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Groundwater Quality in Different Climatic Zones of Sri Lanka: Focus on the Occurrence of Fluoride

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Abstract—**The chemical and physical parameters of groundwater in some areas of dry and wet zones of Sri Lanka are described. Hydrochemical data from 114 water samples are used to evaluate water quality and to identify the processes that control fluoride levels in water. The results indicate that fluoride levels of groundwater in the dry zone are very high (< 8.0 mg/L) compared to those of the wet zone (< 0.8 mg/L). The measured nutrients (NO3- < 15 mg/L, PO43− < 5 mg/L) concentrations and chemical oxygen demand (< 18 mg/L) in both regions inferred that higher fluoride levels reported in the groundwater are not from anthropogenic sources. The pH-Eh stability diagrams show that nitrate, phosphate and iron does not co-exist with fluoride under prevailing redox condition. Physical parameters further show that fluoride values increase in slightly alkaline in pH (7.5-8.2), relatively low EC (1.0-2.5 mS/m) and highly oxidized water (50-200 mV). Groundwater movements of shallow regolith aquifers in the country are mainly controlled on joint and fault system in the partially weathered basement. This study observed that their variation in the dry and wet zones is a major parameter for the control fluoride levels in water. Thus less fractured rocks in the dry zone water discharge areas show higher values and may be due to lesser mixing of water with non-fluoride sources. Conversely, highly fractured rocks are predominant in the wet zone and hence mixing of fluoride poor and rich waters in both recharge and discharge regions in the zone results in low fluoride levels.**

Index Terms—**groundwater, fluoride, physical properties**

I. Introduction

Fluorine is particularly available in rocks, soils, water and biological chains in living organisms. It has higher electronegativity and reactivity. Fluoride occurs naturally in water due to weathering of rocks that contain fluoride rich minerals such as hornblende, biotite, apatite and fluorite [1], [2], [3]. Fluoride can also leach into water f rom anthropogenic sources such as phosphate fertilizers and electronic waste materials [4], [5]. Fluoride-related health problems are common particularly in the arid and semi arid areas of many countries of the world [6], [7], [8]. Optimum contents of fluoride however are essential for the growth of bones and formation of dental enamel, while higher levels (>1.5 mg/L) in drinking water may pose a threat to human health [3], [9], [10]. Further, even at lower concentrations of fluoride, exposure over a long period of time can cause for kidney failures [11].

Abundance of fluoride in water depends on several factors. Fluoride is readily available in water with lower Ca and higher Na [12]. Anthropogenic inputs such as fertilization and farming activities can elevate the concentrations of fluoride in water [4], [13], [14]. On the other hand, as a major component in acidic soils, iron hydroxides serve as an important sink for fluoride in soil resulting into the enhancement of fluoride concentration in water under acidic conditions [15], [16]. Fluorine is not highly redox sensitive and therefore it occurs naturally as fluoride in a wide range of pH and under positive oxidation reduction potential (ORP). However it can be available as HF under strongly acidic conditions [17].

The dry and wet zones are the main climatic divisions in Sri Lanka and the vast majority of people depend on groundwater for their domestic uses. The physical and chemical parameters of water are closely linked to the climatic variations hence the water quality tends to vary in these regions. The dry zone regolith aquifers are well known for higher fluoride concentrations and fluoride-related endemic diseases and chronic renal failures are common problem in such areas [6], [18]. Several studies have been carried out to investigate sources, spatial distribution and movement patterns of fluoride [19], [20], [21], [22]. However, the behavior of fluoride in water in Sri Lanka is still controversial. The scope of this study was to investigate physical and chemical properties of groundwater in high- and low-fluoride regions of the dry and wet zones of Sri Lanka in order to study natural and anthropogenic influences on fluoride chemistry of groundwater.

II. Study Area

A field survey was done in the groundwater discharge regions of Medirigiriya, Eppawala, Talawa, and Padaviya in dry zone in addition to wet zone water discharge and recharge regions of Hikkaduwa, Udunuwara, Kundasale, Alawathugoda, Matale and Naula (Fig. 1a and 1b). Compared to the groundwater in the dry zone, the wet zone is

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characterized by low fluoride in groundwater [19], [21], [22]. The dry zone is typically influenced by the tropical climate with a low mean annual rainfall (< 1500 mm) and average annual evaporation of 1400 mm. Its average annual temperature is 33 ^{0}C, and average annual sunshine is 2555 hrs [23]. The mean annual rainfall in the wet zone is higher than the dry zone (> 2500 mm) and its average annual temperature is low (25 ^{0}C).

A. Hydrological setting

From the hydrological point of view, surface water movement of the country shows centrifugal pattern [24], (Fig. 1a). The central part consists of mountainous forest (> 1000 ft) and receives higher rainfall throughout the year and hence groundwater recharge is extremely high. The region of lower elevation (< 1000 ft) is considered as a water discharge area and receives relatively less rainfall. The river system starts from the recharge area and usually flows through the tank cascade system in the discharge regions (Fig. 1c). The movement of groundwater from recharge to discharge area is structurally controlled by joint and fault system of the basement rocks (Fig. 1d). The rock layers are primarily oriented in a north-south direction and water movement generally follows along the general strike [25]. The main aquifer systems of the recharge area is shallow regolith, alluvial aquifers and are present along the river flood plains and in discharge areas which predominantly consist of shallow and deep regolith aquifers within the tank cascade system [26].

B. Geological setting

More than 90 % of the country is made up of Precambrian high-grad metamorphic rocks consist of four main lithotectonic unites such as Highland Complex (HC), Wanni Complex (WC) Vijayan Complex (VC) and Kadugannawa Complex (KC) (Fig.1b). The Precambrian basement consists of alternative association of meta-sedimentary and meta-igneous formations [25]. Water samples were mainly analyzed from regolith aquifers in HC. The high fluoride sites on HC predominantly consist of alkaline meta-igneous rocks such as metabasites, charnockitic, granitic gneisses and pelitic rocks. The rocks found in the low water-fluoride areas of the HC are meta-sedimentary rocks such as meta-pelites, meta-arkoses and marble. Soil overburdens of the in-situ weathered rocks are extended from few meters to tense of meters. The presence of montmorillonite in dry zone soil indicate the slow weathering processes compared to the wet zone of which composes of kaolinite and laterites as major constituents in soil [27].

III. Methodology

A sampling campaign was undertaken during the intermediate season in March and April 2008. One hundred and fourteen (114) samples were analyzed from d omestic water production wells and agricultural wells drawn from shallow groundwaters. Fluoride, nitrate, phosphate and COD were analyzed using a field pack test method, and pH, DO, ORP and EC were measured using digital multi-electrode.

The field pack test method is well known as a simplified laboratory titration method. Fluoride was determined using 4-pyridine carboxylic acid colorimetric method and nitrate, phosphate, iron and COD are measured using the naphthylethylenediamine method after zinc reduction, molybdenum blue color comparison method, reduction 0-phenanthroline color comparison method and alkaline potassium permanganate oxidation method respectively. Detection limit for the fluoride is 0.2 mg/L and for nitrate, phosphate iron and COD are 1 mg/L, 0.2 mg/L, 0.2 mg/L and 1 mg/L respectively. Horiba multi-parameter meter D-series was used for study the physico-chemical properties. Average accuracy for the analysis for pH, DO, ORP and EC are ±0.1, ±2%, ±20 mV and ±0.5% respectively.

IV. Results And Discussion

Average chemical constituents of the water samples are given in Table 1. Fluoride concentrations in groundwater of both dry and wet zones lie in a range of 0.2 to 8.0 mg/L (avg 0.9 mg/L). Considerably lower concentrations of nitrate, phosphate and iron were measured and values range from 0.5 to 15 mg/L (avg 2.7 mg/L), 0.5 to 5 mg/L (avg 1.0 mg/L) and 0.2 to 8 mg/L (avg 0.5 mg/L) respectively. Values of pH are range from weak acidic to weak basic ranging from 4.0 to 8.2 (avg 7.2). Very low EC values (2.40 mS/m) indicated that dissolved ionic species are very low in the water. Average value of ORP is 113 mV and DO is 4.6 mg/L represent the water is in oxidizing condition and reflect wide range of ORP and DO (Table 1). Average low COD values (6 mg/L) indicate that the amount of organic pollutants is very low.

Dry zone samples show extremely high values of fluoride compared to those of the wet zone (Table 1). Further, relatively higher values of iron reported over in the dry zone. However nitrate and phosphate concentrations are almost similar in both regions. Relatively higher EC values were reported in the wet zone rather than in the dry zone. However pH, ORP, DO and COD values were almost same in the both regions.

A. Relationship between fluoride and physico-chemical properties

Contour plots for fluoride with respect to physico-chemical properties are given in figure 2. The plot of fluoride with pH and EC suggests that higher groundwater fluoride exists at relatively higher pH [6] and lower EC (Fig. 2a). The low values of EC indicate the lesser dissolution of carbonate minerals in water in view of the presence of meta-igneous rocks which consist of less carbonate minerals [28], [29]. This causes lowering of the conditions for the availability of fluoride [6]. The contour plot of fluoride with COD and ORP reflect a wide range distribution, and it is suggested that higher groundwater fluoride is available under oxidizing condition with lower COD levels (Fig. 2b). Oxidized conditions of the water is mainly maintained by well structure [26], most of the wells in Sri Lanka is shallow and open with an average diameter of around 8 feet and hence water contacts directly with atmospheric oxygen which increases DO and ORP [30]. The lower COD values of the studied samples reflect the less dissolved organic carbon in water indicating that water rich in fluoride is less contaminated by organic matter [30]. Hence the contour plot of fluoride with DO and COD shows that the higher fluorides exist under lower COD values with a considerably high DO level (Fig. 2c). The contour plot of fluoride with nitrate and phosphate (Fig. 2d) clearly indicates that higher fluoride in water does not comply with nutrient concentrations. This inferred that the higher fluoride values are not due to artificial contaminations such as fertilizers. Thus fluoride may leach into groundwater due to weathering of fluoride-rich minerals in the basement rocks [19].

B. *Stability relationship of fluoride*

The Eh-pH stability diagram for fluoride is shown in figure 3a. The relationship between pH and ORP of fluoride containing groundwater is observed as a negative correlation (R = - 0.44, n = 114). This is due to fact that higher fluorides exists under slightly alkaline pH and relatively lower oxidizing conditions. The plot reflects that samples of both dry and wet zones lie within the range of groundwater oxidizing zone [17] and under favorable redox conditions for the occurrence of fluoride. The diagram further indicates that fluoride is stable in wide range of pH and both oxidized and reduced conditions due to their higher reactivity and electronegativity. As a result, fluoride can mobilize into both shallow and deep groundwater with some less redox sensitive cations such as Na^+ [3], [16]. The studied samples are obtained from open dug wells and are plotted on the groundwater stability field in the Eh-pH diagram (Fig. 3a). This may indicate that the shallow open dug wells of the fluoride rich area have contact with deep groundwater sources rather than the mixing with surface water sources such as a tank cascade system surrounding them (Fig. 1c).

The local process of water recharge in the dry zone occurs during short the rainy season. Therefore, water percolation through the soil takes place slowly [23]. Hence the water may saturate with fluoride and can percolate down to the deep groundwater through fracture zones. This water gradually releases to wells, especially during the dry season and thus the ORP values are maintained within the range of the groundwater stability field. The pH is closely linked with ORP thus If in case of rains with lower pH fluoride concentration of water can further raise due to dissolution of water soluble metal-fluoride complexes in soils [31], [15], [16], (Fig. 1c). Especially the acid rains reported in dry zone of the country during the November and December (pH, 5.37-7.47) and April to May (avg pH 3.9) [32]. However the effect of lower pH will not*Relationships of fluoride with nitrate, phosphate and iron*

Nutrient levels of studied samples are relatively low although the most of samples are collected from agricultural fields or home gardens. The lower values of nitrate may be a result of denitrification of nitrate by reaction with organic carbon (equation 1) or other denitrification processes [33]. Therefore water samples plot in the N_2 stability field of Eh-pH stability diagram (Fig. 3b). The Eh-pH diagram of phosphorous indicates that the samples lie on the hydrogen phosphate stability field (Fig. 3c). Hydrogen phosphate can react with H_2O to release phosphate and H^+ to water (equation 2), [30], [34], and it might be a result of low pH of some samples (> 4.0). On the other hand due to denitrification H^+ is consumed to maintain weak alkaline conditions (< 8.2) thus pH of t*Distribution of fluoride in different climatic zones*

Distribution of fluoride in the country is diverse and may be controlled by several factors. The studied water samples reflect the higher water fluoride in dry zone as compared to that in the wet zone is a typical observation of the country [8], [11], [21], [22]. Moreover, high fluoride is mainly reported in water discharge areas of the dry zone. However both discharge and rech The Eh-pH diagram for the iron is given in Figure 3d. The fluoride rich minerals such as hornblende, biotite and other ferromagnesian elements are rich in rocks of the study area [25], [29]. However, low values of iron are reported in water samples with fluoride because iron is generally stable in reduced condition and/or acidic pH [34]. However, the stability diagram for iron shows samples are lie in oxidized environment in the field of Fe_2O_3 and it should be the result for poor relationship of iron with fluoride.system of bedrocks of regolith aquifers (Fig. 1d). The joint density of both recharge and discharge regions of the basement in the wet zone is extremely high [35]. As a result the mixing of groundwater is very high due to strong regional water flow. Further, the higher rainfall has a dilution effect on the overall system. Hence even if fluoride rich minerals are available in the basement of the wet zone [29], fluoride values of groundwater are very low and the distribution is identical along the zone. On the other hand the discharge region of the dry zone shows relatively low joint density [35] and lower hydraulic gradient and hence local water flow is highly influenced as compared to the regional flow thus mixing of groundwater is very low between fluoride rich sources and poor sources. Due to dry climatic condition and less rainfall, the saturation of fluoride can readily take place by higher evapotranspiration and accumulation of fluorides with residual alkaline metals [21], [22], [6].

V. Conclusions

Hydrochemical data for groundwater in locations of both dry and wet regions of Sri Lanka show that the higher levels of fluoride are mainly present in the dry zone oxidized water with a slightly alkaline pH. Therefore iron is not stable with fluoride. Lack of correlation of fluoride with nutrients in the studied water indicates that anthropogenic inputs of fluoride are negligible. Lower EC values indicate favorable conditions for the availability of free fluoride in the dry zone due to a lesser impact from carbonate minerals. In contrast higher EC values which can be due to the dissolution of carbonate minerals in the wet zone may have an effect on the fluoride chemistry of the zone. Movements of groundwater have a considerable effect on fluoride distribution and concentration controlled by rainfall and facture density of the basement rocks. Due to higher regional water movement in the wet zone, the fluoride concentration extremely low in contrast to the dry zone where water movements and mixing are limited into localized areas.

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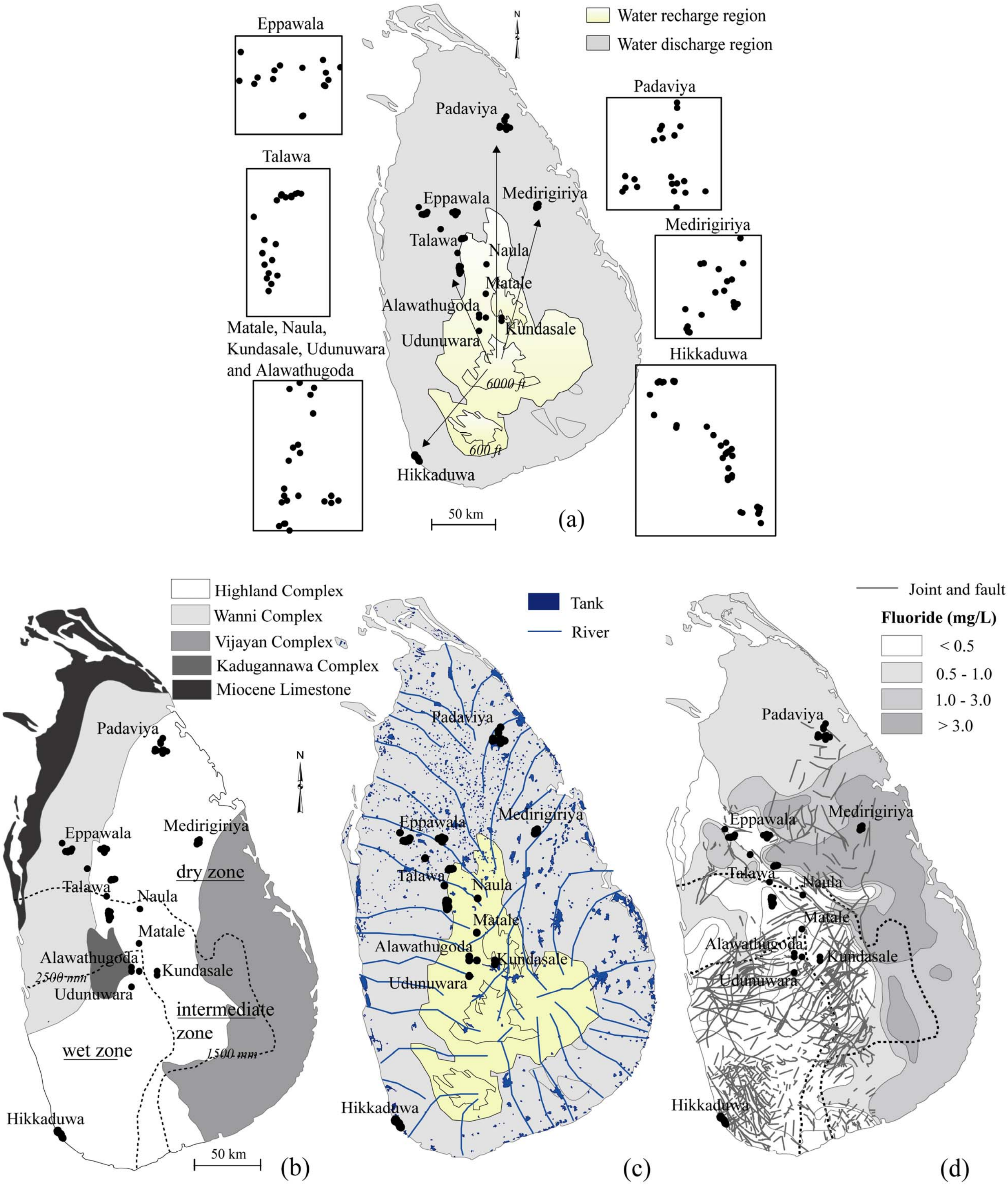

Fig. 1. Maps of the study areas showing (a) sampling locations (filled circles) and groundwater recharge and discharge areas (Groundwater flow directions are indicated by arrows, square denote the detail view of sampling sites) (b) major geological unites and climatic zones (dashed lines denote climatic boundaries) (c) rivers and tank cascade system and (d) distribution of fluoride in groundwater [19] and brittle deformational features (joints and faults) of the basement rocks [35], [25], [36].

TABLE 1. AVERAGE MEASURED VALUES OF GROUNDWATER OF THE DRY AND WET ZONES IN SRI LANKA.

	Tem	pH	EC (mS/cm)	ORP (mV)	DO (mg/L)	COD (mg/L)	NO_3^- (mg/L)	PO_4^{3-} (mg/L)	Fe (mg/L)	F (mg/L)
Total (n=114)										
Avg	30	7.2	2.40	113	4.6	6	2.7	1.0	0.5	0.9
Min	25	4.0	0.16	2	1.8	1	1.0	0.5	0.2	0.2
Max	35	8.2	24.00	223	9.2	18	15.0	5.0	8.0	8.0

SD	3	0.5	3.95	68	1.7	3	3.7	0.9	1.3	1.4
Dry zone (n=69)										
Avg	28	7.2	1.04	111	4.7	6	2.9	0.8	0.5	1.2
Min	25	5.9	0.16	2	1.8	1	1.0	0.5	0.2	0.2
Max	32	8.2	3.25	223	9.2	18	15.0	2.0	8.0	8.0
SD	2	0.4	0.63	71	1.7	3	4.5	0.4	1.5	1.7
Wet zone (n=45)										
Avg	33	7.1	4.69	125	4.2	6	2.4	1.2	0.4	0.3
Min	29	4.0	0.23	15	1.8	1	1.0	0.5	0.2	0.2
Max	35	7.8	24.00	184	6.2	15	8.0	5.0	1.5	0.8
SD	2	0.7	5.77	55	1.6	4	2.3	1.2	0.5	0.1

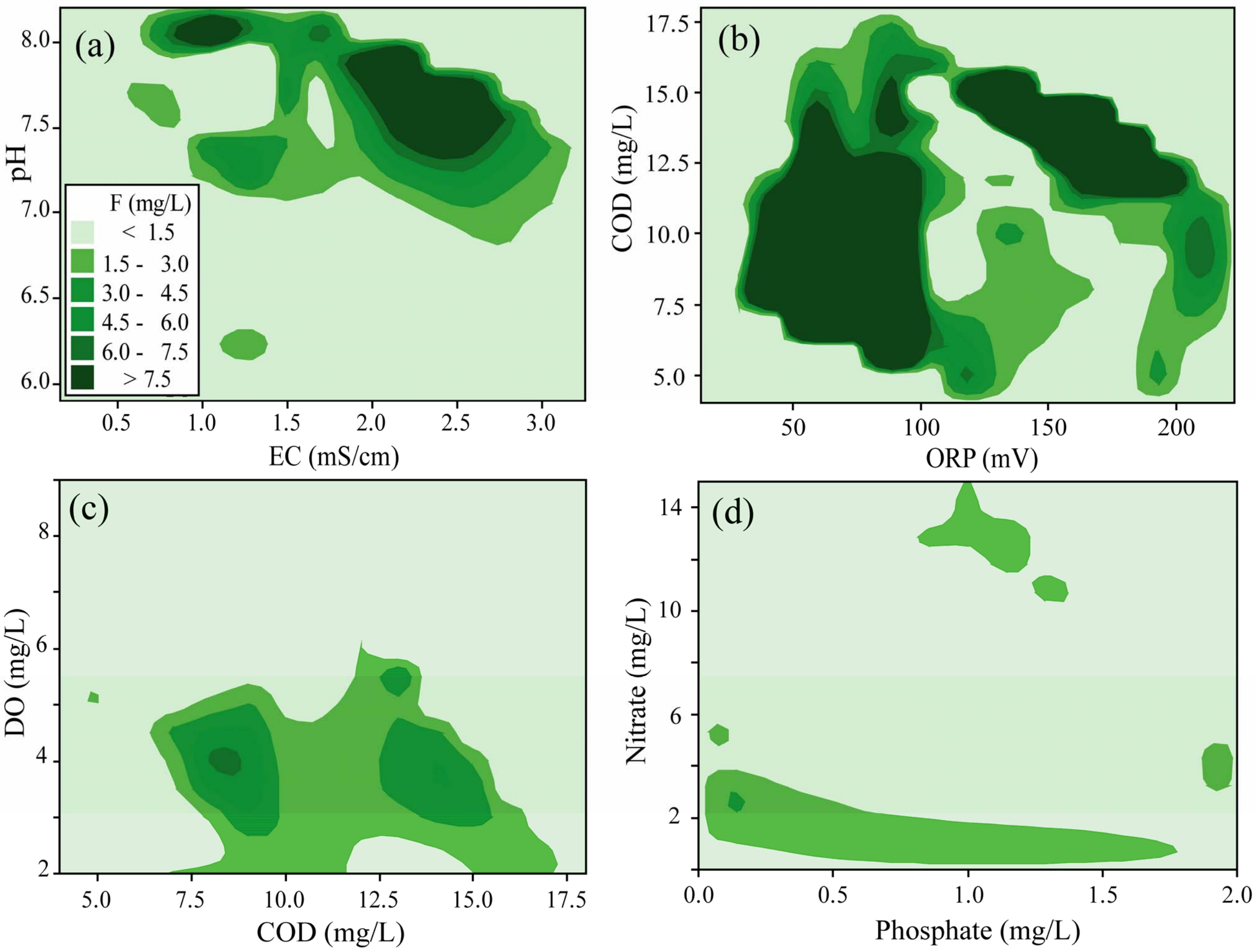

Fig. 2. Contour plots of fluoride with respect to physico-chemical and chemical parameters of water.

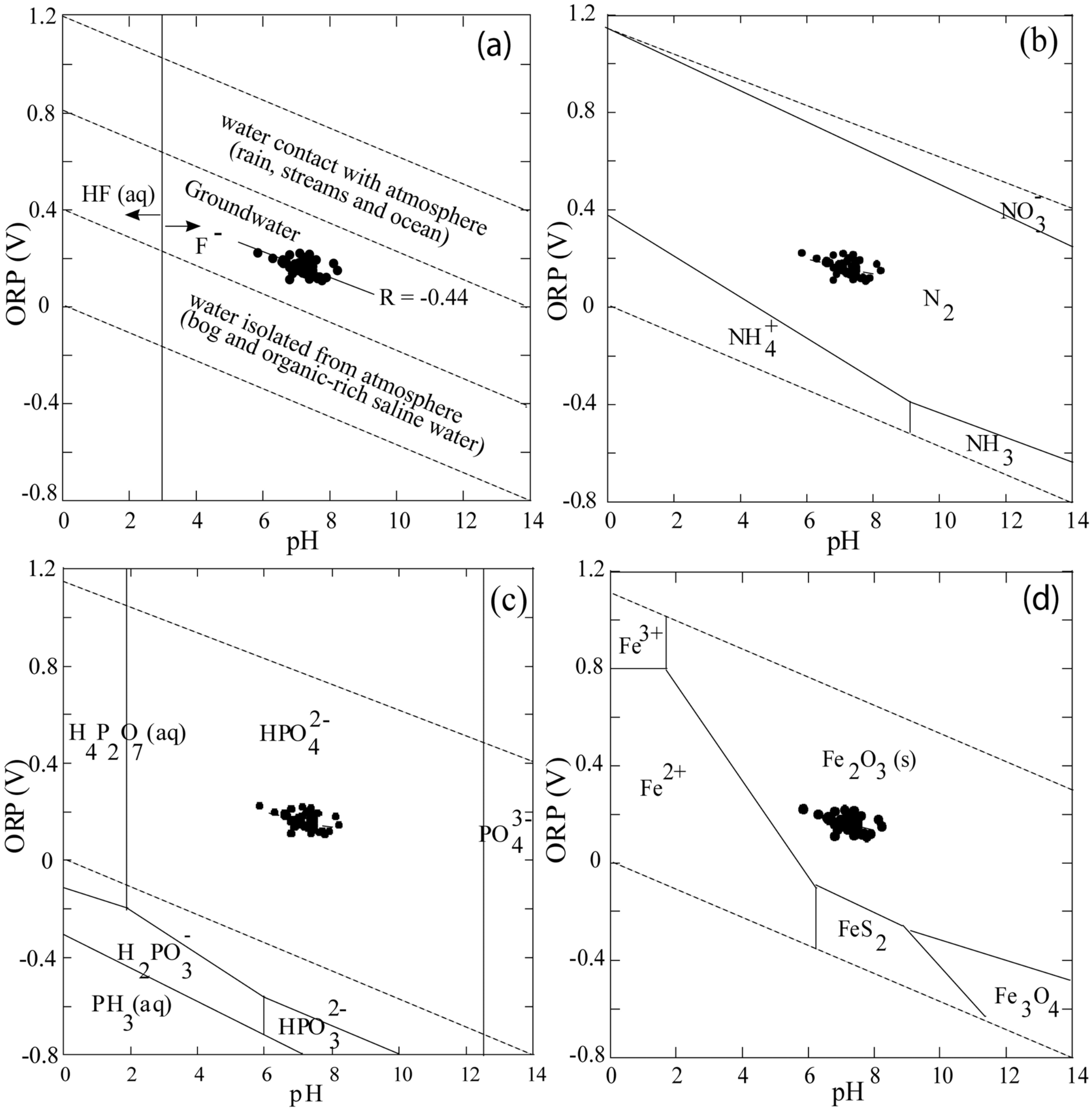

Fig. 3. Composite Eh –pH diagrams for the stability relations of water at 25°C, open circles denote water samples. (a) stability diagram for fluoride with stability limits for natural water at the earth surface [37]; (b), (c) and (d) stability diagrams for nitrogen, phosphorus and iron system respectively [34], [17].