Spatial Distribution and Health Risk Assessment based on Groundwater Fluoride Enrichment in Gaya, Bihar, India

Shaz Ahmad
Department of Civil Engineering, National Institute of Technology Patna, India
shaza.phd19.ce@nitp.ac.in

Reena Singh
Department of Civil Engineering, National Institute of Technology Patna, India
reena@nitp.ac.in
(corresponding author)

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ABSTRACT

Fluoride can be hazardous to the body if consumed in excess. Fluoride exposure in humans can occur through the consumption of contaminated groundwater. The purpose of this study is to determine the amount of fluoride present in the area under investigation and to determine the health risks associated with its ingestion by adults and children. In accordance with the method recommended by the Environmental Protection Agency, the assessment of the health risk was done using the Chronic Daily Intake (CDI) and Hazard Quotient ($HQ_{fluoride}$). The fluoride in groundwater varies from 0.37 - 2.70mg/L in the study area. According to the sampling results of all locations, the $HQ_{fluoride}$ value in adult males ranges from 0.38 to 2.77, 0.34 to 2.45 in adult females, and 0.41–3.0 in children, which is above the permissible limits for most of the sampling locations, indicating significantly higher health risks. The outcomes of this study could be beneficial for organizations in charge of promoting human health.

Keywords-Gaya; fluoride; spatial distribution; health risk assessment

I. INTRODUCTION

Globally, there is a substantial increase in anthropogenic, natural, and man-made contaminants in the water supply in rural regions. Many sources of effluents contribute to freshwater reservoir contamination, including household, agricultural, and industrial effluents, causing global freshwater scarcity [1]. Groundwater fluoride levels are also increasing due to several geological processes, such as geothermal springs, volcanic activity, tectonic processes, weathering, and other geological processes resulting from the contact of rock and water [2]. The mobilization of fluoride through the leaching of rocks and the over-utilization of groundwater is the root cause of fluoride contamination [3]. Moreover, several anthropogenic activities also contribute to fluoride production, namely coal combustion, the use of fertilizers containing phosphates, and cement manufacturing [4]. Industries like semiconductor production, coal-fired power plants, aluminum smelters, etc. emit wastewater enriched with fluoride [5]. The chemical weathering of underground minerals is more likely to cause fluoride contamination in arid and semi-arid regions. Weathering rocks and releasing volcanic particles in the atmosphere also contribute to groundwater fluoride levels [6], which in turn may contaminate agricultural fields [7].

Fluoride-contaminated groundwater is causing adverse effects on human health [8]. A significant amount of fluoride is accumulated in different parts of the body due to the intake of contaminated water [9]. Many areas suffer from fluoride consumption related crippling skeletal fluorosis. There is a possibility that fluoride can adversely affect the immune system of the human body [11]. Chronic kidney diseases have been shown to be accelerated by the continuous ingestion of fluoride [12]. The population eating fish and tea is more likely to be exposed to fluoride because these foods contain higher concentrations [13]. Since excess fluoride causes deter mental effects on human health, various defluoridation techniques have been developed. Out of various techniques, adsorption is cost effective and efficient. Various materials, such as nanomaterials [14], clay [15], chitosan [16], industrial waste [17], carbonaceous [18], alumina [19], calcium [20], and metal oxides [21] have been used as defluoridation adsorbents.

II. MATERIALS AND METHODS

A. *Study Area*

Gaya district covers a total area of 4976km^2 and lies between latitudes 24°30' and 25°06', and longitudes 84°24' and 85°30'. As a result of the continental monsoon, a hostile environment prevails throughout the studied region. Hills surround Gaya on three sides and a river on the other side, which causes the region to experience seasonal temperatures. In the summer months (May-July), the temperature ranges from 20.5°C to 44.5°C. The city receives about 214cm of rain annually between July and October. A temperature range of -4°C to 28°C is expected during winter. Sherghati is located in the Gaya district of Bihar, and the study was conducted in the surrounding area of Sherghati as shown in Figure 1.

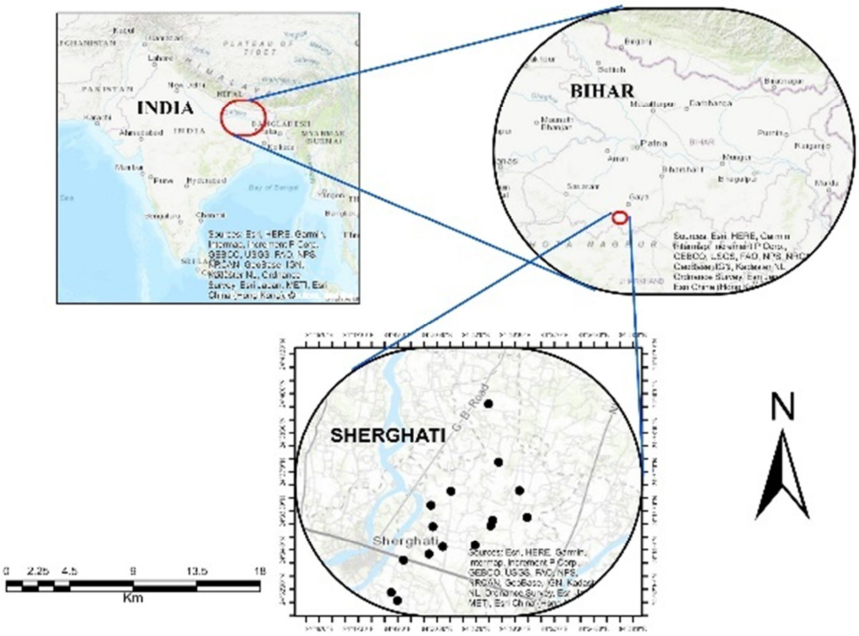

Fig. 1. Map of the study area.

B. *Sample Collection and Analysis*

An analysis of groundwater samples was conducted in 15 villages/towns in the Sherghati region of Gaya (Bihar). The fluoride concentration at each sampling site was determined by the average values of 5 samples taken from each site. Fresh groundwater samples were collected from shallow bore wells and private tube wells between March and May, 2022. The samples were collected using pre-cleaned high-density polythene 500mL bottles. A pair of gloves was worn when handling the sampling bottles to avoid contamination. Water was collected from the bore wells after 5 minutes of draining. The sample bottles were tightly capped to prevent contaminants from entering the bottle and were appropriately labelled. Samples were stored in iceboxes until they were transported to the laboratory, where they were preserved at a temperature of 4°C for further chemical testing. Each step of the sample collection, transportation, and analysis was carefully monitored to ensure that the samples were not contaminated or agitated. Fluoride concentration was determined using the SPADNS method described in APHA-AWWAWPFC (1994).

C. *Statistical Analysis*

The Inverse Distance Weighting (IDW) model was used to interpolate the variation of fluoride concentration in groundwater for the surrounding region of the study area using ArcGIS 10.6 application. The IDW model is a deterministic method to interpolate data, where the weight assigned to perceptual points is inversely proportional to their distances from each other. A decrease in distance will result in a greater weightage in the calculations and vice versa [22].

D. *Fluoride Health Risks*

Hazard Index (HI) values measure the non-carcinogenic risks associated with fluoridated groundwater for children and adults. These values are based on the results of studies conducted on these subjects. As outlined in the USEPA guidelines for assessing health outcomes, any hazard index up to 1 is considered to be within the allowable limit for health risk assessment [23]. Regarding non-carcinogenic risk, HI > 1 indicates a very high fluorosis risk to exposed people, whereas HI <1 indicates a reasonable fluorosis risk. HI<1 is considered acceptable for fluoride [24]. As a part of this analysis, the potency of fluoride in an individual is calculated by determining the Chronic Daily Intake (CDI) [25].

$$\mathrm{CDI} = \frac{\mathrm{IC} \times \mathrm{IR} \times \mathrm{EF} \times \mathrm{ED}}{\mathrm{BW} \times \mathrm{AT}} \quad (1)$$

where CDI is the chronic daily intake (mg/kg body weight/day), IC is the concentration of fluoride as determined by water samples taken from the ground (mg/L), IR is the daily drinking water intake rate (L/day), its value is presently taken as 4L/day for an adult male, 3L/day for an adult female, and 1L/day for a child [25]. EF refers to the total drinking water exposure frequency (365 days/year). ED refers to the duration of exposure which is taken as 64 years for adult males, 67 years for adult females, and 12 years for children [26]. BW refers to the average body weight (65kg for adult males, 55kg for adult females, and 15kg for children) [27]. AT corresponds to the average exposure time (i.e. 23,360 days for an adult male, 24,455 for an adult female, and 4380 days for children).

The present study aims to identify non-carcinogenic health risks resulting from drinking high fluoride contaminated groundwater, that can be estimated using the HQ estimates. The following equation was used:

$$HQ_{fluoride} = \frac{CDI}{RfD} \quad (2)$$

In the case of fluoride, a recommendation is 0.06mg/kg.BW/day as a human's oral reference dose (RfD) [25]. When the $HQ_{fluoride}$ value is above 1, it is considered potentially dangerous. In contrast, values of $HQ_{fluoride}$ less than 1 are regarded as acceptable considering non-carcinogenic risk to individuals.

III. RESULTS AND DISCUSSION

A. Fluoride Spatial Distribution

Among all sources of fluoride ingestion in the human body, drinking water is the most significant contributor. Based on the analysis of all samples, the results show that most of the study area is at high risk of fluoride contamination due to groundwater contamination. The value of fluoride in the study area lies between 0.37 and 2.70 mg/L, exhibiting elevated fluoride content, above the WHO-permissible limit (1.0mg/L). Among the sampled sites, Site-7 had the maximum fluoride level (2.70mg/L) and Site-9 had the minimum fluoride level (0.37mg/L). It is clear from the results of this study that the local groundwater of Sherghati region in the Gaya district is severely contaminated by fluoride due to the geology of the aquifers in this region, since fluoride is endemic in the Indian subcontinent and varies greatly from region to region. Based on these findings, the study categorized fluoride concentrations and their effects into 5 categories. Accordingly, the samples are listed in decreasing order of concentrations/classes: most samples (46.7%) were collected in Class-2 (0.5–1.5mg/L), followed by Class-3 (1.5–4.0 mg/L) with 40%, and Class-1 ($0.5mg/L^{-1}$) with 13.3%. No samples were found in Class-4 (4.0 -10.0mg/L) and Class-5 (> 10 mg/L). The spatial variation of fluoride is shown in Figure 2.

B. Evaluation of Health Effects

CDI for adult males ranged from 0.02 to 0.17mg/kg of body weight/day. The highest CDI value is recorded at Site-7 (0.17) and the minimum at Site-9 (0.02). CDI varied from 0.02 to 0.15 for adult females in this study area, with maximum at Site-7 (0.15) and minimum at Site-9 and Site-14 (0.02). A child's exposure to fluoride usually results in a greater risk than to an adult. Children have a smaller body size, which allows them to accumulate more contaminants per body weight. Children in this region were found to have a CDI value between 0.02 and 0.18. The highest CDI is observed at Site-7 and lowest Site-9. $HQ_{fluoride}$ was calculated for each sampling site based on the sampling results. It was found that the $HQ_{fluoride}$ ranged between 0.38 and 2.77 for adult males. The highest $HQ_{fluoride}$ for an adult male was found at Site-7 (2.77) and the minimum at Site-9 (0.38). The $HQ_{fluoride}$ level for adult females varied from 0.34 to 2.45, with the highest found at Site-7 and the minimum at Site-9. For children, the $HQ_{fluoride}$ varied from 0.41 to 3.0, with the highest value observed at Site-7 and the lowest at Site-9. The CDI and HQ values are shown in Table I. A comparison of the HI values for fluoride in 3 different age groups is shown in Figure 3. The result shows that the mean HI values for children, teenagers, and adults in the present study were greater than the safe limit (HI <1). In accordance with previous studies, it is concluded that children whose HI values exceeded the acceptable threshold were at the most significant risk of fluoride toxicity.

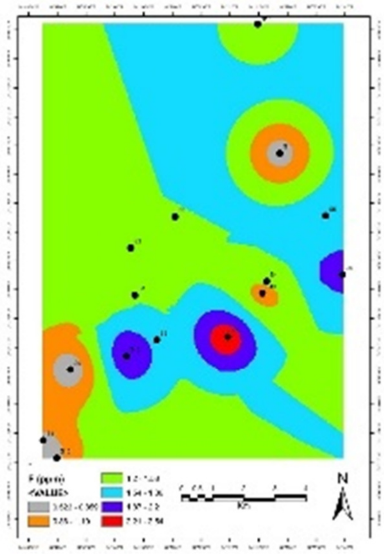

Fig. 2. Spatial variation of fluoride in the study area.

TABLE I. CDI AND HQ VALUES

Site label	F^- (ppm)	CDI_{male} (mg/kg body weight/day)	CDI_{female}	CDI_{child}	HQ_{male}	HQ_{female}	HQ_{child}
Site-1	1.48	0.09	0.08	0.1	1.52	1.35	1.64
Site-2	1.41	0.09	0.08	0.09	1.45	1.28	1.57
Site-3	1.53	0.09	0.08	0.1	1.57	1.39	1.7
Site-4	1.4	0.09	0.08	0.09	1.44	1.27	1.56
Site-5	1.68	0.1	0.09	0.11	1.72	1.53	1.87
Site-6	2.17	0.13	0.12	0.14	2.23	1.97	2.41
Site-7	2.7	0.17	0.15	0.18	2.77	2.45	3
Site-8	1.75	0.11	0.1	0.12	1.79	1.59	1.94
Site-9	0.37	0.02	0.02	0.02	0.38	0.34	0.41
Site-10	0.72	0.04	0.04	0.05	0.74	0.65	0.8
Site-11	0.64	0.04	0.03	0.04	0.66	0.58	0.71
Site-12	2.34	0.14	0.13	0.16	2.4	2.13	2.6
Site-13	0.83	0.05	0.05	0.06	0.85	0.75	0.92
Site-14	0.42	0.03	0.02	0.03	0.43	0.38	0.47
Site-15	1.24	0.08	0.07	0.08	1.27	1.13	1.38

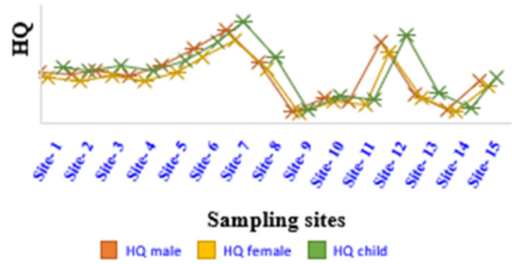

Fig. 3. HQ comparison in the study area.

C. Health Effects of Fluoride

Fluoride is an essential micronutrient that is present in drinking water as well as in various food sources. Although fluoride benefits the body, it can also have detrimental effects. Fluoride concentration in drinking water ranging from 0.6 to 1.2mg/L benefits bone and tooth development [29]. Fluoride can cause serious health problems in infants, children, and adults if the amount accumulated in the body exceeds the permissible limit. A person will not be affected instantly by fluoride, but it will accumulate in the brain and slowly deteriorate the body's ability to function. Fluoride exposure over a long time is associated with widespread health issues [30]. Fluoride plays a vital role in protecting the newly formed teeth of children, and it plays an equally important role in preventing tooth decay in adults. It has been shown that more than 1.5mg/L of fluoride can adversely affect bone homeostasis (skeletal fluorosis) and enamel growth (dental/enamel fluorosis). Nearly 65% of the total cases of endemic fluorosis are caused by water containing excess fluoride [31]. Initially, dental fluorosis can be described by enamel discoloration, which may turn into discrete or aggregate pitting [32]. Fluoride concentrations greater than 1mg/L may cause neurotoxicity, which can interfere with memory and learning. Developing brains are more susceptible to toxicants than mature brains. The presence of fluoride in groundwater above 10mg/L accelerates the occurrence of neurological problems, hypertension, and cancer. Children exposed to fluoride may also suffer from obesity [33]. Other researchers have reported that there is either no effect or a negative correlation between fluoride and obesity [34]. However, the results of these studies are still inconclusive.

It is suggested that 80% of all diseases worldwide are caused by poor water quality. In arid and semi-arid regions, 65% of endemic fluorosis is caused by drinking water with high fluoride levels [35, 36]. Excessive fluoride consumption contributes to tooth decay and bone deterioration [37]. Fluoride intake has also been shown to be positively correlated with children's intelligence levels. Children consuming less fluoride had lower intelligence test scores than those consuming water with greater fluoride [6]. Fluoride has been identified as a carcinogen in some epidemiological studies. There is, however, some evidence that fluoride can cause osteosarcoma (bone cancer) [38, 39]. In children aged between 6 and 8 years, fluoride-caused cancer is more prevalent than in adults [39]. Neurotoxicity can be caused by fluoride concentrations between 2 and 4mg/L [40]. Many researchers have reported that the fluoride concentration adversely affects children and their thinking ability, more that it does to adults. A similar test was conducted to check the IQ of children in the region affected by fluoride and found lower Intelligence Quotient (IQ) scores in individuals who consumed more fluoride. Moreover, fluoride negatively impacts reaction times and visuospatial skills [41]. Those suffering from kidney diseases have a greater risk of developing fluorosis, even if their drinking water contains a permissible amount of fluoride, because their kidneys do not excrete fluoride as effectively. Additionally, it has been shown to affect the reproductive system as well. Fluoride in groundwater/drinking water appears to have a tremendous impact on human health, and its occurrence and distribution have received worldwide attention.

IV. CONCLUSION

There is a chronic fluoride contamination of groundwater in Sherghati, Gaya district. The purpose of this study is to investigate the clinical effects of the chronic exposure to fluoride. Clinical effects consisted of abnormal tooth enamel in children, joint pain and deformities of the limbs and spine in adults. Fluoride slowly accumulates in the body and may damage important organs in a longer stretch of time. A high level of chronic fluoride toxicity has been reported in Sherghati. Fluoride-safe water, education, and awareness of the dangers of fluoride toxicity are all needed in order to tackle the current situation. According to the study results, the area is considered high-risk for fluoride-related problems and requires considerable attention.

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