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Health risk assessment of nitrate and fluoride in ground water of Panchkula Haryana

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Abstract: High nitrate and fluoride contamination in groundwater cause a variety of disorders, including Methemoglobinemia, teratogenesis, and dental and skeletal fluorosis. The present work assesses the non-carcinogenic health risks posed by nitrate and fluoride in infants, children, and adults using the daily water intake (CDI), hazard quotient (HQ), and non-carcinogenic hazard index (HI). Groundwater samples were collected from various wells and boreholes in (Southern and western parts) of the Panchkula district study areas for nitrate and fluoride analysis using ionic chromatography and fluoride selective electrode, respectively. The nitrate concentration in groundwater samples varies from 2 to 200 mg/L, of which 33.33% are above the permissible limit of 45 mg/L for drinking water. The fluoride concentration ranges from 0.2 to 5.0 mg/L, with 28.57% of the samples exceeding the safe value of 1.5 mg/L for drinking water.
Correlation plots demonstrate that the potential of hydrogen (pH), electrical conductivity (EC), total dissolved solids (TDS), Na and HCO_3 are positively correlated with F^-. The Gibbs diagram demonstrates that the interaction between rock and water impacts the groundwater chemistry. Using the method of the United States Environmental Production Agency (USEPA), this study assesses the non-carcinogenic health risk posed by nitrate and fluoride in different age groups (infants, children, and adults). Immediate attention and remedial measures must be implemented to protect residents from the adverse effects of F^- in the study area.

Index Terms – **Nitrate, fluoride, hazard index, groundwater, Panchkula, Haryana.**

I. INTRODUCTION

Particularly in arid and semi-arid areas with limited surface water and precipitation groundwater is an important resource for irrigation and drinking [1]. Groundwater is in greater demand every day due to population growth and a shortage of substitute water sources. For their daily water needs an estimated 2.5 billion people worldwide rely primarily on groundwater [2, 3]. As most people in rural areas believe that drinking groundwater is safe most of them rely on either public or private wells to supply them with water. But the alluvial plains shallow groundwater is susceptible to contamination from a variety of sources. Groundwater pollution is mostly caused by anthropogenic activities such as unscientific effluent discharge excessive fertilizer uses fast industrialization and urbanization and climate change [4]. Other processes that impact the quality of groundwater are precipitation and dissolution of minerals biological activity cation exchange during oxidation or reduction and rainfall composition [5,6]. Furthermore, excessive reliance on groundwater resources has accelerated the weathering of rocks leading to a greater degree of geogenic contamination of groundwater [7]. Long-term consumption of highly contaminated groundwater whether it be organic or inorganic could be detrimental to the health of those living close by [8]. Nitrates and fluorides in groundwater may have detrimental effects on human health. Because of their terrible impact on human health these two toxic ions have drawn attention from all over the world despite the USEPAs listing that they are not carcinogens.
An inorganic ion that is produced naturally as a component of the nitrogen cycle is nitrate (NO_3^-). It is a nutrient that is mostly applied as fertilizer to plants. Human activity is considered to be the main cause of nitrate in groundwater [9,10]. Although there are numerous routes by which nitrates can enter groundwater point and non-point sources account for the majority of them. The most frequent point sources—septic tanks dairy lagoons percolating wastewater effluents and animal waste—through which (NO_3^-). can enter groundwater [11,12]. Applying manure pesticides or fertilizers is one example of a non-point source that can cause elevated nitrate levels in groundwater [13]. The weathering of tobelite nitrate-bearing and nitrite-containing rocks can naturally release nitrates into the groundwater through the interaction of rock and water [14]. Furthermore, legume plants and microbes' nitrogen fixation can also result in nitrate contamination of groundwater [15]. Since plants obtain their nitrogen from nitrate an oxidized form of dissolved nitrogen fertilizers are widely used in agricultural land areas. Soil loses its inherent capacity to hold water when it is heavily farmed. Fertilizers containing nitrogen are frequently used to restore depleted soil nutrients. However, these nitrates are harmful to human health when they get into surface and groundwater waters and the food chain. The Bureau of Indian Standards (BIS) suggested that 45 mg/L be the highest amount of (NO_3^-). that can be legally present in drinking water. High nitrate levels in water can cause Methemoglobinemia or blue infant disorder in infants younger than six months of age. Thyroid dysfunction hypertension non-lymphoma Hodgkins lymphoma stomach tumors and colorectal cancer are among the adult cancer risks that are increased by it [17-22]. In India more than 108.2 million people drink water that has more nitrate than the 100 mg/L allowable limit [18].
Another dangerous material that has the potential to harm both humans and other living things is fluoride (F^-). It is mainly present as a free ion in natural waters. Groundwater typically contains fluoride that has been artificially added or naturally occurring [19].

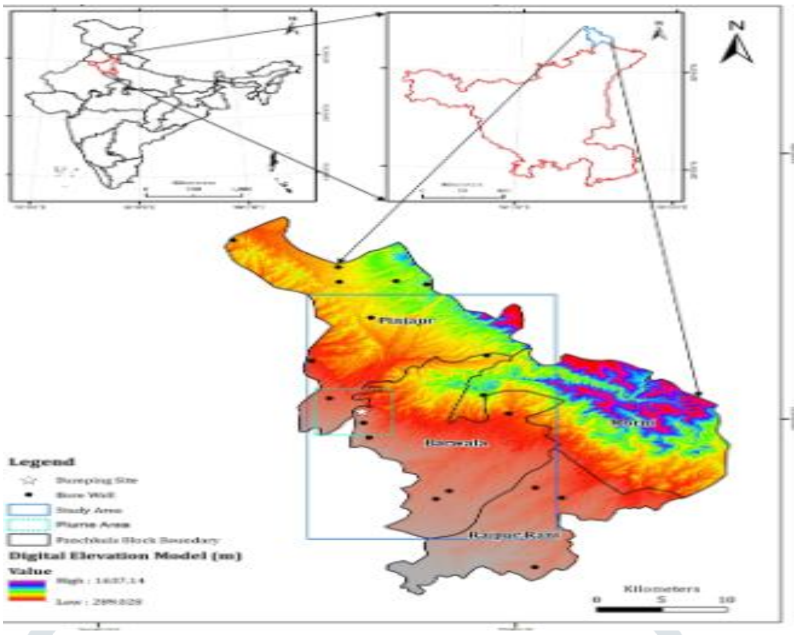
Natural sources of (F^-) include minerals that contain fluoride such as muscovite hornblende biotite amphiboles apatite and fluorite [20, 21]. In granitic terrain these minerals are prevalent [22]. Fluoride is released into the soil when the previously mentioned minerals dissolve. Fluoride is therefore released by this process and infiltrates the groundwater through soil moisture or precipitation. Climate increased bicarbonate pH and sodium concentrations water temperature and host rock composition (the availability of minerals containing fluoride) are some of the many variables that affect the amount of naturally occurring fluoride in groundwater [23]. Among the man-made sources are phosphatic fertilizer factories brick and steel production coal combustion sewage overflow excessive groundwater withdrawal etc. are the main reason why groundwater has higher fluoride levels [24, 25]. The World Health Organization (WHO) considers drinking water with fluoride levels of 0.6 -1.5 mg/L to be safe. Dental caries can develop when the concentration of (F^-) in drinking water is lower than the advised level (0. 6 mg/L). Fluoride concentrations in drinking water above 1. 5 mg/L however have the potential to cause fluorosis (skeletal and dental) [26–28]. Moreover, a high fluoride diet has been connected to thyroid issues osteoporosis arthritis infertility and abortion [29, 30]. About 200 million people worldwide suffer from fluorosis [31].

Numerous reports have been published regarding non-carcinogenic health issues in China [19, 32] India [33–35] Iran [36 37] Mexico [38] and Pakistan [39–40]. Most Indian states have conducted a number of studies on non-carcinogenic health risks. Nawale and associates [41] looked at the health risks that aren't cancer-causing because of the high levels of fluoride and nitrate contamination in the groundwater supplies of the Wardha sub-basin in central India. Duvva & Co. [42] found that the groundwater in Telangana state had higher than average concentrations of (NO_3^-) and (F^-). In addition, they assessed the health risk linked to elevated levels of (NO_3^-) and (F^-) discovering that children in the research region are more susceptible to health issues than adults. Likewise, in northern Indias state of Punjab Singh et al. Children are more susceptible than adults according to research [43] on the possible non-carcinogenic effects of (NO_3^-) and (F^-) on human health through groundwater ingestion. The people in the central and northeastern regions have a higher non-carcinogenic health risk according to Raja and Neelakantans [44] evaluation of the non-carcinogenic hazards posed by (NO_3^-) and (F^-) in the groundwater of Ramanathapuram South India. Jandu and others. [45] investigated the fluoride and nitrate sources in Rajasthans groundwater as well as the associated non-carcinogenic health risk. The primary sources of high nitrate and fluoride levels in groundwater respectively were determined to be anthropogenic and geogenic sources. Moreover, they disclosed that the assessment of non-carcinogenic risks for women men and children went beyond the permitted threshold.

Coimbatore vast textile industry has earned it the moniker Textile City of South India. Ground-water quality in this area has declined as a result of rapid urbanization industrialization intensive farming and mining activities. Textile effluents and sewage are the main sources of groundwater contamination in urban areas [46]. Kom and associates. [47] investigated the chemistry of the groundwater in the current study region and its suitability for agriculture and drinking. The study also found that groundwater system disruption by humans is compromising water quality. In the Tamil Nadu district of Coimbatore Kumar [48] investigated groundwater fluoride contamination and the associated health risks to people. As per the studys findings there is a high concentration of (F^-) in groundwater due to both natural and anthropogenic factors and there are no carcinogenic risks for individuals of any age group. Karunanidhi along with colleagues. examined the effects of fluoride and nitrate contamination on human health at the Sulur Taluk (in the eastern part of the current research area) in [49]. In this research area however no thorough investigation on the non-carcinogenic risks associated with (NO3-) and (F-) contamination of groundwater has been published. The primary goals of this research are to identify the spatial distributions of fluoride and nitrate assess the non-carcinogenic health risk for adult's children and newborns and provide information about the hydro-chemical composition that controls groundwater chemistry.

II. STUDY AREA

Panchkula district is located in Northern part of Haryana (India) and lies between 30°26' - 30°55' N latitudes and 76°46' - 77°10' E longitudes. Total geographical area of the Panchkula district is 898 sq. km. and it is divided into four development blocks namely Barwala, Pinjore, Raipur Rani and Morni. The Ghaggar River is the main river flowing in the region. There are sufficient ground and surface water sources in Panchkula district, however; groundwater is a major source of irrigation in this area. The surface elevation of Panchkula district varies from 289.028 m to 1637.14 m above sea level (a.s.l.). The location of study area, dumping site, and bore-wells are shown in Figure 1. SRTM (Shuttle Radar Topography Mission) Elevation data is used for preparation of elevation map, which has an accuracy of ±15 m at 30 m spatial resolution (NASA-JPL, 2013). The Panchkula dumping site is located at 30°40'50.84" N latitude and 76°52'54.64" E longitude and lies at an elevation of 266 m a.s.l. The solid blue line represents the study area for which the analysis has been carried out using many bore-well dataset and the dotted green line shows the plume area for which contaminant movement is finally shown in results.



III. MATERIALS AND METHODS

3.1. Sampling and analysis

To investigate quality and health concerns related to (NO_3^-) and (F^-) in Panchkula Haryana India we collected a variety of groundwater samples from various locations. In the study area the samples were taken in May and June from a variety of bore wells. The samples were collected using 1.0 L Polyethene bottles. The wells were purged for approximately 10-15 minutes prior to sample collection in order to prevent the negative consequences of standing water. Following sample collection, the potential of hydrogen (pH) total dissolved solids (TDS) and electrical conductivity (EC) of the samples were measured using a portable digital metre (HANNA HI 9828). Samples of groundwater are collected transported and analyzed according to a strict protocol that is advised by the American Public Health Association [50]. Each sample was labeled and kept in a refrigerator at 4ºC prior to chemical analysis. The EDTA titrimetric method was used to estimate the concentrations of total hardness (TH). By using standard solutions of H_2SO_4 and $AgNO_3$ respectively the titrimetric technique was used to measure the levels of chloride (Cl). Fluoride was measured using an ion-selective electrode and NO_3 concentration was estimated using a UV-visible spectrophotometer. Using Microsoft Excel, the chemical data was analysed and ArcGIS 10.5 was used to create the base map and spatial variation maps.

3.2. Health risk assessment (HRA)

The highly toxic chemical concentrations in our drinking water and the ensuing effects on human health can be found using the effective probabilistic technique known as HRA. The USEPA is the organization that created this method [51]. Over the past 30 years it has been extensively utilized to evaluate the non-carcinogenic health risk of contaminants in groundwater [52–56]. Individuals may experience major health issues as a result of drinking tainted water [57]. Nitrate and fluoride are included in the non-carcinogen category of toxic substances while carcinogenic substances are classified as such by the USEPA. However direct consumption and skin contact with water tainted with non-carcinogens can pose a health risk to humans [58]. The most important exposure pathway is oral ingestion [59]. Thus, this study assessed the risk to human health that nitrate and fluoride posed via the oral pathway for three distinct age groups: children (2–16 years) adults (>16 years). The health risks associated with drinking water that are not carcinogenic are calculated using the following formula [60].

$CDI = C \times IR \times F \times ED/BW \times AT$

Where CDI represents the chronic daily intake (mg/kg × day) of (NO_3^-) and (F^-) via oral ingestion, C is the concentration of (NO_3^-) and (F^-) in groundwater in mg/L, IR signifies the ingestion rate, F indicates the exposure frequency, ED denotes the exposure duration, BW represents the average body weight, and AT denotes the average exposure time.

$HQ = CDI/RfD$

Where RfD denotes the reference dose (NO_3^- = 1.6) and (F^- = 0.04 mg/kg × day) [55]. Moreover, the THI values were evaluated using equation (3) to understand the overall non-carcinogenic hazards caused by nitrate and fluoride.

$THI = \Sigma\, (HQ_{\text{fluoride}} + HQ_{\text{nitrate}})$

THI <1 indicates no health risk to humans, while THI >1 signifies a higher level of hazard [61,62].

IV. RESULT AND DISCUSSION

4.1. Physico-chemical parameters of groundwater

Groundwater resources can be studied using hydro-geochemical methods to identify the processes that influence the groundwater chemistry [72]. The hydro-chemical analysis shows that the pH values of groundwater samples vary from 7.32 to 8.7 (average 8.0), indicating alkaline groundwater. Around 23.81% of the samples have a pH value above the BIS permissible limit. Long-term consumption of high pH water causes skin, eye, and mucous membrane irritation. EC concentration is between 250 to 3500 µS/cm, (average 1200 µS/cm). About 56.67% of the groundwater samples are classified as saline water, 11.05% fall into the permissible category, and 10.29% are classified as brackish water. Furthermore, the EC value in 61.43% of samples exceeds the WHO recommended limit of 1500 mS/cm. However, based on the EC classification by Todd (1980), 11.90%, 28.57%, 16.67%, and 42.86% of the samples are classified as good, permissible, doubtful, and unsuitable category, respectively. The EC values may rise in groundwater with the increase of temperature and available geogenic salt. Total dissolved solids (TDS) varied from 160 to 2240 mg/L (mean 768 mg/L), with 13.33% of the samples exceeding the BIS standard value (1500 mg/L). The TDS classification based on Davis and De Wiest shows that 9.52%, 30.95%, 40.48%, and 19.05% of groundwater samples are fall under the desirable for drinking (TDS<500), permissible for drinking (TDS between 500 and 800), useful for irrigation (TDS between 800 and 1500) and unfit for drinking and irrigation (TDS>3000) categories, respectively. The groundwater with high TDS causes intestinal annoyance, kidney stones, heart problems, bad taste, and bad odor. The concentration of total hardness (TH) in the samples ranges between 120 and 1200 mg/L (mean 480 mg/L). Based on Sawyer and McCarty's classification, 9.52% and 90.48% of samples are hard and very hard. The analysis reveals that 25% of samples exceed the BIS standard limit of 600 mg/L for drinking water. The intake of high TH causes body tissue calcification and kidney diseases. The result shows that all samples are within the WHO-recommended calcium limit of 200 mg/L. Coloectal cancer, kidney stones, osteoporosis, and hypertension can be caused by excessive calcium consumption in drinking water. Chloride is the highest concentration among all the anions. It diverges from 21 to 1000 mg/L (averaging 200 mg/L) in the samples, indicating 8.03% of the samples exceed the drinking water limit of WHO (600 mg/L). High concentrations of Cl in groundwater are caused by anthropogenic sources like household waste, landfills, septic tank leaks, and agricultural fertilizers. Another potential source of Cl contamination in the groundwater is the abundant occurrence of clay minerals. Excessive chloride consumption can result in osteoporosis, hypertension, and asthma. Concentrations of Sulfate exhibited a wide range of 10-500 mg/L (mean 120 mg/L). The analysis shows that only 2.76% of samples have sulphate concentrations higher than the BIS recommended level (200 mg/L) for drinking water. However, the mineral sulphate can be found in all types of rock: igneous, sedimentary, and metamorphic, in the form of naturally occurring minerals such as metallic sulphide.

4.2. Nitrate in groundwater (non-carcinogen)

A broad range of nitrate in the groundwater with an average of 40 mg/L and a range of 2 to 200 mg/L. The BIS (2011) states that the maximum amount of nitrate in water is 45 mg/L. Furthermore, the risk of health problems increases dramatically with nitrate levels between 45 and 100 mg/L and higher than that it becomes dangerous. According to the study 15% of the samples were higher than the suggested amount. Though the natural process of contamination is substantially slower than that in orogenic and anthropogenic sources of NO_3 groundwater can be found. The main man-made sources are urbanization septic tank leaks industrial outlets municipal sewage leachate from landfills and agricultural fertilizers. That proves that the eastern and central regions have the highest NO_3 concentration. Plains which are populated mostly by farmers are the areas with the highest NO_3 levels. Since nitrate is carried to the soil by infiltration there may be an inverse relationship between groundwater nitrate concentrations and rainfall. Low rainfall periods can cause undiluted irrigation return flow to raise nitrogen concentrations. Overconsumption of nitrates can be harmful particularly to young children. In newborns the most prevalent syndromes are congenital disabilities and Methemoglobinemia also known as blue baby syndrome. This syndrome is more likely to develop in infants who are fed milk that has high NO_3 levels in it. However adult goiter high blood pressure and stomach cancer are all potential side effects of nitrate contamination in drinking water. Furthermore, the risk of complications is increased in pregnant women who consume large amounts of nitrate-rich groundwater. On the other hand, tissues muscles and bones may become dysfunctional due to low nitrate concentration.

4.3. Fluoride in groundwater

A range of 0.2-5.0 mg/L of Fis found in the samples. According to the WHO (2011), the threat of dental carries rises when the level of Fis less than 0.6 mg/L in drinking water. However, when it stays between 0.5 and 1.5 mg/L (the accepted level), there is no evidence of a health issue. The level of F above 1.5 mg/L is associated with the development of fluorosis. Fluoride in groundwater is primarily derived from the weathering and dissolution of fluoride-bearing minerals, such as fluorite, biotite, apatite, hornblende, and amphibole. These minerals are common in granitic and gneissic terrains, and the geogenic source contributes vast quantities of F to groundwater. The elevated fluoride regions (central, southern, and eastern regions) are dominated by granitic gneiss, charnockite, granite and hornblende- biotite gneiss. Percolation of rainwater through these weathered rocks may contaminate the groundwater with excessive fluoride. It is possible to replace fluoride ions from fluoride bearing minerals with hydroxyl ion (OH) under alkaline conditions.

4.4. Evaluation of non-carcinogenic health risk

The groundwater becomes the most important source to meet the daily requirements in the study area. However, the most prevalent contaminants and toxicants like NO_3 and F^- are present in groundwater samples at concentrations of 15% and 20.57% over the BIS recommended limits. Drinking this contaminated groundwater can be hazardous to human health. As a result, this study examined the health effects of non-carcinogenic contaminants (NO_3 and F^-) on infants, children, and adults, with water consumption as a key exposure pathway. The HQ nitrate concentrations vary between 0.12 and 5.56 (mean 1.95), 5.56 and 2.78 (mean 0.67-1.33), for children and adults, respectively. The results of HQ nitrate show that the exceedance rates are 48.14% and 16.29% for children, and adults, respectively. On the other hand, the HQ fluoride ranges from 0.2 to 5.02 (mean 1.82) 0.29 to 4.17 (mean 0.84) for children, and 0.25 to 0.85 (mean 1.63) for adults. About 80.48% of groundwater samples for 75.71% for children, and 40.24% for adults surpassed the safe limit (HQ fluoride), suggesting the possibility of non-carcinogenic human health hazards. The total hazard index (THI) of nitrate and fluoride ions on human health was evaluated using equation. The recommended safe limit for THI is 1.0, and higher than this specified limit is considered to pose a non- carcinogenic risk to humans. The computed THI values indicate that about 87.86%, and 68.81% of children, and adults, are at risk of non-carcinogens.

Furthermore, the results reveal that infants are more prone to non-carcinogenic causing health hazards than children and adults, which may be due to the lower body weight of children than adults. Based on the intensity of health risk, the study area is primarily categorized into two zones, namely the safe zone (0<THI1) and vulnerable zone (THI>1). Furthermore, the vulnerable zone is subdivided into three: (i) THI between 1 and 2 as moderate health risk zone, (ii) THI between 2 and 3 as high health risk zone, (iii) THI higher than 3 as very high health risk zone. The spatial maps of THI for infants, children and adults show that the safe zone regarding non-carcinogenic health hazards is mainly located in the western and northern portions of the research area. However, the vulnerable intensity zones are concentrated in the southern and central parts. This result corroborates the NO_3 spatial distribution map of NO_3 and F ions in which high and very high levels of NO_3 and F are found in the THI vulnerable intensity zones and vice versa. Similar findings were reported globally by several researchers. A study by Adimalla et al. found that infants and children are more vulnerable to the non-carcinogenic health risks of NO_3 and F contamination in Telangana, India. Karunanidhi et al. evaluated the health hazards posed by nitrate and fluoride in groundwater of Shanmuganadhi basin, South India. They concluded that 87% and 69% of the samples surpassed the standard limit (THI=1) for children and adults, respectively, which indicates that children are more prone to non-carcinogenic risks than adults. Su et al. investigated fluoride and nitrate contamination and the potential health risks in Jiaokou district, China. They found that non-carcinogenic health hazards of NO_3 and F^- are more susceptible to infants and children than teenagers and adults. In another similar study, Qasemi et al. evaluated fluoride and nitrate contamination in the groundwater of Sabzevar city, Iran, and found higher health risks for children and infants than adults.

The present study recommends the following points to improve the quality of drinking water and minimize potential health hazards: (i) a proper water management plan like reverse osmosis treatment, (ii) adequate construction of landfills and septic tanks, (iii) avoiding the excess use of fertilizers and pesticides, and (iv) construction of artificial recharge systems.

V. CONCLUSIONS

The present work highlighted the non-carcinogenic risk of drinking nitrate and fluoride-contaminated groundwater collected from Panchkula Haryana. Our findings indicate that none of the 26 studied boreholes had nitrate levels above WHO guidelines (50.00 mg/L). Differently, 13 out of 36 areas had fluoride levels exceeding the acceptable limit for drinking water (1.5 mg/L). The USEPA method was used to assess the non-carcinogenic health hazards associated with NO_3^- and F^- exposure from drinking water consumption. The ranges of HQ nitrate are 0.12 and 5.56 (mean 1.95). The average values of the HI for adults, and children, were 5.56 and 2.78 (mean 0.67-1.33), for children and adults respectively. According to the USEPA method, the water samples had HI values exceeding the safety level for children, and adults at 75.71% 40.24% respectively. Children are more vulnerable to non-carcinogenic health risks due to consumption of groundwater than adults. In the future, it would be helpful to study different methods of removing fluoride and nitrate from groundwater, choose the best method for a particular location, and enhance drinking water quality for the local population.

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