GEOCHEMISTRY OF GROUNDWATER: IN VIEW OF FLUORIDE AND NITRATE CONTAMINATION IN PARTS OF YAMUNA RIVER BASIN- INDIA

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Introduction

Fluoride and nitrate contamination is a major problem particularly in shallow alluvial aquifers and is increasingly becoming a threat to groundwater quality. High fluoride level in drinking water gives rise to mottled enamel of teeth, skeletal fluorosis and sometimes osteosulerosis whereas deficiency of fluoride cause over acidification of teeth and bones and dental caries (McClure, 1970). The enrichment of fluoride in groundwater is mainly due to the formation of alkaline soil and enrichment of sodium, bicarbonate and sulphates in shallow aquifers (Hem, 1989).

Ekhtesasi, 2004) About-2.5 million people living in 8700 villages in India suffer the problem of excess fluoride (Handa 1975; 1988, Nawalakhe and Bulusu, 1989) Karro et.al (2006) examined the content and spatial distribution of fluoride in drinking waters. Fordyce (2007) reported the fluoride is powerful calcium seeking element and can interfere with the calcified structure of bones and teeth in human body of higher concentration causing dental or skeletal flourosis.

Geology

The study area forms western part of Ghaziabad district of Uttar Pradesh and north-western part in Yamuna river basin. It is located in western Gangetic plain and lies between 28^{o}15’ and 28^{o} 35’ north latitudes and 77^{o} 15’ and 77^{o} 30’ east longitudes. It is highly industrialize, agriculturally developed and is drained by Yamuna River.

Major part of the area is covered by Quaternary alluvium and a small portion in north-western part is occupied by the Precambrian Alwar quartzites of Delhi Supergroup with NNE-SSW strike and easterly dips. The alluvial sediments comprise sequence of clay, silt, different grades of sand, gravel and kankar (calcareous nodules) in different proportions. The depth to water level varies between 3.20 and 21.50 meter below ground level.

Materials and Methods

A total of 30 groundwater samples from shallow hand pumps and deep tube-wells were collected from various locations from the entire area (Fig. 1) during pre-monsoon period (June 2011-12). The samples were analyzed for electrical conductivity, pH and major cations and anions including fluoride and nitrate using standard procedures of APHA (1992) and Trivedi and Goel (1984). Na, K were analyzed using Flame Photometer. F and NO_3 were analyzed or spectrophotometer, SO_4 was analyzed by gravimetric method E.C and pH were determined in the field by water analysis kit (NPC 360 D) and rest of the ions analyzed by titration methods in the geochemical laboratory of the Geology Department A.M.U., Aligarh and National Geophysical Laboratory(NGRI)Hyderabad.

Results and Discussion

Hydro chemical data of the groundwater samples are summarized in table 1. The summary of statistical parameters such as minimum, maximum, mean and standard deviation for the variables of groundwater is given in table 2. It is evident from chemical results and standard deviation that there is wide spatial variation in groundwater quality. Groundwater at most places is hard, alkaline and belongs to either Na-HCO_3 or Na-Cl facies. Hard water is generally having no harmful effects on humans. Cardiovascular diseases have been reported and appear to be confined more to the areas of soft water than to hard water (Crawford, 1972). Geo-statistics methods were used for studying spatial variability of groundwater (Meer, 1993, Nazari et.al, 2006).

The relative major ion concentrations observed in the area during the study period is generally of the order Na>K>Ca>Mg among the cations and $Cl>HCO_3>CO_3$ among the anions. The average values of major ions in different E.C. ranges, which generally show positive correlation with the electrical conductivity of groundwater. In general, higher concentrations of chemical constituents were observed in shallow aquifers. Nitrate, fluoride and chloride contents at some places were found above the maximum permissible limit prescribed by WHO (1993) for drinking water supply.

Fluoride Contamination

The natural environmental pollution due to fluoride in groundwater is a matter of great concern as its effects human health. The analytical results show high fluoride concentration at some places. The values range from 0.10 to 1.97 mg/L averaging at 0.93 mg/L. Potable water should have 0.6 to 1.5 mg/L of fluoride for substantial protection against dental caries and teeth decay (ICMR, 1975). Main sources of fluoride in groundwater are fluoride salts used in industries manufacturing steel, aluminium, bricks, glass, ceramics etc. Phosphate fertilizers extensively used in the area often contain fluoride as an impurity. These cause increase in fluoride contents of soils which on leaching result in enrichment of fluoride levels in groundwater. Also, fluoride bearing minerals such as apophyllite, phologopite, illite, hydromuscovite and biotite are generally found in clay dominating soil (Rai, 1996) and as the aquifers in the area comprise alternate beds of sand, gravel and clay, some quantity of fluoride in groundwater is contributed from hydration of the above minerals present in clay horizons.

Correlation of fluoride with Ca and HCO_3

Fluoride ion shows positive correlation with bicarbonate and negative correlation with calcium due to the following relationship;

$CaCO_3 + H^+ \rightarrow Ca^{2+} + HCO_3^-$ ------------------------------------ (i)

Applying the law of the chemical equilibrium, the equilibrium constant (K_1) is expressed as;

$$K_1 = \frac{[Ca^{2+}]\,[HCO_3]}{H^+} \qquad \text{------------- (ii)}$$

Similarly, from the dissociation reaction of fluoride (CaF_2) dissolved in water, the equilibrium constant (K_2) is expressed as;

$$K_2 = [Ca^{2+}]\,[F^-] \qquad \text{-- (iii)}$$

From equation (ii) and (iii)

$$\frac{K_1}{K_2} = \frac{[HCO_3^-]}{[H^+]\,[F^-]} \qquad \text{--- (iv)}$$

It is apparent from equation (iii) that an increase in fluoride ions would be accompanied by decrease in calcium ion would be accompanied by decrease in calcium ion resulting in negative correlation. From equation (iv), if hydrogen ion activity or Ph is constant, an increase in fluoride ion would be accompanied by corresponding increase in bicarbonate ion resulting in positive correlation.

Nitrate contamination

Chemical analysis of nitrate shows that its concentration at most places is within the permissible limit (45 mg/L, WHO, 1993) prescribed for drinking water and ranges from 1.8 to 86.6 mg/L. Higher concentrations exceeding 45 mg/L were recorded at some places due to localized pollution effect.

The important source of nitrate contamination is the on-site sewage disposal in septic systems (Aravena and Robertson, 1998). Decomposition of soil organic matter, leaching of soluble synthetic or chemical fertilizers (particularly N-fertilizer), human and animal excreta are the potential sources of nitrate in groundwater (Hill, 1982). The chief sources of groundwater nitrate in the area are sewage and animal wastes, industrial effluents, agricultural activities and septic tanks. Among the agricultural sources, the common sources are fertilizers and irrigation with waste water.

Conclusion

Chemical analysis of groundwater samples shows that the groundwater in major part of the area under study is saline and brakish, which is evident from higher E.C. and T.D.S. contents. Discharge of industrial effluents and soil wastes in the open field and utilization of waste water for irrigation are responsible for higher level of salts contents in the groundwater. During summer, the evaporation increases the concentration of ions in soils which on leaching results in the higher contents of salts thereby increasingly the salinity of groundwater. Sixty percent of the water samples have hardness between 150 and 350 mg/L and fall in moderately hard to hard category of water. In terms of hydro chemical facies, water of Na-HCO_3 type is dominant. The enrichment of fluoride in localized areas may cause adverse effect on human health. There is a large spatial variation in the occurrence of nitrate in groundwater. Relative higher contents of NO_3 in residential areas may be due to reasons other than agricultural activity, such as improper sanitary condition, leachate from animal waste, vegetable market waste, sewage and solid waste dump.

In order to improve the quality of water, the industrial effluents must be treated before the discharge, industrial wastes should not be utilized for irrigation and there should be well designed landfill for dumping urban wastes. Defluoridation techniques may be employed to maintain the level of F^- ion in water.

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Table 2. Summary of Statistical Parameters for the Variable of groundwater (n=65)

Variables	Minimum (mg/L)	Maximum (mg/L)	Mean	Standard deviation
Ph	5.50	8.80	7.15	0.65
E.C.(μS/cm)	500.00	6700.00	1993.80	1132.80
T.D.S	321.00	4295.00	1277.40	725.70
Ca^{2+}	6.00	130.30	43.87	28.40
Mg^{2+}	4.90	132.80	36.99	23.60
Na^{+}	6.00	732.00	179.49	167.60
K^{+}	9.00	558.00	44.62	85.60
CO_3^{2-}	8.00	165.00	39.60	------
HCO_3^{-}	81.00	495.00	219.30	86.90
Cl-	26.00	1267.00	244.01	194.10
Hardness	95.00	760.00	257.77	88.30
F-	0.01	1.97	0.92	0.54
SO_4^{2-}	32.00	518.00	241.74	110.90
NO_3^{-}	1.80	86.80	19.93	17.00