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Assessment of fluoride in groundwater and urine, and prevalence of fluorosis among school children in Haryana, India

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Abstract

Considering the health effects of fluoride, the present study was undertaken to assess the concentration of fluoride in groundwater, and urine of school children in Bass region of Haryana state. Fluoride in groundwater was observed to vary from 0.5 to 2.4 mg/l with an average concentration of 0.46 mg/l. On the other hand, F^- in urine ranged from below the detection limit to 1.8 mg/l among girls and 0.17–1.2 mg/l among the boys. Higher average concentration of fluoride in urine (0.65 mg/l for boys and 0.34 mg/l for girls) may be ascribed to exposure to bioavailable fluoride through food, milk, tea, toothpaste, etc., in addition to intake through groundwater. Relatively more intake of water and food by the boys might be the reason for more cases of severe dental fluorosis (44%) among boys compared to girls (29% cases of moderate to severe dental fluorosis). The groundwater quality for drinking was compromised with respect to dissolved solids, hardness, magnesium ions, and dissolved iron. Hydro-geochemical investigation revealed that rock–water interaction, in terms of direct cation exchange, dominantly regulates groundwater chemistry, and groundwater is of Ca-Na-HCO$_3$ type.

Keywords Fluoride · Urine · Fluorosis · Groundwater · School children

Introduction

Fluorine is 13th most common element in the earth's crust and is widely distributed as fluorspar (CaF$_2$), fluorapatite (Ca$_5$(PO$_4$)$_3$), and cryolite (Na$_3$AlF$_6$). These minerals are easily soluble in water and, therefore, higher fluoride levels are observed in groundwater which interacts with fluoride containing rocks during percolation. Consumption of fluoride (0.5–1.0 mg/l) through drinking water is beneficial in terms of dental health preventing dental caries. But, higher levels of fluoride (> 1.5 mg/l) may result in dental or skeletal fluorosis depending on the intensity and duration of exposure. Apart from drinking water, exposure may also occur through bioavailable fluoride present in milk, food, meat (IPCS 2002), toothpaste/dentifrice, tea, tobacco (Yadav et al.

2007), fluoridated salt (García-Pérez et al. 2013), etc. United States Environmental Protection Agency (USEPA) recommended range on fluoride in drinking water is 0.7–1.2 mg/l, effectively setting the maximum level at the lower end (0.7 mg/l) to maximize dental caries protection benefits and to prevent excessive fluoride exposure in the population which receives fluoridated salt. Absorption of ingested fluoride is observed to be 100% on empty/fasting stomach; and around 60% when taken with a calcium-rich diet (WHO 2002). Hector et al. (2009) reported that retention of fluoride in young children may be higher ($\approx$ 70%) on account of higher ingestion through food and dentifrice. Young children swallow a part of dentifrice voluntarily or involuntarily during brushing which may result in enhanced exposure to systemic fluoride. Once ingested, fluoride reaches the blood stream within 30 min to 02 h depending upon ease in digestion of food and absorption of fluoride. Blood/plasma fluoride level is a contemporary biomarker of exposure to fluoride. Apart from blood, bone surface area, saliva, milk is also contemporary biomarkers; and whereas nails and hair are biomarkers of recent exposure, bone and teeth are the biomarkers for historical exposure (WHO 2014). But, till date urine is considered as the most useful biomarker for

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fluoride exposure since it is widely used, relatively simple, and non-invasive (Szymaczek and Lewicka 2005).

Prevalence of dental fluorosis in early age (15–20 years) is an expression of higher exposure to fluoride, and it is fairly important since it affects permanent teeth leading to mild, moderate, or severe level of dental fluorosis. Some studies (García-Pérez et al. 2013; Del Carmen et al. 2016) have reported prevalence of dental fluorosis in school children of younger age groups of 8–12 years and 12–15 years as well. Therefore, monitoring of urinary fluoride concentration is a useful tool to assess the exposure particularly among the children at places with problem of endemic fluoride. Continued exposure to high level of fluoride may lead to osteo/ skeletal fluorosis after the age of 30 years, but it shall be preceded with dental fluorosis. Skeletal fluorosis is accompanied by neurological, excessive tiredness, sleepiness, headache, and loss of sensation, altered reflexes, impotence, and loss of sphincter control. Some studies have reported (Jolly et al. 1974) that skeletal fluorosis is also associated with nervous system damage. Studies in USA had no clear evidence that consumption of water with fluoride level up to 8 mg/l had any association with dental and skeletal fluorosis, but the reports from India and China clearly present an evidence of dental fluorosis, osteosclerosis, and bone fracture at concentration of 6 mg/l (IPCS 2002). Investigations by different authors in China have reported an association between higher level of fluoride in drinking water and lower IQ of children (Zhao et al. 1996; Xiang et al. 2003). Therefore, it becomes important to study the exposure of children, particularly school children, towards higher level of fluoride in drinking water in areas with endemic fluoride problem. Apart from fluoride, it is important to classify the water in respect of suitability for drinking for rest of the chemical constituents (Haritash et al. 2016); and to determine the major geochemical processes responsible for regulating groundwater chemistry (Haritash et al. 2017).

Materials and methods

Haryana is a major agricultural state of India with an area of 44,212 Km^2, and it has regional pockets of high concentration of fluoride in groundwater throughout its stretch (Haritash et al. 2008). Bass village of Hisar district in Haryana was selected as the study area since it has been identified as a water-challenged site by Department of Science and Technology owing to the problem of fluoride in groundwater. The village is classified as exclusive rural area with a total population of about 15,000 people. The sources of groundwater (hand-pump, tube-well, and submersible pump) were identified to collect representative groundwater samples to determine fluoride, since groundwater is the primary source for drinking and irrigation. A total of 25 groundwater

Table 1 Concentration of fluoride in ground water and urine, and prevalence of fluorosis among school children in Bass village

Parameter		Groundwater ($n=25$)	Urinary fluoride (girls) ($n=25$)	Urinary fluoride (boys) ($n=25$)
Range				
	Min.	0.10	BDL	0.17
	Max.	2.40	1.80	1.20
Mean ± SD		0.46 ± 0.47	0.34 ± 0.44	0.65 ± 0.29
Prevalence of fluorosis (%)	Nil		23	32
	Mild		48	24
	Moderate		21	36
	Severe		8	8

samples were collected from various locations within the village and from the fields around the village. The schoolchildren aged between 15 and 18 years were chosen as subjects ($n=50$) with equal number of both the genders to assess the exposure through urinary fluoride levels. The younger age-groups were excluded since it is difficult for the young children to hold urine for about 4 h after first meal of the day. The urine samples were collected during 1:00–2:00 PM in the afternoon to let the excretory concentration of ingested
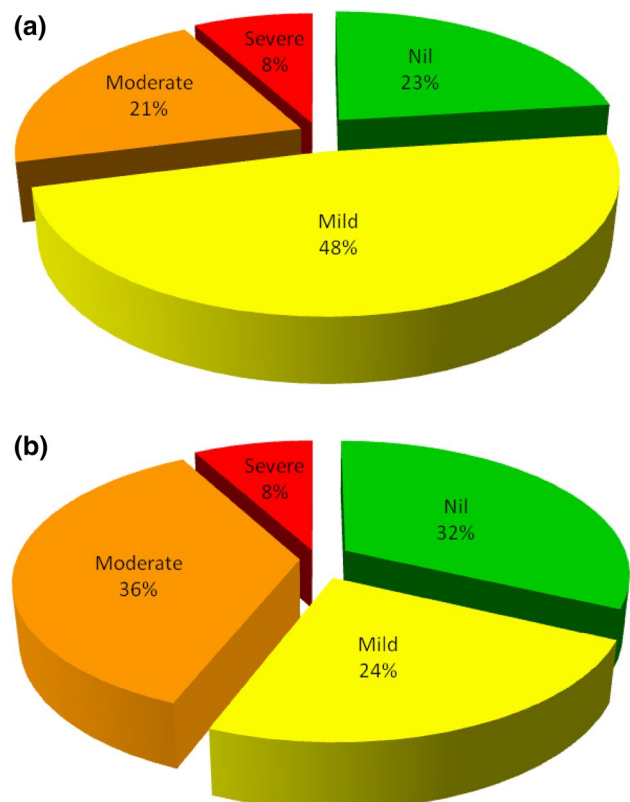

Fig. 1 Prevalence of fluorosis (%) among girls (**a**) and boys (**b**) in the present study ($n=25$ in each)

Table 2 Suitability of groundwater for drinking in the present study

| S. No.	Parameter	Value/concentration		Desirable limit		Suitable samples (%) ($n = 25$)
		Range	Mean $\pm$ SD	BIS (2012)	WHO (2006)	
1	pH	6.8–8.2	7.5 $\pm$ 0.33	6.5–8.5	NM	100[a]
2	Total dissolved solids (TDS) (mg/l)	58–5350	1144 $\pm$ 1192	500	NM	24[a]
3	Total hardness (as CaCO$_3$) (mg/l)	56–2740	526 $\pm$ 522	200	NM	20[a]
4	Sodium (Na$^+$) (mg/l)	9–727	186 $\pm$ 208	NM	200	64[b]
5	Calcium (Ca^{2+}) (mg/l)	8–331	96 $\pm$ 98	75	NM	68[a]
6	Magnesium (Mg^{2+}) (mg/l)	0–463	69 $\pm$ 91	30	NM	32[a]
7	Iron (Fe) (mg/l)	0.83–1.46	0.91 $\pm$ 0.12	0.3	0.3	0[a]
8	Chloride (Cl$^-$) (mg/l)	14–701	171 $\pm$ 226	250	250	88[a]
9	Sulphate (SO$_4^{2-}$) (mg/l)	21–315	173 $\pm$ 81	200	500	56[a]
10	Nitrate (NO$_3^-$) (mg/l)	0.3–10.1	4.9 $\pm$ 3.1	45	50	100[a]
11	Fluoride (F$^-$) (mg/l)	0.05–2.4	0.46 $\pm$ 0.47	1.0	1.5	96[a]

SD Standard deviation, *NM* not mentioned

[a]As per BIS

[b]As per WHO

Fig. 2 Piper-trilinear diagram for geochemical classification of groundwater

1 - CaHCO$_3$ Type
2 - NaCl Type
3 - Mixed CaNaHCO$_3$ Type
4 - Mixed CaMgCl Type
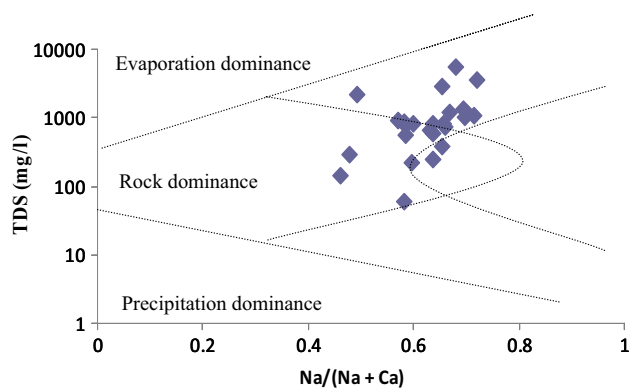5 - CaCl Type
6 - NaHCO$_3$ Type
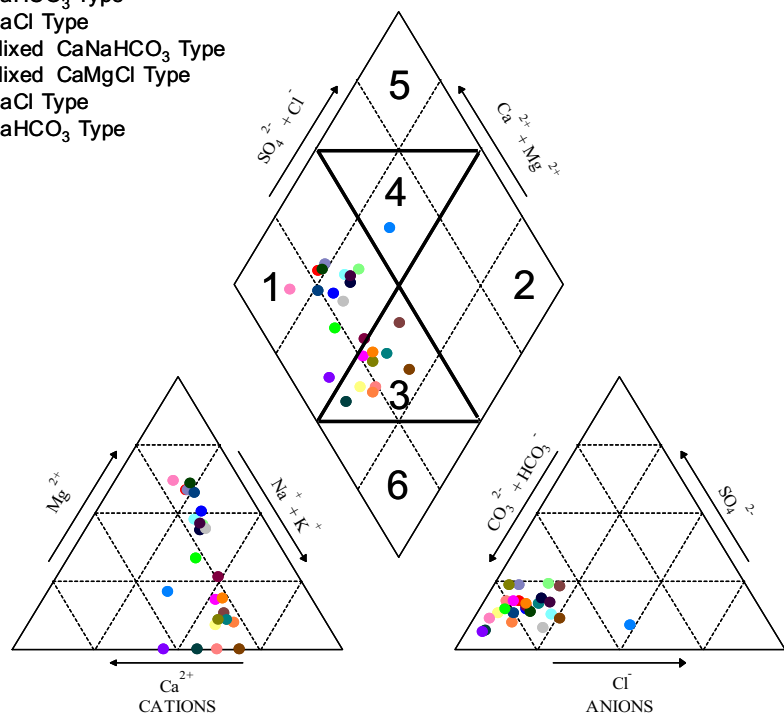
Fig. 3 Gibb's diagram for the factors regulating groundwater chemistry

fluoride reach urine. Spot samples of urine were collected in pre-cleaned polypropylene (PP) bottles of 50 ml capacity. The collection of urine samples was done with prior intimation and consent of the parents; and approval from the institutional committee for ethics and research. Fluoride concentration in groundwater and urine samples was determined using a F^- ion specific electrode (Orion 96-09 BNWP) fitted with an Orion Star A329 ISE Meter, MA, USA. A cross-sectional survey was also carried out (for 50 students) to assess the prevalence of dental fluorosis. Based on the prevalence, dental fluorosis was classified as nil, mild, moderate, and severe as per the Dean's Index (Dean 1942). Physicochemical characterization of groundwater was also performed using standard methods of APHA (Eaton et al. 1995) to get the information about possible soil–water chemistry governing the water quality. The statistical analysis of the results obtained was done on SPSS ver. 20. Analysis of correlation, cluster analysis, and principal component analysis were performed to understand the factors controlling groundwater chemistry.

Results and discussion

Based on the analysis of groundwater, it was observed that fluoride was within the permissible limit for most of the samples. It varied from 0.10–2.40 mg/l with an average value of 0.46 mg/l in groundwater (Table 1). Only one sample exceeded the standard prescribed limit of 1.0 mg/l (BIS 2012). Based on the recorded mean concentration, it may be inferred that exposure to fluoride from drinking water

is not of much concern in the study area. However, spatial variations and observed value of 2.40 mg/l at one location indicates that the health of exposed population may be compromised at few places, particularly if the source is used for drinking water. Moreover, intake of more volume of water during early growing stage may result in fluorosis among children since most of the ingested minerals are absorbed by the body during growing stage (Whitford 1999). Absorption of fluoride is regulated by the diet as well. The population having more of vegetables in food is expected to excrete more of fluoride in urine compared to those who consume more of meat (WHO 2014). In the present study, urinary fluoride concentration varied from below the detection limit (BDL) to 1.8 mg/l for girls and 0.17–1.20 mg/l for boys, with a mean value of 0.34 and 0.65 mg/l, respectively. Low concentration of urinary fluoride in girls may be attributed to relatively less consumption of drinking water and food compared to boys. The average level of fluoride in urine of boys was higher than in drinking water which may be an outcome of additional ingestion of fluoride via intake of tea, milk, vegetables, etc. Similar observations of higher F^- in urine have been made in other studies too (Paez and Dapas 1983; Szymaczek and Lewicka 2005; Rango et al. 2014; Del Carmen et al. 2016) ascribing the reason to additional intake of bioavailable fluoride through food. Based on the survey for dental fluorosis, it was observed that majority of the subjects suffered from dental fluorosis with the degree of expression varying from nil to severe through mild and moderate as per Dean's Index. Prevalence of fluorosis was observed to be 77% among the girls, and 68% among the boys (Table 1). But based on the grading of intensity, most of the girls had mild fluorosis (48%) followed by moderate (21%) and severe (8%). On the other hand, most of the boys suffered from moderate fluorosis (36%) and relatively less from mild (24%) and severe (8%) (Fig. 1). More prevalence of moderate and severe dental fluorosis (44%) among boys (As against 29% of girls) further strengthens the premise that boys had more ingestion of fluoride through food and water, and hence higher urinary fluoride concentration. Statistical evaluation for correlation between urinary fluoride concentration and prevalence of fluorosis, too, revealed relatively stronger correlation for boys (0.64) than the girls (0.44) (p value > 0.01). Calculations of correlation coefficients among different physico-chemical parameters revealed that fluoride is significantly and positively correlated with Na and Ca indicating the possible presence of fluorspar, fluoraptite, cryolite in sub surface soil/rocks. While determining the

Table 3 Concentration of fluoride in groundwater and urine, and prevalence of dental fluorosis in different regions of the world

S. No.	Country	Region	Age group (years)	F$^-$ in drinking water (mg/l)	F$^-$ in urine (mg/l)	Prevalence of dental fluorosis (%)	Criteria for classification of dental fluorosis	Reference (Remarks)
1.	India	Bass Village, Haryana	15–18	0.1–2.4	0.17–1.80	72.0	DI	Present Study
2.	Mexico	(a) Morelos	8–12	0.7–1.5	NA	39.4–60.5	TFI	García-Pérez et al. 2013
		(b) Guanajuato	11–20	4.4	0.50–6.65	91.90	TFI	Del Carmen et al. 2016
3.	Ethiopia	Ethiopian Rift Valley	10–15	1.1–18.0	1.1–39.8	91.00	TFI	Rango et al. 2014
4.	China	(a) Xinhuai	8–13	0.18–0.76	0.37–2.50	NA	–	Xiang et al. 2003 (Lower IQ level related to higher fluoride in drinking water)
		(b) Wamiao		0.57–4.50	0.90–12.50			
5.	Poland	Poznan	22–34	0.40–0.80	0.84–2.22	NA	–	Szymaczek and Lewicka 2005
6.	Venezuela	Maracay, Aragua	1.25–6	0.07–0.09 (*average total intake-mg/kg/day)	0.79–0.92 (average values)	NA	–	Hector et al. 2009
7.	Rajasthan	(a) Banswara (b) Dungarpur (c) Udaipur	17–22	1.5–4.0	NA	77.11	DI	Choubisa 2001
8.	India (Haryana State)	(a) Jhajjar City (b) Dadanpur Village (c) Dariyapur Village	8–15	1.90–2.60 2.53–3.14 1.63–3.33	0.05–2.64 0.69–2.80 0.31–2.50	51.90 94.63 36.84	TSIF	Kumar et al. 2017
9.	India (Haryana State)	(a) Beri Block (b) Bahadurgarh (c) Jhajjar Block (d) Sahlawas Block (e) Matanhail Block	7–15	1.56–3.05 1.53–3.52 1.52–4.00 1.53–3.14 1.52–3.16	NA	59.76 66.32 44.99 58.02 62.10	TSIF	Yadav et al. 2009

DI Dean's Index, *TFI* Thylstrup and Fejerskov Index, *TSIF* Tooth Surface Index of Fluorosis (Horowitz et al. 1984), *NA* not analysed

suitability of groundwater, the only source for drinking, it was observed that the quality was compromised with respect to total dissolved solids (TDS), total hardness (TH), Mg, Fe and SO_4^{2-} chiefly (Table 2), apart from the effect of fluoride. A comparative analysis of concentration of fluoride in drinking water, urine, and prevalence of fluorosis in schoolchildren around the world has been made in Table 3.

Hydro-geochemical classification of the ground water revealed that most of the samples were $CaHCO_3$ type or mixed $CaNaHCO_3$ type. Piper tri-linear classification (Piper 1953) indicated that the alkaline (Na^+ and K^+) species exceeded the alkaline earth (Ca^{2+} and Mg^{2+}); and

weak acids (CO_3^{2-} and HCO_3^-) exceeded strong acids (Cl^- and SO_4^{2-}) (Fig. 2). Based on the values of base-exchange (Matthess 1982) all the samples were found to be sodium sulfate typed (value < 1) except one sample. Negative values of chlor-alkaline index (Schoeller 1977) suggest that most of the sample had direct base-exchange reaction resulting in exchange of Ca and Mg of rocks/ soil with Na and K of water. Gibbs classification (Gibbs 1970) to determine the major regulating factor revealed that the quality of ground water is predominately governed by rock–water interaction (Fig. 3). The average values of ratio of Ca/Na (0.72), Mg/Na (1.15), and HCO_3/Na (3.84)

Table 4 Principal component analysis of observed parameters in the present study

Variables	Component			
	1	2	3	4
pH	−0.50	0.32	**0.66**	0.14
EC	0.33	**0.85**	−0.21	−0.05
TDS	0.33	**0.85**	−0.20	−0.05
TH	**0.85**	−0.45	0.03	−0.18
TA	**0.95**	−0.13	0.21	0.01
Na	**0.86**	0.35	0.27	0.12
K	0.37	−0.45	−0.42	−0.07
Ca	**0.90**	0.28	0.01	−0.04
Mg	**0.77**	−0.56	0.13	−0.07
Li	**0.95**	−0.06	0.06	0.04
Fe	0.15	0.09	−0.07	**0.89**
CO_3^{2-}	**0.73**	−0.38	0.51	0.08
HCO_3^-	**0.93**	0.07	−0.06	−0.05
Cl^-	**0.80**	0.12	−0.18	−0.31
SO_4^{2-}	**0.83**	0.16	−0.23	0.08
NO_3^-	**0.51**	−0.31	−0.21	0.45
PO_4^{3-}	**0.72**	0.04	0.33	0.02
F^-	0.35	**0.70**	0.41	−0.06
Eigen values	9.054	3.301	2.122	1.195
CV (%)	47.66	65.03	76.20	82.49

Statistically significant p values are in bold ($p < 0.05$)

indicated that silicate weathering primarily governs water chemistry (Negrel et al. 1993) followed by carbonate weathering as represented by high average value (5.82) of (Ca + Mg)/(Na + K) ratio.

Statistical analysis

The statistical analysis of the results obtained revealed that a significant positive correlation of fluoride existed with sodium, TDS, and EC indicating to natural dissolution of fluoride from the underlying strata. Among the other ions, significant positive correlation of Na was observed with Ca, Li, CO_3^{2-}, HCO_3^-, Cl^-, SO_4^{2-} etc. and these ions significantly contributed to total alkalinity and total hardness of water. Principal component analysis (PCA) (Table 4) also revealed that total alkalinity, total hardness, sodium, potassium, lithium, calcium, magnesium, carbonate, bicarbonate, chloride, and sulphate regulated the water quality; and indicates that the source is primarily natural. A hierarchical cluster analysis using Centroid Linkage was performed to highlight the inter-relationships among various sites along with the selected water parameters. The dendrogram classified the data into three cluster membership. First cluster includes all the sites except site 21 (second cluster), and site 23 (third cluster) (Fig. 4). Cluster analysis indicated that the water quality is almost the same with the only variation at two sites (Site 21 and 23).

Conclusion

The study concludes that the concentration of fluoride in groundwater in Bass village is within the prescribed limits except a few locations. Although intake through drinking water is limited, ingestion of fluoride through food, tea, milk, toothpaste, etc., may be alternate route for exposure. Depending upon the total daily ingestion, boys are more exposed to fluoride as observed in higher average urinary fluoride level. It has lead to more severe cases of dental fluorosis among the boys than the girls. Significant number of cases of dental fluorosis (72%) despite a relatively lower concentration of fluoride in drinking, as compared to other studies around the world, is a cause of concern. The study recommends extensive screening of groundwater sources for fluoride levels, and a regular monitoring of 24-h urine samples for a bigger sample size to better understand
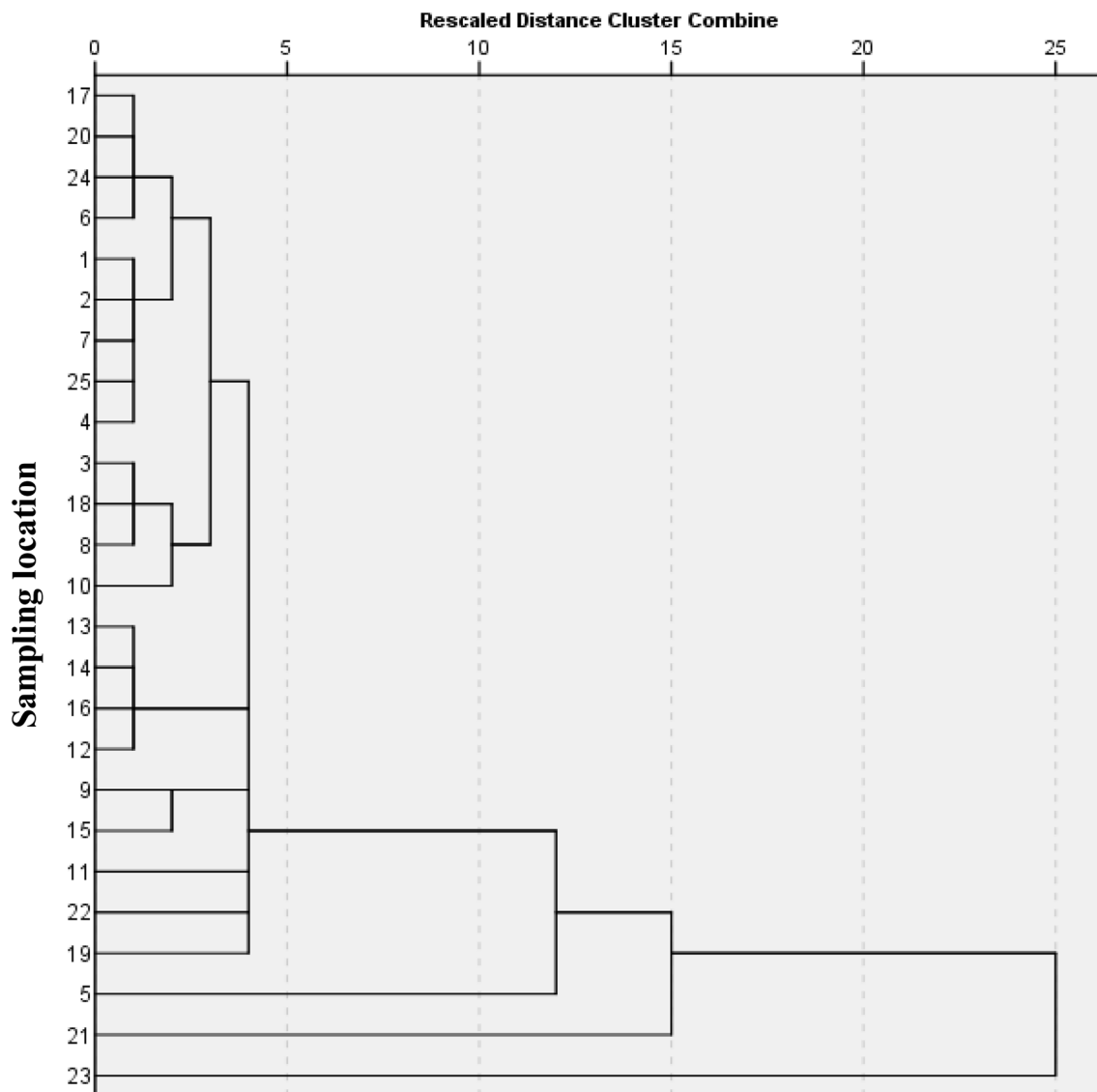
Fig. 4 Cluster analysis of the locations with respect to analysed parameters

the exposure to fluoride, and its control. Further, suitable treatment of groundwater may be targeted to improve water quality especially for dissolved solids, hardness, magnesium ions, and dissolved iron for use in drinking.

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