ASSESSMENT OF FLUORIDE CONTAMINATION IN GROUNDWATER OF DISTRICT MARDAN IN PAKISTAN

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ABSTRACT: If groundwater contains elevated levels of the fluoride ion (F), its consumption for drinking purposes is a major F exposure pathway for humans. Groundwater samples (n=100) were collected from 25 villages of District Mardan of the Khyber Pakhtunkhwa (KP) province, Pakistan, and analyzed for anions including F, chloride (Cl^-), bromide (Br^-), phosphate (PO_4^{3-}), and sulfate (SO_4^{2-}) with ion chromatography. The range of the F concentrations in the groundwater samples was 0.05–10.8 mg/L. The measured concentrations were within the permissible limits of National Environmental Quality Standards (NEQS) of Pakistan for drinking water in 92% of the samples. Groundwater samples of three villages exhibited F concentrations greater than the permissible level of 1.5 mg/L. The F concentration showed a positive correlation with electrical conductivity (EC), total dissolved solids (TDS), and the concentrations of Br^-, PO_4^{3-}, and SO_4^{2-}. To safeguard public health, this study suggests the need for further research in the area for possible sources of F contamination and mitigation measures.

Key Words: Contamination; Fluorosis; Health Risks; Monitoring; NEQS; Water Quality.

INTRODUCTION

Drinking water contaminated with the fluoride ion (F) is a major pathway for human exposure to F, mainly because of its maximum bio-accessibility in the aqueous medium. Although the topical application of F to the teeth is considered to be beneficial for the prevention of dental caries, F is neither an essential trace element nor necessary for the development of healthy teeth and bones and the intake of drinking water with concentrations beyond certain levels can cause health problems in humans.[1-4] Millions of children and adults are facing the risks of dental, skeletal, and non-skeletal fluorosis in regions such as South America to Africa, Asia, and Australia.[5–8]

The risk for the occurrence of fluorosis in human populations depends on the level of F intake which may result from airborne F being inhaled and the consumption of food with high F levels but usually depends on the concentration of F in drinking water and the rate of its consumption.[9,10] Some of the key factors affecting the levels of F in groundwater are the climatic conditions of the region, bedrock composition, the solubility of minerals containing F, pH, rate of groundwater exploitation, and groundwater flow rates.[5,11] F ubiquitously accounts for 0.3 g/kg of the earth crust, and is naturally present in groundwater and plants in the form of various minerals. Some major sources of groundwater contamination of F are rocks and soils rich with minerals like fluorite, cryolite, and fluoroapatite. Anthropogenic sources are dietary products, mining activities, pesticides, fertilizers, coal combustion, and industrial waste.[3,12–15] The second most important pathway for the development of fluorosis

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after exposure to high F-water is the consumption of plants and animals exposed to F-contaminated soils.[16,17]

According to the World Health Organization (WHO) and the National Environmental Quality Standards (NEQS) of Pakistan, the permissible value for the F concentration in drinking water is 1.5 mg/L.[3,5] Prior studies in Pakistan have reported F contamination of groundwater in various areas of the Thar Desert, Sindh province, villages along the rivers Chenab and Mailsi in Punjab province, Mastung, Pringabad, and Manguchar Baluchistan provinces, and Naranji village of District Swabi in Khyber Pakhtunkhwa (KP) province.[11,13,18-27] However, baseline data on the status of F concentration in the groundwater for District Mardan is missing in the literature. Such data are necessary to help guide the local policy making process for drinking water. Therefore, the focus of this work was to investigate the status of F contamination in the groundwater of District Mardan.

MATERIALS AND METHODS

Study area: Drinking water samples of groundwater were collected from 25 villages of District Mardan. In this area, some of the very common minerals present in alkaline bedrock are biotite, epidot, orthpyroxene, clinopyroxene, and hintingsite, while minerals of the acidic bedrock are plagioclase, orthoclase, and ferromagnesian.[18,28] It is likely that these minerals play an important role in the contamination of the groundwater sources of the area. Mardan is an agriculturally intensive area with predominantly rural living. Groundwater is the main source of drinking water and is used without any pre-treatment.

The study area was divided into four zones for sampling and comparative analysis: (i) zone 1 (Katlang zone) which extended from 71.956–72.222 East to 34.442–34.452 North, (ii) zone 2 (Mardan zone) which extended from 71.911–72.208 East to 34.19–34.206 North, (iii) zone 3 (Rustam zone) which extended from 72.147–72.428 East to 34.346–34.383 North, and (iv) zone 4 (Takhtbahi zone) which extended from 71.825–71.996 East to 34.319–34.311 North (Figure 1).

Water sampling: Groundwater samples were collected from 25 villages in clean plastic bottles rinsed three times with the source water.[29] A total of 100 samples were collected randomly from the groundwater sources of the four zones of the district. The number of samples collected from the zones were: zone 1=24, zone 2=24, zone 3=28, and zone 4=24. Geographic coordinates, depth of the well, and elevation of all the locations were recorded at the time of sample collection.

Chemical analysis: All the water samples were analyzed for pH, electrical conductivity (EC), and total dissolved solids (TDS) using Inolab WTW Series inoLab pH 720, Jenway Model 4510 Conductivity Meter and Consort Electrochemical Analyzer C931, respectively. Anions F^- (F), Cl^-, Br^-, PO_4^{3-}, and SO_4^{2-} were analyzed by Ion Chromatography Dionex ICS-1000 (limit of detection 5 μmoles/L) at Arizona Laboratory for Emerging Contaminants, The University of Arizona, USA. All of the analyses were conducted according to the standard methods for the examination of water and waste water.[29]

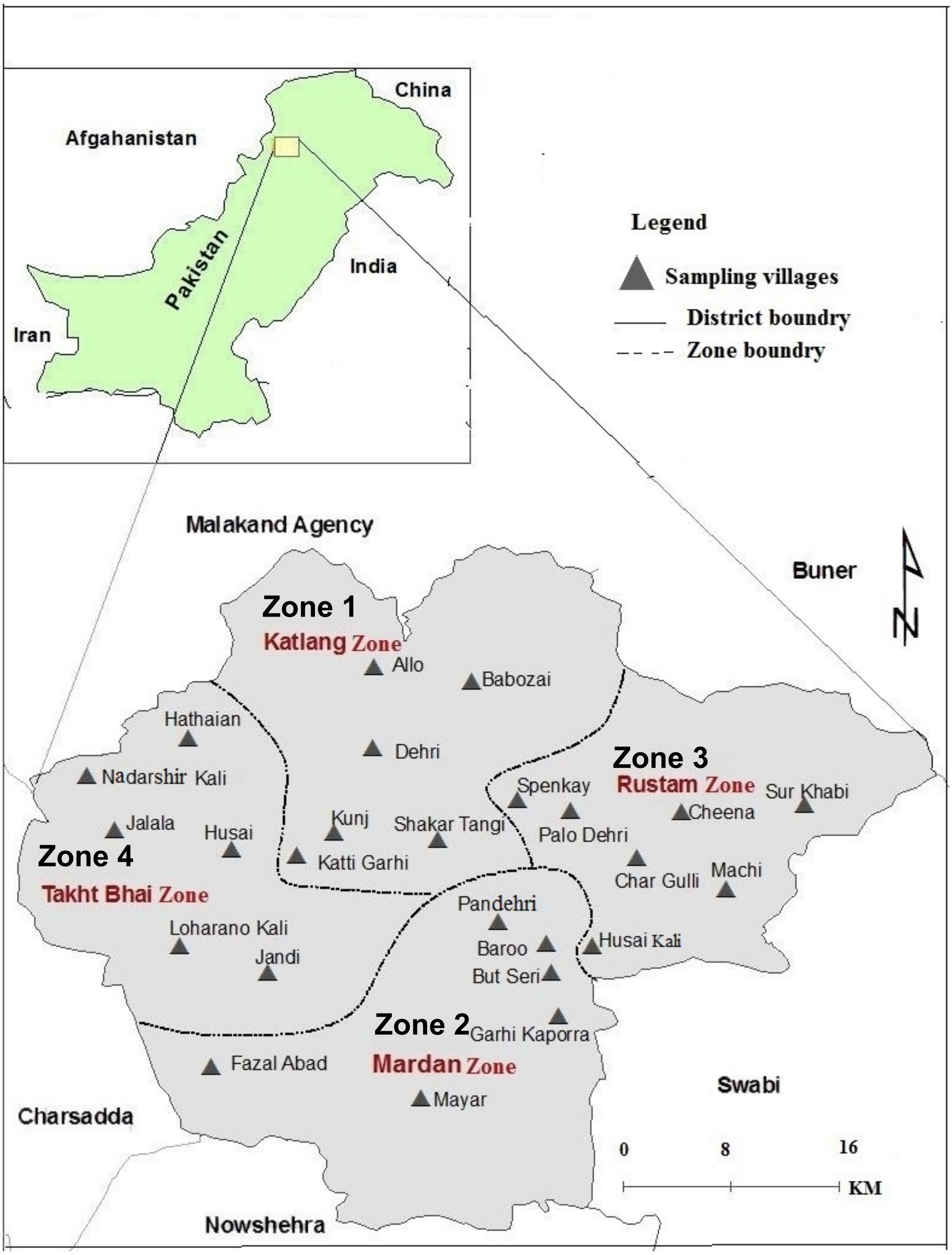

Figure 1. Map of the study area of District Mardan showing the locations of the villages where the groundwater samples were obtained in each of the four zones: (i) zone 1 (Katlang zone) (ii) zone 2 (Mardan zone), (iii) zone 3 (Rustam zone), and (iv) zone 4 (Takhtbahi zone).

Statistical analysis: Descriptive statistics of the data including mean, range, standard deviation, and standard error were calculated using Excel 2016. Prior to correlation analysis, F concentration data were tested for normality. SPSS version 21 was used for normality and Spearman correlation analysis.

RESULTS

Analysis of the data show that the mean values of TDS, Cl^-, and SO_4^{2-} were lower than the guideline values of WHO, and NEQS while the mean values for EC and PO_4^{3-} were higher. The concentration of F ranged from 0.05 to 10.8 mg/L. However, variations were observed in the mean values in the zones. For instance, zone 2 had the highest mean value of 1.57 mg/L followed by zone 3, zone 4, and zone 1 with 0.70, 0.55, and 0.37 mg F/L, respectively, (Tables 1 and 2).

Table 1. Basic groundwater parameters measured for the sampled water of District Mardan comprising the four zones (i) zone 1 (Katlang zone), (ii) zone 2 (Mardan zone), (iii) zone 3 (Rustam zone), and (iv) zone 4 (Takhtbahi zone). (EC=electrical conductivity, TDS=total dissolved solids, NA=not available)

Parameter	WHO guideline	Statistic	District Mardan	Zone 1 (n=24)	Zone 2 (n=24)	Zone 3 (n=28)	Zone 4 (n=24)
Depth of the source (feet)	NA	Range	8–245	8–200	40–245	35–150	38–200
		Mean	86.00	81.41	95.33	82.00	85.00
		±SD	39.00	41.64	44.67	32.00	40.90
Elevation (m)	NA	Range	291–453	350–453	291–333	320–414	306–390
		Mean	355.00	390.60	309.00	366.00	352.00
		±SD	39.00	36.86	11.37	24.00	25.00
pH	6.5-8.5	Range	6.54–8.51	6.82–7.60	6.73–8.51	6.54–8.01	7.18–8.17
		Mean	7.42	7.16	7.52	7.30	7.72
		±SD	0.38	0.16	0.39	0.40	0.29
EC (mS/cm)	0.25	Range	0.23–3.29	0.46–1.82	0.37–3.29	0.23–1.37	0.53–1.25
		Mean	0.91	0.80	1.25	0.70	0.96
		±SD	0.45	0.31	0.66	0.30	0.19
TDS (mg/L)	1000	Range	75–938	152–576	89.50–938	75–386	106–400
		Mean	256.00	243.30	358.90	171.00	263.00
		±SD	137.00	98.15	198.30	77.00	70.80

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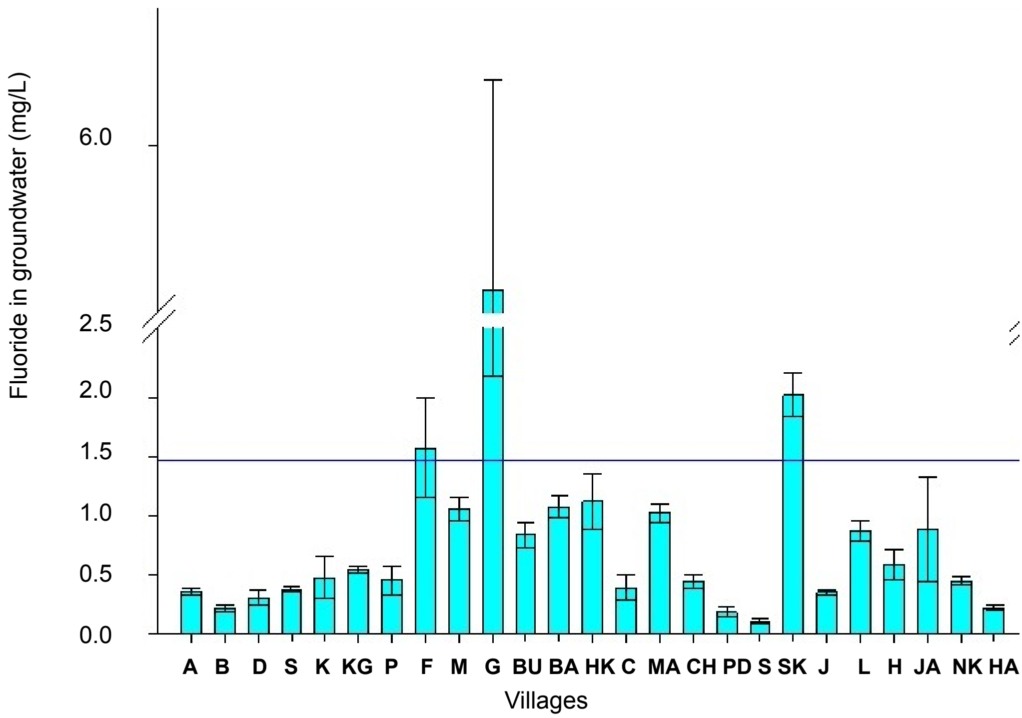
Table 2. Basic groundwater parameters measured for the sampled water of District Mardan comprising the four zones (i) zone 1 (Katlang zone), (ii) zone 2 (Mardan zone), (iii) zone 3 (Rustam zone), and (iv) zone 4 (Takhtbahi zone).

Parameter	WHO guideline	Statistic	District Mardan	Zone 1 (n=24)	Zone 2 (n=24)	Zone 3 (n=28)	Zone 4 (n=24)
Cl^- (mg/L)	250	Range	2.90–619.20	3.60–184	2.20–619.20	3.10–65	4.00–120
		Mean	40.00	22.75	95.80	19.00	25.00
		±SD	86.00	36.71	157.70	15.00	26.50
Br^- (mg/L)	NA	Range	0.00–2.27	0.00–0.48	0.00–2.27	0.01–0.23	0.01–0.20
		Mean	0.15	0.07	0.39	0.10	0.07
		±SD	0.32	0.10	0.58	0.10	0.05
PO_4^{3-} (mg/L)	0.01	Range	0.00–0.86	0.00–0.37	0.00–0.13	0.00–0.86	0.00–0.60
		Mean	0.06	0.06	0.02	0.10	0.09
		±SD	0.14	0.11	0.04	0.20	0.17
SO_4^{2-} (mg/L)	500	Range	5.10–409	10.90–152	10.82–409	5.10–122	8.44–171
		Mean	51.00	42.89	88.00	27.00	49.20
		±SD	57.00	38.97	92.00	23.00	36.00
F^- (mg/L)	1.5	Range	0.05–10.80	0.14–1.00	0.09–10.80	0.05–2.33	0.18–2.19
		Mean	0.79	0.37	1.57	0.70	0.55
		±SD	1.20	0.18	2.15	0.60	0.43

F contamination in the groundwater samples was further classified according to the concentration range: >1.5, 1–1.5, and <1 mg/L, respectively. Out of a total of 100 samples, 8% had F concentrations above the permissible level of 1.5 mg/L and 14% were in the range of 1–1.5 mg/L. Among the zones, zone 3 showed F concentrations beyond the WHO and NEQS permissible level of 1.5 mg/L in 14% samples, followed by zone 2 (12%) and zone 4 (4%). Some of the villages exhibiting higher F contamination than the NEQS and WHO guidelines included Fazal Abad Gharbi and Dolatzai area in Garhi Kapura (zone 2), Husai Kali and Sur Khabi (zone 3), and Jalala Kali camp area (zone 4) with 4,000, 20,000, 3,000, 700, and 9,000 inhabitants, respectively (Figure 2).



Code for villages:
A=Alo
B=Babozai
D=Dehri
S=Shakar Tangi
K=Kunj
KG=Katti Garhi
P=Pandheri
F=Fazal Abad
M=Mayar
G=Garhi Kapura
BU=But Seri
BA=Baroo
HK=Husai Kali
C=Cheena
MA=Machi
CH=Charguli
PD=Palo Dehri
S=Spenkay
SK=Sur Khabi
J=Jandai
L=Loharano Kali
H=Hussai
JA=Jalala Kali
NK=Nadarshir Kali
H=Hatyan

Figure 2. Fluoride concentration in the groundwater samples of the 25 villages in District Mardan

Correlation analysis: The concentration of F in the groundwater samples for District Mardan, comprising all four zones, exhibited a significant positive correlation (p=0.01) with EC, TDS, Br^{-}, PO_4^{3-}, and SO_4^{2-} (r=0.314, 0.314, 0.277, 0.401, 0.276, respectively) (Table 3). However, some variation was present in the correlation values for the four individual zones. For instance, pH had a significant positive correlation with F concentration of the groundwater for zone 1 (r=0.572, p=0.01, n=24), zone 4 (r=0.510, p=0.05, n=24) and a negative correlation (r= –0.413, p=0.05, n=28) for zone 3. The well depth was negatively correlated with the F concentration (r= –0.406, p=0.05, n=24) for zone 4. A positive correlation of the F concentration was present with Cl^{-} for the groundwater samples of zone 2 (r=0.434, p=0.05, n=24).

Table 3. Spearman correlation of the F concentration with the other tested water parameters of District Mardan comprising the four zones (i) zone 1 (Katlang zone), (ii) zone 2 (Mardan zone), (iii) zone 3 (Rustam zone), and (iv) zone 4 (Takhtbahi zone). (r= Spearman correlation coefficient, n=number of groundwater samples, DM=District Mardan)

Area		Depth	pH	EC	TDS	Br^-	$PO4^{3-}$	Cl^-	SO_4^{2-}
DM	r	–0.041	0.185	**0.314†**	**0.314†**	**0.277†**	**0.401†**	0.171	**0.276†**
	n	100	100	100	100	100	100	100	100
Zone 1	r	0.017	**0.572†**	0.358	0.359	0.150	0.045	0.347	0.057
	n	24	24	24	24	24	24	24	24
Zone 2	r	–0.221	0.272	0.149	0.193	–0.203	0.038	**0.434***	–0.021
	n	24	24	24	24	24	24	24	24
Zone 3	r	0.034	**–0.413***	0.014	0.071	0.045	0.233	0.260	0.198
	n	28	28	28	28	28	28	28	28
Zone 4	r	**–0.406***	**0.510***	0.072	0.070	0.085	0.076	0.101	–0.090
	n	24	24	24	24	24	24	24	24

*Correlation is significant at the 0.05 level (2-tailed);
†Correlation is significant at the 0.01 level (2-tailed).

DISCUSSION

Groundwater of District Mardan is reported to have high concentrations of HCO_3^-, Na^+, K^+, Ca^2, and Mg^{2+}.[27] F concentrations in groundwater are relatively higher in the South plains and the North East hilly areas of the district. The calculated Spearman correlation values of the measured F levels in the groundwater samples with the various parameters measured in the current study are consistent with the available literature. The solubility of F in water, for instance, is pH dependent and the presence of various salts of K^+, Ca^{2+}, Na^+, Cl^-, Mg^{2+}, HCO_3^-, and SO_4^{2-} help maintain the required conditions.[22,24,30-33] At high pH, the presence of bicarbonate dissociates F from granite rocks which then contaminates groundwater sources.[34,35] Some studies have noted that high pH facilitates the exchange of OH^- with F^- present in various rocks of the aquifers.[21] In contrast, according to a very recent study carried out in Iran, the F concentration in the groundwater has a negative correlation with pH and HCO_3^-.[36] In addition, the presence of high levels of alkalinity, as a result of carbonate and bicarbonate ions, is responsible for the weak correlation of the F concentration with the pH in the groundwater.[37]

Apart from the pH, the F concentration in groundwater depends on the TDS, the EC, and the depth of the sources.[38] Some studies have reported a negative correlation of F concentration with the depth of the groundwater sources. This would be expected for anthropogenic sources of F such as irrigation and fertilizers.[5,8,13,27,31,39] Such situation is quite likely in the study area as most of the land is used for agriculture. This is a likely explanation for the observed negative correlation of F concentration with groundwater depth for zone 4.

However, according to a study carried out in southern Pakistan, the temperature and the depth of the source showed no significant impact on the concentration of F in the groundwater.[40] On the other hand, F concentrations in zone 3 appear to be due to the presence of alkaline rocks of Ambela and Koga complex in Rustam area.[18] This may explain why a negative correlation between F concentration and groundwater depth was not observed for zone 3.

Health risks: Analysis of the data showed that 8% of the water samples were above the permissible level of 1.5 mg/L F concentration. However, this does not mean necessarily that the remainder of the areas are free from the risks and health effects of F in groundwater. Among the zones, 41% of samples from zone 2 had F in the range of 1–1.5 mg/L. Reviewing the literature shows that concentrations less than the permissible values can still pose health risks in regions of the world where people consume more water. The occurrence of mild dental fluorosis, which has been considered to be simply “a cosmetic effect)[41] and not “cosmetically obvious to the children or their parents,”[5] is likely for children in temperate areas at approximately 1 mg/L F in the water. Therefore, it is imperative to decrease the levels of F for drinking water in such areas to minimize the associated health risks.[42-44] Consequently, optimum levels of 0.3 to 0.6 mg/L have been suggested for countries like Pakistan, India, Sudan, and other temperate regions of the world.[1,45,46] Some studies have suggested a minimum recommended value of 0.5 mg/L and a maximum of 1 mg/L of F concentration for the groundwater of such regions.[3,43]

CONCLUSION

The present study was a first attempt to characterize the baseline of groundwater F concentrations for District Mardan. F concentrations in most of the groundwater samples were below the upper permissible levels of WHO and NEQS. However, F concentrations in some of the villages were above the permissible levels and thus putting the residents at the risks of fluorosis with its varied effects on health. Since no industrial activity has been reported in the proximity of the sampling locations, the use of fertilizers in agriculture and natural bedrock sources are mainly responsible for the presence of F in the area. Residents of the villages need to be educated on fluorosis and other associated risks from the consumption of groundwater. Government agencies should help ensure that drinking water sources meet health guidelines for the affected communities.

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