



Hydro-chemical characterization of groundwater and evaluation of health risk assessment for fluoride contamination areas in the eastern blocks of Purulia district, India

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Abstract

Purulia is a hard rock terrain where water scarcity as well as water quality degradation has been a major threat for the past few decades. The prolong use of fluoride contaminated groundwater causes serious health issues in human. The study was performed during the post-monsoon period for better understanding of the general geochemistry along with the fluoride contamination of groundwater and analysis of the health risk factor for the local dwellers. The physico-chemical parameters were analysed during the field work and rest in the laboratory using the standard procedures. The statistical mean values of the cations Ca^{+2} ; Na^{+} ; Mg^{+2} ; K^{+} ; and Fe^{+2} are 91.53, 42.3, 31.76, 3.58 and 0.93 mg/l and for anions HCO_3^{-} , Cl^{-} , SO_4^{-} , NO_3^{-} and F^{-} are 231.67, 106.81, 82.83, 31.52 and 4.06 mg/l, respectively. Fluoride is one of the important trace elements in groundwater, and the value ranges from 1.30 to 7 mg/l with an average of 4.06 mg/l in the study region. According to the piper plot, the water type is 80% Ca– HCO_3 , 19% mixed CaMgCl and 1% CaCl type. Gibbs diagram indicates that the rock–water interaction is the most dominating mechanism prevailing in this region. The health risk assessment is revealed based upon the values of hazard quotient for ingestion (HQ_{in}) and dermal pathway (HQ_{de}) for the different age groups. The results from the research show that 6–12 months babies are more exposed to health risk through direct consumption of contaminated water in the study region.

Keywords Groundwater chemistry · Fluoride contamination · Gibbs diagram · Chloro-alkaline indices · Health risk assessment

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1 Introduction

Groundwater occurs beneath the earth surface almost everywhere either connected to one single or many other local aquifer systems possessing similar characteristics (Vasanthavigar et al., 2010). Extraction of large quantity of groundwater has led to severe water crisis in different parts of the world (Asoka et al., 2017; Das, 2019; Rodell et al., 2009; Zolekar et al., 2021). Degradation of groundwater level has its negative impact on groundwater quality also (Bera et al., 2020, 2021; Biswas et al., 2020). Groundwater quality has been investigated by many researchers (Abdul-Wahab et al., 2020; Chakraborty et al., 2021; Chitsazan et al., 2019; Gaikwad et al., 2020; Islam et al., 2018; Jalees et al., 2020; Kalawapudi et al., 2019; Singaraja et al., 2016; Thivya et al., 2013). The contamination of groundwater is a major issue nowadays occurring both in rural and urban areas with large number of potential sources (Jayapraskh et al. 2008; Vasanthavigar et al., 2010). Fluoride ion occurs in all waters as a trace element mostly (Gaciri et al. 1993; Farooq 2018). It is an incompatible lithophile element and electronegative in nature (Faure, 1991). The drinking water which contains high concentrations of fluoride shows adverse effect on human health that has been reported from many different terrains of the world (Adimalla & Qian, 2019; Emenika et al., 2017; Narsimha & Sudarshan, 2018a, 2018b; Shaji et al., 2007). The impact of fluoride toxicity in human health depends mainly on its concentration in drinking water and the period for which the water has been exposed to the body surface in the different age groups independently (Dregne, 1967; Viswanthan et al., 2009). Frencken (1992) reported that the fluoride concentration ranges from 100 to 1000 mg/l of water in different rocks types—igneous, metamorphic and sedimentary rocks. International regions of Africa, Syria, Jordan, Sudan, Kenya, Uganda, Mexico, Turkey, China, Korea, India are highly been affected by fluorosis (Grech, 1966; Tekle-Haimanot et al., 1987; Gizaw, 1996; Carrilo-Rivera et al., 2002; Oruc, 2003; Guo et al., 2007; Chen et al., 2017; Adimalla et al. 2018b; Adimalla, 2019; Adimalla & Li, 2019).

From the view of geochemistry, it has been observed that certain minerals contain fluoride in their structures which are apatite $[\text{Ca}_5(\text{PO}_4)_3\text{F}]$, fluorite $[\text{CaF}_2]$, apophyllite, topaz $[\text{Al}_2\text{F}_2(\text{SiO}_4)]$, sphene, cryolite $[\text{Na}_3\text{AlF}_6]$, villiaumite $[\text{NaF}]$, some silicates such as amphiboles like hornblende $[(\text{Ca},\text{Na})_{2-3}(\text{Mg},\text{Fe},\text{Al})_5(\text{Al},\text{Si})_8\text{O}_{22}(\text{OH},\text{F})_2]$ and clay minerals like muscovite, biotite, kaolinite $[\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4]$, vermiculite $[(\text{Mg},\text{Fe},\text{Al})_3(\text{Al},\text{Si})_4\text{O}_{10}(\text{OH})]$, montmorillonite (Subba Adimalla & Qian, 2019; Chae et al., 2006; Rao & Devadas, 2003; Sivasankar et al., 2016). The occurrence of fluoride in groundwater is mostly due to geological sources with lower concentrations of anthropogenic sources either from agricultural fertilizers or industries sewages (Das et al., 2016; Kumar et al., 2016, 2020; Singh et al., 2020). Fluoride concentration depends mainly upon factors such as temperature, pH, cationic–anionic exchange with the aquifer materials, characteristics of the geological formations, solubility of minerals possessing fluoride in their structure and dissolution of those minerals through some geochemical processes (Adimalla & Qian, 2019; Raju et al., 2009). Naseen et al. (2010) reported that granite acts as a good source of fluoride in many Archean hard rock terrains, where during weathering processes granite leaches to form kaolin and other clay minerals which enriches the fluoride concentration in the groundwater.

In India during early 1930s, only 4 states were identified with higher fluoride concentration in the waters leading to endemic fluorosis, but now at present nearly 21 states are highly been affected (Adimalla et al., 2018a, 2018b, 2018c; Narsimha & Rajitha, 2018). An estimation of about 60 million people which includes children of about 6 million suffers from dental and skeletal fluorosis due to the consumption of fluoride rich water (Raju

et al., 2009). The problem of fluorosis is increasing day by day throughout India within a very short span of time, and it is leading to an alarming health risk factor in the country (Adimalla & Venkatayogi, 2017; Adimalla et al., 2018a, 2018b, 2018c; Ayoob & Gupta, 2006). In West Bengal, 43 blocks in 7 districts are highly affected by higher fluoride concentration (> 1.5 mg/l) (Mandal & Sanyal, 2019). According to WBPHEd 2006, the rural population of the state are at high risk consisting nearly 7.41 million which is about 12% of the total rural populations. Chakrabarti and Ray (2013) reported that over 65 blocks from 8 districts are at high risk due to higher fluoride concentrations. Purulia is a drought-prone arid to semi-arid region of Archean granitic terrain which is affected by the endemic fluorosis. According to Chakraborti and Ray (2013), the fluoride value in the groundwater varied from 0.126 to 8.16 ppm over the blocks of Hura, Purulia 1 and Purulia 2. The exact number of people and the age groups mostly facing the risk of fluoride is unknown due to lack of systematic survey and understanding of the spatial distribution over Purulia (Jha et al., 2013).

Researches related to the fluoride contaminated water and the health risk on humans in Purulia have been done previously by some researchers. Mondal et al. (2013) studied the different water quality parameters and stated the fluoride contaminated zones in Purulia district. Samal et al. (2015) stated about the fluoride intake through the food chain in adults and infants and also the effect of elevated fluoride in the soil structure. Bhattacharya (2016) discussed about the concentration of fluoride in water and soil in the six different blocks (Hura, Pancha, Kashipur, Santuri, Raghunathpur-I and Manbazar-I) of Purulia. Farooq et al. (2018) discussed about the spatial distribution of fluoride concentrations in the groundwater of Purulia-I and Purulia-II blocks. Mandal and Sanyal (2019) with the help of GIS techniques studied the spatial distribution of fluoride concentrations in the waters of Purulia. Researches related to the use of the health risk model for fluoride contaminated water and its effects of on the different age groups of humans in detailed manner have not been performed earlier in Purulia.

The main objectives of this research are to understand the general groundwater quality based upon the different physical, chemical parameters and identification of the potential sources that leads to the higher proportions of different cations–anions in water. Secondly, identification of the fluoride vulnerable areas and assessment of the health risk factor for the different age groups- babies, young children, teenagers, adults and aged people both by consumption and dermal contact pathways. The results obtained from this study would further help the policy makers and the planners for preparing groundwater management programs and scientific techniques to improve the health of the local dwellers in the coming future.

2 Study area

The study area is situated in the eastern and the north-eastern bocks of Purulia district which has an area of about 1545 km². The area lies within the latitudes of 23°31'44'' N and 23°39'47'' N and the longitudes between 86°32'57'' E and 86°51'50'' E. Purulia is a hard rock terrain over which the tropic of cancer passes. It shows semi-arid tropical monsoonal climate. The area is a drought-prone region of West Bengal which is a semi-arid in nature (Archarya & Nag, 2013). The selected study area consists of Para, Kashipur, Raghunathpur-I, Raghunathpur II, Santuri and Neturia blocks of Purulia (Fig. 1). The climate in Purulia varies from hot, dry summer to cold winters with scanty rainfall. The average

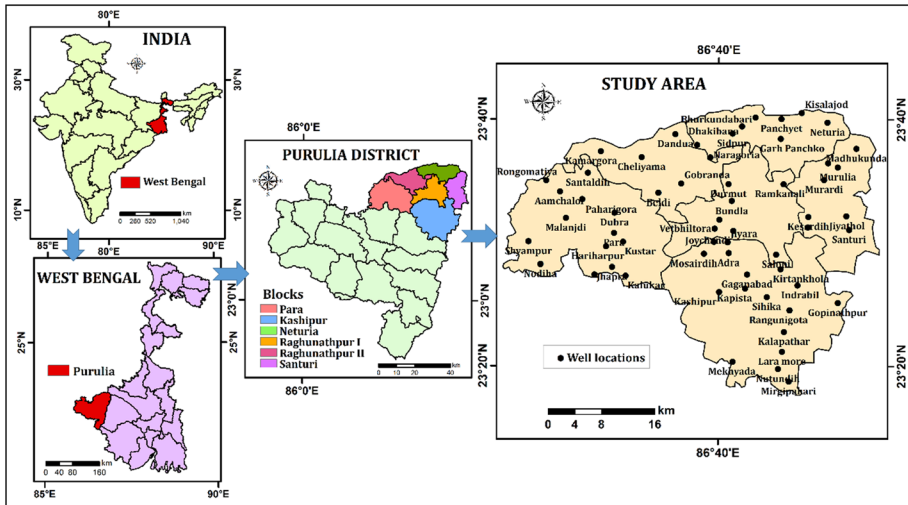


Fig. 1 Location map of the study area

temperature of the area ranged from 7 °C during winter to 38 °C in summers (Fig. 2). The annual rainfall varies from 1100 to 1500 mm. The area mainly consists of the Pre-Cambrian highly metamorphosed rocks which act as the basement rocks and a small portion of Gondwana sedimentary rocks in the north-eastern parts of the district. The metamorphic rocks mainly include the granite gneiss, biotite granite gneiss, calc-granulite, ultrabasics, meta-basics, meta-sedimentary rocks, pegmatites and quartz veins (Baidya, 1992). Generally, the E-W trending strike of the formations is predominant with moderate to steep northerly and southerly dipping beds (Acharya et al., 2014). Aquifers are recharged during the monsoon through precipitation. According to CGWB, the modes of occurrence of groundwater in Purulia are: (i) weathered mantle and (ii) fractured zone of the hard rock. Near the lithological boundaries of biotite gneiss, phyllites, micaceous bodies and phyllites

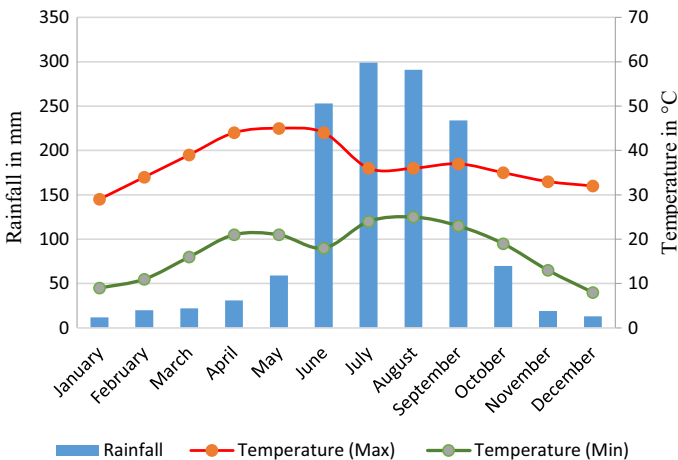


Fig. 2 Temporal distribution of rainfall and temperature of the study area

exist higher permeability zones as small pockets (Saha, 1997). According to the total number of population for the 6 blocks of Purulia under study is 812196 and the population density is 525/km². The population density in the Para block is comparatively high (642/km²), and Santuri block shows comparatively low population density (437/km²). Purulia is one of the economically backward districts of West Bengal. Agriculture is the main occupation for the majority of the population. The rivers in this region are mainly rainfed. Therefore, during the monsoon seasons the water levels are quite high in the rivers, whereas during the dry seasons the rivers mostly dry out and the farmers have to depend on the reservoirs and the groundwater irrigation for farming. Water scarcity is still a major threat in the land of Purulia during the summer seasons. Even after 73 years of independence, the people of Purulia still have to struggle for the basic amenities of life which are food and water.

3 Materials and methods

Groundwater level determines the water status in an area. The water level fluctuates with season as it depends mainly on the availability of rainfall. The investigation was carried out in the month of December 2019 for the post-monsoon period. Depth to water level was measured for 60 cylindrical fitted tube wells around different villages in the blocks of Para, Kashipur, Santuri, Neturia, Raghunathpur-I and Raghunathpur-II (Fig. 3). The physical parameters like the electrical conductivity (EC), pH, temperature, total dissolved solids (TDS) are measured for each sample using Hanna Multi-parameter waterproof meter (HI98194) in the field itself. The total alkalinity for each sample was measured in the field using AQUASOL Alkalinity kit (AE-214). The samples of water were collected from the 60 tube wells in 500 ml bottles for testing in the laboratory. The water was collected in fresh bottles after pumping it for 7–8 min, so that the water stored in the casing is lost and we get the fresh sample from the aquifer itself. The samples were analysed by standardized methods for different cationic and anionic concentrations which are Mg²⁺, Na⁺, K⁺, Ca²⁺, HCO₃⁻ + CO₃⁻, Cl⁻, SO₄⁻², NO₃⁻, Fe^{+2/+3}, F⁻, pH, EC, TDS and temperature using different instruments. Sodium and potassium are measured by digital flame photometer (Systronics 128), chloride by argentometric method, calcium and magnesium by titrimetric method. Iron is estimated by using Move-100 with Spectroquant reagents. Fluoride (F⁻) concentration in groundwater is detected using the ion-selective electrode method (APHA, 1995). The different cationic and anionic values and the physical parameter values are given in Table 1 for the study area. The analysed geochemical data are plotted in the Hill Piper diagram (Piper, 1944) which helps in understanding the groundwater facies. The analysed values of the above-mentioned parameters were compared with standardized values recommended by WHO (2011) guidelines. Gibbs diagram (Schoeller, 1967) for the analysed samples was plotted to determine the mechanism or the processes which controls the geochemistry of the water.

For validating the water quality, charge balance error (CBE) is calculated for the samples using the following equation

$$\%CBE = \frac{\Sigma Cations - \Sigma Anions}{\Sigma Cations + \Sigma Anions} \times 100 \quad (1)$$

Here, all the cationic and anionic values are expressed in meq/l. The CBE for all the collected samples is found to be within the acceptable limit, i.e. within $\pm 10\%$.

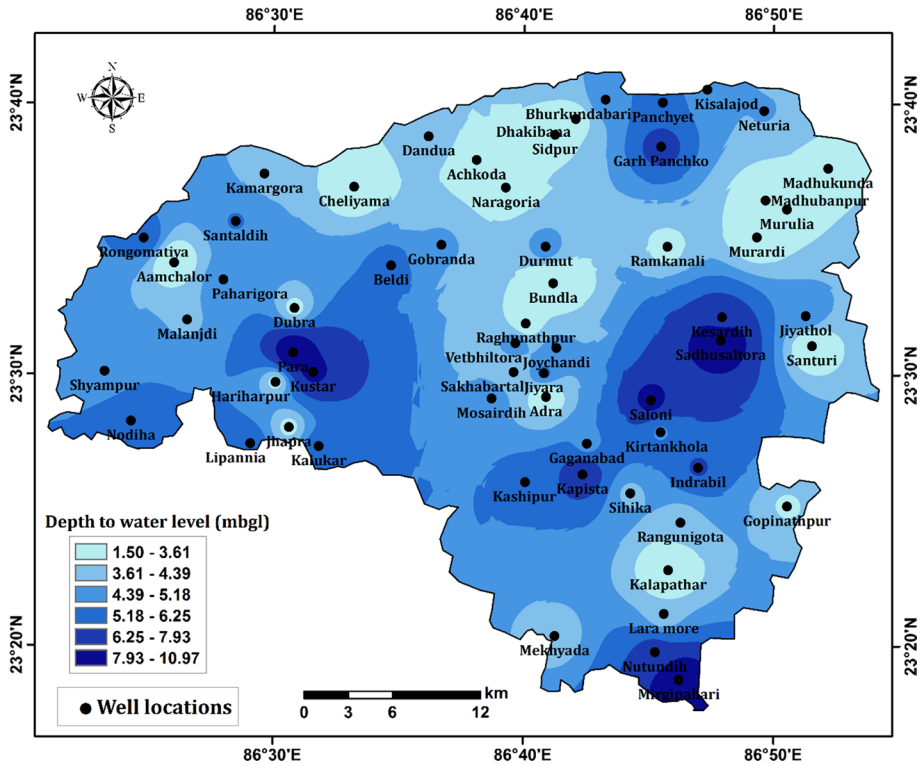


Fig. 3 Spatial distribution of depth to water level in the study area

3.1 Health risk assessment

Health risk assessment model is utilized for understanding the impact of higher or lower concentration of different ions on the human health (Adimalla & Qian, 2019; Adimalla et al., 2018a, 2018b, 2018c). The United States Environmental Agency (US EPA, 1989 and Moya et.al, 2011) has introduced the health risk model for evaluating the potential impact of harmful chemicals and its risk on different age groups of mankind through different pathways either directly or indirectly. The calculation of the hazard quotient (HQ) for the 60 different locations on the different age groups: 6–12 months, 1–5 years, 5–10 years, 10–15 years, 15–18 years, 18–21 years, 21–65 years, >65 years, was performed to get a detailed evaluation. In this model, the assessment is computed based on the results associated with the hazard quotient (HQ) for the different age groups both by direct consumption, i.e. ingestion and dermal contact pathway using Eqs. (2)–(5):

$$CDD_{in} = \frac{C_{fw} \times IR_w \times EF_r \times ED}{BW \times AT} \tag{2}$$

$$CDD_{de} = \frac{C_{fw} \times EF_r \times K \times ESA \times ED \times CF}{BW \times AT} \tag{3}$$

Table 1 Physico-chemical parameters of groundwater samples in the study area (EC in $\mu\text{S/cm}$, remaining mg/l, except pH)

| Location | pH | EC | TDS | TH | $\text{HCO}_3 + \text{CO}_3$ | Cl^- | SO_4^{2-} | NO_3^- | F^- | Ca^{+2} | Na^+ | Mg^{+2} | K^+ | Fe^{+2} |
|---------------|------|------|--------|--------|------------------------------|---------------|--------------------|-----------------|--------------|------------------|---------------|------------------|--------------|------------------|
| Mekhyada | 6.3 | 512 | 327.68 | 272.84 | 300 | 85.08 | 30 | 50 | 5 | 42 | 22.62 | 40.82 | 1.94 | 0.5 |
| Mirgipahari | 6.9 | 1000 | 640 | 431.62 | 300 | 170.16 | 150 | 75 | 5 | 119.2 | 46.15 | 32.56 | 3.26 | 0.4 |
| Kashipur | 6.6 | 705 | 451.2 | 245.81 | 400 | 99.26 | 70 | 0.5 | 4.5 | 65.6 | 54.01 | 19.93 | 2.69 | 2 |
| Kapista | 7 | 292 | 186.88 | 173.91 | 200 | 42.54 | 50 | 0.5 | 3 | 30 | 24.7 | 24.06 | 1.11 | 1 |
| Gaganabad | 7.6 | 699 | 447.36 | 333.75 | 300 | 56.72 | 40 | 1 | 4 | 83.2 | 31.3 | 30.62 | 2.84 | 0.3 |
| Kirtankhola | 7.1 | 406 | 259.84 | 737.76 | 200 | 28.36 | 20 | 1.5 | 4.5 | 67.2 | 15.91 | 138.51 | 0.75 | 0.3 |
| Indrabil | 6.9 | 1200 | 768 | 458.63 | 200 | 198.52 | 120 | 70 | 4 | 122.8 | 75.93 | 36.94 | 2.64 | 0.3 |
| Sihika | 7.1 | 1177 | 753.28 | 506.56 | 400 | 148.89 | 120 | 1.5 | 3.2 | 140 | 75.73 | 38.15 | 3.24 | 2 |
| Rangunigota | 7.3 | 759 | 485.76 | 383.74 | 400 | 42.54 | 20 | 2.5 | 2 | 83.2 | 44.5 | 42.77 | 1.99 | 0.4 |
| Kalapathar | 7.2 | 658 | 421.12 | 236.78 | 200 | 92.17 | 90 | 1 | 5 | 68 | 52.84 | 16.28 | 2.2 | 1.5 |
| Lara more | 7 | 1940 | 1241.6 | 877.32 | 400 | 340.32 | 130 | 50 | 5.2 | 226.4 | 76.9 | 75.82 | 3.33 | 0.3 |
| Kustar | 6.9 | 1401 | 896.64 | 681.52 | 300 | 205.61 | 100 | 60 | 5 | 158.8 | 40.23 | 69.26 | 7.89 | 0.5 |
| Beldi | 6.9 | 414 | 264.96 | 245.80 | 300 | 35.45 | 20 | 2.5 | 6 | 60.8 | 20.75 | 22.84 | 1.55 | 0.3 |
| Nutumdh | 7.02 | 364 | 232.96 | 224.90 | 200 | 35.45 | 30 | 2.2 | 1.4 | 33.2 | 17.36 | 34.51 | 8.26 | 1.5 |
| Ramkanali | 6.3 | 1680 | 1075.2 | 423.65 | 200 | 177.25 | 130 | 50 | 5.7 | 108 | 67.06 | 37.42 | 6.24 | 0.4 |
| Garh panchkot | 6.6 | 557 | 356.48 | 307.71 | 200 | 35.45 | 20 | 1 | 2.5 | 88 | 9.3 | 21.38 | 3.08 | 2 |
| Panchyet | 7.07 | 550 | 352 | 309.74 | 200 | 28.36 | 40 | 2.5 | 1.5 | 88.4 | 17.64 | 21.63 | 1.9 | 1.5 |
| Kisalajod | 7.38 | 829 | 530.56 | 325.75 | 200 | 56.72 | 90 | 2.5 | 1.4 | 76.8 | 67.52 | 32.56 | 1.72 | 0.05 |
| Neturia | 7.4 | 1000 | 640 | 269.80 | 200 | 48.63 | 120 | 7.5 | 6 | 68.4 | 27.06 | 24.06 | 3.61 | 0.05 |
| Madhukunda | 6.86 | 977 | 625.28 | 443.61 | 300 | 163.07 | 180 | 1 | 5.5 | 128.4 | 103.68 | 29.89 | 3.03 | 6 |
| Murulia | 7.15 | 1473 | 942.72 | 362.68 | 300 | 70.9 | 600 | 6.5 | 5 | 106.8 | 30.1 | 23.33 | 1.18 | 0.4 |
| Madhubanpur | 6.83 | 1500 | 960 | 759.31 | 200 | 248.15 | 140 | 80 | 7 | 231.2 | 87.2 | 44.23 | 17.06 | 0.3 |
| Murardi | 6.85 | 1406 | 899.84 | 301.77 | 300 | 92.17 | 110 | 70 | 2.9 | 72.8 | 45.17 | 29.16 | 2.49 | 1 |
| Jiyathol | 6.8 | 490 | 313.6 | 140.93 | 100 | 92.17 | 80 | 2.5 | 1.5 | 18.8 | 26.06 | 22.84 | 0.63 | 2 |
| Santuri | 7.38 | 864 | 552.96 | 238.85 | 200 | 42.54 | 40 | 1 | 1.4 | 49.2 | 17.23 | 28.19 | 1.98 | 3 |
| Saloni | 6.95 | 789 | 504.96 | 682.35 | 400 | 361.59 | 140 | 50 | 3.5 | 219.2 | 107.39 | 32.81 | 7.3 | 0.3 |
| Kesardih | 7.3 | 595 | 380.8 | 144.93 | 200 | 35.45 | 40 | 40 | 3 | 28 | 15.74 | 18.23 | 2.27 | 0.1 |

Table 1 (continued)

| Location | pH | EC | TDS | TH | HCO ₃ + CO ₃ ⁻ | Cl ⁻ | SO ₄ ²⁻ | NO ₃ ⁻ | F ⁻ | Ca ⁺² | Na ⁺ | Mg ⁺² | K ⁺ | Fe ⁺² |
|---------------|------|------|---------|---------|-------------------------------------------------|-----------------|-------------------------------|------------------------------|----------------|------------------|-----------------|------------------|----------------|------------------|
| Sadhusaltora | 7.2 | 944 | 604.16 | 228.83 | 180 | 56.72 | 60 | 50 | 6.8 | 50 | 23.14 | 25.27 | 1.35 | 0.4 |
| Kalukar | 7.3 | 241 | 154.24 | 114.93 | 200 | 28.36 | 30 | 50 | 4 | 26.4 | 18.67 | 11.91 | 1.6 | 0.7 |
| Nodaha | 7.1 | 704 | 450.56 | 354.70 | 200 | 85.08 | 50 | 60 | 1.5 | 98 | 32.35 | 26.73 | 3.25 | 1 |
| Malanjdi | 6.5 | 941 | 602.24 | 447.58 | 200 | 177.25 | 70 | 60 | 6 | 132 | 36.21 | 28.67 | 2.79 | 1 |
| Aamchalor | 7 | 370 | 236.8 | 191.87 | 200 | 85.08 | 20 | 50 | 5 | 45.2 | 16.12 | 19.2 | 1.16 | 0.3 |
| Lipannia | 7.18 | 1491 | 954.24 | 339.76 | 200 | 106.35 | 110 | 50 | 5.4 | 80.4 | 50.13 | 33.78 | 2.17 | 1 |
| Rongomatiya | 6.9 | 457 | 292.48 | 215.83 | 200 | 163.07 | 30 | 50 | 4 | 51.6 | 34 | 21.14 | 1.83 | 0.5 |
| Para | 6.7 | 816 | 522.24 | 294.95 | 200 | 113.44 | 50 | 50 | 4.7 | 96.8 | 61.22 | 12.94 | 1.53 | 2 |
| Hariharpur | 6.8 | 371 | 237.44 | 187.83 | 100 | 56.72 | 40 | 60 | 4 | 54 | 25.87 | 12.88 | 1.08 | 1 |
| Dubra | 7.1 | 1636 | 1047.04 | 1273.76 | 300 | 432.49 | 120 | 75 | 5 | 406 | 82.36 | 63.18 | 10.37 | 2 |
| Paharigora | 7.1 | 750 | 480 | 374.74 | 300 | 77.99 | 70 | 50 | 4 | 88 | 45.82 | 37.67 | 3.14 | 0.5 |
| Shyampur | 6.4 | 478 | 305.92 | 222.83 | 100 | 77.99 | 80 | 2.5 | 2.5 | 53.2 | 39.97 | 21.87 | 1.12 | 1.5 |
| Santalidih | 7.3 | 861 | 551.04 | 343.73 | 200 | 92.17 | 80 | 7.5 | 4 | 87.2 | 69.17 | 30.62 | 9.72 | 0.4 |
| Kamargora | 6.9 | 1577 | 1009.28 | 764.23 | 400 | 212.7 | 160 | 75 | 4.5 | 251.2 | 49.8 | 33.29 | 3.49 | 1 |
| Cheliyama | 7 | 1247 | 798.08 | 503.60 | 300 | 148.89 | 130 | 65 | 6 | 131.2 | 77.36 | 42.77 | 39.74 | 0.3 |
| Dandua | 7.1 | 486 | 311.04 | 205.83 | 100 | 49.63 | 80 | 50 | 3.8 | 47.2 | 36.57 | 21.38 | 1.7 | 0.3 |
| Gobanda | 6.7 | 538 | 344.32 | 164.91 | 100 | 113.44 | 80 | 40 | 6 | 28.8 | 17.73 | 22.6 | 3.55 | 0.7 |
| Raghunathpur | 7.09 | 153 | 97.92 | 256.79 | 20 | 70.9 | 110 | 40 | 7 | 68.4 | 37.7 | 20.9 | 1.3 | 1.5 |
| Bundla | 7.04 | 1023 | 654.72 | 278.74 | 200 | 56.72 | 40 | 50 | 4.8 | 82 | 15.61 | 17.98 | 3.9 | 0.3 |
| Durmut | 6.34 | 342 | 218.88 | 195.88 | 200 | 42.54 | 40 | 10 | 1.4 | 35.6 | 13 | 26 | 2.39 | 1.5 |
| Naragoria | 6.95 | 179 | 114.56 | 85.93 | 100 | 21.27 | 30 | 2 | 3.5 | 16.8 | 14.9 | 10.69 | 1.03 | 0.5 |
| Achkoda | 6.7 | 216 | 138.24 | 131.91 | 100 | 28.36 | 20 | 30 | 5.5 | 24.4 | 15.57 | 17.25 | 1.48 | 0.4 |
| Sidpur | 6.8 | 475 | 304 | 210.84 | 200 | 49.63 | 50 | 20 | 4.8 | 52.8 | 31.25 | 19.2 | 2.73 | 1 |
| Bhurkundabari | 7.13 | 623 | 398.72 | 281.80 | 400 | 28.36 | 20 | 5 | 5.8 | 66.4 | 50.77 | 28.19 | 4.09 | 0.3 |
| Dhakibana | 6.3 | 238 | 152.32 | 87.96 | 200 | 14.18 | 40 | 50 | 4 | 12.8 | 8.02 | 13.61 | 1.51 | 1 |

Table 1 (continued)

| Location | pH | EC | TDS | TH | HCO ₃ ⁻ + CO ₃ ⁻ | Cl ⁻ | SO ₄ ²⁻ | NO ₃ ⁻ | F ⁻ | Ca ⁺² | Na ⁺ | Mg ⁺² | K ⁺ | Fe ⁺² |
|--------------|------|------|---------|--------|--------------------------------------------------------------|-----------------|-------------------------------|------------------------------|----------------|------------------|-----------------|------------------|----------------|------------------|
| Vet bhiltora | 6.47 | 1406 | 899.84 | 806.39 | 300 | 283.6 | 110 | 50 | 6.7 | 191.6 | 60.34 | 79.7 | 1.56 | 1 |
| Adra | 7.04 | 1351 | 864.64 | 541.51 | 300 | 163.07 | 160 | 50 | 2.8 | 153.6 | 52.53 | 38.39 | 1.28 | 0.3 |
| Joychandi | 7.2 | 1000 | 640 | 256.84 | 200 | 49.63 | 40 | 70 | 1.4 | 45.2 | 31.62 | 34.99 | 1.48 | 0.5 |
| Sakhabartal | 7.5 | 604 | 386.56 | 190.88 | 100 | 28.36 | 50 | 0.5 | 4.5 | 43.6 | 14.36 | 19.93 | 0.68 | 2.5 |
| Mosairdih | 6.94 | 1306 | 835.84 | 295.74 | 200 | 70.9 | 50 | 30 | 1.3 | 85.2 | 46.67 | 20.17 | 1.39 | 1 |
| Jiyara | 7.15 | 874 | 559.36 | 215.82 | 200 | 35.45 | 40 | 5 | 1.4 | 61.6 | 27.05 | 15.07 | 1.09 | 1 |
| Jhapra | 6.95 | 1663 | 1064.32 | 806.40 | 400 | 311.96 | 150 | 0.5 | 4 | 198 | 86.09 | 75.82 | 2.27 | 0.7 |
| Gopinathpur | 7.3 | 945 | 604.8 | 160.88 | 200 | 42.54 | 40 | 0.5 | 2.5 | 43.6 | 29.36 | 12.64 | 1.02 | 2.5 |

$$HQ_{in} = \frac{CDD_{in}}{RfD} \quad (4)$$

$$HQ_{de} = \frac{CDD_{de}}{RfD} \quad (5)$$

where CDD_{in} —chronic dose via ingestion pathway in daily basis (mg/kg/day); CDD_{de} —chronic daily dose via dermal exposure pathway (mg/kg/day); C_{fw} —concentration of fluoride in drinking water (mg/l); EF_r —frequency of exposure (days/years); ED —exposure duration (years); BW —body weight (kg); AT —resident time (days/years); ESA —exposed skin area (cm^2); K —skin adherence factor; CF —conversion factor (l/cm^3); and RfD —reference dose of fluoride (0.06 mg/kg/day) taken from the International Risk Information System (IRIS) and (US EPA 1989). The detailed value of the parameters used for the calculation is given in Table 2 (US EPA 1989; Chen et al., 2017; Adimalla Li & Qian 2018; Emenike et al., 2018; Adimalla & Qian, 2019).

4 Results and discussion

4.1 Quality based on the general parameters

The basic physio-chemical parameters for each location are given in Table 1 which are analysed individually and systematically for understanding the general water quality. The analysed data are compared with the standard ranges of WHO (2011) to assess the water quality and its suitability for drinking purpose.

pH acts as an indicator of water strength. It also determines the capacity of water in reacting with the acidic and alkaline materials present within. The pH value ranges from 6.3 to 7.5 with an average value of 6.96. Most of the water samples are neutral to weakly alkaline in nature. According to WHO (2011), the acceptable range of pH varied between 6.5 and 7.5. All the samples show that the values are within the acceptable limits for drinking purpose.

TDS ranges from 97.92 to 1241.60 mg/l with a mean of 539.12 mg/l in the study area. According WHO (2011), the highest permissible limit is 500 mg/l, but 1500 mg/l is accepted as the maximum extended desirable limits for drinking purpose. 51.67% of the samples are within 500 mg/l, and 48.33% are within the maximum desirable limits. Todd (1980) classified the groundwater as freshwater when the TDS value is less than 1000 mg/l; brackish water when TDS value ranged from 1000 to 10,000 mg/l; saline water when TDS ranged from 10,000 mg/l to 1,000,000 mg/l and lastly as brine water when TDS ranged greater than 1,000,000 mg/l. In the study area, 80% of the samples are fresh and 20% are brackish in nature.

Electrical conductivity (EC) is the electric current conveyed by the water. EC values ranged from 153 to 1940 $\mu s/cm$ with an average value of 834.38 $\mu s/cm$. EC can be classed into three types—type I (low) when the EC is less than 1500 $\mu s/cm$; type II (moderate) where EC ranges from 1500 to 3000 $\mu s/cm$; and lastly as type III (high) where EC value is greater than 3000 $\mu s/cm$ (Sarith Logeshkumaran et al., 2015; Prasanth et al., 2012). All the samples are within the WHO desirable limits of 1500 $\mu s/cm$. Higher EC values in the groundwater are an indication of rock–water interaction mechanism dominance.

Table 2 Different values of the parameters used for the health risk assessment

| Parameters | Units | 6–12 months | 1–5 years | 5–10 years | 10–15 years | 15–18 years | 18–21 years | 21–65 years | > 65 years |
|---------------------------------------------|-------------------|---------------|-----------|------------|-------------|-------------|-------------|-------------|------------|
| <i>Ingestion pathway</i> | | | | | | | | | |
| Ingestion rate (IR _w) | L/day | 1 | 1.16 | 1.32 | 1.82 | 1.78 | 2.34 | 2.94 | 2.73 |
| Exposure frequency (E _f) | Days/year | 365 | 365 | 365 | 365 | 365 | 365 | 365 | 365 |
| Exposure duration (E _D) | Year | 6 | 6 | 6 | 6 | 6 | 30 | 30 | 30 |
| Body weight (BW) | kg | 9.1 | 16.6 | 29.3 | 54.2 | 67.6 | 67.6 | 78.8 | 80 |
| Average time (A _{ti}) | Days | 2190 | 2190 | 2190 | 2190 | 2190 | 10,950 | 10,950 | 10,950 |
| Concentration of element (C _{fw}) | mg/l | Present study | | | | | | | |
| <i>Dermal pathway</i> | | | | | | | | | |
| Skin surface area (SA) | cm ² | 4500 | 6800 | 10,500 | 15,700 | 18,000 | 19,550 | 19,800 | 19,400 |
| Exposure time (E _T) | h/event | 0.54 | 0.54 | 0.54 | 0.54 | 0.54 | 0.71 | 0.71 | 0.71 |
| Exposure frequency (E _f) | Days/year | 350 | 350 | 350 | 350 | 350 | 350 | 350 | 350 |
| Exposure duration | Year | 6 | 6 | 6 | 6 | 6 | 30 | 30 | 30 |
| Conversion factor (CF) | L/cm ³ | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 |
| Body weight (BW) | kg | 9.1 | 16.6 | 29.3 | 54.2 | 67.6 | 67.6 | 78.8 | 80 |
| Average time (A _{ti}) | Days | 2190 | 2190 | 2190 | 2190 | 2190 | 10,950 | 10,950 | 10,950 |
| Skin adherence factor (K _p) | cm/h | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 | 0.001 |
| Concentration of element (C _{fw}) | mg/l | Present study | | | | | | | |

Total hardness (TH) ranged from 87.96 to 1273.76 mg/l with a mean value of 359.27 mg/l. The most desirable limit is 100 mg/l, and the maximum allowable limit is 500 mg/l as given by WHO (2011). 80% of the samples are within the maximum allowable limits, while rest 20% show very high hardness value which is within 1500 mg/l. The location Dubra shows the highest value of total hardness where the Ca and Mg concentrations are very high. The higher concentration of total hardness can cause scaling in the pots and boilers used in different industries; clogging in the irrigation pipes and sometimes causes health problems like kidney failure (Vasanthavigar et al., 2010). The total hardness is determined using the following equation (Todd, 1980);

$$\text{TH}(\text{CaCO}_3)\text{mg/l} = (2.497)\text{Ca} + (4.115)\text{Mg} \quad (6)$$

According to the mean value of the cationic and anionic concentrations in the collected samples from the study area are arranged in the following decreasing order of occurrences- for cations $\text{Ca}^{+2} > \text{Na}^+ > \text{Mg}^{+2} > \text{K}^+ > \text{Fe}^{+2}$ and anions as $\text{HCO}_3^- > \text{Cl}^- > \text{SO}_4^- > \text{NO}_3^- > \text{F}^-$.

The Ca^{+2} value ranges from 16.8 to 251.2 mg/l with a mean value of 91.53 mg/l in the study area. 10% of the samples show very high concentrations of Ca^{+2} exceeding the limit of 200 mg/l given by WHO (2011). The rest remaining 90% samples are within maximum allowable limits. According to WHO, the most desirable limit for calcium is within 75 mg/l and 58% of the samples are within the desirable ranges; rest falls within the maximum allowable limits. The higher concentration of Ca^{+2} can be predicted due to the lithology which is mostly granitic gneiss and amphibolite schist.

The Na^+ concentration ranges from 9.3 to 107.39 mg/l with an average of 42.3 mg/l which are within the maximum allowable limit of 200 mg/l for the drinking purposes as given by WHO (2011). The chief sources of sodium are the Archean gneisses and the weathering of the crystalline rocks.

The value of Mg^{+2} ranged from 10.62 mg/l to 138.51 mg/l with a mean of 31.76 mg/l. The maximum allowable limit is 150 mg/l, and all the samples show values which are within the limit. The major Mg^{+2} sources in the water are the magnesium-bearing minerals in the country rock, domestic and industrial wastes (Adimalla & Qian, 2019; Adimalla & Venkatayogi, 2017; Marghade et al., 2011). Ca^{+2} and Mg^{+2} are very much essential for human body, but when taken in excess, it can cause adverse effect on human health (Adimalla & Qian, 2019).

Potassium (K^+) ranges from 0.75 to 39.75 mg/l with an average of 3.58 mg/l in the study area. The location Cheliyama shows the highest value of 39.75 mg/l. 98% of the samples are within the desirable limits of 12 mg/l. According to He and Macgregor (2008), the K^+ ion in the groundwater is present as a very necessary trace element to maintain stability in the human body.

Iron is present generally in the groundwater as a trace element. According to WHO (2011), the acceptable limit is 0.3 mg/l, but in the study area most of the samples show higher concentrations of iron. The value ranges from 0.2 to 2.5 mg/l of a mean value of 0.93 mg/l. Higher concentrations of iron lead to reddishness in the water colour and kidney problems and deteriorate the quality of utensils leaving a stain behind. The probable sources of iron are the iron–magnesium-bearing amphiboles, pyroxene gneisses and biotite schist. Rock–water interaction and weathering of these iron-rich minerals and rocks lead to higher concentration iron.

Bicarbonate (HCO_3^-) value ranges from 100 to 500 mg/l with a mean value of 231.67 mg/l. No direct negative effect on human health has been reported yet due to HCO_3^- .

Cl^- concentration ranged from 14.18 to 432.49 mg/l with an average value of 106.81 mg/l. Excess of Cl^- causes deleterious effect on human body and also causes pollution (Adimalla & Venkatayogi, 2017; Adimalla et al., 2018a, 2018b, 2018c; Marghade et al., 2011). Here, it is the second most dominating anion, where 90% of the samples are within the desirable limit of 200 mg/l, but most of the samples are within the maximum desirable limit of 600 mg/l. Tiwari et al., 2014; Adimalla and Li (2019) stated that the higher chloride (Cl^-) concentration is mostly due to the effluents from domestic wastes, wastes from leaked septic tanks and breakdown of the chloride bearing minerals.

Sulphate (SO_4) in the study region ranges from 20 to 600 mg/l, with an average of 82.83 mg/l. The maximum allowable limit is 250 mg/l (WHO). The highest value is observed in Muradi which is 600 mg/l.

The NO_3^- concentration in the samples ranged from 0.5 to 80 mg/l with a mean value of 31.52 mg/l. According to WHO, the desirable limit is 45 mg/l and only 40% of the samples exceed the desirable limit. In several studies, it has been observed that there are some correlations between nitrate concentration in the water and practice of agricultural pattern (Adimalla & Li, 2019; Debernardi et al., 2008). Zang et al. (2018) stated that NO_3^- pollution in groundwater has been spread all over the world. Diseases like methemoglobinemia, gastric cancer, birth malfunction, hypertension are mostly caused due to higher concentrations of nitrate (Bao et al., 2017; Fan, 2011; Spalding & Exner, 1993). The source of nitrate has been identified by many workers to be the chemicals used as fertilizers, leakage of the septic tanks and sewage systems (Datta & Tayagi 1996; Subba Rao, 2003). Mostly during post-monsoon period, agricultural activities are favourably done in the study region; therefore, use of nitrogenous fertilizers is high which causes higher concentration of NO_3^- at some places.

4.2 Fluoride contamination in groundwater and its spatial distribution

Fluoride is an incompatible lithophile, electronegative element which forms complexes with polyvalent metal ions present in the water (Faure, 1991). Fluoride is one of the important elements found in the groundwater as trace. The concentration of fluoride in the study area ranged from 1.30 to 7 mg/l with an average value of 4.06 mg/l. The concentration is mostly high in almost all locations of the study area which should be within 1.5 mg/l for drinking water stated by WHO. Edmunds and Smedley (2013) stated the best range suited for fluoride in the drinking water is 1 mg/l. Fluoride concentration in water can be classified into the following classes: < 1 mg/l as low fluoride; 1–1.5 mg/l acceptable limit of fluoride; and > 1.5 mg/l as high fluoride. It is observed that 16.67% of the collected samples are within the acceptable limits, whereas the rest 83% shows higher values of fluoride up to a range of 7 mg/l.

Jiang et al. (2014) stated that the geochemistry of water possessing higher elevated concentrations of fluoride is mostly associated with weakly alkaline pH, moderate TDS and higher concentrations of HCO_3^- and Na^+ . Alkaline condition promotes dissolution of minerals that contains fluoride in their structure like CaF_2 in groundwater. Chen et al. (2016) stated the following equations in an alkaline to weakly alkaline groundwater, and increased OH^- concentration causes the reaction (7), then to the corresponding dissolution of CaF_2 in reaction (8)



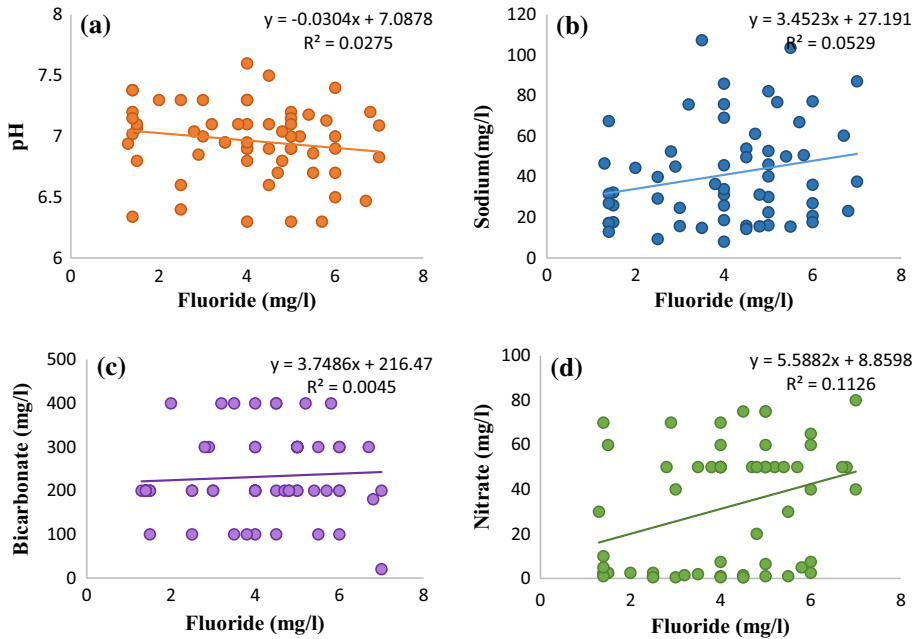


Fig. 4 a–d The graphs depicting the correlation between fluoride and the other physico-chemical parameters like pH, sodium, bicarbonate and nitrate

Figure 4a–d depicts the correlation between the different physio-chemical parameters in the study area. Fluoride shows a positive correlation with HCO_3^- ($r^2=0.0045$); Na^+ ($r^2=0.052$) and NO_3^- ($r^2=0.112$). The sources of fluoride are considered to be muscovite, biotite and granite gneisses which are present through the area. Researchers have predicted that the probable reason for elevated fluoride concentrations is mainly due to the dissolution reaction and the exchange reactions (Jack et al., 2000; Liu et al. 2015).

The spatial distribution of fluoride concentration in the study area is prepared using the IDW Interpolation technique, with the help of spatial analysis tool of ArcGIS 10.0, and Jenks's Natural Break method has been used to classify them. Using Jenk's data segmentation method, best arrangement value of different classes can be determined (Zhang et al., 2012). This method reduces intra-class variation and increases inter-class variation (Jenks, 1967).

The area is distributed into 6 zones—less than 1.5; 1.5–2; 2–3; 3–4; 4–5; and 5–7 mg/l (Fig. 5). The ranges vary from 1 to 7 mg/l, and most of the villages show ranges > 3 mg/l. Only in few villages 10 stations show concentration within the safe permissible limit. This distribution is made to study the region more minutely and accurately.

4.3 Piper plot

Piper plot helps in understanding the chemistry of groundwater and its evolution processes. It determines the dominant water type (Piper, 1944). The diagram consists of one diamond-shaped field at the centre and two triangular fields at the two sides. The cations and anions are expressed as percentage of the total cations and anions present in mg/l. The left triangle

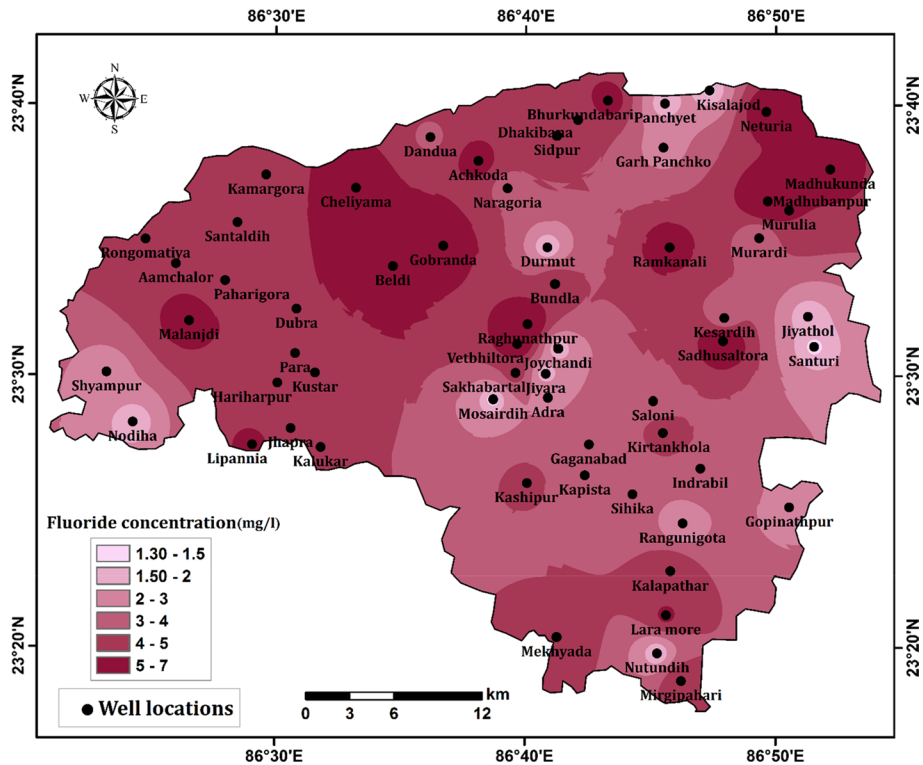
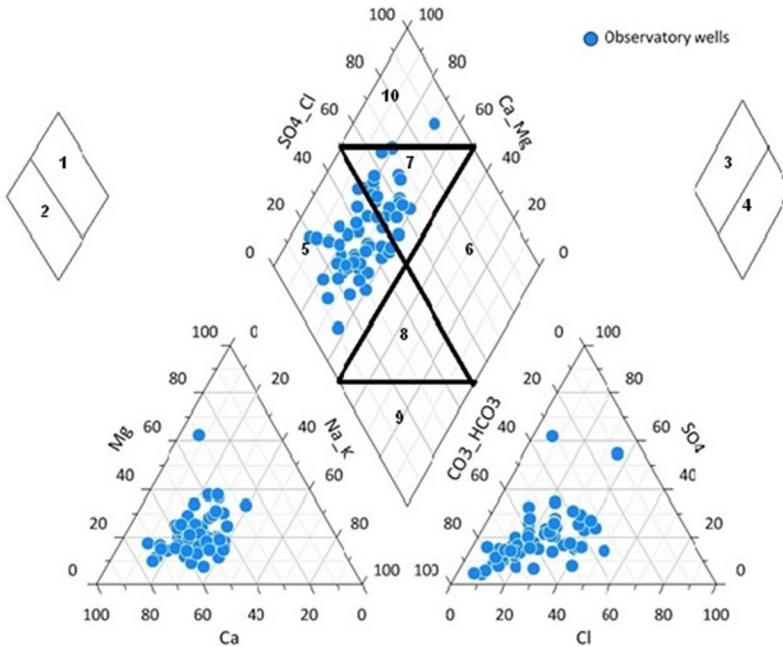


Fig. 5 Spatial distribution map of fluoride concentrations in the study area

represents the cationic plot, whereas the right triangle represents the anionic plot. The anions are Cl^- , SO_4^{2-} and $\text{HCO}_3^- + \text{CO}_3^{2-}$, and the cations are Ca^{+2} , Mg^{+2} and $\text{Na}^+ + \text{K}^+$ which are projected into the central diamond-shaped field that determines the water type or the water facies. From Fig. 6, it is observed that 95% of the sample shows alkaline earth metals ($\text{Ca}^{+2} + \text{Mg}^{+2}$) exceed the alkali cations ($\text{Na}^+ + \text{K}^+$) in zone 3 and 85% of the samples are the weak acids ($\text{HCO}_3^- + \text{CO}_3^{2-}$) represented as zone 2 over the strong acids ($\text{Cl}^- + \text{SO}_4^{2-}$). Therefore, based upon the cation and anion distribution the dominant water types are 80% Ca-HCO_3 ; 19% mixed CaMgCl ; and 1% CaCl type. Ca^{+2} and Mg^{+2} are dominant as the result of weathering of silicate rocks like granite gneiss (Adimalla et al., 2018a, 2018b, 2018c). These samples also show high EC with high TDS which determines the major role of rock–water interaction along with longer residential time (Dehbandi et al., 2018; Edmunds & Smedley, 2013).

4.4 Gibbs plot demonstrating the chemical weathering

Gibbs diagram (Gibbs, 1970) helps in understanding the relationship possesses between the aquifer lithology and the water composition. It consists of three different distinct fields that are evaporation dominance, rock–water or rock dominance and the precipitation dominance. For cations, TDS and $(\text{Na}^+ + \text{K}^+)/(\text{Na}^+ + \text{K}^+ + \text{Ca})$ are plotted, whereas for anions TDS and $(\text{Cl}^-/\text{Cl}^- + \text{HCO}_3^-)$ are plotted to determine the



The characterisation of the different zones in piper plot-

- | | |
|------------------------------------|-----------------------------------------------|
| 1- Strong acids exceed weak acids | 8- Mixed Ca-Na-HCO ₃ type of water |
| 2- Weak acids exceed strong acids | 9- Na-HCO ₃ type of water |
| 3- Alkaline earths exceed alkalies | 10- Ca-Cl type of water |
| 4- Alkalies exceed alkaline earths | |
| 5- Ca-HCO ₃ type water | |
| 6- Na-Cl type water | |
| 7- Mixed Ca-Mg-Cl type of water | |

Fig. 6 Piper plot for the collected water samples

prevailing mechanisms leading to the variable concentrations of the ion in the groundwater and its impact on the surrounding country rock. From Fig. 7, it is observed that the samples fall mostly in the rock–water interaction dominance field. Hard rock regions possessing low rainfall and high temperature mostly show rock–water interaction dominance (Adimalla & Li 2018; Subba Rao et al., 2017; Adimalla et al., 2018a, 2018b, 2018c). Therefore, it can be inferred that higher rate of residence time or slow water percolation intensely enhances the ionic concentration in the groundwater (Chen et al., 2017; Subba Rao et al., 2017; Adimalla 2018).

Furthermore, a plot has been made for better understanding of the weathering pattern. Ca + Mg and HCO₃ + SO₄ values have been plotted (Fig. 8) which indicates that 88% of the samples fall in the silicate weathering region and the rest in carbonate weathering region. The process that is dominant in this region is the rock–water interaction along with silicate weathering (Fig. 9).

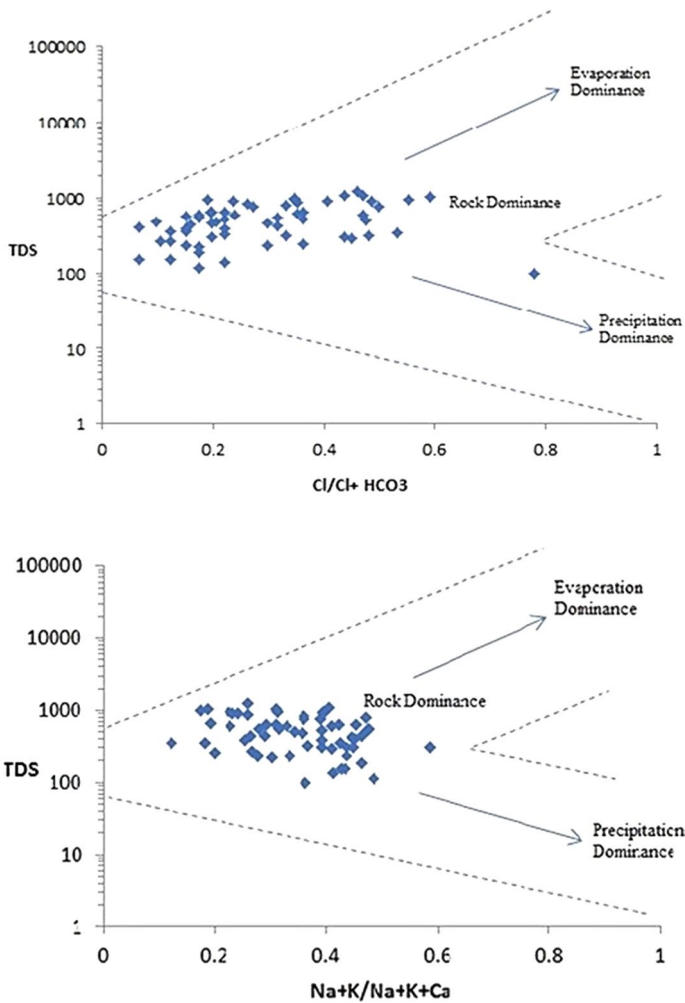


Fig. 7 Gibbs plot for the collected water samples

4.5 Ion exchange by hydrogeochemical analysis

Schoeller (1967) proposed a mathematical relation for better understanding of the ionic exchange taking place between the groundwater and the aquifer host rock material is called the chloro-alkaline indices (CAI). This method provides a quantitative analysis about the direct and reverse ion exchange processes which tells us about the groundwater chemistry. CAI (I) and CAI (II) are calculated using the following equations which are expressed in meq/l (Adimalla et al., 2018a, 2018b, 2018c).

$$\text{CAI(I)} = [\text{Cl}^- - (\text{Na}^+ + \text{K}^+)/\text{Cl}^-] \quad (9)$$

$$\text{CAI(II)} = [\text{Cl}^- - (\text{Na}^+ + \text{K}^+)/(\text{SO}_4^{2-} + \text{HCO}_3^- + \text{CO}_3^{2-} + \text{NO}_3^-)] \quad (10)$$

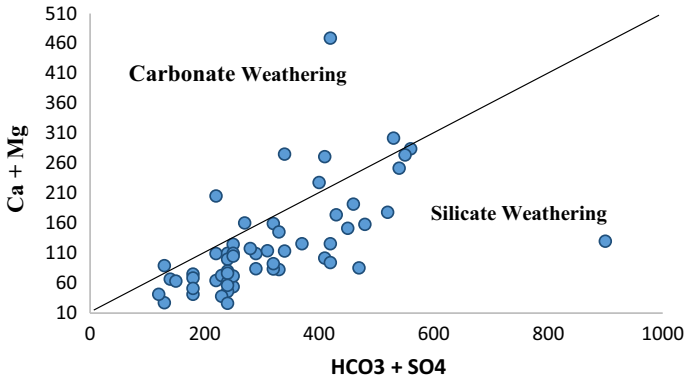


Fig. 8 The scatter plot between (HCO₃ + SO₄) and (Ca + Mg)

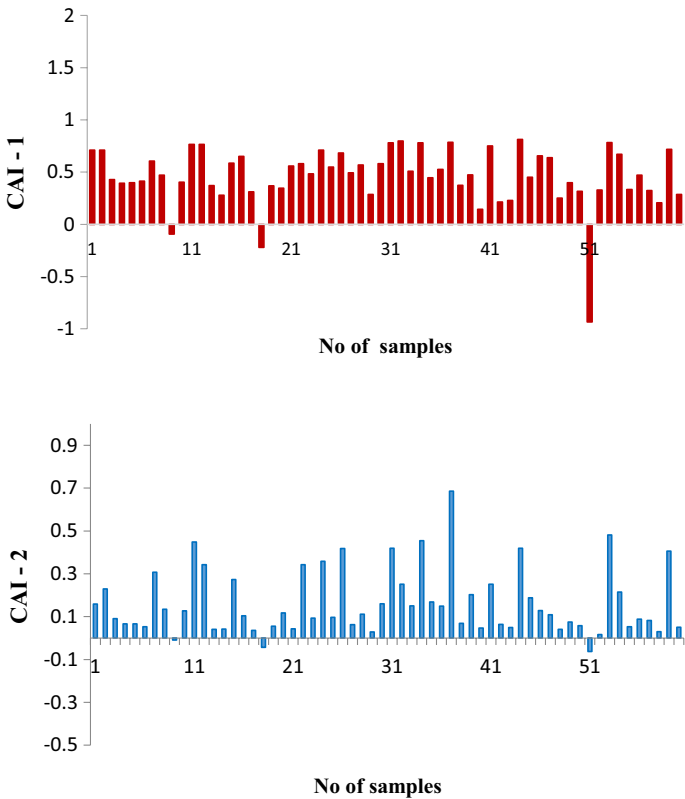


Fig. 9 The CAI (I) and CAI (II) values for the different locations

When CAI (I) and CAI (II) indices are positive, then the ionic exchange is between (Na⁺ + K⁺) of the groundwater and (Ca⁺² + Mg⁺²) of the aquifer material or the host rock, and then, it is called reverse exchange method. On the other hand, in a direct ion exchange

method both the CAI (I) and CAI (II) indices are negative when ($\text{Na}^+ + \text{K}^+$) cations of the aquifer material/host rock exchange with ($\text{Ca}^{+2} + \text{Mg}^{+2}$) of the groundwater (Adimalla et al., 2018a, 2018b, 2018c; Schoeller, 1967).

The CAI (I) and CAI (II) indices for the study area range from (− 0.934 to 0.796) and (− 0.062 to 0.686), respectively. The values are plotted in bar graphs in Fig. 7. The plots reveal that 5% of the sample shows reverse ionic exchange, whereas 95% of the samples show direct ionic exchange process. Therefore, the dominant ionic exchange that governs the chemistry of the groundwater is reverse exchange or chloro-alkaline equilibrium.

4.6 Fluoride health risk factor assessment

The health risk assessment is done based upon the model given in USEPA (Moya et al., 2011; USEPA, 1989). This model is used to calculate the non-carcinogenic (Adimalla & Li, 2019; Chen et al., 2017) effects of fluoride for the different age groups which include the babies, children, teenagers, adolescence, grown-up adults and the aged people, i.e. 6–12 months; 1–5 years; 5–10 years; 10–15 years; 15–18 years; 18–21 years; 21–65 years; and >65 years for more detailed investigations. Fluoride is very much essential for the protection of teeth from demineralization which are caused by the acids produced by the different bacteria and sugar present in the mouth. Children require fluoride to protect the newly formed teeth, and adults need for lowering the decay of teeth. If fluoride is taken within 1.5 mg/l, it helps in demineralization and enamel strengthening (Ahada & Suthar, 2019). Many hard rock regions of India are affected by the excess of fluoride concentrations in drinking water which causes some of the epidemic diseases like dental and skeletal fluorosis (Brindha et al., 2011; Adimalla & Qian, 2019). Apart from dental and skeletal fluorosis, the intake of excess of fluoride causes other acute disorders such as respiratory failure, general paralysis, lowering of blood pressure, weight loss, anaemia and adverse effects on reproduction (Dissanayake 1991; Freni 1994; Chen et al., 2017; Adimalla & Qian, 2019). Guo et al. (2007) discussed on the effects of fluoride in the male fertility level and also stated that children consuming > 2 mg/l of fluoride showed poor intelligence test than the ones consuming within the standard limit of fluoride. Similarly, many towns and municipalities of Purulia with high population are highly been effected by the elevated concentrations of fluoride in groundwater (Mandal & Sanyal, 2019). This study considers the concentration of fluoride consumed on daily basis through drinking and dermal contact as bathwater source or other domestic purpose. The health risk is computed for the different age groups based on the HQ values through ingestion and dermal pathway. The health risk is computed for the different age groups based on the HQ values through ingestion and dermal pathway. The calculated HQ values for each age group are plotted using simple Kriging interpolation method, and the indexes are prepared ranging between the highest and lowest HQ values, to understand the distribution of health risk in the study area (Figs. 10 and 11).

4.7 Ingestion pathway

USEPA Exposure Factor Handbook (Moya et al., 2011; USEPA, 1989) provides the values of the different key parameters given in Table 2, which are used to compute the health risk with analysed fluoride concentrations for the different villages in Purulia. The estimated HQ_{in} values are calculated using the formulas (2–5) as described under material and methods for the different age groups—6–12 months; 1–5 years; 5–10 years; 10–15 years;

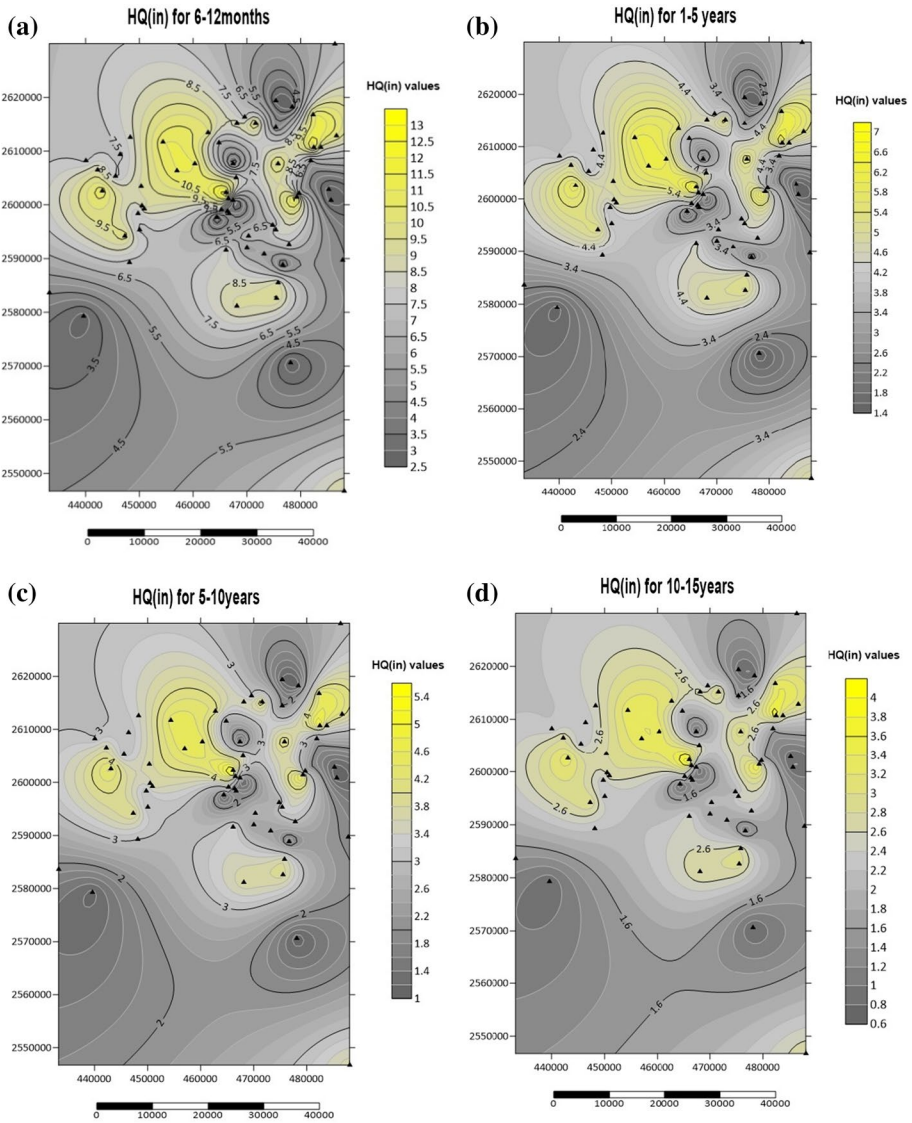


Fig. 10 a–h HQ (in) values for the different age groups through ingestion pathway in the study area

15–18 years; 18–21 years; 21–65 years; and > 65 years, which are given in Table S1, see supplementary file, and shown in Fig. 10a–h. The HQ_{in} value ranges from 2.38 to 12.82 with an average of 7.4 for 6–12 months babies/infants. The HQ_{in} value for the age group 1–5 years young children varies from 7.02 to 1.30 with an average value of 4.1. The HQ_{in} value for the age group 5–10 years children varies from 0.97 to 5.25 with a mean value of 3.0. The HQ_{in} value ranges from 0.72 to 3.91 with an average of 2.3 for the age group 10–15 years teenagers. The HQ_{in} value for the age group 15–18 years young people varies from 0.57 to 3.07 with a mean value of 1.7. The HQ_{in} value ranges from 0.75 to 4.03

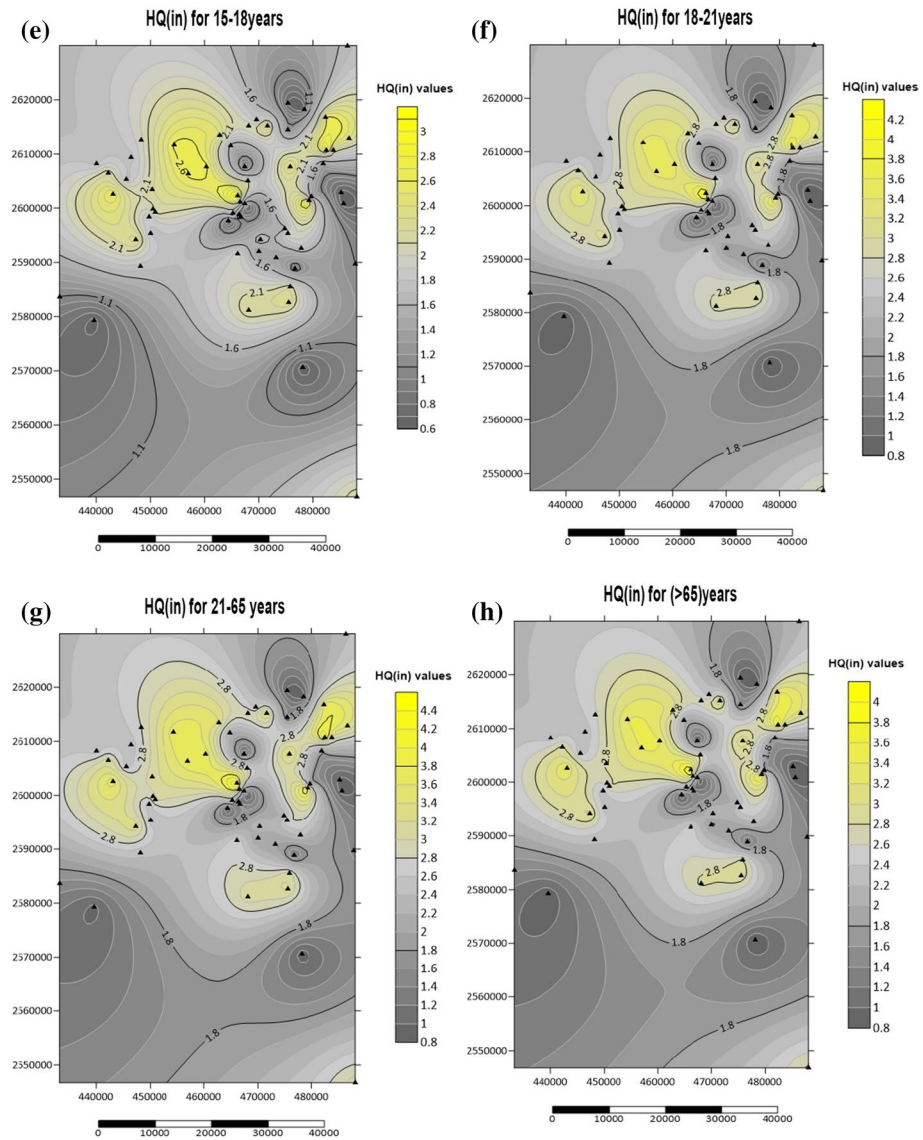


Fig. 10 (continued)

with an average of 2.3 for the age group 18–21 years young adults. The HQ_{in} value for the age group 21–65 years adults ranges from 0.80 to 4.35 with a mean value of 2.5. The HQ_{in} value ranges from 0.73 to 3.98 with an average of 2.3 for the age group greater than 65 years aged people.

The acceptable limit or the safe limit for HQ_{in} is below 1, but most of the locations show much higher value some up to 12.8. Based upon the mean HQ_{in} values of the non-carcinogenic fluoride health risk on the different age groups can be arranged in the following decreasing order: 6–12 months > 1–5 years > 5–10 years > 21–65 years > 18–21 years > abo

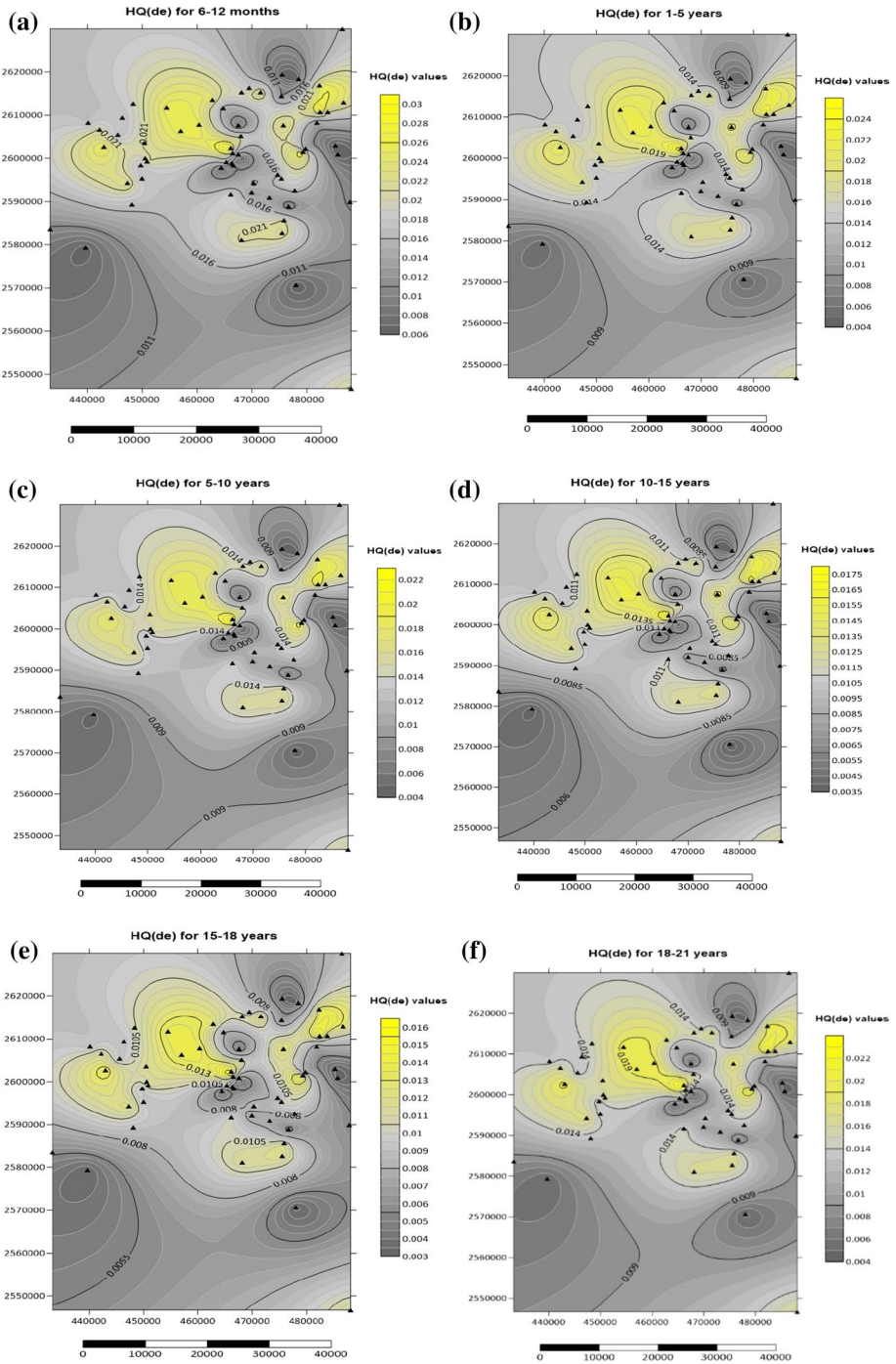


Fig. 11 a–h HQ(de) values for the different age groups through ingestion pathway in the study area

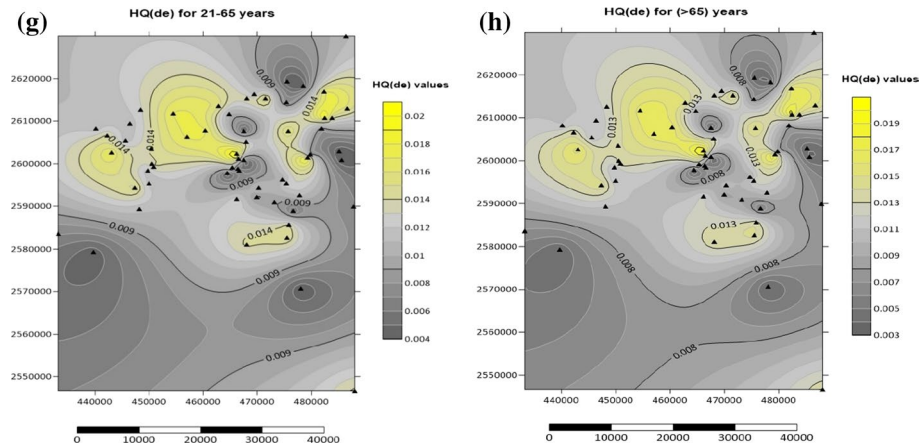


Fig. 11 (continued)

ve 65 years > 10–15 years > 15–18 years. From the observation, it can be inferred that the age groups 6–12 months babies and the 1–5 years children are most likely to suffer from the health complications and diseases due to direct consumption of groundwater with high concentration of fluoride in the long run. The health risk factor of 1–5 years is 2.4, 3.2, 4.2, 3.2, 2.9 and 3.2 times greater than the age groups 5–11 years; 11–16 years; 16–21 years; 21–65 years; and > 65 years, respectively, due to consumption of the elevated fluoride contaminated water. Nearly 85% of health problems arises due to consumption of uncleaned and contaminated water; therefore, ingestion pathway would be responsible for 70% of global fluorosis (Dissanayake 1991; Freni 1994). The age group 6–12 months babies is at high health risk than the other age groups in the study area, so proper planning is necessary for water management. Many workers from India (Adimalla & Qian, 2019; Singh et al., 2018) and globally (Chen et al., 2017) reported similar results in which children are more prone to health risk. The probable reason could be due to the lower body weight of the children and constant exposure to fluoride contaminated water through ingestion. So, proper planning is necessary for water management for the proper and healthy future.

4.8 Dermal pathway

The estimated HQ_{de} values are calculated using the formulas (2–5) as described under material and methods for the different age groups: 6–12 months; 1–5 years; 5–10 years; 10–15 years; 15–18 years; 18–21 years; 21–65 years; and > 65 years, which are given in Table S2, see supplementary file, and the HQ_{de} values are shown in Fig. 11a–h. The HQ_{de} value ranges from 0.005 to 0.029 with an average value of 0.017 for 6–12 months babies. The HQ_{de} value varies from 0.004 to 0.024; 0.004 to 0.021; 0.003 to 0.017; 0.002 to 0.016; 0.004 to 0.022; 0.003 to 0.019; and 0.003 to 0.019 with the mean values of 0.014; 0.012; 0.010; 0.009; 0.013; 0.012; and 0.011, respectively, for the age groups: 1–5 years; 5–10 years; 10–15 years; 15–18 years; 18–21 years; 21–65 years; and > 65 years, respectively. All the HQ_{de} values for the age groups are within the safe limit which is within 1. The dermal contact pathway effect is quite lower than the direct consumption ingestion pathway. Therefore, it can be concluded that the principal pathway for non-carcinogenic fluoride in the human body is through direct ingestion or consumption of the groundwater

with elevated fluoride concentrations. The calculated HQ_{in} and HQ_{de} values can act as baseline information for the different authorities for making the policies related to water sources and its management.

5 Conclusion

Water is the basic need in life, so the hydro-chemical characterization of groundwater is very much essential for understanding its suitability for drinking and other purposes. Groundwater samples were collected from the eastern portion of Purulia district for estimating the health risk in the different age groups due to high fluoride concentrations. It is observed that the general trend of the ionic concentrations based on their mean values is arranged as $Ca^{+2} > Na^{+} > Mg^{+2} > K^{+} > Fe^{+2}$ for cations and $HCO_3^{-} > Cl^{-} > SO_4^{-} > NO_3^{-} > F^{-}$ for anions. The water is weakly alkaline in nature with moderate TH and TDS value in the overall area. The concentration of iron and fluoride varies from 0.2 to 2.5 ppm and 1.30 to 7 mg/l which deteriorates the drinking water quality. The mechanism which mostly controls the groundwater geochemistry is the rock–water interaction and the weathering of the silicate minerals. 95% of the samples show ionic exchange through direct process and the rest 5% by reverse ionic exchange. The health risk assessment for non-carcinogenic fluoride contaminated water reveals that the pathway through which people are been affected is direct consumption or ingestion pathway. It is observed that the HQ_{in} values have exceeded the safe limit (less than 1) for all the age groups. 6–12 months babies are more prone to health complications related to elevated concentrations fluoride. Therefore, proper and necessary steps must be taken to reduce the health risk and reduce the contamination effect on human health. However, this study would help the governmental and the non-governmental associations for making rules and regulations for groundwater managements on the scientific background. This would in the long run improve the health of the people in this region and would provide safe drinking water.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s10668-021-01911-1>.

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