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MONITORING OF FLUORIDE CONTAMINATION IN SOUTHERN REGION OF CHHATTISGARH, INDIA: CORRELATION WITH PHYSICO-CHEMICAL PARAMETERS

Sahin Rubina[1], Tapadia Kavita[2]
[1] Research Scholar, Department of Chemistry, National Institute of Technology, Raipur, India, CG, 492010, Lecturer, NMDC DAV Polytechnic, Dantewada (C.G) India
[2] Assistant Professor , Department of Chemistry, National Institute of Technology, Raipur, India, CG, 492010
Email.ID- rsm141209@gmail.com,
akavita9@rediffmail.com, ktapadia.chy@nitrr.ac.in,

ABSTRACT:
Groundwater quality is a very sensitive issue, which transcends national boundaries. It is influenced by many factors including atmospheric chemistry, the underlying geology, the vegetation (organic matter decay) and anthropogenic agents. In this paper, monitoring of the concentration of fluoride in groundwater of South-East region of Chhattisgarh had been studied. Fluoride is very much essential for healthy growth of teeth and bones if it present between 0.6 mg/l to 1.5 mg/l in drinking water. But if the level is higher than 1.5 mg/l then causes dental and skeletal fluorosis, decalcification, digestive and nervous disorders. Fluoride concentrations in groundwater samples were determined in twelve samples of south-east region. Forty two groundwater samples were collected from different location in May 2012 and analysis is done for Fluoride content along with Physico-Chemical parameters such as pH, alkalinity, Total Dissolved Solid (T.D.S), Electrical Conductivity (E.C), Total hardness (T.H), Nitrate(NO_3^-), Sulphate(SO_4^{2-}), Phosphate(PO_4^{3-}), Chloride(Cl^-), Calcium (Ca^{2+}), Magnesium (Mg^{2+}) and Sodium (Na^+). Fluoride concentration varies from 0.12 mg/l to 5.05 mg/l. Fluoride concentration is not uniform throughout this region. Physico-chemical condition like dissociation, decomposition, subsequent dissolution and agrochemical, might be responsible for leaching of fluoride into the groundwater resources. From correlation analysis of fluoride concentration (F^-) in groundwater with Physico-Chemical parameters, it was found that though most of the samples were within the permissible limit but showed positively correlated with pH, SO_4^{2-}, Cl^-, Na^+ and alkalinity and negatively correlated with Ca^{2+}, Mg^{2+} and total hardness.
Keywords: *Fluoride, Correlation, Groundwater Quality, Physico-Chemical Parameters, Permissible limit.*

1. INTRODUCTION

Fluorine is the most electronegative element. It is the lightest element of the halogen group. As compared to other halogen its properties are quite different and it is also reflected in ground water. Fluoride is the reduced form of fluorine. Fluoride commonly in the forms of minerals and salts are found on both earth's crust and groundwater. CaF_2 is a common fluoride mineral. It is considered as one of the minor constituents of natural waters, but it is an important parameter in ascertaining the suitability of water for potable purposes. Fluoride intake of 1 mg/l per day is very much essential for healthy growth of teeth and bones, but level higher than the permissible limit of 1.5 mg/l is dangerous to health [Ambade et al. (2012)] Fluoride contamination of groundwater has now became a serious geo-environmental issue in many parts of India due to its toxic effects on long terms consumption. Contamination of groundwater due to fluoride is becoming a serious issue for various states of India, among all, Chhattisgarh is one of the state [Apambire et al.(1997)] The South- East parts of Chhattisgarh are mostly suffering from this problem. Now a day's groundwater of three district of this region is affected a lot [Arif et al. (2012)].

Deficiency of fluoride leads to dental caries and higher concentration leads to dental and skeletal fluorosis [Behera et al. (2012)]. Fluorosis was first reported from India by Short et al., in 1937. Fluoride easily enters the cell membrane and also circulates in blood and effect fetus, nerves and heart. Fluoride reduces secretion of thyroid gland by affecting Iodine in the body which may lead to monogolism [Dutta et al.(2010)] High fluoride intake over a period of time can cripple one for life[Edward Groth and Saxena et al.(2003)].

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Corresponding Author: Dr.(Mrs.) KavitaTapadia, Assistant Professor , Department of Chemistry, National Institute of Technology, Raipur, India, CG, 492010, Email.ID- akavita9@rediffmail.com, ktapadia.chy@nitrr.ac.in, Mo.: +91-9977987702

From study it is found that groundwater bearing fluorides are too high in sodium and alkalinity due to bicarbonate and low in calcium hardness. It is chemically distinctive in that, it is soft, has high pH and contains large amount of Silica [Govardhan,et al.(1994)].

2. STUDY AREA

The three district of Southern region of Chhattisgarh taken under study. The total area covered by these three districts is18,727.98 sq.km**.** The Southern part of Bijapur touches the border of Andhra Pradesh. The eastern part of Bastar district touches the border of Orissa. This region basically comes under Bastar plateau [Arif et al. (2012)]. The average rainfall recorded is 1233 mm. and the average temperature in summer season is 33.25° C and in winter season it is 22.12° C. Approximately 75% of the land are covered with forests. Among the available land 84% are used for agriculture purpose. The paddy is the most common cultivation crop. In Dantewada district there is open cast mines of iron, mostly have mined hematite ore.

3.0 MATERIALS AND METHODS

The 40 groundwater samples were collected from different locations of three districts shown in Table 1. The sources of the water samples were manually operated hand pump and bore well. Hand pump and bore well were operated at least 10-15 minutes before collection to flash out the stagnant water inside the tube and to get fresh groundwater. The water samples were collected in clean 500 ml Poly propylene bottles and stored in box [Handa et al. (1975)]. Table 1 showing the place of sampling which was done in the month of May-June 2012. The sampling was done by both in random way and systematic way. Means in systematic manner collection of water done kept the distance of approximately 2 km between two locating point. Ground Water samples from three districts were distinguish as such, GW-1, 3, 6, 8, 10, 11, 12, 15, 18, 19, 25, 32, 35 and 38 (total 14) groundwater samples from Dantewada district. Similarly 14 sampling point in Bastar district which were GW-2, 4, 7, 13, 16, 20, 21, 22, 23, 24, 31, 36, 39 & 40 and 12 sampling point in Bijapur district which were GW-5, 9, 14, 17, 26, 27, 29, 30, 33, 34, 37 & 40.

3.1 *Sample Analysis*

pH of the samples was measured at the site of collection by using pH meter (Elico Model 1012) . The temperature also taken at the time of sampling by using of thermometer. Electrical Conductivity was calculated by using conductivity meter (Model No-304 Systronics). Hardness of water & alkalinity were calculated by titrimetric method using Eriochrome Black T &phenolphthalein as indicator. Sodium(Na^+) and Potassium (K^+) in mg/l were determined by flame photometer (Elico CL-361). Chloride (Cl^-), Nitrate (NO3), Sulphate(SO_4^{2-}), Phosphate(PO_4^{3-}), &Fluoride(F^-) were determined by using of NOVA 60 spectrophotometer. The fluoride concentration was also determined electrochemically, using ion selective electrode (APHA 1991).The electrode used was an Orion fluoride electrode, coupled to an Orion electrometer. Gravimetric method used for the determination of TDS & TSS in water samplers. Apart from instrumental analysis all reagents used for analysis are from Merck Company with Purity of 99%.

4. RESULTS AND DISCUSSION

All the groundwater samples collected from different location of three district of Chhattisgarh, India. The data obtained after analysis of all the samples was compiled and presented in Figure parameters wise. A total of 40 samples, 13 from each district are taken in the study area. Fluoride concentration in the study area is depicted in Fig. 1. Fluoride concentration in the study area varied from 0.09 to 5.05 ppm or mg/l. Out of 40 samples above 1.5 ppm fluoride (above WHO guideline) is 35 % i.e 14 samples. The GW- 39 which have highest fluoride contents, its contain 5.05 mg/l after that GW-21 having contain 4.85 mg/l, GW-26 contain 4.68 mg/l ,GW-16 contain 4.21 mg/l, GW-20 contain 3.89 mg/l, 2.96 mg/l, 2.98 mg/l, 2.68 mg/l, 2.63 mg/l, 2.54 mg/l, 2.05 mg/l , 2.01 mg/l , 1.96 mg/l and 1.56 mg/l of GW-30,GW-28,GW-01,GW-40,GW-05, GW-09, GW-31,GW-6 and GW-37 respectively. The concentration of fluoride in three district are different but the interesting thing among all three district is that entire fluoride affected region is in the same geological set up.[14]It seems more appropriate that rocks rich in fluoride content of ground water during course of weathering of rock types

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fluorspars, rock phosphate and phosphate. In general relatively high pH conditions have a tendency to displace fluoride ions from the minerals surface[18]. From the correlation analysis it was observed that high fluoride concentration and high pH values. This correlation was finding of Sanjay Kumar et.al[19]. Among these districts people are commonly involved in agriculture profession. But they used manure and compost rather than using of fertilizers. But for the protection of crops they used Pesticides and Insecticides which might be dilute with rain water and goes to groundwater aquifer [Nemade et al. (1996)].

The correlation of some selected ions and other parameters with fluoride. (Fig. 2 a-k). The ions Ca^{2+} , Mg^{2+} , Total Hardness, PO_4^{-3} and NO_3^- showed negative correlation with fluoride contents. The correlation coefficients were -0.425, -0.580, -0.561, -0.397 and -0.123, respectively. The negative correlation of fluoride with Ca^{2+} & Mg^{2+} is as expected due to low solubility of fluoride of these ions[20] Generally from various studies it is found that water with fluoride more than 1.5 ppm has hardness less than 200ppm[20]. The phenomenon of decrease in hardness concentration contributing to higher fluoride concentration contributed to calcium complexion effect. [22] Fluoride shows positive correlation with sulphate, pH, E.C, Alkalinity, Sodium and Chloride. Figure 5 shows it clearly. The correlation coefficients were 0.079, 0.442, 0.720, 0.754, 0.552 and 0.020, respectively.

The pH values varying from 6.68 to 7.96 (Fig. 3). The permissible limit of pH in drinking water is 6.5 to 8.5 Indian Standards[1] .Maximum pH values 7.96 are found in sample GW-39 and minimum value is found 6.68 in sample number GW- 02. The pH has no direct effect on human health; all the biological reactions are sensitive to variation of pH. For most of the reactions as well as for human beings, pH value 7.0 is considered as best and ideal.

The electrical conductivity varying from 268 µS/cm -1020 µS/cm (Fig. 4**).** All the samples showed E.C below the permissible limit. For drinking water (1500 µS/cm) as per BIS [Ambade et al. (2012)] . Maximum values found in sample number GW-16 which is1020 µS/cm and minimum value 268 µS/cm in sample number 27. Conductance is a function of water, hence a standard temperature, usually 25◦C, is specified in reporting conductivity [Hem, (1998)]. High the concentration of electrolytes in water, and the more is its electrical conductance. E.C positively correlates with pH and correlation coefficient is 0.291.

All the samples analysed showed the total alkalinity varying from125 mg/l to 654 mg/l (Fig. 5). The permissible limit of alkalinity for drinking water is 200 ppm. The maximum value of total alkalinity is 654 mg/l in sample number GW-39 and minimum value is 125 mg/l in sample number GW-17. The alkalinity basically depends upon the dissolved salt might be it polyvalent ions present in it. The high concentration of alkalinity in some water sample is due to dissolution of polyvalent metallic ions from soils minerals, sedimentary rocks. In the present study alkalinity is positively correlated with chloride and nitrate with correlation coefficient is 0.0277 and 0.170 respectively.

The chloride concentrations in all the analysed samples are within permissible limit i.e. 250 ppm. The range of chloride concentration varies from 12.0 mg/l to 74.0 mg/l(Fig. 6). The maximum concentrations found in the sample number GW-05 and the minimum value is found in the sample number GW -27.

The concentration of sodium is varied among 40 samples is 4.2 mg/l to 35.2 mg/l (Fig. 7). The concentration of sodium is maximum in sample number GW-20 and minimum value is in sample number GW- 38. It is found from the study that fluorine element easily combines with sodium forming minerals called fluoride. The concentration of sodium is positively correlated with fluoride concentration in water samples and its correlation coefficient is 0.552.

The variation of the phosphate concentration is shown in the (Fig. 8**)**. The phosphate concentration varied from 0.1 mg/l to 2.54 mg/l. The maximum concentration of the phosphate is found in the sample number GW-15 which is 2.54 mg/l and minimum concentration is found in sample number GW-2 which is 0.1.The phosphate ion in the water samples is due to anthropogenic activities, geogenic formation and dilution of minerals from rocks and soil. [Nemade et al. (1996), Sharma et al. (1992), Susheela (2001)] .

All the samples analysed showed the total hardness varying from 98 mg/l to 409 mg/l (Fig. 9). The permissible limit of T.H for Drinking water is 300 ppm [1].The maximum value of T.H is 409 mg/l in sample number GW-14 and minimum value is 98 in sample number GW-28. The hardness of water is not depend upon by a single

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substance but by a variety of dissolved polyvalent metallic ions, predominantly calcium and magnesium cations [Govardhan (1994), Handa (1975)]. The high concentration of T.H in some water samples is due to dissolution of polyvalent metallic ions from soils, sedimentary and ingenious rocks and run off from soil. In this study T.H is positively correlated with calcium and magnesium with correlation coefficient is o.447 and 0.826.

The concentration of calcium varied from 9.0 mg/l to 98 mg/l. The highest concentration of calcium is found in the sample number GW-05 is 98 mg/l and minimum concentration in sample number GW-09 is 9.0 mg/l. The concentration of the calcium ion is shown in the (Fig. 10). The permissible limit is 200 ppm. In fact 98% of all world ground water are dominated by calcium and bicarbonate ions due to limestone weathering in the catchments and underground beds.

The concentration of the magnesium is from 23 mg/l to 265 mg/l. (Fig. 11) .The maximum concentration 265 mg/l is found in sample number GW-29 and minimum concentration found is 23 mg/l in sample number GW-39. The permissible limit of magnesium concentration is 200 ppm.

The sulphate concentration Fig.12 is varied from 23 mg/l to 152 mg/l. The water samples analysed showed lower concentration then the prescribed permissible limit for drinking water. The BIS permissible limit of sulphate is 200 ppm. Maximum concentration of 152 mg/l is found in sample number GW-24 and the minimum concentration 23 mg/l in the sample number GW-29. Sulphate shows weak negative correlation with Ca^{2+} and weak positive correlation with Mg^{2+} which indicates in the water samples calcium sulphate may be absent and magnesium sulphate may be present. Sulphate in the water samples is due to oxidation of sulphur compounds used for agriculture.

The nitrate concentration is varied from 3.5 mg/l to 36.3 mg/l (Fig. 13). The highest concentration of nitrate is 36.3 in sample number is GW-04 and minimum concentration is 3.5 mg/l in sample number is GW-07. All the 40 samples contain nitrate in permissible limit .i.e. 45ppm. Nitrate shows positive correlation with Mg^{2+} which indicates that the Mg (NO_3) present in the water samples.[11] Nitrate ion may be leaching to the water sources from fertilizers used in the agriculture.
Among complete study of Physico-Chemical parameters of water and correlation of these above mentioned parameters with the fluoride concentration. It finally concludes that each parameters correlates with each other.

5. CONCLUSION

Fluoride distribution is associated with pH, Calcium, Magnesium, Phosphate, Nitrate, Sulphate. Positive correlation is observed with pH, E.C and Sulphate and Chloride and negative correlation is observed with total hardness, phosphate, calcium, magnesium and nitrate [Krishnaraj et al. (2010), Tewari et al. (2012)]. The linear correlation is very useful to get fairly accurate idea of the quality of drinking water or nature of water. In this study it is clear that there is lots of variation in concentration of fluoride. Measures of fluoride monitoring should be taken where alternative sources for direct use is not feasible and diet of rich calcium and phosphate are suggested where high level fluoride is found. From the statistical evaluation of different parameters in groundwater samples of south-east region of Chhattisgarh indicate the variation in different region. The maximum variation found in case of fluoride concentration which is 193.4 it indicate that dilution of fluoride is not uniform throughout the region. The concentration of fluoride is might be high in that region where favourable condition like pH, temperature, velocity of flowing groundwater along with alkalinity provides dilution of fluoride minerals and rocks with groundwater [Nagendre et al.(2003), Susheela et al. (2001), Manish (2012)].

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Table1. Showing sampling point of southern region of Chhattisgarh, India

PLACE OF SAMPLING	SAMPLE NO.	PLACE OF SAMPLING	SAMPLE NO.
BAKAWAND	GW-1	KHANDSARA	GW-21
AWARABATTA	GW-2	KILEPAL	GW-22
BANGAPAL	GW-3	KORENAR	GW-23
BASTANAAR	GW-4	KUMAHARPARA	GW-24
BELNAR	GW-5	KUNDENAAR	GW-25
BOGAM	GW-6	MARGHAT, BARAMGARH	GW-26
BUS STOP JAGDALPUR	GW-7	MATHWADA	GW-27
CHITALANKA	GW-8	NESALNAAR	GW-28
DANAPAL	GW-9	NIMED	GW-29
DHURLI	GW-10	PANDEMURGA	GW-30
FARASPAL	GW-11	PARPANAKA	GW-31
GANJENAR	GW-12	PATRARASH	GW-32
GARENGA	GW-13	PINKONDA	GW-33
HALUR	GW-14	PUSNAR	GW-34
HARAMPARA	GW-15	RAJENDRA NAGAR	GW-35
JAIBELL	GW-16	SHANTI NAGAR	GW-36
JANGLA	GW-17	SOLVI	GW-37
JAWNAGA	GW-18	TEKANNAAR	GW-38
KATYARASH	GW-19	MAIN ROAD JAIBELL	GW-39
KESARPAL	GW-20	TURPURA	GW-40

Table2. Statistical Evaluation for different parameters in groundwater samples in South-East region of Chhattisgarh

Parameters	Present Range	BIS	WHO	Mean	Standard Deviation	Coefficient of variation (%)
Temperature	24-9-29.5	28-30	28-30	27.02	2.347	8.6
pH value	6.68-7.96	6.5-8.5	6.5-8.5	7.33	5.113	69.7

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Alkalinity, mg/l	125-586	200	200	328.93	133.109	40.4
Electrical Conductivity(µmho/cm)	268-1020	1500	1500	565.8	223.369	39.4
Total Hardness(as$CaCO_3$)	98-409	300	300	252.1	87.946	34.8
Magnesium Hardness, mg/l	23-265	200	200	106.43	52.697	49.5
Calcium Hardness ,mg/l	9.0-98	200	200	49.55	25.118	50.6
Total Dissolved Solid, mg/l	115-582	500	500	309.4	106	34.2
Sulphate(SO_4^{-2}) ,mg/l	23-152	200	200	60.1	30.351	50.5
Phosphate (PO_4^{-3}) mg/l	0.1-2.54	5	5	0.961	0.707	73.5
Nitrate(NO^-_3),mg/l	3.5-36.3	45	45	16.4	10.952	66.7
Fluoride, mg/l	0.09-5.05	1.5	1.5	1.29	2.496	193.4
Chloride, mg/l	12.0-74.0	250	250	35.7	16.211	45.4
Sodium mg/l	4.2-35.2	…	…	15.02	9.826	65.4

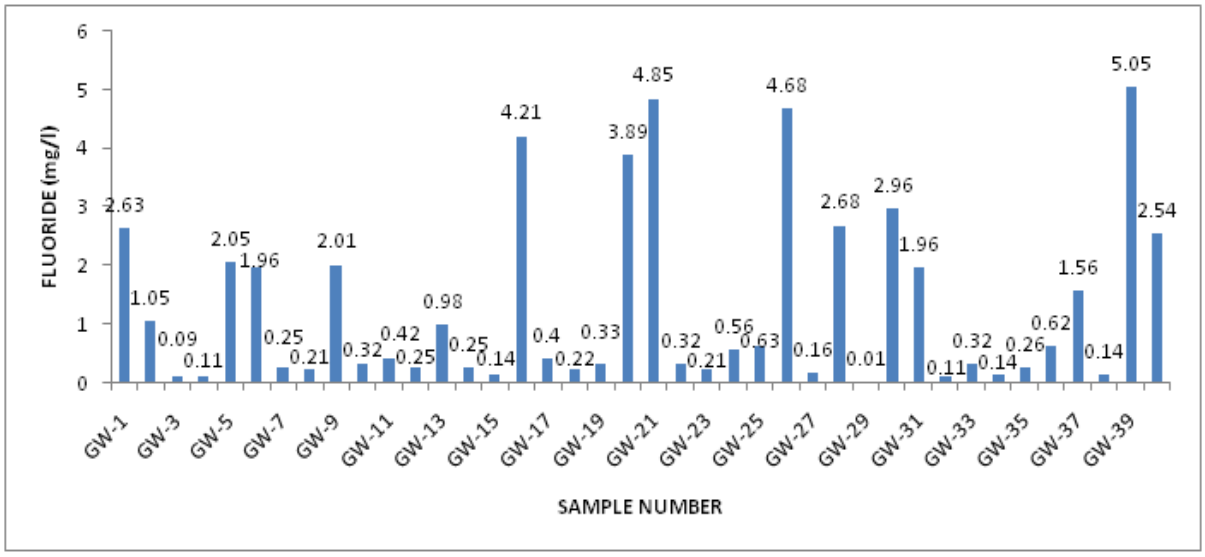

Fig.1. Fluoride Concentrations in the water samples of three district of Southern part of Chhattisgarh.

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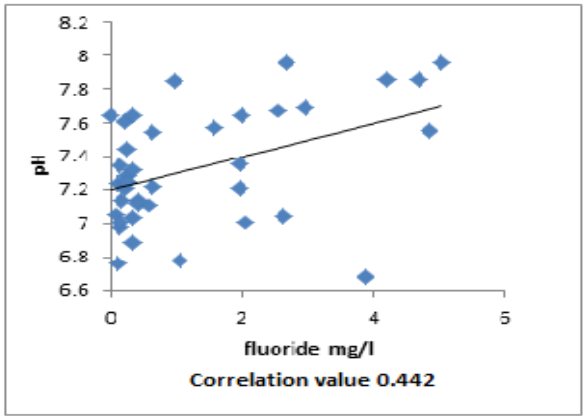

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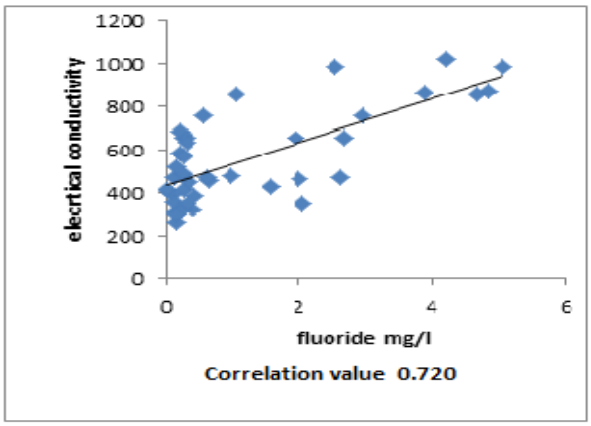

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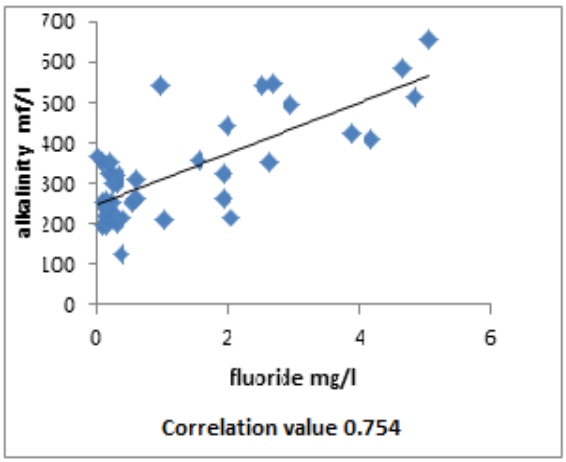
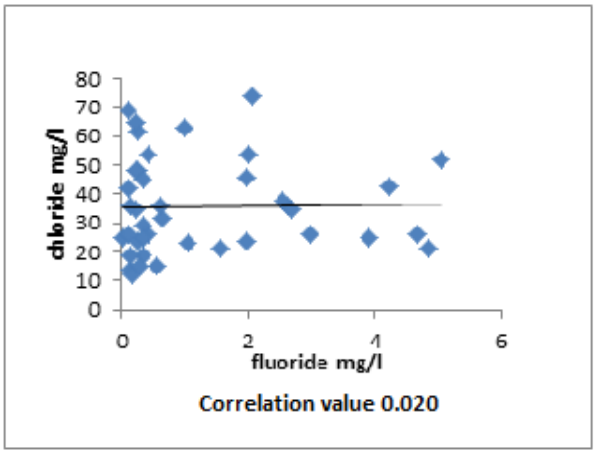


(c)



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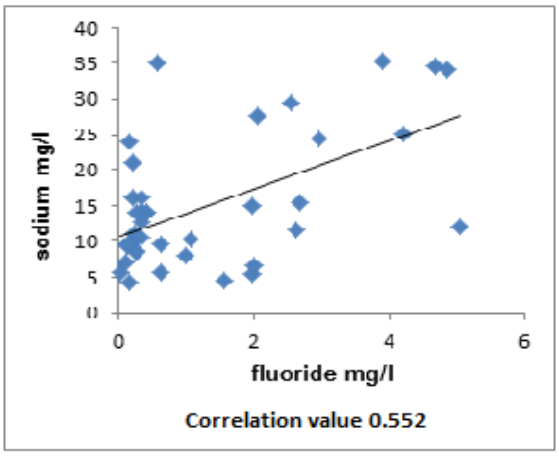

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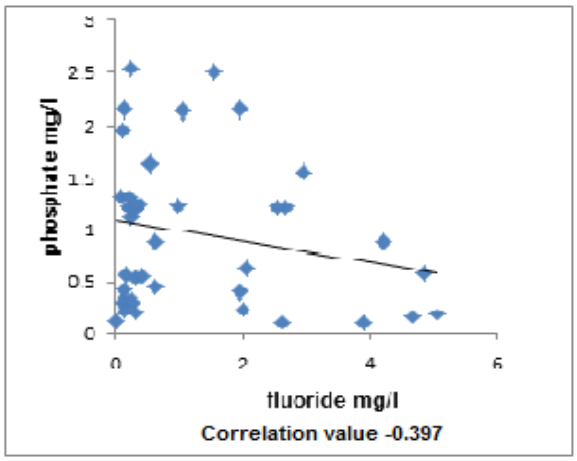

(f)

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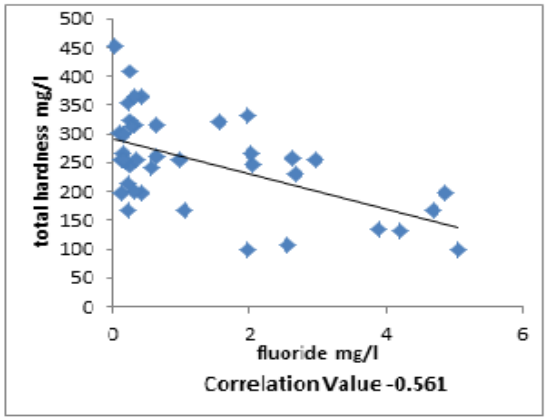

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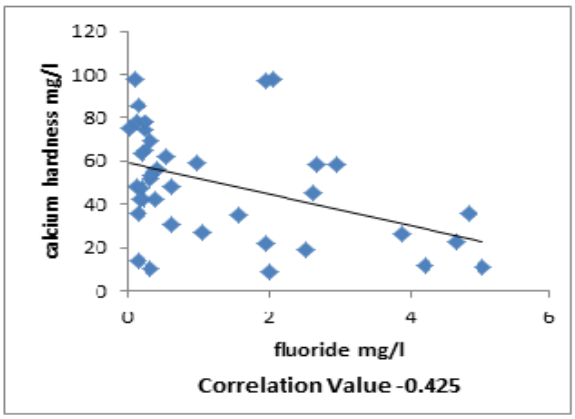
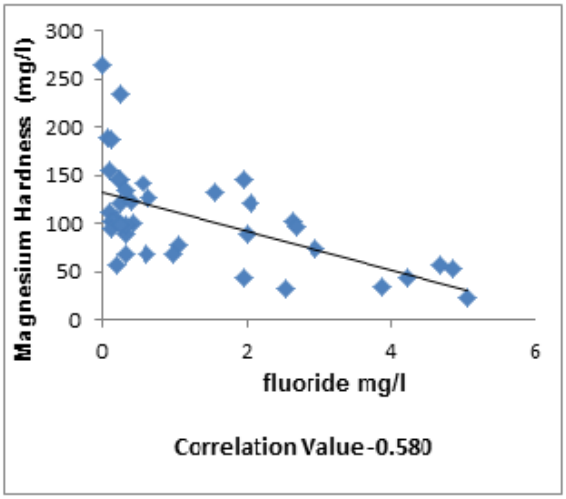
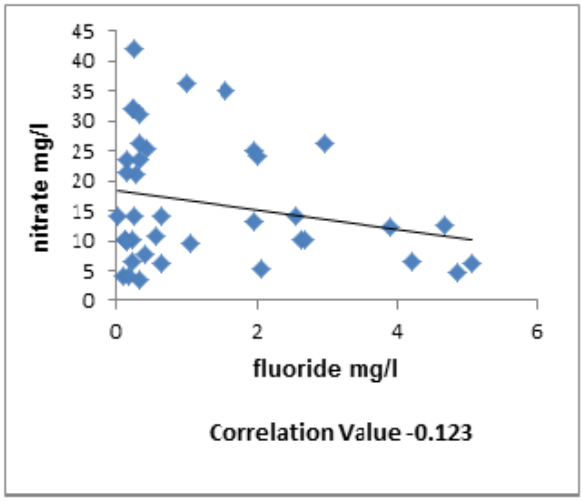


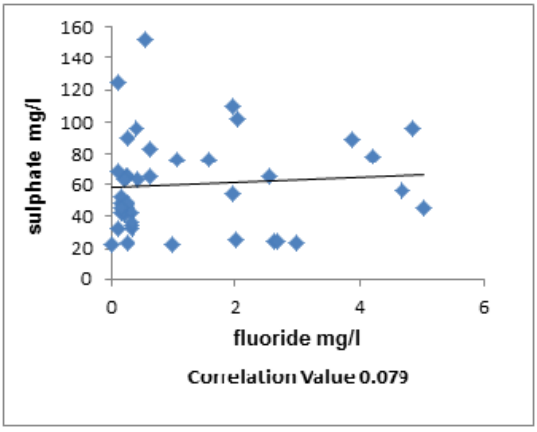
(h)



(i)



(j)



(k)

Fig.2. (a-k). Correlation of different parameters and ions with fluoride concentration in water samples selected for three district of South- East region of Chhattisgarh (a) pH vs F^- (b) EC vs F^- (c) Alkalinity vs F^- (d) Cl^- vs F^- (e) Sodium vs F^- (f) Phosphate vs F^- (g) T.H vs F^- (h) Cavs F^- (i) Mg vs F^- (j) Nitrate vs F^- (k) Sulphate vs F^-

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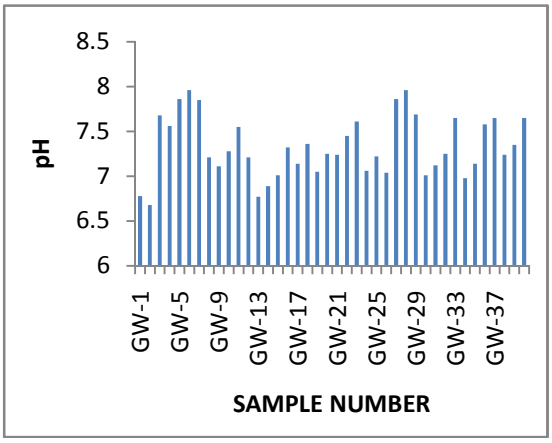

Fig.3. pH concentration in the water samples.

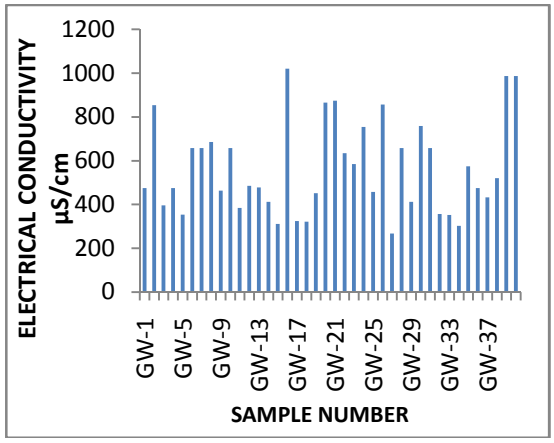

Fig. 4.Electrical Conductivity in the water samples.

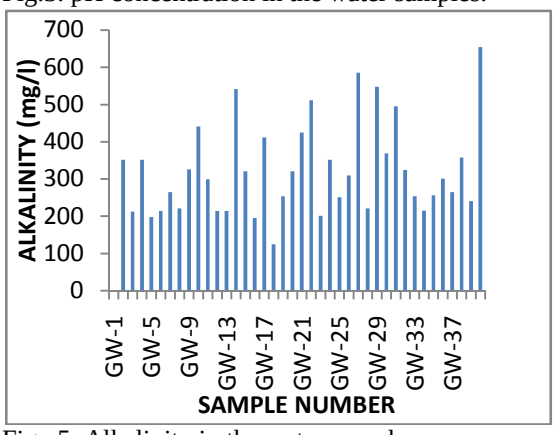

Fig. 5. Alkalinity in the water samples.

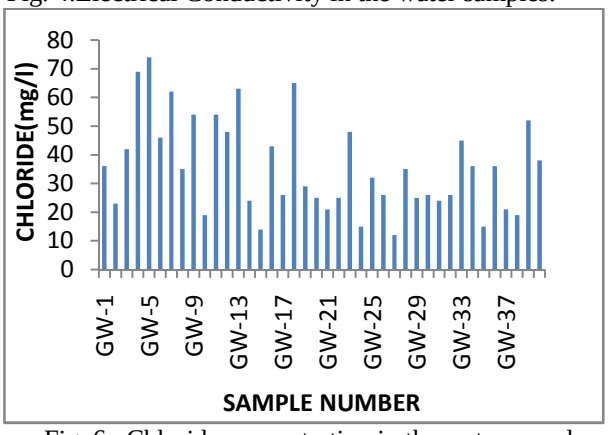

Fig. 6. Chloride concentration in the water samples.

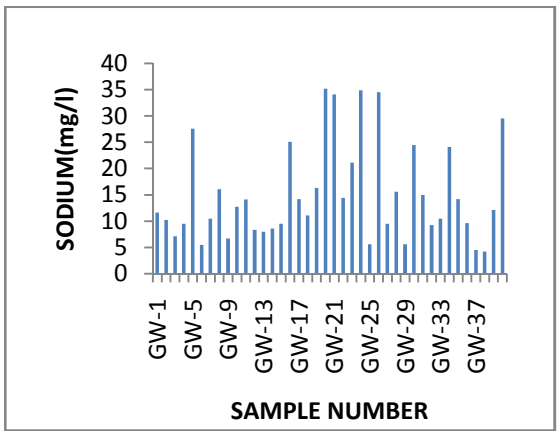

Fig.7. Sodium Concentration in the water samples.

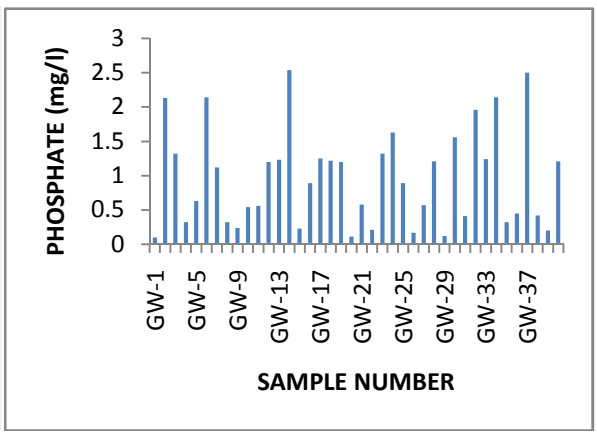

Fig. 8. phosphate concentration in the water samples.

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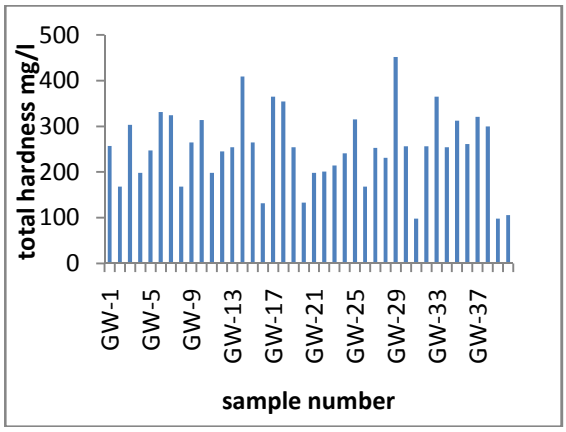

Fig. 9.Total hardness in the water samples.

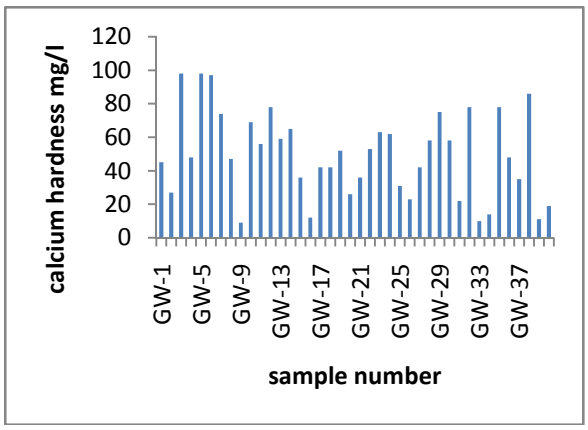

Fig. 10.Calcium hardness in the water samples

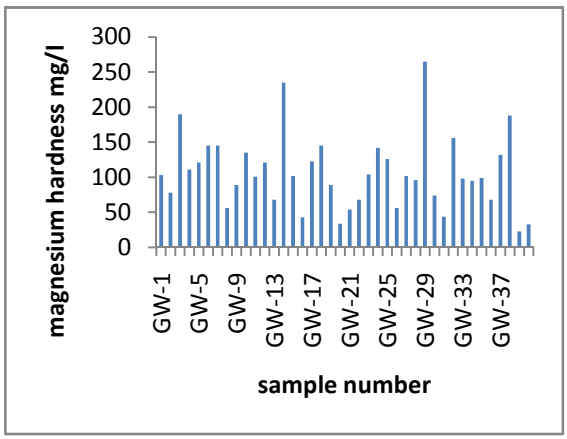

Fig. 11.Magnesium hardness in the sample water.

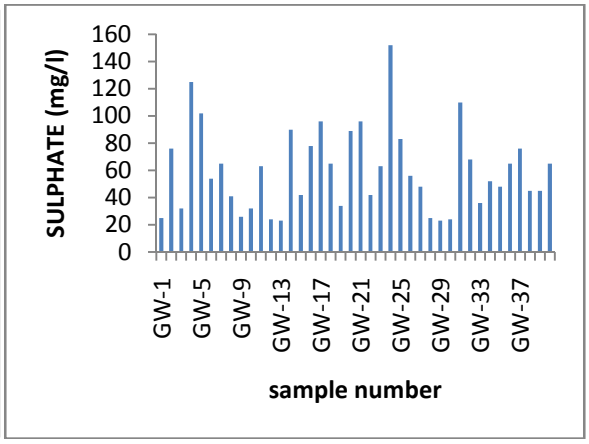

Fig. 12.Sulphate concentration in the samples.

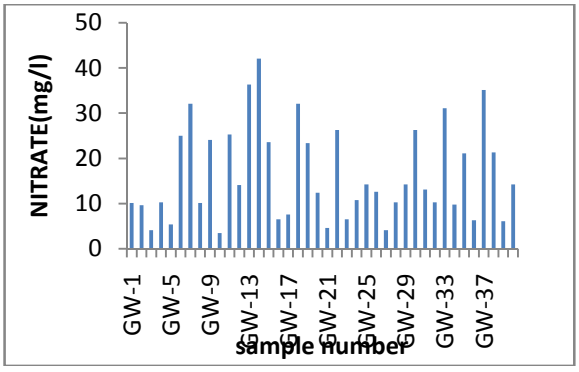

Fig.13. Nitrate concentration in the water samples.