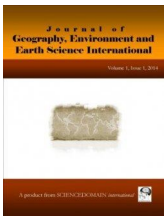

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Origin and Spatial Distribution of Fluoride in Aquifers of Ambalapuzha Basin, Alappuzha District, Kerala, South India

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Authors' contributions

All Authors collaborated for the completion of this work. Author PNA designed the study, ran the model for all cases, performed the data analysis, prepared and revised the manuscript. Author AVG provided valuable suggestions for the study design and supervised the data analysis. Author VBR revised and modified the manuscript according to the suggestion. All authors read and approved the final manuscript.

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ABSTRACT

The present study is an attempt to find out the origin, occurrence and distribution of fluoride content in deep aquifers of Ambalapuzha river basin of Ambalapuzha Taluk. Comparison of the past tube well data with present data, at various localities of the study area shows a drastic increase in fluoride to hazardous level in recent years. Critical analysis of the data reveals that out of 15 tube wells, in all the 14 tube wells, fluoride is much above permissible limit. When cations and anions of the tube well samples were plotted in the piper diagram, replacement of K by Na is clearly indicated. Water type identified with in the study area is predominantly rich in sodium with sodium ion dominance. From ANOVA test, it is revealed that a significant effect of sodium, pH, TDS,

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Alkalinity, bicarbonate and chloride on fluoride. From the study it is found that the large scale withdrawal of groundwater from the clay bed resulted in increase of Na in groundwater. This in turn promoted leaching of fluoride bearing minerals such as flour apatite, apatite, hornblende etc in to the clay beds at deep sources and liberation of fluoride by the process of anion exchange and increased alkalinity which in turn results increase of pH. The study made it clear that the fluoride enrichment is insitu and it is better to avoid development of deep aquifers in Ambalapuzha taluk or it is to be utilized in a very judicious manner by the town planners and authorities of public water supply agencies.

Keywords: Ambalapuzha; fluoride; tube well data; anion exchange.

1. INTRODUCTION

Fluoride is an essential element which is good for teeth of the animal at the same time in in higher concentration it can lead to fluoride contamination in groundwater resources and it is a major problem in many parts of the world. Fluoride is one of the critical chemical parameter, which influences the quality of ground water. Excess intake of fluoride through drinking water has resulted in fluorosis on humans in many states of India [1]. People living in such area were drinking groundwater severely affected by high fluoride content. In India, this problem is noticed in almost all the states such as Andhra Pradesh, Bihar, Chhattisgarh, Gujarat, Madya Pradesh, Punjab, Rajasthan, Tamil Nadu Uttar Pradesh and Kerala [2]. Geologically, Ambalapuzha Taluk is covered by unconsolidated sediments of recent age, underlined by semi consolidated formations of tertiary age. These sedimentary formations are grouped in to Warkalai, Quilon, Vaikom and Alleppey beds. Laterite is seen overlying the tertiary and crystalline formation towards the south eastern parts of Alappuzha district, Analysis of bore hole data indicate, that sediments attain maximum thickness of 600 m in Ambalapuzha area.

The alluvium forms the phreatic surface. Open dug wells tapping this recent alluvium has depth varying from 1 to 2 m below ground level. Filter point wells which are also widely developed in the study area, tap this alluvium based phreatic aquifer. Potential aquifers are seen in tertiary sediments Warkalai and Vaikom formations.

From the detailed study of different chemical parameters revealed that, the hydrochemistry is prevalent in concerned aquifers of study area [3]. Fluoride concentration in deep aquifers can be correlated with the concentration of certain major dissolved constituents [4-6]. Fluoride is inversely related to dissolved calcium and magnesium is normally present in water as F ion [7] and it may also exist in the form of HF, SiF4 or SiF6 under very low pH (3.5) conditions [8]. Water with sufficient concentration of calcium, will be of low F ions concentration [9,10]. Hence higher concentrations of F ions are expected in water with low calcium concentration.

2. MATERIALS AND METHODS

2.1 Study Area

The study area, Ambalapuzha Taluk lies between 90° 19′ 14″ and 90° 36′ 04″ N latitudes and 76° 19′ 44 ″ and 76° 24′ 22 ″ E longitudes (Fig. 1). It is one of the most populated taluks of Kerala. The area under investigation is blessed with abundant groundwater resources that occur in phreatic semi confined and confined conditions. These potential aquifers are widely tapped by dug wells, filter point wells and deep tube wells. The deep tube wells are confined to Warkalai and Vaikom formations. The area covered by 13 villages and 1 Municipality extending from Mararikulam in the north to Thottapally in the south. The main geological features identified are sand and clay formations, with bedrock at a depth of 600 meters. The Warkalai formation and Quilion formation also attains maximum thickness within the study area [11]. In general, the elevation of the area is less than 5 meters above MSL [12].

Among the various lithostratigraphic units, the recent unconsolidated formations along the coastal tracts and flood plain deposits of the interior regions of Ambalapuzha Taluk, are of potential phreatic aquifers. Water table is generally shallow having a depth of 1.5 meters. Depth of filter point wells range between 6 to 15 meters. Soil type of study area is classified in to sandy loam, alluvial and laterite and annual rainfall of the area is 3025 mm.

2.2 Methodology

In order to establish the origin and occurrence of fluoride in the deeper aquifers of Ambalapuzha Taluk, the stratigraphy and distribution of various formations within the study area were studied in detail. Water samples from 15 tube wells of different locations of the study area were collected on 15th of every month, for a period of 22 months. These tube well samples were subjected to critical analysis and interpretation. Samples from dug wells and filter points well were excluded in the present study because it is clearly established that there is no fluoride contamination up to a depth of 60 meters in hazardous level with in the study area [12] and [13]. A comparative study of the past data 1995 to 2000 [13] Table 1 and present data Table 2, Table 3 and Table 4 revealed that a drastic increase in fluoride content in the observation wells.

Table 1. Comparison of Fluoride (mg/l) for the years 1987, 1999, 2000 and 2006 of observation wells in the study area

Place	1987	1999	2000	2006
Milma Punnapra	1.60	1.51	1.75	
Medical College, Vandanam	1.50	1.30	1.44	3.08
Alissery I	1.60	1.45	1.7	3.28
Chudukad I	1.90	1.79	1.93	3.16
Chandanakavu	1.60	1.56	1.58	3.84
Thukkukulam	1.12	1.32	1.45	2.52
Pazhaveedu	1.30	1.45	1.64	2.48
Purakkad	1.30	1.40	1.52	
Kommadi	2.70	1.90	2.24	

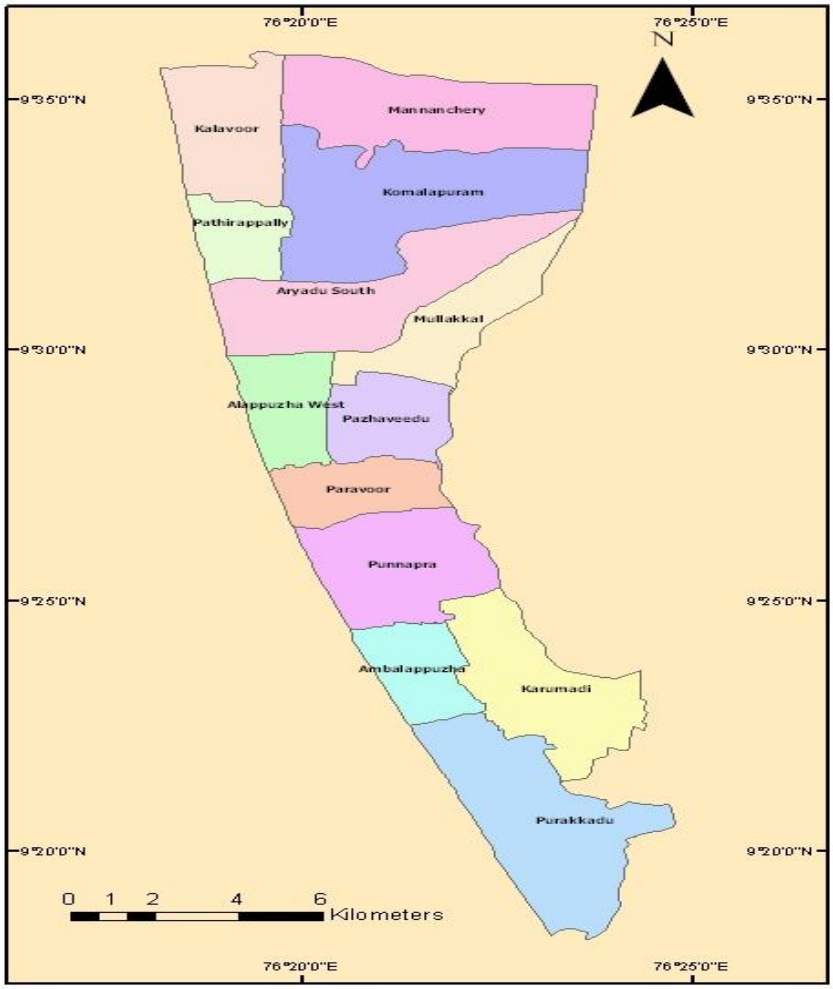

Fig. 1. Location map of Ambalapuzha Taluk

Table 2. Distribution of fluoride in the tube wells of the study area

Well no	Hamlet	Apr-05	May-05	Jun-05	Jul-05	Aug-05	Sept-05	Oct-05	Nov-05	Dec-05	Jan-06	Feb-06	Mar-06	Apr-06	May-06	Jun-06	Jul-06	Aug-06	Sept-06	Oct-06	Nov-06	Dec-06	Jan-07
QS-03	Kattoor	2.95	2.95	2.70	2.20	2.46	2.59	2.22	2.17	2.83	2.55	2.41	2.50	1.60	2.30	2.2	1.70	3.40	2.10	3.40	2.70	2.40	2.10
QS-06	Chettikad	2.36	2.80	2.72	2.80	2.97	3.03	2.87	2.39	2.65	3.20	2.85	3.20	3.80	3.40	3.5	3.40	3.5	3.0	3.5	2.50	2.60	3.00
QS08A	Thumboli	0.74	0.85	1.00	0.25	0.58	0.34	0.88	0.60	0.48	0.54	0.55	0.50	0.37	0.13	0.44	0.90	0.16	0.28	0.90	0.40	0.50	0.90
QS-10	Vazhicherry	1.76	1.80	1.63	1.97	2.60	2.68	2.85	1.50	1.80	2.10	1.76	1.96	1.96	1.99	2.10	1.68	3.08	1.32	3.60	1.70	2.00	3.60
QS-11	Pazhavangadi	3.41	3.20	3.26	2.80	2.15	1.50	2.00	1.00	1.37	2.87	1.91	1.75	2.79	1.99	0.04	2.08	2.72	3.20	1.60	1.30	1.50	1.60
QS-12	Chandnakavu	2.87	0.65	2.80	3.20	3.22	3.30	3.30	3.00	2.04	3.60	2.75	2.95	4.23	3.02	2.69	3.44	2.68	3.84	2.10	3.10	2.80	2.10
QS-13	Chudukadu	2.18	1.90	2.15	1.90	1.97	1.46	2.20	2.10	1.93	2.19	1.93	2.55	2.85	2.30	1.94	2.08	3.28	3.16	1.45	2.50	1.90	1.50
QS-14	Thookukulam	1.26	1.27	1.39	1.39	1.36	1.26	1.19	1.57	1.46	2.06	1.96	1.36	2.01	1.61	1.36	1.88	1.72	2.52	1.20	1.80	1.50	1.20
QS-17	Vandanam	2.00	1.90	1.27	1.04	2.20	1.47	1.79	1.43	1.84	1.99	2.13	1.52	1.94	2.04	1.42	2.04	2.36	3.08	2.40	2.50	2.00	2.40
QS-18	Kanjipadam	1.13	1.27	1.31	1.18	1.18	1.14	1.00	1.35	1.23	1.32	1.35	1.96	2.61	1.70	1.54	1.84	5.84	2.58	1.50	1.50	1.60	1.50
QS-22	Thottapally	1.46	1.43	1.01	1.24	1.25	1.21	1.50	1.45	1.55	1.74	1.72	1.66	2.44	1.85	1.66	1.65	1.34	1.55	1.60	1.10	1.20	0.60
QS-26	Komalapuram	1.46	1.27	0.95	1.23	1.30	1.81	1.32	1.59	1.43	1.95	2.30	2.12	1.95	2.04	1.70	2.24	2.20	2.24	1.60	1.90	1.80	1.60
QS-28	Pazhaveedu	1.58	1.72	1.65	2.00	1.27	1.74	2.00	2.05	1.90	2.05	1.97	2.36	1.86	2.50	1.72	2.68	3.04	2.48	1.90	1.90	1.90	1.90
QS-29	Alissery	1.95	1.85	1.45	1.80	2.50	2.23	2.30	2.20	1.83	2.28	2.49	2.58	2.63	2.29	2.41	2.04	3.12	3.28	1.50	2.40	1.90	1.50
QS-30	Paravoor	1.65	1.00	2.20	1.75	1.57	1.62	1.76	1.80	1.23	1.37	1.33	1.35	2.12	1.17	1.15	1.45	1.30	1.65	1.15	1.40	1.30	1.20

During April 2005 to January 2007

3. RESULTS AND DISCUSSION

Critical analysis of tube well data from the study area exhibits that except at Thumboli (QS-08A), in all other tube wells, the concentration of fluoride is much above the permissible limit. Exceptionally high values of 2.45 ppm, 2.94 ppm and 2.81 ppm were observed (Table 4) at (QS-03), (QS-06) and (QS-12) respectively and it is represented in in Fig. 2 and Fig. 3.

Apart from QS-08A, QS-22 and QS-30 all the tube wells are characterized by high fluoride content ie >1.50 ppm (Table 4) (Fig. 4). Out of these 3 wells, well No QS-08A, locates at Thumboli was within permissible limit of 1.5 ppm throughout the study period. Out of the 27 samples collected, well No.QS14 (9 samples), QS 22 (10 samples) and QS18 (8 samples) shows fluoride values higher than 1.5 ppm (Table 1, Table 2 and Table 3). In all other tube wells, fluoride content was above the permissible limit (Fig. 4). Out of 15 tube wells, well No. QS-06 (Chettikad) shows the highest value of 2.94 ppm. Wells at Kattoor (QS-03) Pazhavangadi (QS-11), Chandanakavu (QS 12) and Chudukad (QS-13) have almost identical values that range from 2.16 to 2.8 ppm (Table 5) throughout study period.

3.1 Piper Diagram

Different values of cations and anions of 13 tube well samples for the month of May 2006 were plotted in piper diagram (Fig. 4). The water types identified are sodium - bicarbonate – chloride, sodium – magnesium – chloride – bicarbonate and sodium – bicarbonate – chloride – types (Table 6). This indicates dominance of sodium ions throughout the study area. Fig. 6 and Fig. 7 are the diagrammatic representation of cations and anions present in the water samples of well No. QS-12 and QS 06 and it revealed maximum fluoride value for the month of May 2006. ANOVA test was carried to fund out influence of chemical parameters Na, pH, TDS, Ca and on the concentration of fluoride.

Table 5. Average value of fluoride in the study area for the period April 2005 to January 2007

Well no	Hamlet	Average (ppm)
QS-03	Kattoor	2.45
QS-06	Chettikad	2.94
QS-08A	Thumboli	0.45
QS-10	Vazhicherry	2.01
QS-11	Pazhavangadi	2.12
QS-12	Chandanakavu	2.81
QS-13	Chudukadu	2.16
QS-14	Thookukulam	1.55
QS-17	Vandanam	1.90
QS-18	Kanjipadam	1.52
QS-22	Thottapally	1.42
QS-26	Komalapuram	1.73
QS-28	Pazhaveedu	2.01
QS-29	Alissery	2.20
QS-30	Paravoor	1.42

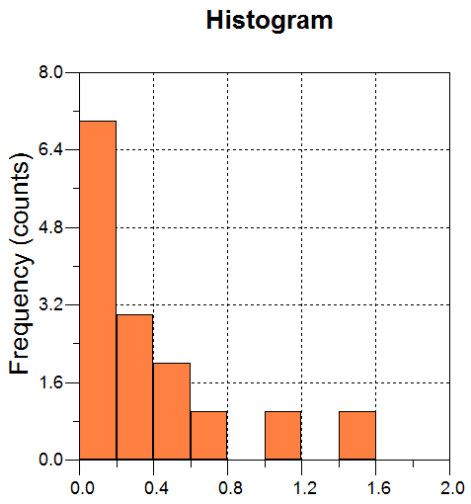

Fig. 2. Variation of fluoride in open wells

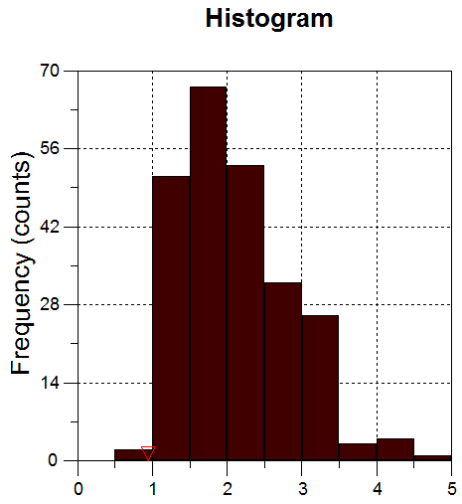

Fig. 3. Variation of fluoride in tube wells

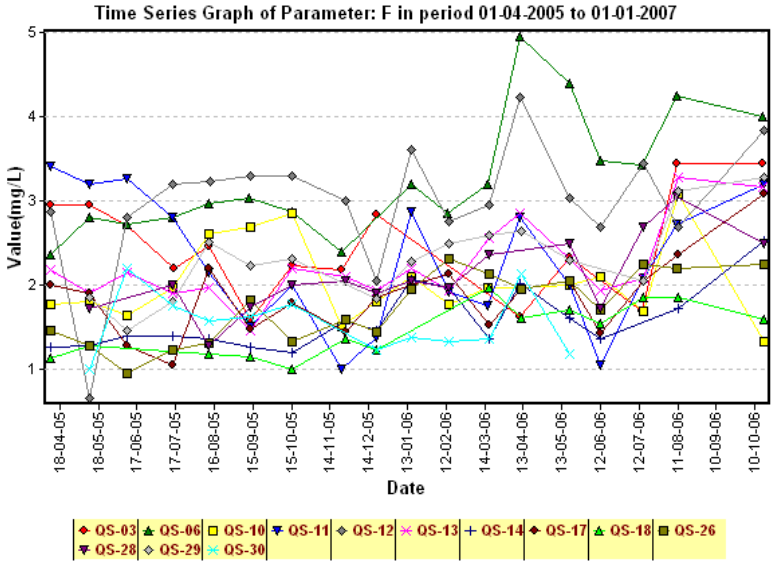

Fig. 4. Variation of fluoride in different wells of the study area

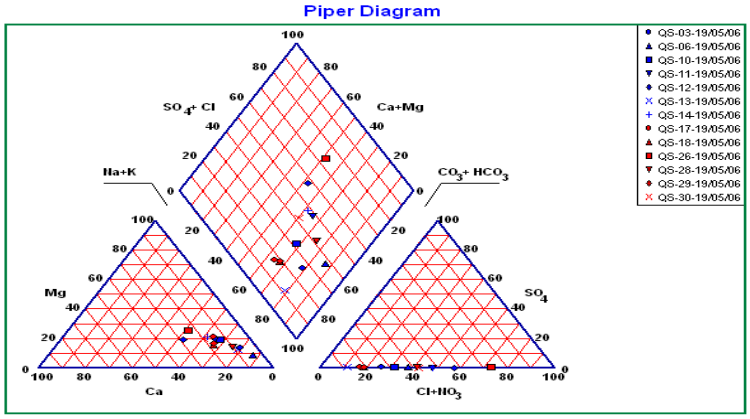

Fig. 5. Water types of study area during May 2006

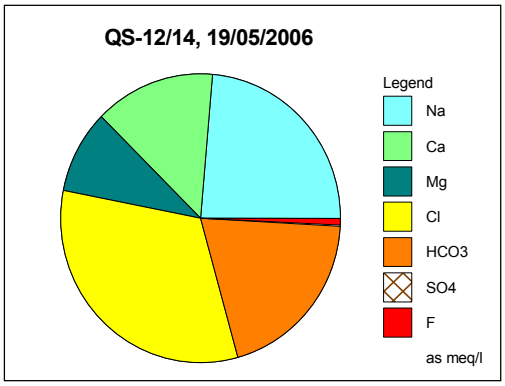

Fig. 6. Pie diagram showing different parameters of QS 1 during May 2006

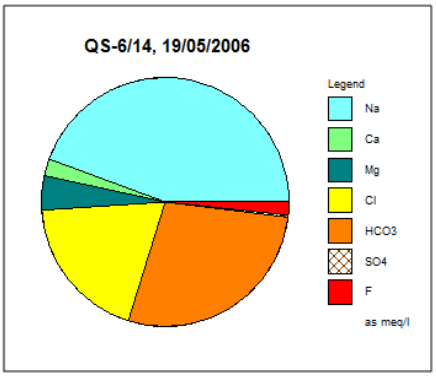

Fig. 7. Pie diagram showing different parameters of QS 6 during May 2006

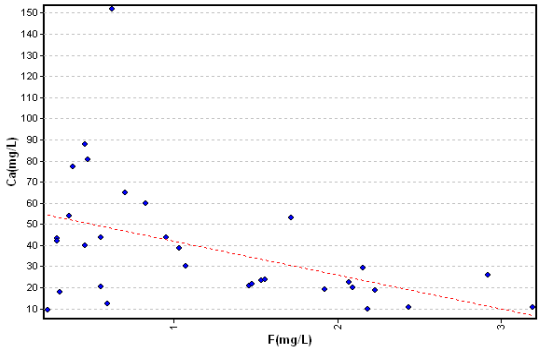

Fig. 8. Graph represents inverse relationship of fluoride with calcium in the study area

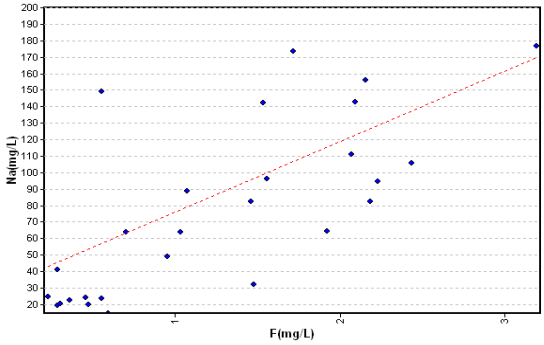

Fig. 9. Graph represents inverse relationship of fluoride with sodium in the study area

Table 6. Water types of study area based on sodium ions

Well No.	Sample Id	Date	Water Type
QS-03	QS-3/14	19/05/2006	Na-HCO3-Cl
QS-06	QS-6/14	19/05/2006	Na-HCO3-Cl
QS-10	QS-10/14	19/05/2006	Na-HCO3-Cl
QS-11	QS-11/14	19/05/2006	Na-Cl-HCO3
QS-12	QS-12/14	19/05/2006	Na-Ca-Cl-HCO3
QS-13	QS-13/14	19/05/2006	Na-HCO3
QS-14	QS-14/14	19/05/2006	Na-Mg-Cl-HCO3
QS-17	QS-17/14	19/05/2006	Na-Mg-HCO3
QS-18	QS-18/14	19/05/2006	Na-HCO3
QS-26	QS-26/14	19/05/2006	Na-Mg-Ca-Cl
QS-28	QS-28/14	19/05/2006	Na-HCO3-Cl
QS-29	QS-29/14	19/05/2006	Na-HCO3
QS-30	QS-30/14	19/05/2006	Na-HCO3-Cl

3.2 Relationship between Fluoride and Ca, Na, HCO3 and pH

In the present study fluoride is inversely related to dissolved Ca (Fig. 8). High fluoride is found only when the concentration of Ca is less than 20 ppm [14]. Fluoride has a direct correlation with dissolved sodium, bicarbonate concentration and pH. [15]. In the present study fluoride increases with higher values of sodium (Fig. 9). The association of higher bicarbonate and lower calcium with dissolution of fluoride bearing minerals explains the correlation of fluoride and bicarbonate (Fig. 10). The situation usually prevails, when fluoride occurs in confined aquifer condition [16]. Another contributor of bicarbonate in the confined aquifer is oxidation of organic matter to dissolved carbon dioxide [17]. Formation of bicarbonate from the carbon dioxide produces acidity, which further derives the dissolution of carbonate minerals or buffers. The pH increase from carbonate solution is caused by the decrease in calcium and magnesium from cation exchange [18]. The wide band of increasing bicarbonate with increasing fluoride is as represented in the graph (Fig. 10) is probably related to both process. A well pronounced positive correlation was observed between pH and fluoride. Source of additional fluoride at higher pH could be the exchange of hydroxyl ions for fluoride ions in liable positions on fluorine containing phyllosilicates, clays and weathered mica such as biotite and muscovite that are present in the fine grained sediments [19]. pH value greater than 8.5 are probably necessary for this process to be a substantial fluoride source relative to dissolution of fluoride bearing minerals. This is because the hydroxyl ion concentration would be too low at lower pH values. The highest pH occurs in the confined aquifers as expected from the greater cation exchange and the carbonate dissolution in the concerned aquifer environment.

3.3 The Hydro Chemical Process Involved in Fluoride Enrichment

The enrichment of fluoride in deeper aquifer is unique since it is not seen in shallow or phreatic aquifers of the present study area. Thus the aquifer materials and the chemical reactions will have a very important role in the enrichment of fluoride in deeper aquifers. The change in the degree and type of mineralization of the water in the Warkalai aquifer from south to north direction can be explained as, recharging fresh water through the sediments deposited under marine conditions. By ion exchange from clay minerals, the calcium rich water moving northward, exchanges calcium for sodium and thereby it became sodium rich and results in Na-HCo_3 type. This alkaline water exchange fluoride from minerals like Fluor apatite with hydroxide, and it suggest beyond doubt that the fluoride enrichment is insitu. Fluoride ions in liable positions of clays and micas present in the argillaceous sediments in the aquifer system. Relationship of fluoride with pH is represented in Fig. 11.

Continuous withdrawal from tube wells causes physical and chemical changes around the well. Thus results in the enrichment of fluoride ions. First of all, a low pressure zone will be created around the well that will result in the vertical leakage of brackish water from the confined clay layers enriched with fluoride ions. This causes release of fluoride ions to fresh water as a consequence of cation exchange of calcium for sodium. Fluoride enrichment may also result from mixing of Ca – HCO_3 type water with NaH-CO_3 or Nacl type water to $NaHCO_3$ type and fluoride is being released as the alkaline water, which is depleted in calcium.

Fluoride values are almost similar during the study period. This indicates that Na enrichment due to heavy pumping facilitates the occurrence of F in water. Fluoride values of tube well water during 1995 were between 1 and 1.26 ppm. By 1999 this increased to 1.11 – 1.86 ppm and became 1.04 to 1.58 in 2000 [20]. This steady increase in fluoride content over a period of time is due to large scale withdrawal of water, which in turn resulted in the increase of sodium, finally leading to increase of fluoride. The observed values in the study period do not show much variation apart from the phenomenon that the monsoon recharge slightly increases F content. It is a generally accepted fact that the Warkalai aquifer is affected by heavy pumping of water by a number of tube wells. This facilitates sodium concentration leading to the enrichment of fluoride content in water. Water having pH above 8.5 permits hydroxyl ion exchange that result in increase of fluoride. In the present study most of the samples have pH above 8.5. So enrichment of fluoride in tube well water is also controlled by its observed pH value. Maximum value of fluoride i.e. 3.80 ppm was reported in well QS- 06 (Chettikad) during April 2006. Well No. QS -12 (Chandanakavu) is also equally contaminated. GIS zonation map of fluoride for the pre monsoon period (Fig. 12) shows that the minimum value of fluoride occurs in Punnapra area and the maximum at Alappuzha, Pazhaveedu and Pathirapally area. The maximum value reported was 3.85. During monsoon period (Fig. 13) there was a slight decrease in the maximum value than the pre monsoon (maximum 3.54) and during post monsoon, the maximum value reported was 2.57 (Fig. 14).

The average value of fluoride of each tube well from 1/4/2005 to 31/1/2007 and it comparison with the study area average the maximum fluoride during the study period was observed in tube well of Chettikad (QS-06) and the minimum at Thumboli (QS-08A) (Fig. 15).

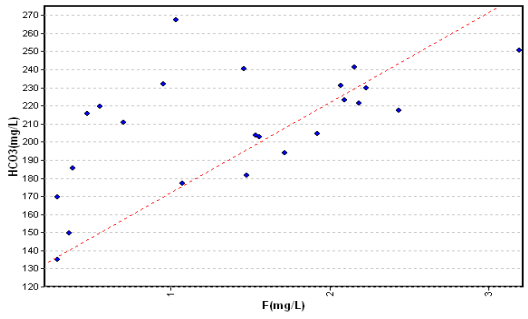

Fig. 10. Graph represents direct relationship of fluoride with bicarbonate in the study area

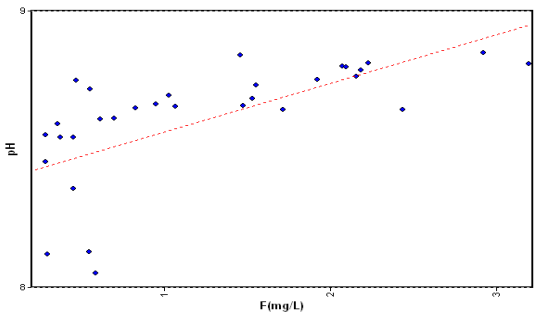

Fig. 11. Graph represents direct relationship of fluoride with pH in the study area

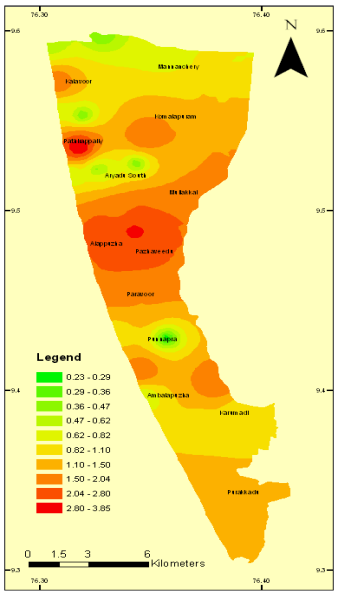

Fig. 12. Zonation map of flouride during pre monsoon monsoon

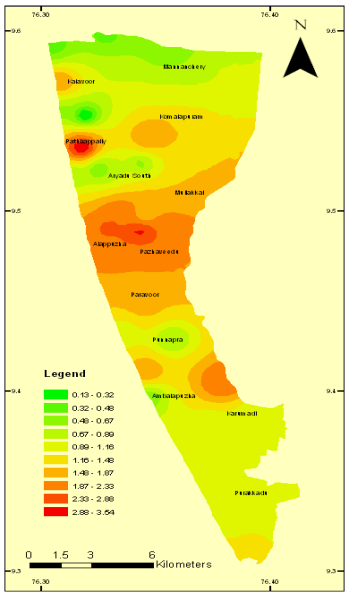

Fig. 13. Zonation map of flouride during monsoon

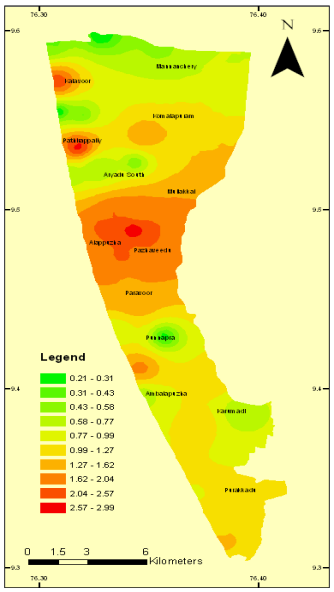

Fig. 14. Zonation map of fluoride during post monsoon

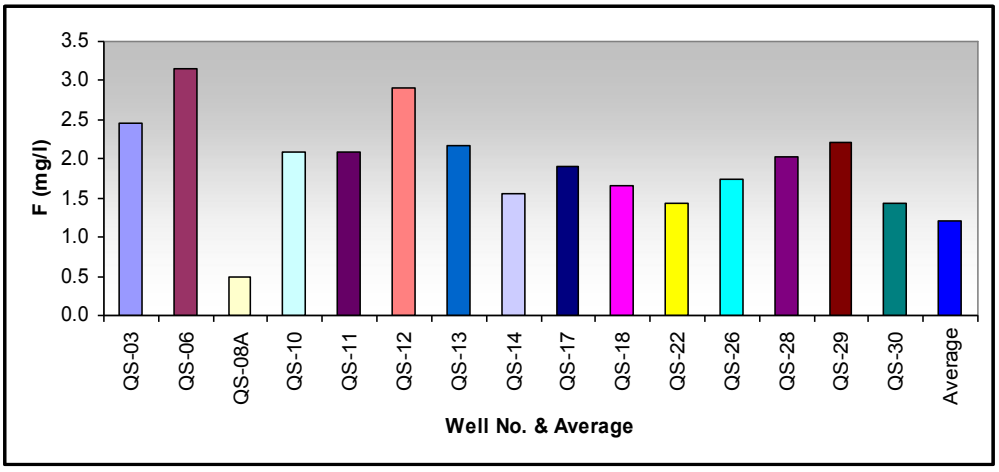

Fig. 15. Graph represents the comparison of average value of fluoride of each tube wells from 01/04/2005 to 31/01/2007 with the study area average

4. CONCLUSION

The major water supply of the Ambalapuzha taluk is by tube wells, and this water is having high fluoride content. Comparison of fluoride content of the present data with the past data confirms that, drastic increase in fluoride content in these wells. Critical evaluation and assessment of data revealed that large scale, non-judicious withdrawal of groundwater piercing the clay beds has resulted in increase of sodium in water which in turn promotes leaching of fluoride from fluoride bearing minerals in to the clay beds. Thus the high fluoride content can be attributed to liberation of fluoride by the process of anion exchange and increased alkalinity, which results in increase of pH. Hence the present large scale consumption of groundwater from deeper aquifer has to be discouraged and judicious use of these aquifers is to be promoted by public water supply agencies.

CONSENT

It is not applicable.

ETHICAL APPROVAL

It is not applicable.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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