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ENVIRONMENTAL CHEMISTRY, POLLUTION & WASTE MANAGEMENT | RESEARCH ARTICLE

GIS-based inverse distance weighting spatial interpolation technique for fluoride distribution in south west part of Nagaur district, Rajasthan

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Abstract: In Merta block of Nagaur District, 54 groundwater samples were collected and analyzed to determine fluoride concentration. It was observed that 64.8% villages have fluoride concentration within the acceptable limit (i.e. 1.5 mg/l) and are safe for drinking in respect of fluoride concentration. As prescribed by Bureau of Indian Standard, 35.2% villages have fluoride concentration above the acceptable limit and habitants in these villages are under threat of fluorosis. The fluoride-contaminated area is further divided into three categories. About 27.8% villages have fluoride concentration between 1.5 and 3.0 mg/l which is less problematic to consumers. In Dhadhlas Uda and Bhensra Khurd village, fluoride concentration in groundwater is above 3.0 mg/l and below 5.0 mg/l which is problematic in respect to fluorosis. In the entire survey village, Chawadiya Kalan and Akhawas are in the highly problematic category. In these villages, fluoride concentration is above 5.0 mg/l. Due to the higher fluoride level in drinking water, several cases of dental and skeletal fluorosis have appeared at an alarming rate in the region.

ABOUT THE AUTHORS

Mohammed Arif is a PhD scholar working on fluoride and its effect of flora and fauna. He has 25 publications so far.

Ikbal Husain has been working on water quality since 1997. His study contributes in natural and industrial pollution of water resources of Rajasthan. He has 68 publications so far.

Jakir Hussain has been working on water quality since 1999. He studied on natural and industrial pollution of groundwater resources of Rajasthan. He has 78 publications so far.

Manoj Kumar Sharma has been working on groundwater quality since 1997. He has a great contribution in assessment of fluoride in western Rajasthan.

Sudesh Kumar is working as an assistant professor. He has experience in water quality monitoring and assessment special reference to fluoride.

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PUBLIC INTEREST STATEMENT

Fluorosis affects the ability of people to work efficiently and in some cases, they become totally unproductive. The inability to work can reduce a family to a state of utter economic despair. The study will help people to identify the area of high fluoride. The GIS-based map produced by data marks the area with high and low fluoride. The use of alternate drinking water source with low fluoride concentration or treated groundwater may prevent them from fluorosis. The paper also suggests the mitigation tactics both modern and traditional. The tradition tactics are well proved and experienced from years. These are mostly prevention based and suggest nutritional interventions. The modern tactics deal with treatment of high fluoride water.

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1. Introduction

One of the greatest challenges of the twenty-first century is to provide an adequate supply of safe water for household consumption to everyone, everywhere. But, whereas water demand is constantly on the rise, the quality of the water resources, which are unevenly distributed over the earth's surface, is deteriorating due to anthropogenic activities. Therefore, even countries with vast water resources could suffer from scarcity of water in the future.

Fluorine is the most electronegative and reactive of all elements, and is present as fluoride in drinking water. It occurs as fluoride naturally in soils and natural waters due to chemical weathering of some F-containing minerals (Teotia & Teotia, 1985). Nearly 12 million of the 85 million tons of fluoride deposits on the earth's crust are found in India. It is not surprising; therefore, the fluorosis is endemic in 17 states of India (UNICEF, 2008). The most seriously affected areas are Andhra Pradesh, Punjab, Haryana, Rajasthan, Gujarat, Tamil Nadu, and Uttar Pradesh (Arif, Hussain, Hussain, & Kumar, 2013a, 2013b, 2014; Arif, Hussain, Hussain, Kumar, & Bhati, 2014; Arif, Hussain, Hussain, & Neyol, 2011; Arif, Hussain, Hussain, Sharma, & Kumar, 2012a, 2012b; Arif, Joshi, & Kumar, 2012; Arif, Yadav, & Garg, 2013; Hussain, Arif, & Hussain, 2012; Hussain, Hussain, & Sharma, 2010; Hussain, Hussain, Sharma, & Ojha, 2002; Hussain, Sharma, & Hussain, 2003, 2004, Hussain et al., 2005; Hussain, Sharma, Ojha, & Hussain, 2000). In Indian continent, the higher concentration of fluoride in groundwater is associated with igneous and metamorphic rocks. It is estimated that around 200 million people, from among 25 nations the world over, are under the dreadful fate of fluorosis. India and China, the two most populous countries of the world, are the worst affected. Estimation finds that 65% of India's villages are exposed to fluoride risk (UNICEF, 2008). High groundwater fluoride concentrations associated with igneous and metamorphic rocks such as granites and gneisses have been reported from India, Pakistan, West Africa, Thailand, China, Sri Lanka, and Southern Africa (World Health Organization [WHO], 2006). The dissolution of fluorite, apatite, and topaz from local bedrocks leads to high fluoride concentration in groundwater.

The dietary source of fluoride is drinking water. Fluoride in drinking water is known for both beneficial and detrimental effects on health. The intake of fluoride in permissible limits of 0.5–1.0 mg/l is known to be beneficial for human health in production and maintenance of healthy teeth and bones (WHO, 2005), while excessive intake of fluoride causes dental and skeletal fluorosis, which is a chronic disease manifested by mottling teeth in mild cases, softening of bones, and neurological damage in severe cases. According to WHO (1997), the permissible limit for fluoride is 1.5 mg/l, whereas United States Public Health Service (USPHS, 1962) has set a range of allowable concentrations for fluoride in drinking water for a region depending on its climatic conditions because the amount of water consumed and consequently the amount of fluoride ingested are being influenced primarily by the air temperature. Accordingly, the maximum allowable concentration for fluoride in drinking water in Indian conditions comes to 1.4 mg/l, while as per Indian standard it is 1.5 mg/l. The fluoride-related problems are closely associated with climate; in hot tropical part of the world, people consumed more water and consequently the risk of fluoride accumulation increases.

Rajasthan is the largest state of India having 342,239 km^2 area and with relatively low population density i.e. 165 persons per square kilometer. According to physiographic divisions, the north and western parts of the state are under The Great Plain of north India while, south and middle as well as eastern parts are classified under the Peninsular Plateau. The selected part for this study is situated in northern most part of the state where groundwater is a major source of drinking water. A bibliographic survey has shown that yet no studies have been undertaken in the study area with regard to fluoride and fluorosis problem. So, the objective of this study was to investigate the quality of drinking water (underground water) with special reference to the concentration of fluoride in rural habitations of Central Rajasthan, India.

1.1. Indian scenario

In India, fluoride was first detected in drinking water at Nellore district of Andhra Pradesh in 1937 (Ayoob & Gupta, 2006). Since then, considerable work has been done in different parts of India to explore the fluoride-laden water sources. At present, it has been estimated that fluorosis is prevalent in 17 states of India, indicating that endemic fluorosis is one of the most alarming public health problem of the country, especially in Rajasthan, Madhya Pradesh, Andhra Pradesh, Tamil Nadu, Gujarat, and Uttar Pradesh. At present, in India, endemic fluorosis is thought to affect ~1 million people (Sneha, Yenkie, Labhsetwar, & Rayalu, 2012). Districts known to be endemic for fluoride in various states of India and the ranges of fluoride in drinking water are given in Table 1.

1.2. Guidelines and standards

According to WHO's guidelines for drinking water, a fluoride level of 1.5 mg/l is the desirable upper limit. India reduced the upper limit of fluoride in drinking water from 1.5 to 1.0 mg/l with a rider that less is better (Bureau of Indian Standards [BIS] 10500, 2012). This is due to extremes in climatic

State	District	Range
	Table 1. District showing fluoride concentration > 1.5 mg/l in groundwater in India in 2010	
Assam	Goalpara, Kamrup, Karbi Anglong, and Nagaon	1.45–7.8
Andhra Pradesh	Adilabad, Anantpur, Chittoor, Guntur, Hyderabad, Karimnagar, Khammam, Krishna, Kurnool, Mahbubnagar, Medak, and Nalgonda	1.8–8.4
Bihar	Aurangabad, Banka, Buxar, Jamui, Kaimur(Bhabua), Munger, Nawada, Rohtas, and Supaul	1.7–2.85
Chhattisgarh	Bastar, Bilaspur, Dantewada, Janjgir-Champa, Jashpur, Kanker, Korba, Koriya, Mahasamund, Raipur, Rajnandgaon, and Surguja	1.5–2.7
Delhi	East Delhi, North West Delhi, South Delhi, South West Delhi, West Delhi, Kanjhwala, Najafgarh, and Alipur	1.57–6.10
Gujarat	Ahmadabad, Amreli, Anand, Banaskantha, Bharuch, Bhavnagar, Dohad, Junagadh, Kachchh, Mehsana, Narmada, Panchmahals, Patan, Rajkot, Sabarkantha, Surat, Surendranagar, and Vadodara	1.6–6.8
Haryana	Bhiwani, Faridabad, Gurgaon, Hissar, Jhajjar, Jind, Kaithal, Kurushetra, Mahendragarh, Panipat, Rewari, Rohtak, Sirsa, and Sonepat	1.5–17
Jammu and Kashmir	Doda, Rajauri, and Udhampur	2.0–4.21
Karnataka	Bagalkot, Bangalore, Belgaun, Bellary, Bidar, Bijapur, Chamarajanagar, Chikmagalur, Chitradurga, Davangere, Dharwad, Gadag, Gulburga, Haveri, Kolar, Koppal, Mandya, Mysore, Raichur, and Tumkur	1.5–4.4
Kerala	Palakkad, Palghat, Allepy, Vamanapuram, and Alappuzha	2.5–5.7
Maharashtra	Amravati, Chandrapur, Dhule, Gadchiroli, Gondia, Jalna, Nagpur, and Nanded	1.51–4.01
Madhya Pradesh	Bhind, Chhatarpur, Chhindwara, Datia, Dewas, Dhar, Guna, Gwalior, Harda, Jabalpur, Jhabua, Khargaon, Mandsaur, Rajgarh, Satna, Seoni, Shajapur, Sheopur, and Sidhi	1.5–10.7
Orissa	Angul, Balasore, Bargarh, Bhadrak, Bandh, Cuttack, Deogarh, Dhenkanal, Jajpur, Keonjhar, and Sonapur	1.52–5.2
Punjab	Amritsar, Bhatinda, Faridkot, Fatehgarh Sahib, Firozepur, Gurdaspur, Mansa, Moga, Muktsar, Patiala, and Sangrur	0.44–6.0
Rajasthan	Ajmer, Alwar, Banaswara, Barmer, Bharatpur, Bhilwara, Bikaner, Bundi, Chittaurgarh, Churu, Dausa, Dhaulpur, Dungarpur, Ganganagar, Hanuman-garh, Jaipur, Jaisalmer, Jalor, Jhunjhunun, Jodhpur, Karauli, Kota, Nagaur, Pali, Rajsamand, Sirohi, Sikar, SawaiMadhopur, Tonk, and Udaipur	1.54–11.3
Tamil Nadu	Coimbatore, Dharmapuri, Dindigul, Erode, Karur, Krishnagiri, Namakkal, Perambalur, Puddukotai, Ramanathapuram, Salem, Sivaganga, Theni, Thiruvannamalai, Tiruchirapally, Vellore, and Virudhunagar	1.5–3.8
Uttar Pradesh	Agra, Aligarh, Etah, Firozabad, Jaunpur, Kannauj, Mahamaya Nagar, Mainpuri, Mathura, and Mau	1.5–3.11
West Bengal	Bankura, Bardhaman, Birbhum, Dakshindinajpur, Malda, Nadia, Purulia, and Uttardinajpur	1.5–9.1

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conditions and the diet being deficient in essential nutrients (calcium, vitamins C, E, and antioxidants) in the rural communities of India. So, according to Indian Standards, the maximum desirable limit of fluoride in drinking water is 1.0 mg/l and maximum permissible limit 1.5 mg/l. As the amount of water consumed and consequently the amount of fluoride ingested are influenced primarily by air temperature, USPHS (1962) has set a range of concentrations for maximum allowable fluoride in drinking water for communities based on the climatic conditions as shown in Table 2.

2. Study area
Merta block is located between latitude 26°26′18″ and 26°51′0″N and longitude 73°41′45″and 74°17′24″E in Nagaur district. Hydrogeologically, the Merta block can be classified into three formations viz. consolidated formation, semi-consolidated formation, and unconsolidated formation. The study area shown in Figure 1.

2.1. Consolidated formation
The consolidated formations comprise schist, gneiss, quartzite, and phyllites of Precambrian metamorphic rocks and limestone and sandstone of Marwar Super Group. Nagaur sandstone is coarse-to-fine grained, loosely cemented with gravel at basal part which act as good aquifer, and occupies mainly the part of Merta blocks. The associated rocks are siltstone and shales. Its thickness varies from 140 to 240 m.

2.2. Semi-consolidated formation
These include only Palana sandsotene consisting of very coarse grained, gravelly sand with intercalation of clay with kankar and lignite. Ground water occurs under phreatic to confined condition and saturated thickness of 40 m constitutes a potential aquifer. This mainly occurs in part of Merta block. Its thickness varies from 100 to 250 m.

2.3. Unconsolidated formation
Quaternary alluvium is the main aquifer which comprises unconsolidated to loosely consolidated fine-to-coarse grained sand having intercalations and intermixing silt, clay with "kankar". Ground water occurs under unconfined to semi-confined conditions, Quaternary alluvium covers part of Merta block. Its thickness is limited to 200 m (Census, 2001).
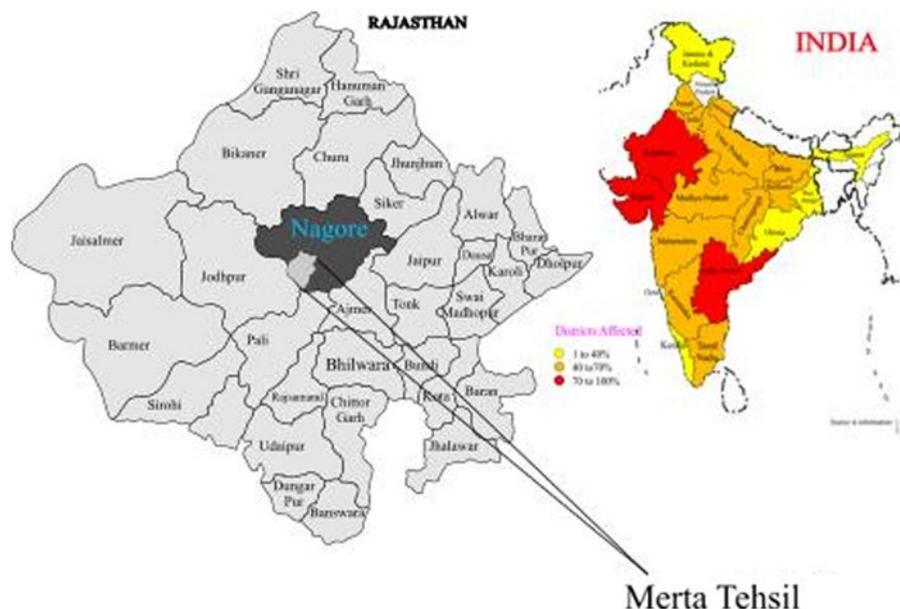
3. Materials and methods
Groundwater samples of 56 villages located in Merta tehsil of Nagaur district were collected in pre-cleaned polythene bottles with necessary precautions (Brown, Skougstad, & Fishman, 1974). The samples were collected, in year 2013, from manually operated public hand pumps and public walls in residential localities of studied habitations. The fluoride concentration in water was determined electrochemically, using fluoride in selective electrode (American Public Health Association [APHA], 2012). This method is applicable to the measurement of fluoride in drinking water in the concentration range of 0.01–1,000 mg/l. The electrode used was an Orion fluoride electrode, coupled to an Orion Ion

Annual average of maximum daily air temperature (°C)	Recommended fluoride concentration (mg/l)			Maximum allowable fluoride concentration (mg/l)
	Lower	Optimum	Upper	
10–12	0.9	1.2	1.7	2.4
12.1–14.6	0.8	1.1	1.5	2.2
14.7–17.7	0.8	1	1.3	2
17.8–21.4	0.7	0.9	1.2	1.8
21.5–26.2	0.7	0.8	1	1.6
26.3–32.5	0.6	0.7	0.8	1.4

Table 2. USPHS recommendation for maximum allowed fluoride in drinking water

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Figure 1. Study area.

meter. Standards fluoride solutions (0.1–10 mg/l) were prepared from a stock solution (100 mg/l) of sodium fluoride. As per experimental requirement, 1 ml of Total Ionic strength Adjusting Buffer Grade III (TISAB III) was added in 10 ml of sample. The ion meter was calibrated for a slop of −59.2 ± 2 (APHA, 2012). The composition of TISAB solution was as 385.4 g ammonium acetate, 17.3 g of cyclohexylene diamine tetraacetic acid, and 234 ml of concentrate hydrochloric acid per liter. All the experiments were carried out in triplicate, and the results were found reproducible with ±2% error.

3.1. Spatial distribution of fluoride using Inverse distance weighted method

Geographic Information System (GIS) is a computerized data-based system for capture, storage, retrieval, analysis, and display of spatial data. GIS is a general purpose technology for handling geographic data in digital form, and satisfying the following specific needs, among others. The Inverse distance weighted (IDW) interpolation explicitly implements the assumption that things that are close to one another are more alike than those that are farther apart. To predict a value for any unmeasured location, IDW uses the measured values surrounding the prediction location. The measured values closest to the prediction location have more influence on the predicted value than those farther away. IDW assumes that each measured point has a local influence that diminishes with distance. It gives greater weights to points closest to the prediction location, and the weights diminish as a function of distance, hence the name IDW.

4. Results and discussion

The fluoride concentrations in 54 water samples from different villages were collected and analyzed following APHA ion selective electrode method (APHA, 2012). In the study area, fluoride concentration in drinking water samples varied 0.4–8.6 mg/l with highest at Akhawas (8.6 mg/l) and lowest at Nokha Chandawata (0.40 mg/l). Considering the health impact associated with high fluoride intake, all villages were categorized into following four categories:

(1) Safe: Fluoride < 1.5 mg/l,

(2) Less problematic: Fluoride > 1.5 to 3.0 mg/l,

(3) Problematic: Fluoride > 3.0 to 5.0 mg/l, and

(4) Highly problematic: Fluoride > 5.0 mg/l.

Number of villages along with their name and fluoride concentration is shown in Table 3. It was observed that 64.8% of the villages are within the permissible limit in the absence of alternative source (i.e. 1.5 mg/l). These villages are safe for drinking in respect of fluoride concentration. As prescribed by BIS, 35.2% villages have fluoride concentration above the acceptable limit (2012) and the villagers in these villages are under threat of fluorosis. The affected area is further divided in three categories and 27.8% villages have fluoride concentration between 1.5 and 3.0 mg/l which is less problematic to consumers. Dental fluorosis is common in these villages. At this concentration, teeth lose their shiny appearance and chalky black, gray, or white patches develop known as mottled enamel (Dean, 1942). In some cases, the pre-stage of skeletal fluorosis may occur after 45-year age (Choubisa, Choubisa, & Choubisa, 2009).

In Dhadhlas Uda and Bhensra Khurd village, fluoride concentration in groundwater is above 3.0 mg/l and below 5.0 mg/l, and this fall in problem in respect to fluorosis. Per day intake of fluoride by an adult in these villages is very high. About 6% population of these villages may have all degree of dental fluorosis (Mild, moderately, moderately severe, severe fluorosis) including skeletal fluorosis after 30-year of age. However, the probability of second-stage skeletal fluorosis age may be common after the age of 45 (Choubisa, Choubisa, & Choubisa, 2001).

In the entire survey, the Chawadiya Kalan and Akhawas were the village falls in highly problematic category. In these villages, fluoride concentration is above 5.0 mg/l, which may result all types of fluorosis in inhabitants. In the second clinical stage, the affected persons may have pain in bones, which causes further calcification in ligaments. It has been reported that under such, persons may suffer from stiffness in joints. At this concentration, the vertebrae partially fuse together and crippling the patient which is known as "crippling skeletal fluorosis." It has also been reported that high doses of fluoride are toxic to man and causes health hazards including hemorrhagic gastroenteritis acute toxic nephritis and degree of injury to the liver and heart muscles (Suttie, 1980). In such cases, plasma epinephrine and fluoride-induced hyperglycemia are significantly increased (Agency for Toxic Substances and Disease Registry [ATSDR], 2003). Fluorosis is accompanied by adverse effect on the other systems and organs of the body namely liver, kidney, muscles, heart, lungs, blood, and the hormonal functions (WHO, 2002; US Department of Agriculture, 2005; US Environmental Protection Agency, 2010). The kidney is the principal organ through which maximum concentration of fluoride is excreted.

Based on GIS technique, spatial distribution of fluoride using IDW method was plotted and presented in Figure 2. The figure shows the fluoride distribution in block. From the figure, it is concluded that the fluoride in south eastern part is greater than other areas. It decreases on moving toward west direction in the block.

Table 3. Fluoride concentration in different villages of Merta block of southwest part of Nagaur district			
Safe (Fluoride < 1.5 mg/l)	**Less problematic (Fluoride > 1.5 to 3.0 mg/l)**	**Problematic (Fluoride > 3.0 to 5.0 mg/l)**	**Highly problematic (Fluoride > 5.0 mg/l)**
Sangvo ki Dhani (0.5), Karwasaron Ki dhani (0.6), Harsolao (0.5), Rol Chandawata (0.5), Nokha Chandawata (0.4), Dadhawara (1.4), Oladan (1.3), Deshwal (1.2), Dhawa (1.4), Kumpras (0.5), Riya Shyamdas (0.6), Bakaliyas (0.8), Tukliya (1.3), Gothan (1.0), Talanpur (0.8), Dhanapa (1.4), Lamba Jatan (0.8), Sirsala (1.1), Gathiya (0.6), Gagrana (1.5), Poondloo (1.1), Beetan (1.1), Bayadc (1.3), Katyasani (1.3), Basni Nathoo (1.1), Dotalai (1.3), Dookiya (1.5), Khakharki (0.6), Bargaon (1.4), Phalki (1.2), Medas (0.5), Rohisi (1.3), Jalwana (1.5)	Looniyas (2.7), Dangawas (1.7), BasniKachhawa (2.0), Leeliya (2.6), Surpura (2.6), Jaswantabad (3.0), Jasnagar (2.6), Sooriyas (2.9), Lampolai (2.4), Padukalan (2.0), Khera Dhoonawala (2.3), Riyan Bari (1.9), Beejathal (2.2), Jatawas (1.6), Ren (1.8)	Dhadhlas Uda (4.4), Bhensra Khurd (4.3)	Chawadiya Kalan (6.8), Akhawas (8.6)

Figure 2. Fluoride areal distribution map of Merta block, Nagaur use of GIS-based inverse distance weighting method.
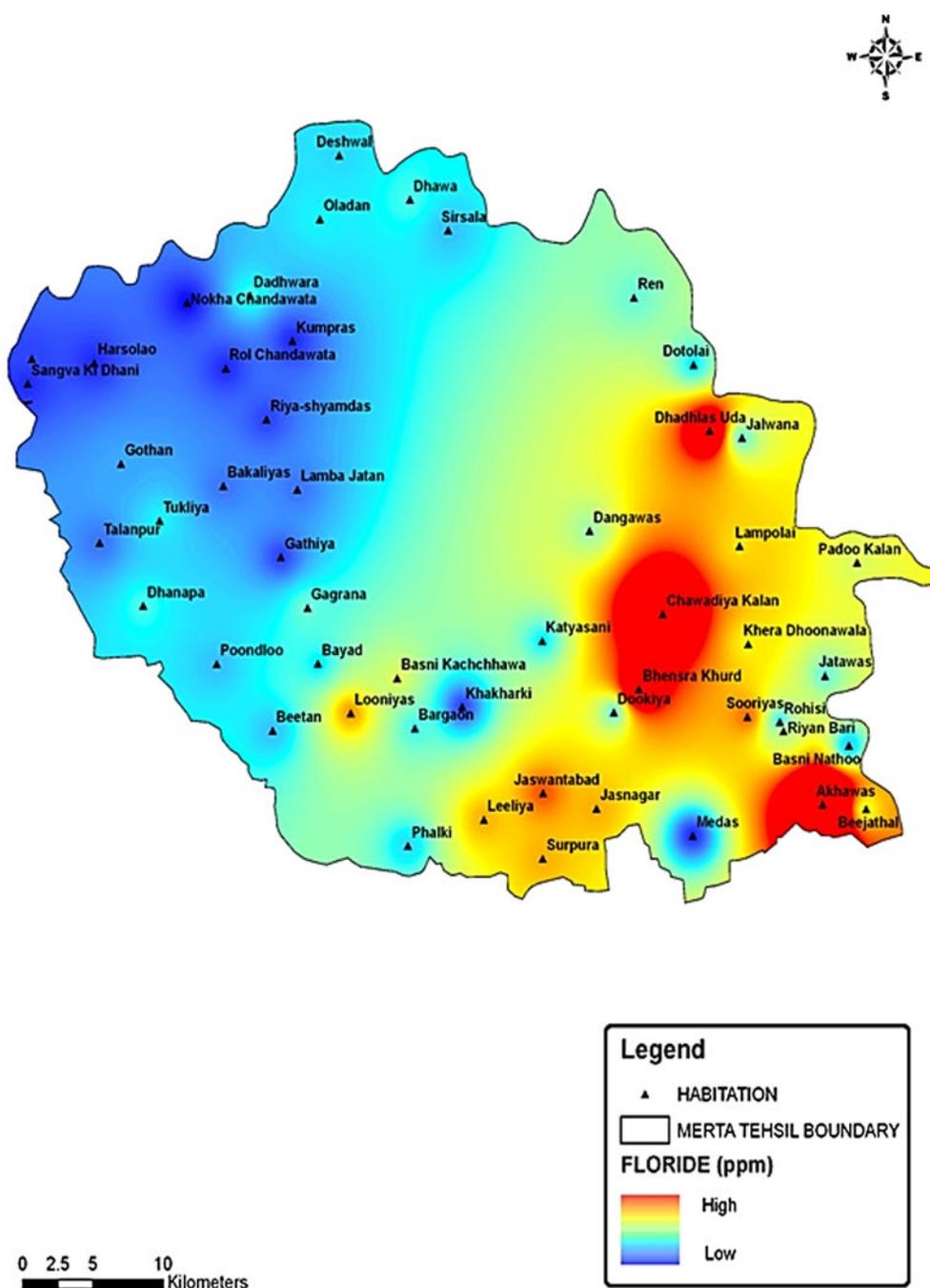
4.1. Technical fluoride mitigation practices introduced in area

For fluoride mitigation in the area, two technical practices were introduced. First one is based on co-precipitation methods (Nalgonda technique) and the second is based on adsorption methods using Activated Alumina.

4.2. Co-precipitation method (Nalgonda technique)

In this method, Alum (Alumina Ferric) is used as a coagulant. The fluoride available in drinking water will get adsorbed on the floccs and settle down at bottom of pot. The supernatant water is collected in another pot and been used for drinking purposes. This method has very low cost and do not require any technical skills, but people in the area do not adopted this technique so far due to flowing reasons:

(1) It requires alum dose calculation based on Alkalinity and fluoride concentration in raw water. Addition of excessive alum leads to change in taste due to decrease in pH value,

(2) Monitoring of activated alumina for its working is difficult task, and

(3) It is a daily job to be carried out and requires at least 1.5 h.

4.3. Adsorption method (activated alumina technique)

In this method, granules of activated alumina are used. Activated alumina has a capacity to adsorb fluoride on its surface. After adsorption to a certain level, the further adsorption reduce/stops. In such condition, the activated alumina exhausted is recharged/regenerated by alkali and then neutralized by acid. Activated alumina may be regenerated 5–6 times and needs to be replaced. The kit was distributed by government on subsidized rate and NGOs were appointed to establish regeneration center and IEC. After all these, this technique was also not adopted due to following reasons:

(1) Activated alumina is costly, and government facilitate them one time and then they have to purchase from market and

(2) Regeneration is a difficult task and needs skilled persons. Once the NGO closed his regeneration center, the further use of technique stopped due to the exhaustion of media.

During the study of these, it was found that these techniques are not effective in the area; however, people were found to use traditional fluoride mitigation practices, and by using them, they are preventing/delaying fluorosis.

4.4. Traditional fluoride mitigation practices used in area

To mitigate fluorosis, persons in the area established some rules/practices by their experience of years and hence well proved. These practices are based on two basic principles:

(1) Selection of least contaminated source for drinking purposes and

(2) Change in dietary habits.

4.4.1. Selection of source

In the area with high fluoride in groundwater, people were found to use least contaminated sources without any knowledge of fluoride availability in water and its ill effects. On studying further, it was found that there are three ways to select least contaminated source:

(1) By the experience of years, local people categorized the source into ill source and healthy sources irrespective of fluoride examination in water. The ill sources are unsafe and if used results in illness regarding bones deformities. The healthy sources are safe and were used by community,

(2) Groundwater sources near to surface water bodies are being used by community. These sources also provide least contaminated water, and

(3) In some areas, surface water is being used by community after treatment. Surface water has no fluoride contamination and community saved themselves by fluorosis.

4.4.2. Change in dietary habits

In the area people have changed their dietary habits by increasing use of calcium rich products in daily diet. They were found to use more calcium products in daily diet. In some area there is a proverb regarding daily diet, "even eat some less but take a bowl of curd". Curd has a good quantity of calcium, and intake of calcium prevents from fluorosis even when high fluoride water has been used.

5. Conclusion

In recent years, there has been an increased interest in fluoride research because excess concentration fluoride in groundwater causes adverse impact on human health. In order to mitigate excess fluoride in groundwater, it is essential to determine and monitor the casual for the casual factors of

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enrichment of fluoride concentration groundwater in time and space. The facts of the above study indicate the distribution of higher concentration of fluoride ion in the eastern part of the block. The higher concentration of fluoride in ground water may be due to the deeper water levels in the aquifers. The increase in distribution of the samples and the frequency of sampling will still aid to get a

vivid picture on the variation in the concentration and spatial distribution of this ion. Regular intake of fluoride-rich waters seems to be the main cause for high incidence of fluorosis in the region. Dilution by blending, artificial recharge, efficient irrigation practices, and well construction are some common cost-effective fluoride control measures which can be adopted to improve the health status of the community.

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