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Contamination of Water, Dust, Soil, Rock and Urine with Fluoride in Central India

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Abstract

At least 15% of 0.1 million people residing in 117 villages of Tamnar block (Chhattisgarh, central India) are suffering from fluorosis diseases. In this work, the contamination of F^- in the environment (*i.e.* water, soil, rock and urine) of the Tamnar block is described. The concentration variations of F^-, Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+}, Ca^{2+}, Al, Mn, Fe and Zn in the groundwater are reported. The F^- content in the water was ranged from 1.7 - 17 mg/L with mean value of 9.0 ± 3.7 mg/L. Fluoride was enriched up to 3-, 54-, 69- and 244-folds in the urine, soil, dust and rock, respectively. The cluster and factor analysis models were used to apportion sources of F^- and other elements in the water.

Keywords

Fluoride, Contamination, Groundwater, Soil, Rock, India

1. Introduction

Fluorine is a highly reactive element, and it has an important role in precipitation of various elements as minerals [1]. Fluorine contents in the soil vary between 10 - 150 mg/kg, and the majority of fluorine occurs naturally in combined forms in various rocks, soils, waters, plants, other living organisms, slag, fluxes, etc. The fluoride

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in the ground water is severely extracting from the bed rock causing a disease known as "fluorosis", which continues to be an endemic problem in most parts of the world [2]. India is among the 23 nations around the globe, where fluorosis health problems (*i.e.* dental, skeletal and/or non-skeletal) are continuing to exist mainly due to the consumption of contaminated water [3]-[21]. Most of the minerals and coal of the country are reserved in the Chhattisgarh state, India. The vast exploitation of the natural resources and overuse of ground water leads to depletion of the ground water quality in several parts of the state [22]-[23]. The endemic fluoride toxicity in the Tamnar block, India has been observed [24].

Therefore, in this work, concentration, variation and sources of F^- in the ground water, urine, dust, surface soil and rock of the Tamnar block (Raigarh, CG, India) are described for the evaluation of the risks associated with the water, dust and soil exposures.

2. Materials and Methods

2.1. Study Area

Tamnar block includes Tamnar town and 116 villages with population of 0.1 million, situated in the Raigarh district, Chhattisgarh, India. The area of the Tamnar block is ≈250 km^2 (22˚05'N - 22˚15'N and 83˚20'E - 83˚30'E). It lies at an elevation of 215 m above sea level, situated in a passing point of the Kelo River. A ≈ 285 MT coal is reserved in this area and an open cast mine of capacity ≈5.25 MT coal/Yr is in operation. The thermal power plant of capacity, 600 MW is running by pouring the industrial effluents in the environment. The area is composed of Gondwana subgroup of rocks consisting of sand stone, shale, carbonaceous shale, clay and coal seams. These rocks were subjected to deformation and crustal movement during Paleozoic times with evidence of folding and faulting.

2.2. Sample Collection

Twenty two groundwater samples from 6 villages (*i.e.* Bandhapali, Dholnara, Kunjemura, Mudagaon, Pata and Saraitola) were collected during pre and post monsoon period of years, 2008 - 2012 as prescribed in the literature [25]. The sampling locations are shown in **Figure 1**. The road dust, surface soil (0 - 10 cm depth) and stone from six villages (*i.e.* Bandhapali, Dholnara, Kunjemura, Mudagaon, Pata and Saraitola) were collected in the post monsoon period, December 2008 [26]. The first morning urine sample (100 mL) was collected in plastic bottles containing 0.2 g EDTA in year 2008. Total 36 (6 × 1 × 6)) urine samples of human, cattle, buffalo, sheep and goat were collected and shipped to the laboratory in insulated container and stored at −20˚C until use.

2.3. Sample Preparation

The groundwater sample was collected into cleaned 1-L polyethylene container. The sample was divided into two portions. The 1st portion was used for analysis of physical parameters, ammonia and anions. The 2nd portion was acidified with a few drops of ultrapure nitric acid (E. Merck) for the analysis of metals. All water samples were refrigerated at 4˚C. The soil samples were dried, crushed and sieved out of particles of mesh size < 0.1 mm. A 5.00 g soil sample was mixed with 50 mL of hot (50˚C) deionized water in an ultrasonic bath for 6 hr. The extract was used for the measurement of the pH and ions. A weighed amount of crushed granite sample (0.25 g) was placed in a 50 ml Pt-crucible by adding 2.0 g NaOH [27]. The crucible was kept in a muffle furnace and slowly raising the temperature up to 600˚C. The sample was fused up to 30 min, and the residue was dissolved with hot water. The pH of the extract was adjusted to 9.0 to precipitate the interfering ions *i.e.* Fe, Al, Mn. Then, it was filtered and diluted to 100 mL in a polyethylene volumetric flask for the F^- analysis.

2.4. Analysis

The physical parameters *i.e.* temperature (T), pH, dissolved oxygen (DO), reduction potential (RP) and electrical conductivity (EC) values were determined by HANNA made sensors. The total dissolved solid (TDS) value was determined by evaporation of the filtered water sample (through glass fiber filter) by drying at constant weight [28]. The F^- content was monitored with Metrohm ion meter-781, equipped with fluoride ion selective electrode and calomel electrode. The ion meter was calibrated with the standard solution over range of 2.0 - 25.0 mg/L F^- containing acetate buffer into 1:1 volume ratio (58 g NaCl and 57 mL acetic acid with de-ionized water and

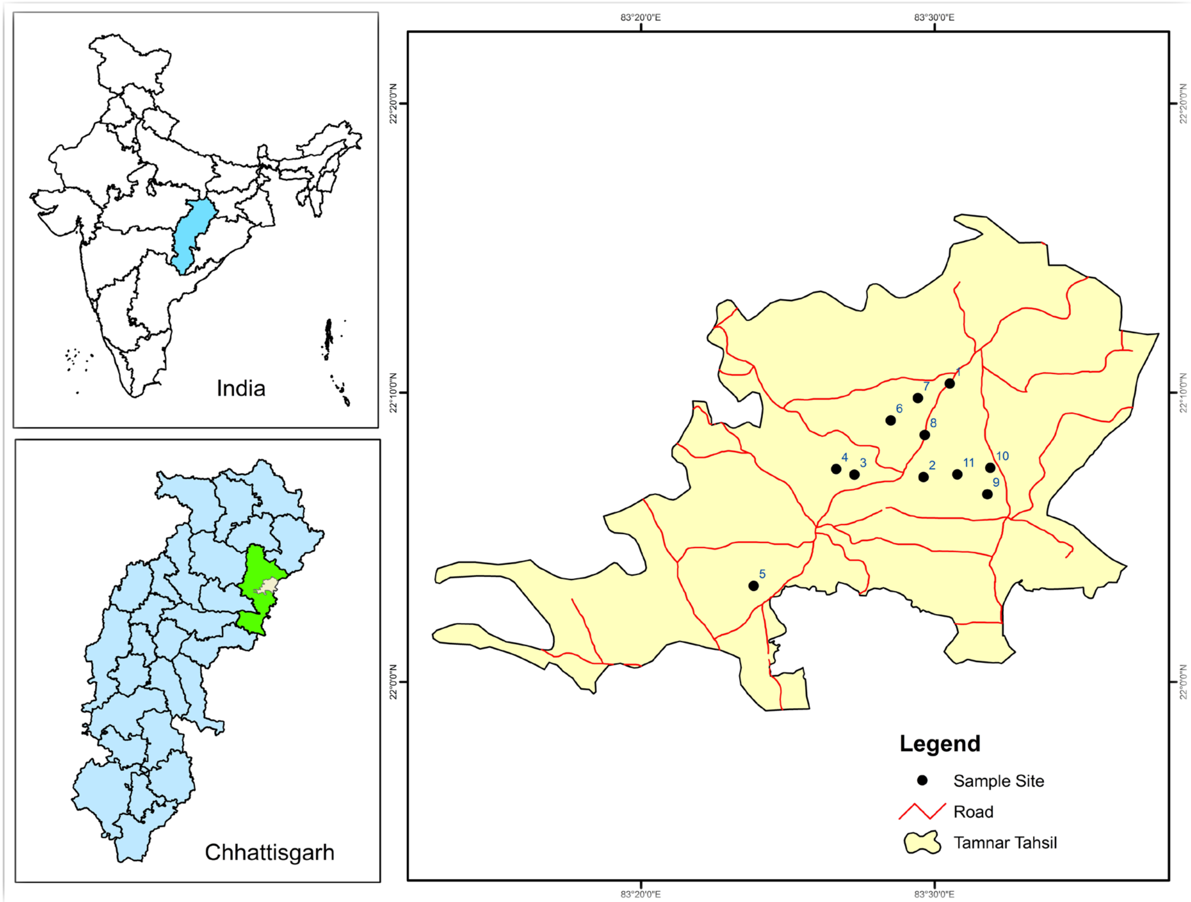


Figure 1. Representation of sampling locations.

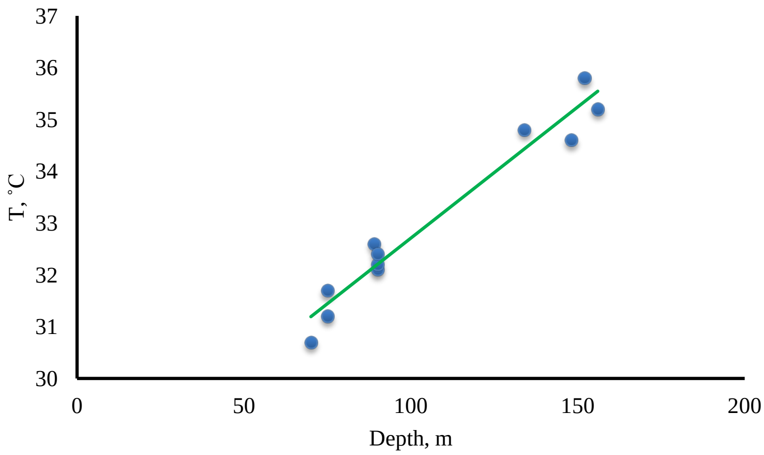
neutralized with 5 M NaOH solution to the pH value of 5.5 into final dilution of l liter). Similarly, 10 mL of the sample solution was mixed with 10 mL buffer in a 50-mL plastic beaker in a 1:1 volume ratio and F^- content was monitored. The content of Cl^-, NO_3^-, SO_4^{2-}, Na^+, K^+, Mg^{2+} and Ca^{2+} was analyzed by Dionex-DX1100 ion chromatography, equipped with anion and cation separation columns and conductivity detector. The hardness and alkalinity were determined by the titration methods [29]. The metals were analyzed with the AA280FS atomic absorption spectrophotometer, equipped with VGA-77 (plasma flow: 15 L/min, auxiliary flow: 1.5 L/min, power: 1KW, PMT voltage: 650 V).

Multivariate statistical analysis such as factor analysis (FA) and hierarchical cluster analysis (HCA) were used for the source apportionments [30]-[32]. The statistical Windows software STATISTICA 7.1 was employed for the multivariate statistical calculations. The enrichment factor (Ef) was determined by using Al as reference metal [33].

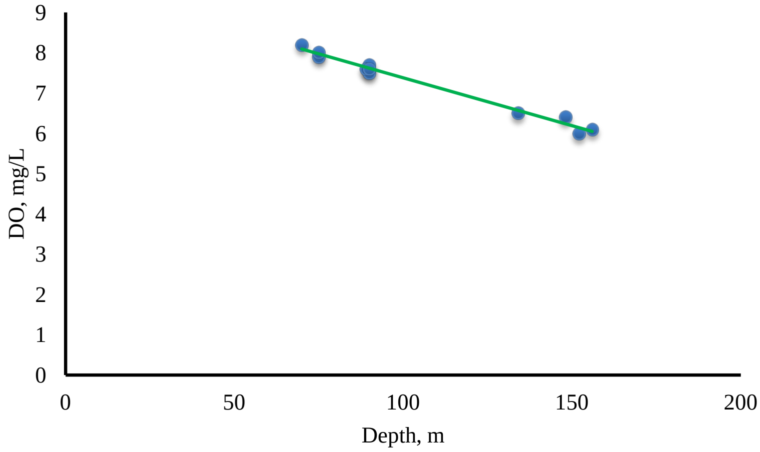
3. Results and Discussion

3.1. Physical Characteristics of Tube Well

The physical parameters of the tube well and groundwater are summarized in **Table 1**. The depth and life of tube well (n = 11) was varied from 70 - 156 m and 2.0 - 21 Yr with mean value of 106 ± 20 m and 7.7 ± 3.0 Yr, respectively. The temperature and pH values of groundwater of 11 tube wells were ranged from 30.7˚C - 35.8˚C and 6.4 - 7.5 with mean value of 33.0˚C ± 1.0˚C and 7.1 ± 0.2, respectively. The pH value of all groundwater was found to be neutral due to higher content of ions *i.e.* Mg^{2+}, Ca^{2+}, HCO_3^- and CO_3^{2-}. The $[\Sigma_{anion}]/\Sigma_{cation}]$ ratio was ranged from 0.89 - 1.28 with a mean value of 1.1 ± 0.1. The increasing T value of the water with respect to tube well depth was marked due to the geothermal energy, **Figure 2**. The DO value of the water was



(a)



(b)

Figure 2. Correlation of tube well depth profile with the T and DO of the water.

Table 1. Geophysical characteristics of water and tube well.

S. No.	Location	Depth of tube well, m	Life of tube well, Yr	Water table, m	T, ˚C	pH	DO, mg/L	RP, mV
1	Dholnara	70	9	10	30.7	6.4	8.2	169
2	Kunjemura	90	10	11	32.1	7.5	7.7	167
3	Pata	75	12	15	31.2	7.3	7.9	149
4	Pata	90	6	15	32.2	7.5	7.5	155
5	Bandhapali	89	6	16	32.6	7.3	7.6	145
6	Mudagaon	75	21	18	31.7	6.9	8.0	168
7	Mudagaon	156	9	16	35.2	7.1	6.1	158
8	Mudagaon	134	3	16	34.8	7.3	6.5	163
9	Saraitola	148	3	20	34.6	6.9	6.4	168
10	Saraitola	90	4	19	32.4	6.8	7.6	170
11	Saraitola	152	2	20	35.8	6.9	6.0	169

ranged from 6.0 - 8.2 mg/L with mean value of 7.2 ± 0.8 mg/L. The higher DO value of the water than the recommended value of 4.0 mg/L was observed, may be due to dense vegetation coverage. The DO value was found to decrease with respect to depth profile of the tube well, **Figure 2**. However, the T and DO value of the water was found to be negatively ($r = -0.57$) and positively ($r = 0.58$) correlated with the life of the tube well, respectively, **Figure 3**. The lower RP value of the water of the studied area was recorded, ranging from 145 - 170 mV with mean value of 162 ± 5 mV, being ≈<3.5-folds less than recommended value of 600 mV. The EC and TDS values of the water was ranged from 316 - 576 µS/cm and 193 - 499 mg/L with mean value of 391 ± 51 µS/cm and 312 ± 54 mg/L, respectively. The EC value was found to be well correlated with the TDS and total ionic concentration ($r = 0.92 - 0.97$), **Figure 4**.

3.2. Chemical Characteristics of Groundwater

The concentration of F^- and other elements in the ground water of the Tamnar block (n = 11) during post monsoon period (December, 2008) is summarized in **Tables 2-3**. The value of HCO_3^-, CO_3^{2-}, TH and TA was ranged from 121 - 302, 0.9 - 6.6, 61 - 218 and 159 - 397 mg/L with mean value of 190 ± 30, 2.4 ± 1.3, 102 ± 27 and 250 ± 40, respectively. The concentration of F^-, Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Mg^{2+}, Ca^{2+}, Al, Fe, Mn and Zn was ranged from 0.7 - 17.3, 6.0 - 61, 2.8 - 9.2, 0.5 - 39, 1.2 - 4.6, 5.9 - 43, 6.3 - 32, 6.6 - 25, 11 - 50, 0.28 - 0.34, 1.4 - 13, 0.07 - 0.26 and 0.8 - 1.7 mg/L with mean value of 9.0 ± 3.8, 18 ± 9, 6.0 ± 13, 6.7 ± 6.7, 2.6 ± 0.6, 30 ± 7, 15 ± 5, 12 ± 4, 21 ± 7, 0.30 ± 0.01, 5.6 ± 2.4, 0.16 ± 0.03 and 1.17 ± 0.19 mg/L, respectively. The metals *i.e.* K, Mg, Ca, Fe, Mn and Zn were highly enriched in the water with Ef value of 135 ± 55, 259 ± 71, 197 ± 60, 43 ± 18, 90 ± 20, 152 ± 24 and 100 ± 20, respectively.

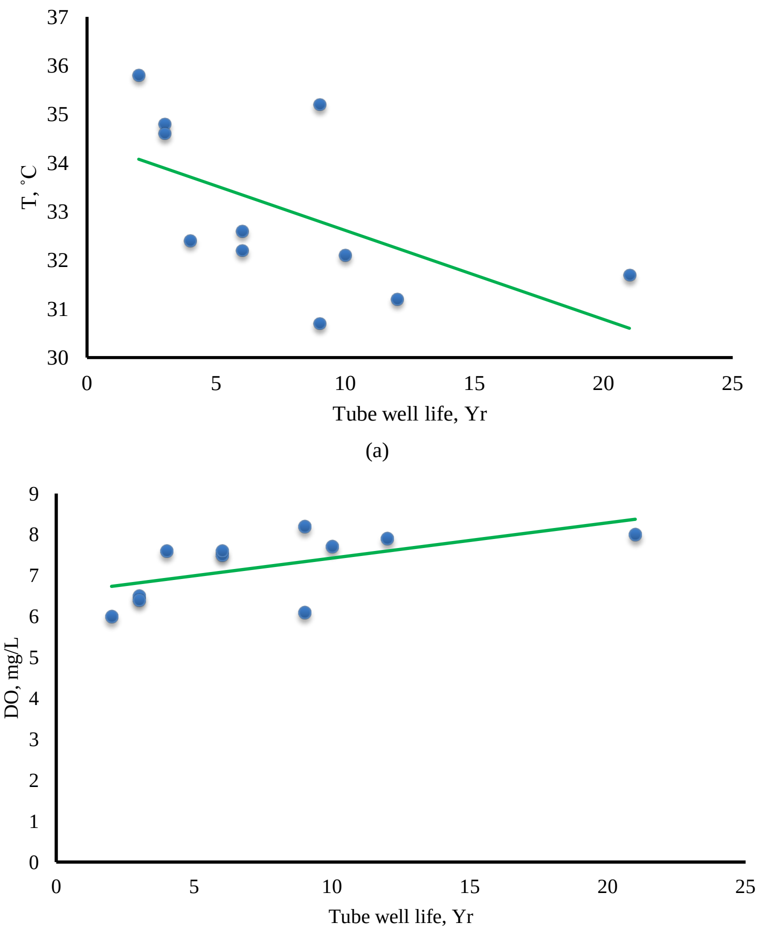


Figure 3. Correlation of tube well life with the T and DO of the water.

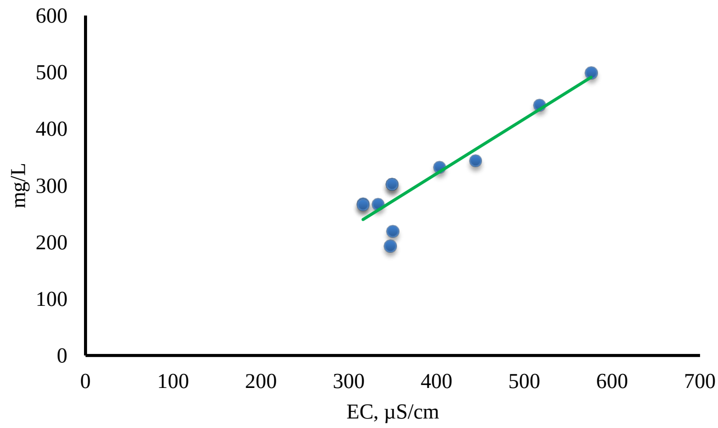


Figure 4. Correlation of EC and [$\sum_{11\ ions}$].

Table 2. Chemical characteristics of groundwater during post monsoon period, 2008, mg/L.

S. No.	EC, µS/cm	TDS	TA	TH	F^-	Cl^-	NO_3^-	SO_4^{2-}	HCO_3^-	CO_3^{2-}
1	347	396	159	90	1.7	6.7	4.1	12	121	1.2
2	349	492	272	105	2.2	13.9	5.4	0.8	208	1.5
3	517	716	397	145	3.1	13.5	7.3	4.5	302	3.3
4	444	568	270	124	3.8	18.8	8.5	4.3	201	6.6
5	576	806	317	218	2.8	61.3	8.3	39	242	1.5
6	333	450	210	61	10.1	16.7	2.8	5.1	161	0.9
7	403	535	270	82	13.4	16.6	6.7	0.5	201	6.6
8	349	491	246	71	17.3	14.9	5.8	0.6	188	1.2
9	316	443	211	78	13.2	13.6	9.2	5.9	161	1.5
10	350	494	176	62	16.7	6.0	4.3	0.9	134	1.2
11	316	399	220	81	14.3	11.7	3.1	0.5	168	1.2

Table 3. Concentration of cations and metals in groundwater during post monsoon period, 2008, mg/L.

S. No.	NH_4^+	Na^+	K^+	Mg^{2+}	Ca^{2+}	Al	Fe	Mn	Zn
1	1.6	5.9	7.4	9.9	21	0.29	3.9	0.11	1.1
2	2.3	13	18	14	23	0.29	3.9	0.16	1.3
3	3.5	30	22	21	32	0.29	3.2	0.16	1.5
4	3.1	24	32	14	28	0.30	7.2	0.22	1.6
5	2.7	39	27	25	50	0.31	3.4	0.26	1.7
6	1.2	42	7.8	7.7	12	0.34	13	0.07	0.9
7	2.4	43	16	11	15	0.28	1.4	0.18	1.0
8	1.7	41	11	8.0	12	0.29	4.0	0.15	0.9
9	1.9	33	6.3	8.8	14	0.28	6.4	0.21	1.0
10	4.6	26	7.2	6.6	11	0.29	13	0.15	0.8
11	3.1	30	11	8.7	14	0.29	2.1	0.10	0.8

The increased content of F^- and other ions was found in the deeper tube wells lie along the Digi Nala (stream) due to the higher weathering. The content of F^- and other elements (except NO_3^-, NH_4^+ and K^+) was enhanced from the monsoon to pre monsoon period, may be due to decreasing of the water level (up to ≈50 m) and increasing of water temperature (up to ≈35˚C) (**Figure 5**). The temporal variation in the ion and metal contents during post monsoon period of year 2008-2012 is presented in **Figure 6**. The concentration of most of the chemical species was enhanced at the rate of >5%, may be due to increasing anthropogenic activities viz. coal mining, coal burning, transportation, agriculture, etc. in this region.

The Ward's method was used for grouping of groundwater dataset. Three cluster groups-I (observations 3 and 5), group-II (observations 2, 4, 6 - 9 and 11) and group-III (1 and 10) are presented in **Figure 7**. The discriminating parameters observed were EC, TDS, Fe, Mn, K^+, F^-, Cl^-, SO_4^{2-}, NO_3^- and alkalinity. The parameters such as life of tube well, Mg^{2+}, Ca^{2+} and hardness had the higher median value in the group-I than the group-II and -III.

Factor analysis was executed on 22 variables for the 11 sample sites, in order to identify factors controlling the groundwater quality. In reference to the Eigenvalues, five factors were extracted after varimax rotation as they had eigenvalues greater than 1. The variable loading, eigenvalues, explained variance and cumulative variance are presented in **Table 4**. Calcium, chloride or sulfate with positive strong loading values, and hardness or TDS with moderate loading values in factor-I characterized the inorganic salts and groundwater hardness due to bicarbonates and carbonates. Life of tube well and Al contents had a strong loading value in factor-II. Aluminum content in groundwater was seemed to be more important if the life of tube well was high. Parameters *i.e.* water table, sodium and fluoride content had strong loading values in factor-III, which could be related to cation

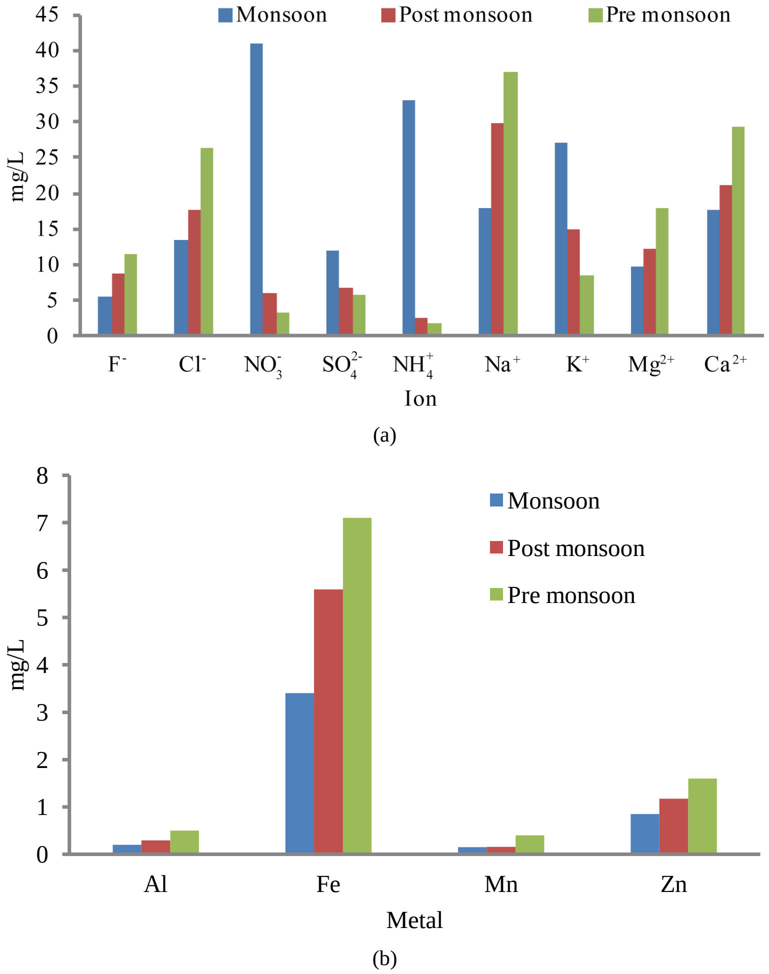


Figure 5. Seasonal variation of ions and metals in groundwater.

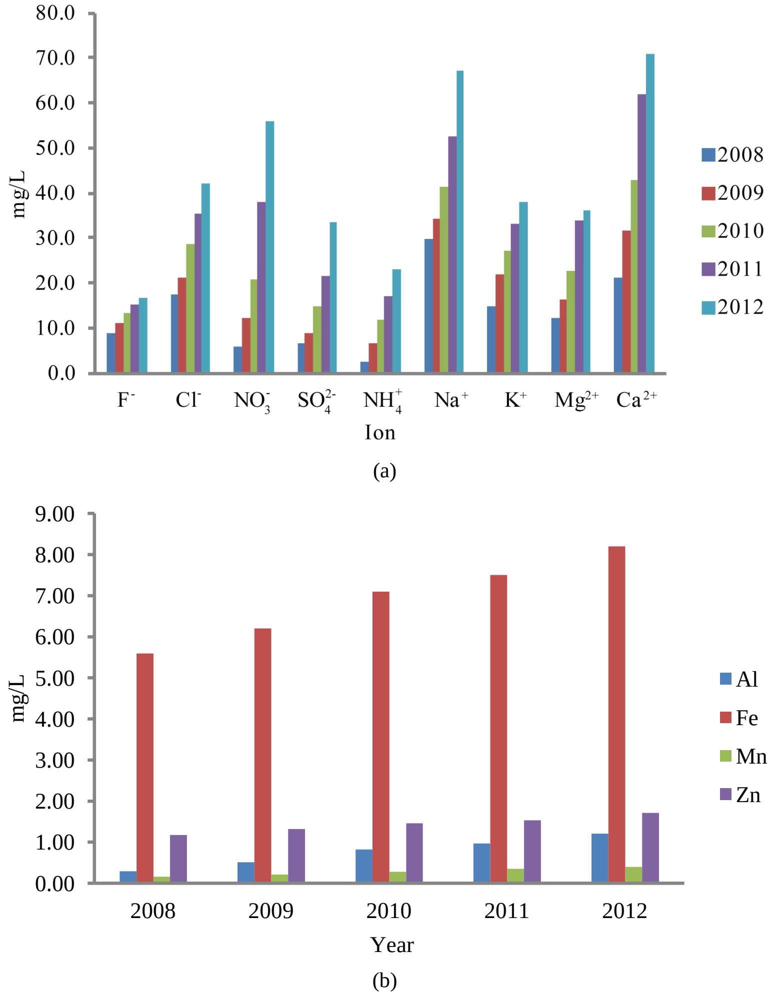


Figure 6. Temporal variation of ions and metals in groundwater.

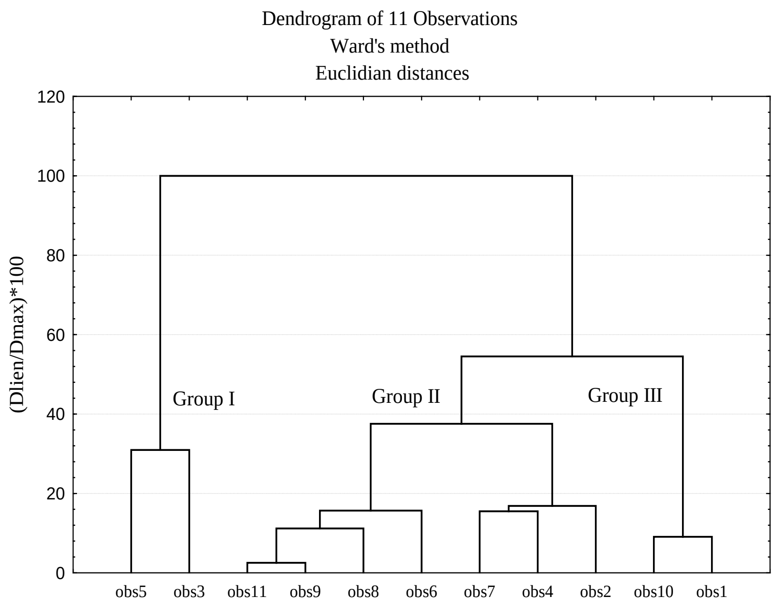


Figure 7. Ward's dendrogram of the sample sites in the study area.

Table 4. Factor loadings after varimax rotation.

Parameter	Factor-I	Factor-II	Factor-III	Factor-IV	Factor-V
T (˚C)	−0.19	0.27	0.05	**−0.86**	−0.23
pH	0.15	0.03	0.06	0.07	**0.84**
EC	0.14	−0.05	0.02	0.02	**0.97**
TDS	**0.74**	−0.05	0.07	0.02	0.65
Water table	−0.04	0.05	**0.88**	0.34	−0.15
Depth of tube	−0.21	0.57	0.65	−0.36	−0.02
Life of tube	−0.11	**−0.90**	−0.19	−0.13	0.17
Al	0.27	**−0.92**	0.13	0.13	−0.04
Fe	−0.14	−0.50	0.20	0.59	−0.39
Mn	0.63	0.50	0.07	0.12	0.42
Zn	0.60	0.01	−0.43	0.14	0.64
Na^+	0.16	−0.19	**0.89**	−0.17	0.29
K^+	0.41	0.07	−0.21	0.20	**0.78**
Mg^{2+}	0.69	0.01	−0.28	0.08	0.63
Ca^{2+}	**0.81**	0.01	−0.33	0.12	0.46
NH_4^+	−0.05	0.38	0.01	**0.81**	0.18
F^-	−0.40	0.20	**0.82**	0.01	−0.30
Cl^-	**0.92**	−0.09	0.17	−0.05	0.27
NO_3^-	0.44	0.47	0.07	0.01	0.53
SO_4^{2-}	**0.99**	−0.09	−0.09	−0.03	−0.02
Alkalinity	0.23	0.00	−0.04	0.02	**0.92**
Hardness	**0.76**	0.01	−0.31	0.11	0.54
Eigenvalues	9.99	3.54	2.62	1.97	1.80
% Explained variance	45.45	16.09	11.94	8.99	8.2
% Cumulative of variance	45.45	61.54	73.48	82.47	90.67

exchange between Ca^{2+} and Na^+. Ammonium ions with high positive loading value and T(˚C) with a negative loading value were related to Factor-IV. This could highlight the impact of agricultural fertilizers. Factor-V characterized the mineralization of the groundwater and the importance of carbonic acid on the groundwater alkalinity which increased the pH value.

The F^- content of the water had fair positive correlation (r = 0.69 0 0.75) with the geological characteristics (*i.e.* water table and depth) of the tube wells likely to Na^+ and Fe (r = 0.25 0 0.56). However, almost all ions had negative correlation with the tube well geological parameters. The F^- content with the pH, alkalinity, HCO_3^-, Mg^{2+} and Ca^{2+} values had negative correlation (r = −0.22 0 0.72). The pH value was dependent on OH^- concentration rather than HCO_3^- and CO_3^{2-} concentration. The negative correlation of the F^- content with the hardness was expected due to the precipitation of $CaCO_3$. The positive correlation (r = 0.56) of Na^+ with the F^- indicated its release in the ground water from weathering of silicate minerals. The cation exchange (Na^+ for Ca^{2+}) accompanied with anion exchange (OH^- for F^-) of mica and clay was expected as important processes in release of Ca^{2+} and F^-. Fluoride ions in the ground water had fair positive correlation only with Fe. While with other species, F^- had negative correlation. Most of the ions *i.e.* Na^+, K^+, Mg^{2+}, Ca^{2+}, NH_4^+, F^-, Cl^-, NO_3^- and SO_4^{2-} among themselves had positive correlation unlikely to F^-.

3.3. Chemical Characteristics of Dust, Soil and Rock

The dusts and soils were yellowish colored with mean pH value (n = 6) of 7.2 ± 0.5 and 7.4 ± 0.6, respectively. The mean water soluble content (n = 11) of F^-, Cl^-, NO_3^-, SO_4^{2-}, NH_4^+, Na^+, K^+, Mg^{2+} and Ca^{2+} in the soil extracts was found to be 490 ± 19, 1615 ± 512, 417 ± 55, 805 ± 98, 108 ± 46, 3610 ± 712, 1010 ± 250, 502 ± 51 and 3118 ± 276 mg/kg, respectively. The enhanced content of ions in the road dust samples was achieved due to higher anthropogenic stresses, **Figure 8**. The distribution pattern of the ions in the surface dust and soil had almost similar trend as in the ground water. The F^- content in the granite (n = 6) was ranged from 1798 - 2510 mg/kg with mean value of 2200 ± 154 mg/kg, respectively, may be due to presence of fluoride minerals *i.e.* fluorite (CaF_2), apatite [$Ca_5(Cl,F,OH)(PO_4)_3$], etc. The F^- content in the dust and soil of this region was found to be higher than other regions [34]-[36].

3.4. Toxicities

Two types of water: ($Na\text{-}Ca\text{-}Mg\text{-}HCO_3$ and $Na\text{-}Ca\text{-}Mg\text{-}HCO_3\text{-}Cl$) was observed in the study area (**Figure 9**). The majority of groundwater belonged to the $Na\text{-}Ca\text{-}Mg\text{-}HCO_3\text{-}Cl$ type. The water in the studied area becomes turbid when exposed to the air. The tolerance limit of F^-, Fe, Mn and Al in drinking water reported is 1.5, 0.30, 0.10 and 0.03 mg/L, respectively [37] [38]. The water of all tube wells was found to be contaminated with F^-, Fe, Mn and Al beyond the permissible limits. The elevated concentration of F^- in the urine samples of humans and domestic animals were observed, **Table 5**. The F^- concentration (n = 6) in the urine samples of male(MU), female(FU), cattle(CU), buffalo(BU), sheep(SU) and goat(GU) was ranged from 8.1 - 12, 8.8 - 13, 20 - 29, 22 - 28, 20 - 26 and 43 - 66 mg/L with mean value of 9.9 ± 0.9, 11 ± 1, 24 ± 2, 25 ± 2, 23 ± 2 and 52 ± 6 mg/L, respectively. Among them, the highest F^- concentration was seen in the GU, may be due to intake of higher the contaminated green food. The sevear dental and skeletal fluorosis in human and domestic animals *i.e.* cattle, buffalo, sheep and goat were seen, mainly due to intake of fluoride contaminated water.

4. Conclusion

The whole environment in the Tamnar block is contaminated with F^- at excessive levels. The groundwater was found to be contaminated with F^-, Fe, Mn and Al at levels above permissible limits. The deeper tube well water lie along the stream was contaminated with higher content of F^-. The increased fluoride toxicity in the studied area is recorded due to occurrence of relatively lower concentration of the sequestrating agents *i.e.* Al, Mg and Ca, in the water. The groundwater of the Tamnar area was found to be unsuitable for drinking purposes.

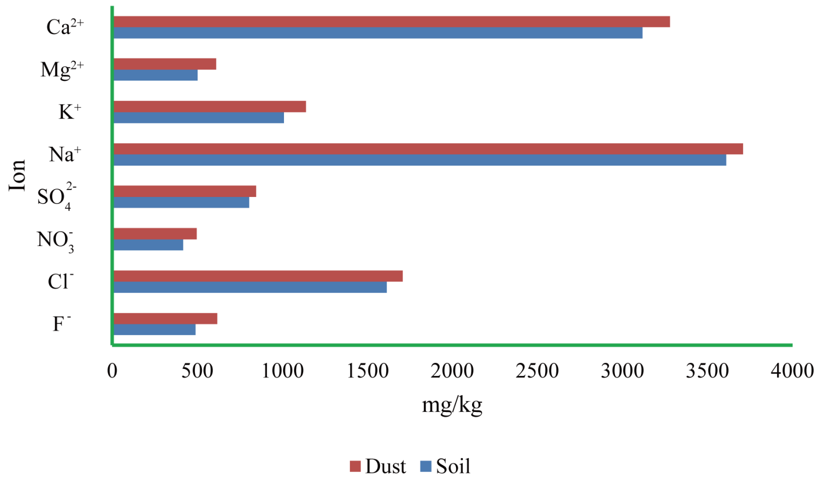


Figure 8. Distribution of water soluble ions in road dust and soil.

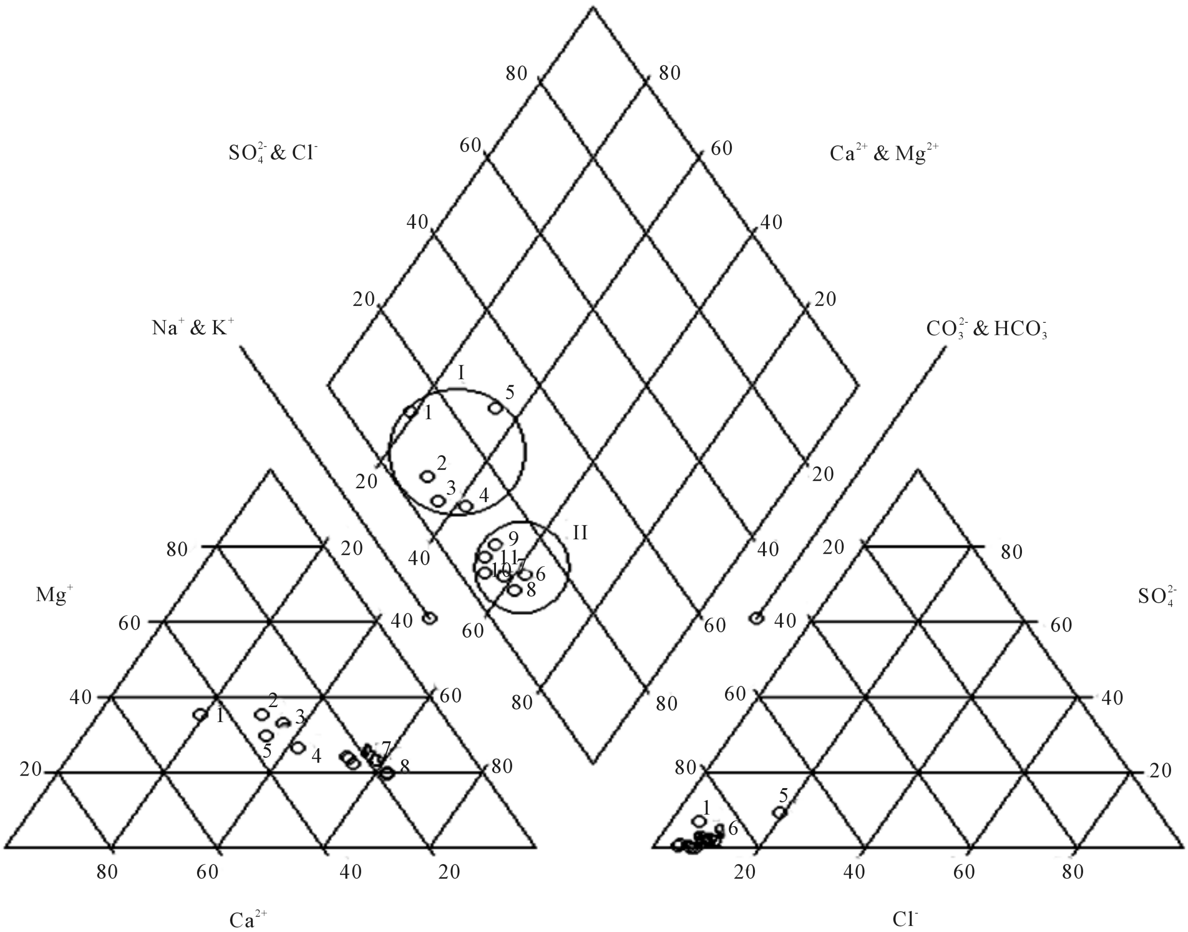


Figure 9. Piper diagram of the general water types in the study area.

Table 5. Mean fluoride concentration in urine, mg/L.

Location	MHU	FHU	CU	BU	GU	SU
Dholnara	8.1	9.2	23	22	49	22
Kunjemura	9.2	10	20	24	43	24
Pata	11	13	24	27	49	23
Bandhapali	12	12	29	26	66	26
Mudagaon	9.8	9.8	21	28	49	20
Saraitola	9.6	8.8	25	22	55	25

MHU, MFU, CU, BU, GU and SU = Male human, female human, cattle, buffalo, goat and sheep urine.

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