Delhi, groundwater quality is a major concern in addition to groundwater overexploitation.[25,29,30] The YFP aquifers are the most potential aquifers of Delhi.[13,29] An insight into the geological and hydrogeological aspects of the study area is as follows.

Computers in Earth and Environmental Sciences
https://doi.org/10.1016/B978-0-323-89861-4.00046-4

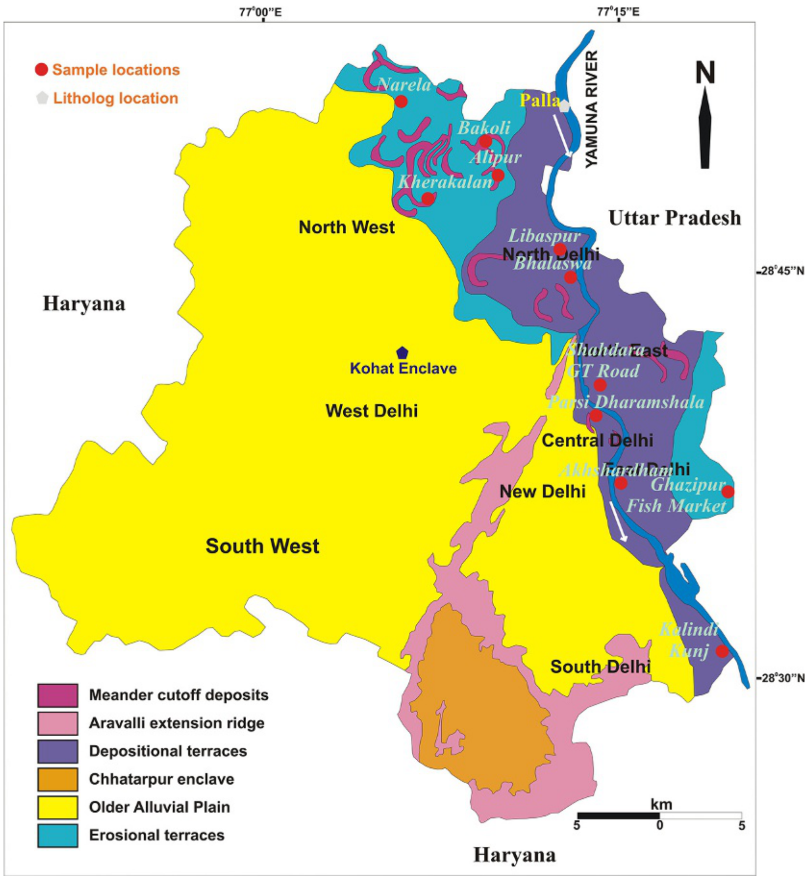

FIG. 1 Geological map showing groundwater and sediments sampling locations on Yamuna flood plains.

2.1 Geology

The Yamuna flood plain (YFP) was deposited on both sides of the Yamuna River in the region. The YFP is broadly divided into the Older Yamuna flood plain and the active Yamuna flood plain (Fig. 1). Gray color medium sand, silt, and clay define the lithology of the area.[31] The occurrence of coarse sand mixed with kankars (calcareous nodules) at a shallow level with yellowish-brown sand at a deeper level was also observed by author SA in older Yamuna flood plains.

2.2 Hydrogeology

The aquifer along the river is predominantly composed of fine-to-medium unconsolidated sand. The average discharge of tube wells of newer alluviums ranges from 150 to 300 m^3/h, with an average transmissivity of 730–2100 m^2/d.[29] The aquifer is unconfined in nature and hence receives in situ rainfall/flood recharge by direct infiltration.[32,33]

During peak floods, groundwater velocity in the sands of flood plains can reach up to 2.12 m/d.[14] The depth to water level (DTWL) map suggests a shallow level of groundwater in the study area, with the DTWL varying in the range of 10 m below ground level (mbgl) (Fig. 2). The inhabitants of Delhi have their drinking wells on the plain and drink water without any treatment.

3 Material and methods

3.1 Groundwater and sediment sampling

In this study, a total of eleven groundwater samples were collected from the YFP. The groundwater samples were acidified by nitric acid during the field to maintain a water pH below two for the analysis of major cations. Unacidified

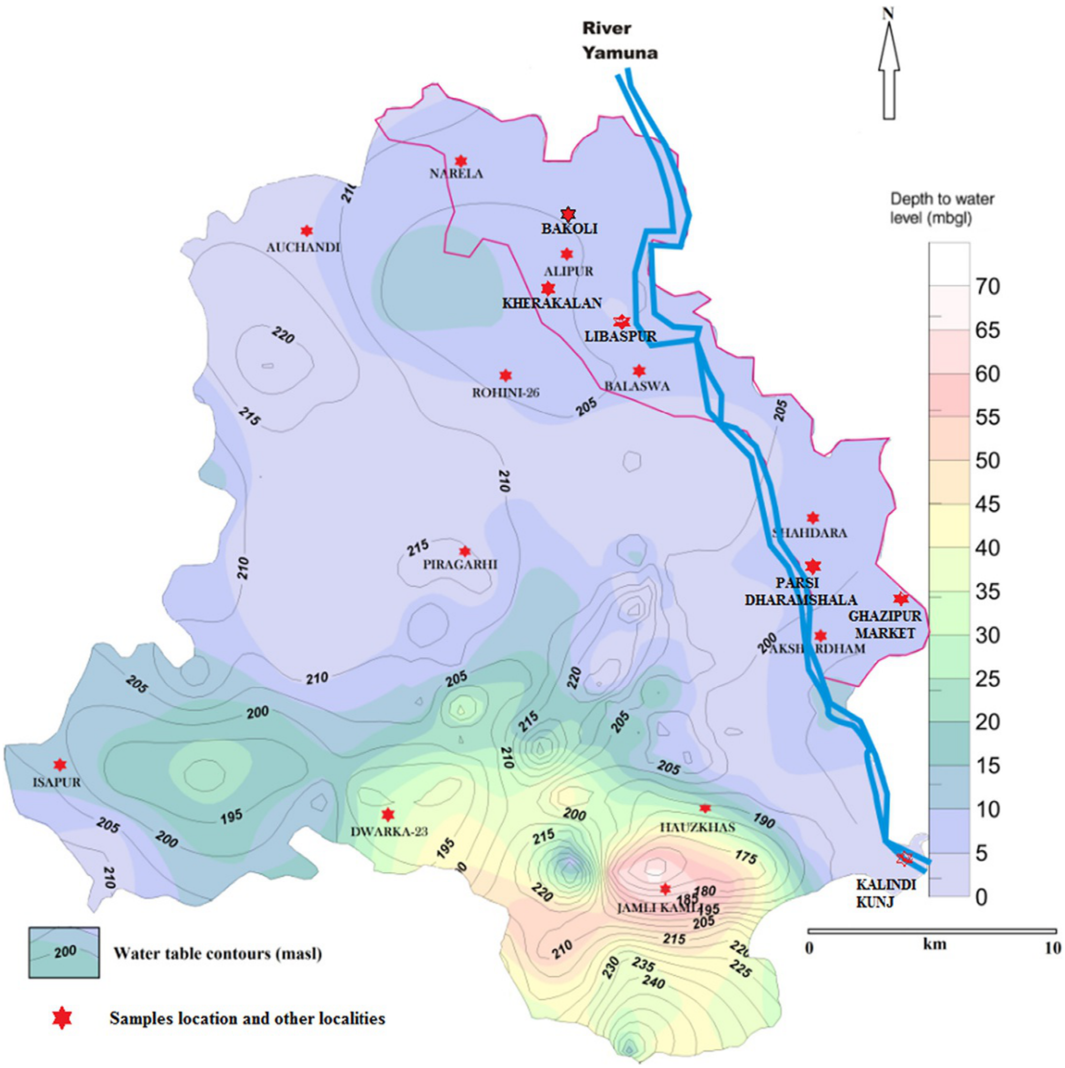

FIG. 2 Water table and depth to water level map of Delhi for non-monsoon (May) season.[34] The *redline* in the figure shows the area under the Yamuna flood plain. Sampling stations and other important locations are shown in the figure.

samples were collected for analysis of major anions. Therefore, samples were collected in duplicates in poly-lab bottles. The bottles were thoroughly rinsed three times with water for analysis. The sediment along the river was collected at a depth of 0.3 m from the river bed in a transparent zip-lock bag from the Palla locality (Fig. 1) and opened only during the bulk sediment analyses by XRD.

3.2 Lab analysis

Eleven groundwater samples were collected along the Yamuna stretch during the pre-monsoon (dry) season in 2016 and analyzed for major ions such as Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, HCO_3^-, and CO_3^{2-} at IIT Bombay, India. F^- Concentration was measured at the University of Delhi using a fluoride meter. Electrical conductivity (EC) and pH were measured during the field using an EC and a pH meter (Hanna). The geographical location was mapped using GPS and an Android phone app (Samsung 7562). For confirmation, the EC was again measured in the lab at IIT Bombay. The major cations, such as Na^+, K^+, Ca^{2+}, and Mg^{2+}, were analyzed using ICP-AES (Perkin-Elmer, France) at IIT Bombay. The SO_4^{2-} concentration was measured using a spectrophotometer (Shimadzu UV-visible spectrophotometer 160), alkalinity by titration, and chloride (Cl^-) using an Expandable Ion Analyzer 940A with a combination of the Orion ion plus 9817 BN available at IIT Bombay. For XRD, bulk sediments of the Yamuna flood plain were mounted on a sample holder using the back-loading technique and scanned from 5° to 70° (2θ) with a step size of 0.01° and a scan speed of

38 s/step, using Cu-Kα radiation from an Empyrean X-ray diffractometer (PANalytical) equipped with a Pixel 3D detector.

3.3 Human health risk assessment

The non-carcinogenic human health risk assessment due to the consumption of F^--enriched groundwater expressed as hazard quotient (HQ_{ORAL}), was calculated using Eq. (1).

$$HQ_{ORAL} = EDI_{ORAL}/RfD_{ORAL} \tag{1}$$

HQ_{ORAL} was calculated by estimating the daily intake due to the ingestion of F^- enriched groundwater (EDI_{ORAL}) and RfD_{ORAL} (oral reference dose). EDI_{ORAL} (Eq. 2) was estimated using the concentration of F^-(c), ingestion rate (IR), exposure frequency (EF), exposure duration (ED), body weight (BW), and average time (AT) for children and adults.

$$EDI_{ORAL} = (C \times IR \times EF \times ED)/(BW \times AT) \tag{2}$$

The RfD_{ORAL} values of the Delhi region were obtained from Ali et al.[19] An estimated HQ_{ORAL} value greater than 1 was considered unsafe.[19,35]

The method used in this study is summarized as a flow chart (Fig. 3).

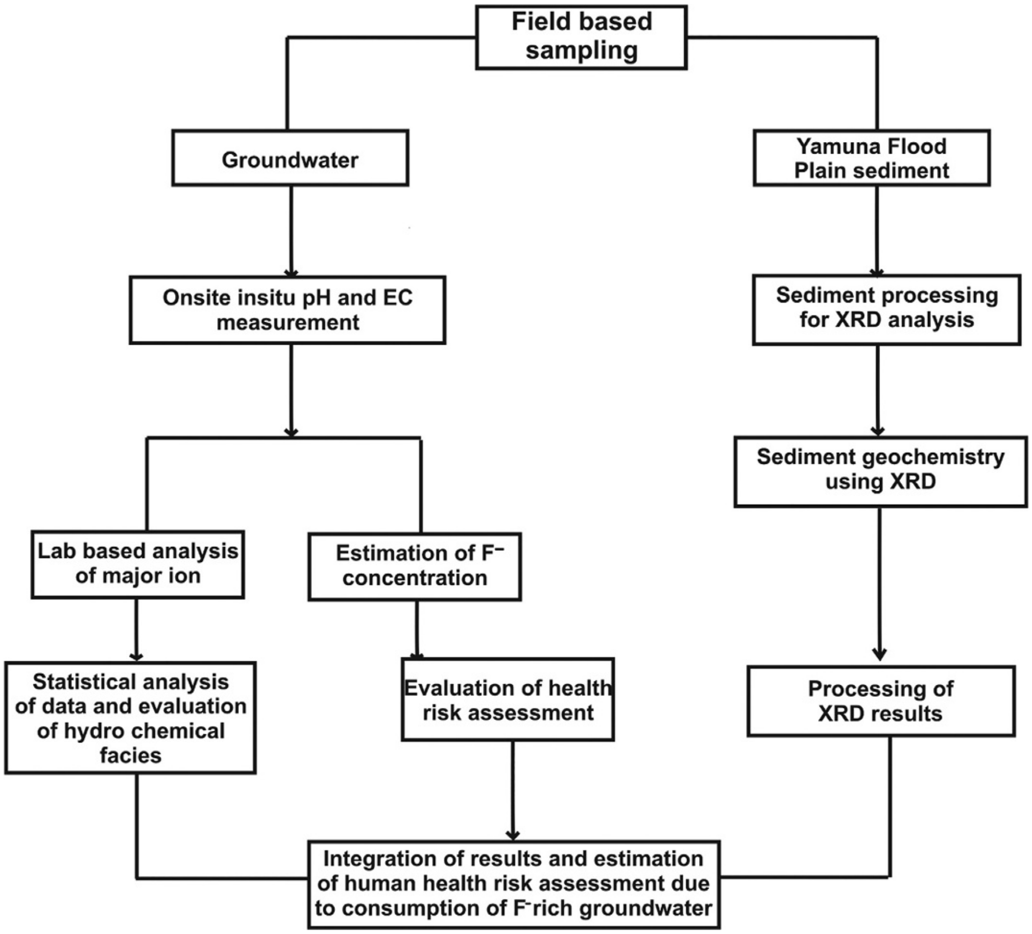

FIG. 3 A flow chart elucidating the methods used in the present study.

4 Results and discussion

4.1 Major ions chemistry and hydro-chemical facies

The Na^+ and Cl^- ions in the groundwater of the study area were comparatively higher than those of the other ions and varied from 97 to 667 mg/L (average: 382 mg/L) and 150 to 1350 mg/L (average: 750 mg/L; Fig. 4, Table 1).

Except for groundwater from the Bakoli locality (Fig.1), the concentration of SO_4^{2-} ions are comparatively lower than the groundwater of the Older Alluvium Plains (OAP) and ranges from 3.9 to 904 mg/L.[26,36] Similarly, Ca^{2+} (35–202 mg/L) and Mg^{2+} (23.7–137 mg/L) ion and electrical conductivity values of the groundwater in the Yamuna flood plains are comparatively lower than the OAP.[36] However, the HCO_3^- ion concentration was comparatively higher than that of the OAP groundwater.[26] Very high K^+ (153.2 mg/L) and HCO_3^- concentrations were observed in the Bhalaswa sampling locality near the landfill site (Fig. 1; Table 1). Thus, the major ions in the landfill locality clearly show their effect on the quality of groundwater.

The hydro-chemical facies of the Yamuna floodplain groundwater suggests the dominance of the Na-Cl facies (Fig. 5). In addition, groundwater also showed the Na-Ca-Cl-HCO_3 water type. The variation of facies in the groundwater in the area is obvious owing to different water-sediment interactions.

4.2 Statistical analysis

Statistical correlation is a powerful tool and is best known for evaluating the relationship between various parameters (Table 2). The correlation suggests a positive correlation between F^- and pH and a negative correlation with Ca^{2+} and Mg^{2+}. This indicates that the high pH of the groundwater is responsible for triggering F^- from sediment to the aqueous phase in the study area. A similar relationship was also observed in earlier studies.[17]

4.3 Bivariate plot

After statistical correlation, bivariate plots were created for a deeper understanding of the interrelationship between the major ion and F^- (Fig. 6). The bivariate plot of F^- with pH showed a positive correlation, while F^- with HCO_3^- and Ca^{2+} showed a negative correlation.

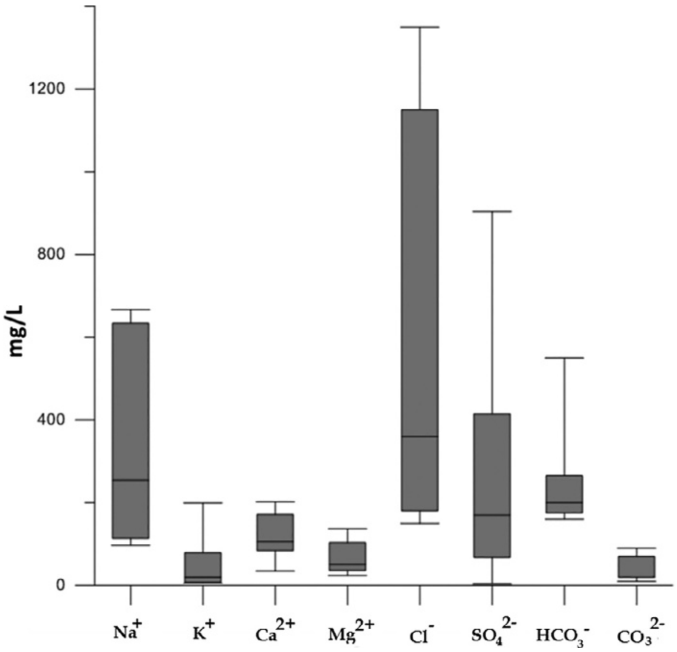

FIG. 4 Box and whisker plot of major ion of groundwater of Yamuna flood plains.

TABLE 1 Major ions chemistry and F^- level in groundwater along with the aquifers of River Yamuna stretch, Delhi.[36]

S. No	Locality	Latitude	Longitude	Source	Depth[a]	EC	pH	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	CO_3^{2-}	F^-	Water type
					m	μS/cm							mg/L				
1	Akshardham	28.616	77.273	SS	75	968	7.97	114	6.7	105.8	36	180	54	200	20	0.24	Na-Ca-Mg-Cl-HCO_3
2	Alipur	28.797	77.134	HP	7.5	2233	7.79	236	199	131	61.6	360	144	255	50	0.65	Na-Ca-Cl-HCO_3
3	Bakoli	28.815	77.146	HP	NA	4880	7.71	654	11.3	202	135.9	1080	904	175	10	0.18	Na-Ca-Mg-Cl-SO_4
4	Bhalaswa	28.741	77.166	HP	4.5	4440	7.94	634	153.2	171.8	137	1150	109	550	90	0.37	Na-Mg-Cl-HCO_3
5	Ghazipur Fish Market	28.628	77.327	SS	90	2422	7.98	343	7.6	93	63	1350	176	160	20	0.44	Na-Mg-Cl
6	Kalindi Kunj	28.549	77.303	SS	24	917	7.88	102	10.2	84	36	150	3.9	250	10	0.53	Na-Ca-Mg-Cl-HCO_3
7	Kherakalan	28.772	77.115	HP	15	1720	8.15	254	74	67	44.7	220	170	310	80	1.2	Na-Ca-Cl-HCO_3-SO_4
8	Libaspur	28.75	77.143	TW	24	4660	7.69	559	20	180	103.7	1215	447	185	20	0.6	Na-Mg-Cl
9	Narela	28.842	77.088	SS	42	3100	8.25	667	5.5	35	23.7	430	415	180	20	1.57	Na-Cl-SO_4
10	Parsi Dharamshala	28.638	77.24	SS	60	1647	8.18	119	36	99	51	230	172	165	30	0.7	Na-Ca-Mg-Cl-SO_4
11	Shahdara GT Road	28.673	77.283	HP	15	1085	7.96	97	79	114	43	150	68	265	70	0.26	Na-Ca-Mg-Cl-HCO_3

[a] *Depth information was noted during the field by nearby residents, which may not be accurate.* NA, *not available;* SS, *submersible;* HP, *hand pump;* TW, *tube-wells.*

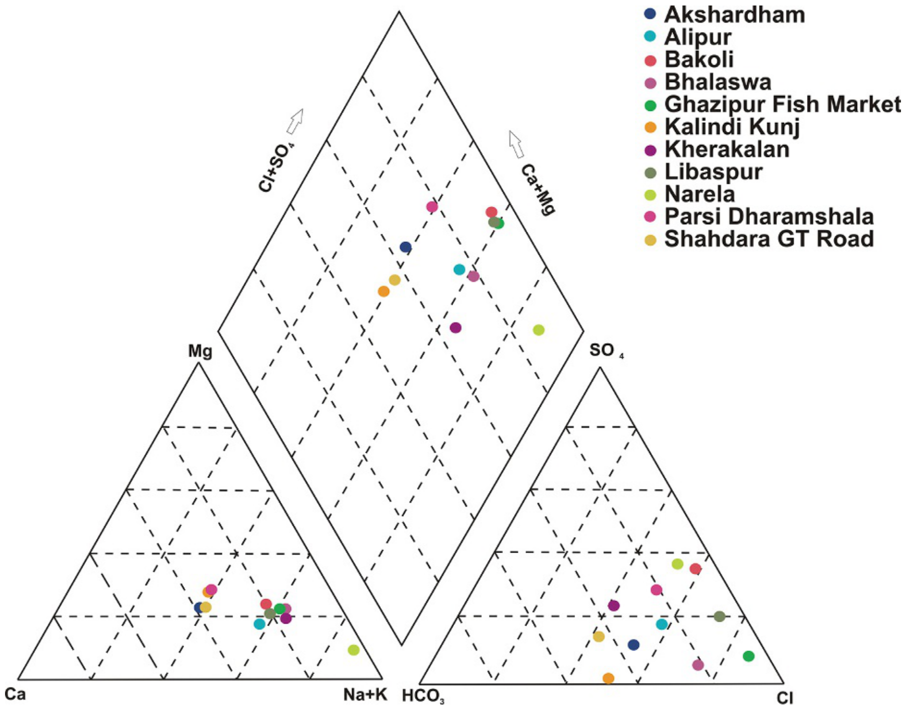

FIG. 5 Hydro-chemical facies of groundwater of Yamuna flood plains for the non-monsoon season (2016).

TABLE 2 Statistical correlation of major ion and other parameters of non-monsoon season groundwater.

	EC	pH	Na^+	K^+	Ca^{2+}	Mg^{2+}	Cl^-	SO_4^{2-}	HCO_3^-	CO_3^{2-}	F^-
EC	1										
pH	−0.46	1									
Na^+	**0.92**	−0.19	1								
K^+	0.10	−0.16	0.01	1							
Ca^{2+}	**0.72**	**−0.82**	0.41	0.24	1						
Mg^{2+}	**0.85**	**−0.63**	**0.63**	0.27	**0.92**	1					
Cl^-	**0.80**	−0.47	**0.69**	−0.03	**0.62**	**0.76**	1				
SO_4^{2-}	**0.73**	−0.35	**0.70**	−0.29	0.49	**0.53**	0.46	1			
HCO_3^-	0.21	0.00	0.21	**0.66**	0.23	0.42	0.12	−0.34	1		
CO_3^{2-}	0.09	0.22	0.08	**0.75**	0.04	0.23	−0.01	−0.35	**0.84**	1	
F^-	−0.15	**0.69**	0.13	−0.02	**−0.72**	**−0.55**	−0.38	−0.05	−0.09	0.18	1

4.4 Control of bulk sediments chemistry on major ions

The XRD patterns of the bulk sediments of the YFP sediments reveal the dominance of quartz and biotite minerals with albite and fluorite as an accessory (Fig. 7). The high amounts of quartz and biotite in the sediments were due to the presence of sandy soils.

From sediment chemistry, it was observed that the Na^+ ions in the groundwater may be due to the chemical weathering of albite ($NaAlSi_3O_8$), whereas biotite ($KMg_3AlSi_3O_{10}(OH)_2$ weathering may contribute to Mg^{2+} and K^+ ions in the water.[26] The irrigation practices in the area are confined only to the Palla locality (Fig. 1), while the entire study area is residential. Except for Bakoli, it is assumed that the anthropogenic pollution of SO_4^{2-} ions are not sufficient to push the SO_4^{2-} concentration beyond the safe limits (Table 1).

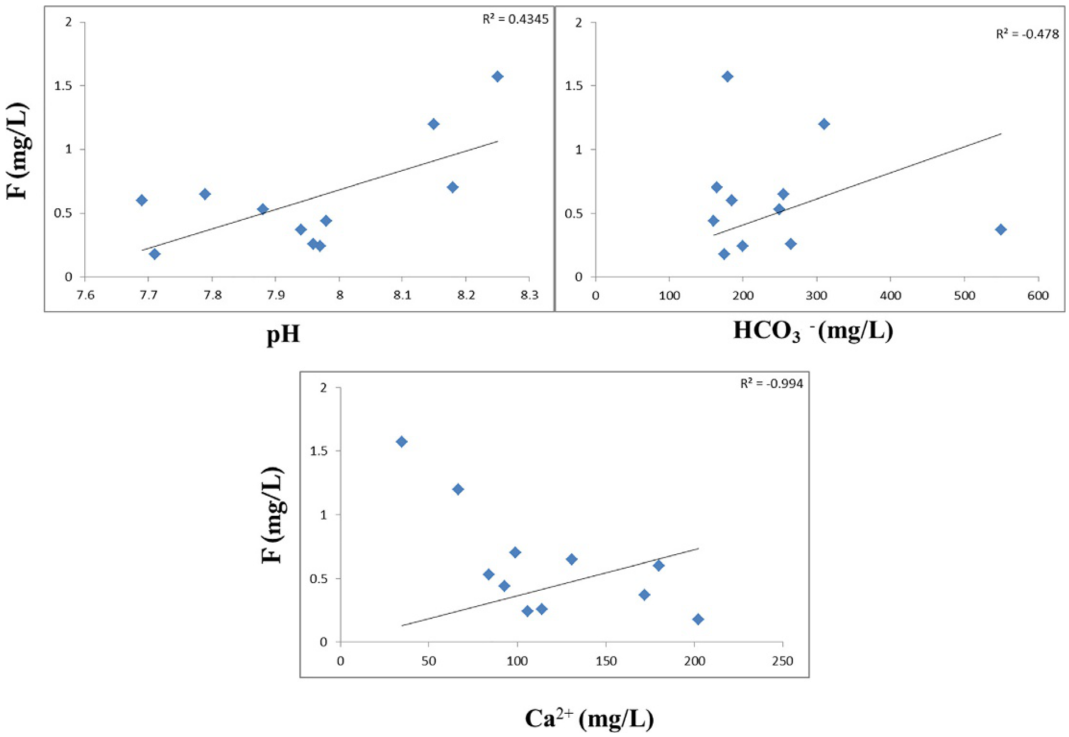

FIG. 6 Bivariate plots between F^- with pH, HCO_3^- and Ca^{2+} with zero intercept.

4.5 Fluoride level and human health risk assessment

The F^- level in the Narela drinking well was observed to be 1.57 mg/L, while in the rest of the samples, the F^- concentration was below 1.5 mg/L (permissible limit). This shows that in the majority of drinking wells, the F^- level is within the safe limit. This is further substantiated by the low to moderate EC values of groundwater, which suggest shorter water-sediment interactions. The significant positive correlation between pH and F^- indicates that a high pH triggers F^- in groundwater.

In this study, F^- exposure was observed in children and adults in the two age groups. The HQ_{ORAL} of children and adults are given in Table 3 and shown as box and whisker diagrams in Fig. 8.

The estimated HQ_{ORAL} shows that children drinking groundwater in Kherakalan and Narela (Fig. 1) are likely to have adverse health effects (HQ_{ORAL} more than 1[35]), while children and adults in other localities are safe (Table 3 and Fig. 8). The HQ_{ORAL} values for children are higher than those for adults because of their lower weight. This has also been observed in many other studies conducted on the non-carcinogenic health risk assessment of F^-.[37–41] This indicates that the problem is more serious for the health of children than for adults.

Fig. 9 explains the possible causes for the low level of F^- in YFP.

The Yamuna River, like all other major rivers, flows through a valley, which under normal circumstances is a natural sink for surface and groundwater flow. Flood plain sediments are dominated by sand with higher values of hydraulic conductivity. This facilitates regular flushing of the groundwater system and annual recharge from the monsoon floods, thereby resulting in low F^- and EC concentrations in the groundwater of the study area. Such a hydrogeological setting makes the YFP a safe option for water supply during stress periods.

5 Conclusions

The F^- level in the groundwater from the study area ranges from 0.18 to 1.57 mg/L. Statistical correlation and bivariate plots revealed that the alkaline water in the study area was responsible for triggering F^- release in the groundwater. However, because of continuous flushing and recharge through the permeable formations, the sediment-groundwater interaction is for a limited duration, leading to a lower F^- concentration in the groundwater. The bulk sediments of the Yamuna flood plain suggest the dominance of quartz and biotite minerals with albite and fluorite as accessory minerals. The existing sediment chemistry is significant for controlling the hydro-chemical facies in the area.

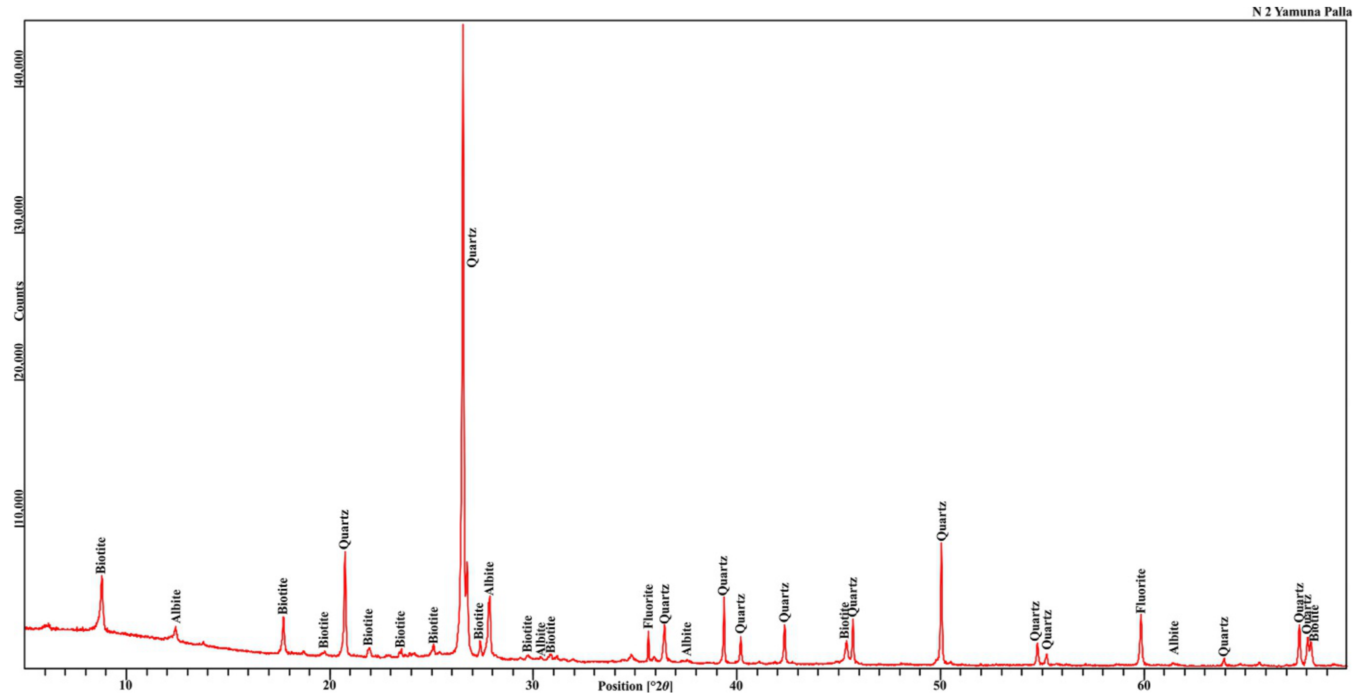

FIG. 7 XRD of bulk sediment of the River Yamuna flood plain from 0.3 m depth.

TABLE 3 HQ_{ORAL} for children and adults in the study area for all sampling locations.

S. No.	Locality	$HQ_{CHILDREN}$	HQ_{ADULTS}
1	Akshardham	0.24	0.13
2	Alipur	0.64	0.34
3	Bakoli	0.18	0.09
4	Bhalaswa	0.37	0.19
5	Ghazipur Fish Market	0.44	0.23
6	Kalindi Kunj	0.52	0.28
7	Kherakalan	**1.19**	0.63
8	Libaspur	0.59	0.31
9	Narela	**1.55**	0.82
10	Parsi Dharamshala	0.69	0.37
11	Shahdara GT Road	0.26	0.14

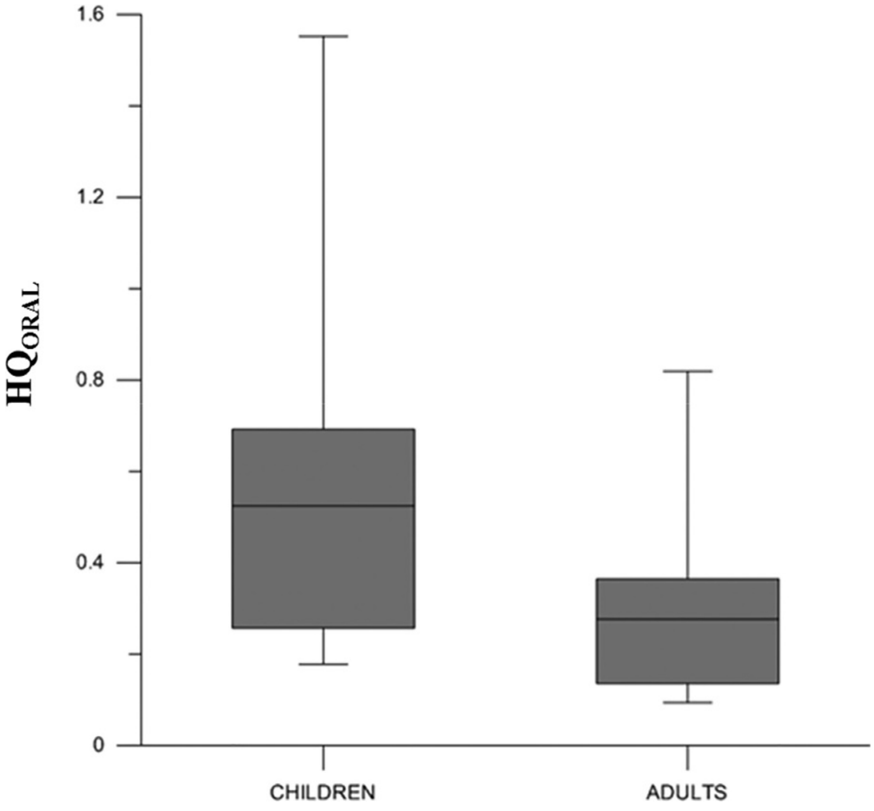

FIG. 8 Hazard quotient (HQ_{ORAL}) via drinking for children and adults.

The HQ_{ORAL} values suggest that the children in the Kherakalan and Narela localities are at adverse health risks due to the intake of groundwater ($HQ_{ORAL} > 1$). In other localities, no adverse health risks were observed ($HQ_{ORAL} < 1$). In the context of F^--related health hazards, this study suggests that except for two localities, the use of groundwater is safe for human consumption. Thus, it is advised to undertake scientific planning for the exploitation of the flood plain groundwater resources and formulate strategies for the adoption of a bank filtration approach to augment the drinking water needs of Delhi.

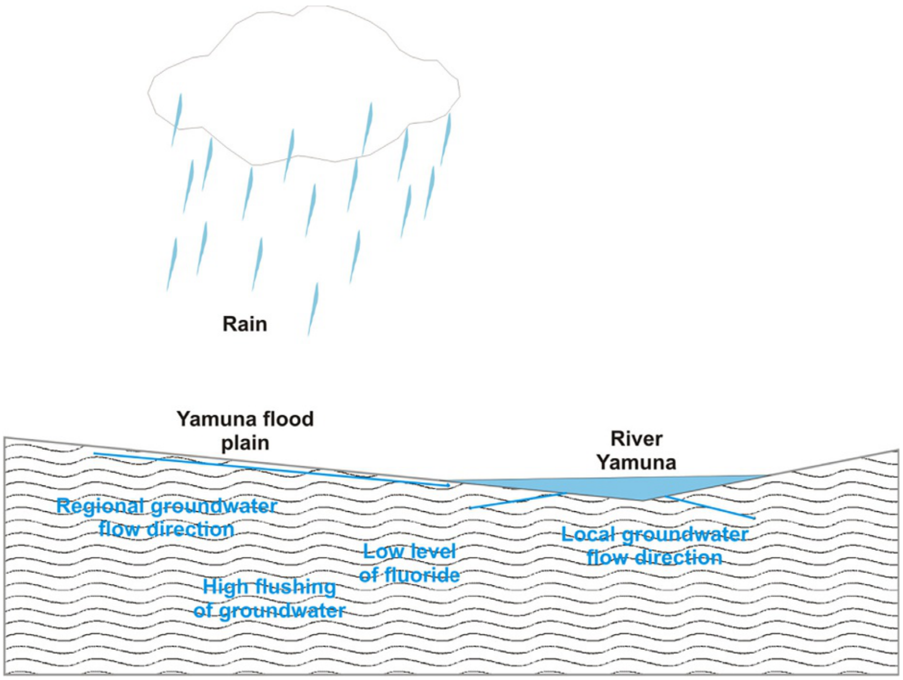

FIG. 9 A schematic figure explaining the possible causes for a low level of F̄ in the YFP.

Acknowledgments

It is duly acknowledged that the present work is a part of the Ph.D. of SA. This research was based on the datasets of Ph.D. and M.Phil. of SA. This work was supported by the R&D project (2015-16) of the University of Delhi granted to SS and is duly acknowledged. SA acknowledges the non-NET fellowship provided by the University of Delhi. It is to acknowledge that an abstract entitled "Major ions chemistry and non-carcinogenic health risk assessment of fluoride in groundwater of the alluvial plains of River Yamuna, Delhi, India" using partially the dataset of the article was accepted for the 2nd International Conference on Water Resources in Arid Areas, Muscat, Oman" but could not be presented due to disruptions caused by the COVID-19 pandemic.

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