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Groundwater Fluoride Contamination and Probabilistic Health-risk Assessment in Fluoride Endemic Areas of the Lower Satluj Basin, Punjab, India

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Groundwater fluoride contamination and probabilistic health-risk assessment in fluoride endemic areas of the Lower Satluj Basin, Punjab, India

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Abstract

Groundwater fluoride contamination in some parts of the study region was long recognized as a water quality issue. The results of WQI showed that 85% and 15% of sampling sites fall within the class of "good" and "poor" quality, respectively. Non-carcinogenic health risk (NCHR) assessment (using Hazard quotient (HQ)), and sensitivity analysis for three age groups were also carried out using the Monte Carlo simulation technique. The estimated levels of HQ_{oral} were greater in magnitude than those estimated from HQ_{dermal}, thus the main source for fluoride toxicity is oral ingestion. Amongst the three age groups studied, children >female >male were found to be more prone to NCHR with HQ_{oral} ranging from (0.13-5.45), (0.07-2.97), and (0.06-2.51), respectively. Sensitivity analyses indicated that fluoride concentration, ingestion rate, shower exposure time, and fractional skin contact with water were the most relevant variables in the model to reduce the potential health effect.

Keywords: Dermal absorption; Endemic; Fluoride; Health risk; Oral ingestion

Highlights

1. No studies in the region deal with fluoride contamination and its assessment of human health risks.
2. The WQI of 15% of the sampling locations have fluoride above acceptable limits (>1.5 mg/L) and falls under "poor" water class.
3. Deterministic risk assessment of fluoride exposure was carried out for three age groups ($HQ_{Fluoride:}$ children>female>male).
4. Based on the sensitivity analysis, fluoride concentrations, daily ingestion rate, fractional surface skin contact with water, and shower exposure time were the most relevant parameters.
5. Public education and understanding of the impact of water quality and fluorosis on human health should be high.

Introduction

Groundwater is the world's most valuable resource and plays a key role in socio-economic development. Due to less rainfall and shortage of surface water supplies, significant areas of rural communities, particularly in semi-arid regions, rely on this source for drinking, irrigation, and industrial purposes. In the last few decades, it is constantly being over-exploited and contaminated by the rapid increase in anthropogenic activity in the agricultural and industrial sectors (Chaudhry *et al*. 2019). Geogenic activities also result in groundwater contamination. Groundwater contaminants cover a broad range of physical, chemical (organic and inorganic), radioactive, and bacteriological parameters. When contaminated, groundwater remains in an unusable state for quite a long time or even for centuries. So, the concerns of groundwater contamination have gained the attention of social activists and researchers worldwide.

Recently high fluoride contamination in groundwater is becoming a major global issue and has been reported from various parts of the world (Kundu and Mandal 2009; Edmunds and Smedley 2013). Natural groundwater concentrations of fluoride are documented to be high (>30 mg/L) in more than 30 countries, including India (Ahada and Suthar 2019). Over 0.2 billion people use fluoride-concentrated drinking water having concentrations above the acceptable limits (>1.5 mg/L as per World Health Organization (WHO)) in over 20 industrialized and developing countries including India (WHO 2004; Amini *et al.* 2008;

Vithanage and Bhattacharya 2015). Around 80 percent of the world's diseases are due to poor drinking water quality, 65 percent of which are due to endemic fluorosis (Brindha and Elango 2011; Ali *et al.* 2016). Geogenic processes such as rock-water interaction, the accumulation of volcanic particles in the atmosphere, leaching of fluoride-bearing minerals, and marine aerosols often add fluoride to the groundwater (Gleeson *et al*. 2012; Karunanidhi *et al.* 2020). Major anthropogenic processes [Figure 1S] include climate change, irrigation of dry land without sufficient drainage, increase in the use of phosphate fertilizer, animal manures, domestic and industrial wastewater discharge on the surface, automobile exhaust increase the fluoride contamination in groundwater (Genxu and Guodong 2001). Fluoride's effects on human health, however, are not immediate but chronic, so long-term exposures to high doses of fluoride are obligatory (Jia *et al.* 2019). The key routes of human ingestion of fluoride are fluoride-containing groundwater used for drinking purposes, food compounds, and toothpaste (Das *et al*. 2020). Fluoride can be absorbed from the gastrointestinal tract after ingestion, following inhalation, and through the skin. Inhalation and skin/dermal absorption can be important for some occupational exposure situations but are not important exposure routes for fluoride in drinking water (Zhang *et al*. 2019; Guth *et al*. 2020). Intake of low fluoride concentrations (≈1 mg/L) in drinking water can stop the occurrence of dental caries. However, surplus fluoride ingestion for long period may lead to dental and skeletal fluorosis, and other adverse health effects like diarrhea, retardation in growth, Alzheimer’s, reduced mobility, DNA structure, and functional changes of soft tissues like the liver, lung, and kidneys, reduction in children's IQ and even death if doses exceed very high levels (≈250 mg/L) (Shomar *et al.* 2004; Reddy *et al.* 2010; Choi *et al.* 2012; Salifu *et al.* 2012; Razdan *et al.* 2017). A United Nations International Children's Emergency Fund (UNICEF 1999) report shows that 65% of the rural population of India including Punjab has been directly exposed to fluoride-related health problems. They use untreated groundwater for drinking and cooking due to the lack of knowledge, safe and clean water, sanitation, and lower socio-economic standards and are thus exposed directly to the high level of fluoride (Mor *et al.* 2009).

The health risk assessment (HRA) is one such method that helps in determining adverse health effects, based on input data such as fluoride or any other parameter concentration to the risk model. This method of assessment will look at the real risk, particularly in areas where low risk is considered and is widely used by various researchers to assess the non-carcinogenic health risk (NCHR) due to fluoride exposure in water (Ahada and Suthar 2019; Mukherjee and Singh 2018; Narasimha and Ranjitha 2018; Gao *et al.* 2019; Saeed *et al.* 2019). However, most of these studies have used a single point value for every input model parameter to assess human health risks that may lead to intervention, error, and uncertainty in the results of the model. Probabilistic methods such as the Monte Carlo simulation (MCS) include a more practical risk assessment that accounts for the uncertainty, sensitivity to various exposure mechanisms, and the consequences of different intervention scenarios (Yuan *et al.* 2020). Thus, in this study, MCS evaluated possible adverse health effects from groundwater fluoride exposure through oral ingestion and dermal contact pathways. The sensitivity analysis of the HRA parameters was also performed to understand the parameters that influence the three age groups (i.e. children (0-16 years) and adults (males and females having age greater than 16 years) studied (Soleimani *et al.* 2020).

There has been a paradigm shift from “groundwater development” to “groundwater management” in the past decade in the study region. This is mainly due to rapid industrialization, urbanization, excessive pumping for agronomic activities, and an increase in anthropogenic activities in the region. Most industries dump their wastewater (directly or indirectly) in neighboring areas, open pits, or depressions at the field, which contributes to groundwater contamination, with little treatment (Purandara & Varadarajan 2003). Some earlier studies have indicated an increase in the spatial variability of fluoride content in the study region. Singh *et al.* (2011) reported a significant range of fluoride (0.05-0.65 mg/L) in the region. Chaudhry *et al.* (2019a) reported that fluoride in the region is due to an increase in anthropogenic activities and geogenic processes such as weathering and the breakdown of rocks. In the study conducted by the Geological Survey of India (GSI) for the northern region (GSI 2017), metamorphic and igneous rocks (i.e. such as gneisses and granites) have been reported in the study region; indicating fluoride to be an endemic problem in the region. Yet, no studies in the study region deal with fluoride contamination and its assessment of human health risks. Comprehensive research was therefore conducted to elucidate the possible effects of fluoride trends through time and also to assess the possible non-carcinogenic health risk (NCHR) from the ingestion and dermal absorption of fluoride-exposed drinking water for three age groups as no evidence has been found between fluoride and carcinogenic health risks after decades of research (American Academy of Pediatrics 2020). Furthermore, the drinking water quality index (WQI) was evaluated for the quantification of groundwater quality in the area. Using MCS and sensitivity analysis to find the hazard quotient (HQ), the uncertainties in the risk estimates have been quantified too.

Study Area

Rupnagar district (1414 km^2 areal extent) lies in the eastern region of the Punjab state (76°16'26" E-76°43'21"E, 30°44'21" N-31°25'53"N) and is a part of the Indo-Gangetic basin [Figure 1(a-b)]. The region is surrounded by the Himachal Pradesh districts in the north and north-east, Fatehgarh Sahib, and Mohali in the south and south-east, Hoshiarpur, Nawanshahr, and Ludhiana in the west. The major source of the economy in the region is Agriculture (covering nearly 55% of the region). The main crops grown are rice and wheat. Most of the fallow and uncultivated land is barren with few shrubs and bushes. In agricultural activities, various kinds of phosphate fertilizers and other agrochemicals are used to increase farm yields (Department of Agriculture and Farmer Welfare (DAFW), 2020). The area has semi-arid weather with warm summers (April-June) and cold winters (November-February). River Satluj is the main and principal source of water in the region. Drinking water, in the study region, is obtained from both surface and groundwater supplies. The district is influenced by the southwest monsoon (July-September), which constitutes about 78% of the total precipitation. The area is primarily filled by quaternary sediments, mainly of fluvial nature. The rock formations ranging in age from Pleistocene to recent are exposed. They are represented by siwaliks and alluvium deposits. Soil consists predominantly of four main types of soil, namely, ustochrepts, ustorthents, ustipsamments, and ustifluvents (Chaudhry *et al.* 2019b). Based on the piper trilinear plot, it was found that the governing cations and anions in the region are calcium (Ca^{2+}), sodium (Na^+), magnesium (Mg^{2+}), and bicarbonate (HCO_3^-) [Figure 2S]. The groundwater samples in the area are mainly of the form Ca^{2+}, Na^+ / Mg^{2+}-HCO_3^- and control groundwater chemistry.

Materials and Methods

Data procurement and descriptive statistics

Central Groundwater Board (CGWB) is a statutory body that works under Water Resources Ministry, India. All data regarding the country's physico-chemical parameters are monitored and analyzed by them in their chemical laboratories using standard methods given in the Bureau of Indian Standards (BIS) IS: 3025 (2004). For the present study, thirteen parameters for the 14 sampling locations (mainly bore and dug wells) covered by the study area have been chosen which have consistency in their data set for over 28 years (1990-2018). All the data was procured from CGWB. Table 1 lists the descriptive statistics and acceptable limitations as per BIS IS 10500 (1991) and WHO (2011) for all the parameters being evaluated. It has been evident from the results of the descriptive statistics that the concentrations of nitrate (NO_3^-), total hardness (TH), magnesium (Mg^{2+}), and fluoride (F^-) are well above the acceptable limits of BIS IS 10500 (1991). The ionic-balance-error (IBE, the ratio of ((ΣCation-ΣAnion)/(ΣCation+ΣAnion) x 100)) was evaluated for each groundwater sample to ensure accuracy of the study. The determined IBE was within the acceptable ±10% limit (Pastén-Zapata *et al.* 2014).

Trend analysis and its quantification

The Mann-Kendall trend test was used in the analysis to assess the trend in time series data of physico-chemical parameters over more than 28 years (1990-2018) at 14 sampling locations. It is a non-parametric test and unlike parametric tests, it doesn't necessitate data to be independent and normally distributed. It is also useful if the data set has some seasonal components in it (Mosmann *et al.* 2004). The theoretical framework related to the methodology of the test is omitted here for brevity and can be found in many books related to statistical hydrology (Haan 1977; Naghettini 2016). Sen's slope method (Sen 1968) an extension of the Theil method (1950), has been used to quantify increasing and decreasing trends in all the physico-chemical parameters.

From the trend analysis results (Table 2, we displayed only fluoride results for brevity), it has been elucidated that, there is an increasing trend (p-value ≤ 0.05, $\alpha = 0.05$) in the fluoride parameter at the sampling locations. Fluoride exhibited the presence of increasing trends at two sites (i.e. Chak Dera and Soara), while no trend was observed at the rest of the sites. Thus, it can be concluded that the significant increasing trends at two sampling locations may be attributed to various anthropogenic factors. Besides this, natural factors could attribute to no trends in the rest of the study area (Machiwal and Jha 2015).

Water quality index (WQI)

The WQI is a mathematical equation used to turn multiple groundwater quality data into a single number by the policymakers, health care staff, planners, and the general public. It is determined from the human consumption point of view as it is useful in evaluating drinking water quality and the impact of various sources, i.e. natural and anthropogenic on it. The BIS IS: 10500 (1991) considered the water norm for each physico-chemical parameter for computing WQI. The weight values for each parameter were calculated in terms of their relative value (w_i) for overall drinking water quality to measure WQI values at each sampling site [Table 1S]. A weightage (W_i) value of 5 was assigned to the parameters NO_3^-, and F^-;

for pH, EC, and SO_4^{2-} the weight of 4 was assigned taking parameters' importance in water quality into account along with expert opinions. Due to their lesser importance in water quality, Cl^-, TH, Ca^{2+}, Mg^{2+}, and HCO_3^- were given the minimum weight of 3 (Sener *et al.* 2017). The index developed for rating each parameter and the methodology used in calculating WQI is shown in equation 1 (Sharma *et al.* 2018).

$$WQI = w_i w_r \tag{1}$$

$$w_r = \text{water quality rating} = 100\left(\frac{A_V - I_V}{S_V - I_V}\right) \tag{2}$$

where A_v is the actual value present in the water sample; I_v is the ideal value which is taken as 0 for all the parameters except for pH which is taken as 8.5; S_v is the standard value. Based on the WQI results, groundwater quality can be classified into five categories, namely, excellent (WQI<50), good (50-100), poor (100-200), very poor (200-300), and unsuitable for human consumption (WQI>300) (Wang *et al.* 2019).

Fluoride non-carcinogenic health risk assessment for oral and dermal absorption

HRA has been carried out in the present study to determine the adverse health effects of fluoride exposure in groundwater resources. Human exposure to contaminants can occur across three main pathways, namely oral, dermal, and inhalation. In the present study, only two modes were considered (i.e. oral ingestion and dermal absorption). The daily exposure dose (EDI) of fluoride can, therefore, be determined by oral ingestion and dermal absorption using equations (3), and (4), respectively (U.S. Environmental Protection Agency (USEPA 2010); Yuan *et al.* 2020):

$$EDI_{oral} = \frac{F_C * D_W IR * E_F * E_D}{B_W * T_{Avg}} \tag{3}$$

$$EDI_{dermal} = \frac{F_C * SS_A * K_D * FS_W * S_{ET} * E_F * E_D * 10^{-3}}{B_W * T_{Avg}} \tag{4}$$

where EDI_{oral} estimates daily fluoride intake per day from drinking water and EDI_{dermal} estimates the amount of fluoride received by skin absorption (mg/kg/day); F_C is fluoride concentration in drinking water (mg/L); $D_W IR$ is drinking water intake rate (L/day); E_F is the frequency of exposure (day/year); E_D is the duration of exposure (year); B_W is body weight (kg); T_{Avg} is the average lifespan (days); SS_A is skin surface area (cm^2); K_D is the coefficient of skin permeation (cm/h); F_W is fractional surface skin contact with water (dimensionless), and; S_{ET} is the shower exposure time (h/day). Parameters used for HQ assessment are given [Table 3]. HQ for oral intake is calculated using equation 5, where $R_f D_0$ refers to reference dosage or minimal risk level (MRL) is taken as 6E-2 mg/kg/day for oral ingestion (USEPA 1993).

$$HQ_{oral} = \frac{EDI_{oral}}{R_fD_0} \quad (5)$$

HQ for dermal/skin absorption is calculated using equation 6, where reference skin/dermal absorption dosage (R_fD_{dermal}) is taken as 5.82×10^{-2} mg/kg/day (Staff 2001; Mukherjee *et al.* 2019). The complete NCHR for a high concentration of F^- present in groundwater is estimated in terms of the Total Hazard Index (THI) by using equation 7.

$$HQ_{dermal} = \frac{EDI_{dermal}}{R_fD_{dermal}} \quad (6)$$

$$THI = HQ_{oral} + HQ_{dermal} \quad (7)$$

If THI is > 1, possible non-carcinogenic effects on health may occur while THI < 1 means that people in the area are less likely to be at risk for their health due to fluoride exposure (USEPA 2010).

Monte Carlo simulation (MCS) and sensitivity analysis

High ambiguity is found during the evaluation of health risks. For instance, if individual point values are used for calculating the risk of a given population, a high degree of uncertainty is obtained. Thus, MCS has been used in this analysis to minimize uncertainty (Miri *et al.* 2018). In MCS, a set of variable values is used instead of using an individual point value of parameters, and the measurement is repeated many times, and results are obtained with different degrees of confidence (1-99 percent).

Sensitivity analysis has been performed in the MCS to evaluate the parameters which have the greatest effect on the risk assessment outcome. Crystal Ball software (version 11.1.1.1 Oracle, Inc., USA) has been used in this analysis for MCS and conduct sensitivity analysis using 10000 trails. The parameters used during the analysis are listed in Table 3.

Results and Discussion

Spatial distribution of fluoride

From the spatial distribution map of fluoride (prepared using inverse weighted distance technique in ArcGIS 10.5 software) [Figure 3S], it can be elucidated that the northern and central region is worst affected with fluoride contamination, while the rest of the regions were well within the acceptable limits of 1.5 mg/L. The fluoride concentration in the region ranged between 0.12 and 4.90 mg/L with a mean concentration of 0.78 mg/L. Weathering of fluoride-bearing minerals is the most significant geogenic source (GSI 2017). Given that no other natural source of fluoride exists in the study region, various anthropogenic activities also contribute to increasing the fluoride concentration in the region (i.e. brick kilns, industrial and domestic wastewater discharge, increase in the use of fertilizers, mining activities, and extreme irrigation practices) (Chaudhry *et al.* 2019a). The broad gap between the groundwater residence time and the charging area alongside Ca^{2+} insufficient groundwater level is responsible for Na^{+}-HCO_3^{-} level groundwater in the region. This is also one of the major causes of fluoride contamination in the region (Hounslow 1995). Since the study is related to fluoride assessment, spatial distribution maps of other parameters are avoided here for brevity. Readers can refer to Chaudhry *et al.* (2019) for details.

WQI-based water quality assessment

One of the main objectives of this work was to analyze the drinking water quality in the region. For this, 10 physico-chemical parameters [pH, EC, Cl^{-}, NO_3^{-}, SO_4^{2-}, TH, Ca^{2+}, Mg^{2+}, HCO_3^{-}, and F^{-}] have been taken and the WQI method was used.

The computed WQI values range from 55.44-139.40 [Table 4]. The region's water quality is in the "poor" to "good" range. Out of the fourteen sampling locations, two sampling locations (Ahmedpur and Bhalan) fall under the “poor” water class while the rest of them fall under the “good” class. The spatial distribution map of the final WQI has been prepared and presented [Figure 1b]. From the map, it can be elucidated that the northern and central region has poor water quality mainly due to input of municipal and industrial waste and/or discharge of agricultural activities in the area (Yidana and Yidana 2010).

HQ based human health risk assessment

In the current study, a health risk assessment was performed to evaluate the NCHR effects of fluoride in the area using MCS for each site and age group. The EDI_{oral}, EDI_{dermal} of fluoride [Table 2S and 3S, respectively] as well as HQ_{oral} and HQ_{dermal} for male, female, and children have been determined to assess the THI of the fluoride in drinking water [Table 5]. The table describes that fluoride exposure due to oral ingestion was higher than the exposure due to dermal absorption by several orders of magnitude. Oral ingestion is thus the main route for fluoride exposure of the three age groups in the region. NCHR in children due to oral ingestion of fluoride was maximum in Ahmedpur district (HQ_{oral} = 5.45) followed by Bhalan (HQ_{oral} = 1.73) while it was least in Dumewal (HQ_{oral} = 0.13). This heterogeneity can be due to fluoride concentration differences at various locations. Amongst different age groups studied, children were found to be at maximum risk (0.13-5.45) followed by females (0.07-2.97), and males (0.06-2.51) through

HQ_{oral}. However, NCHR associated with HQ_{dermal} for each population group at each site is close to zero (≈0), hence negligible.

Several other public health studies also indicate that the NCHR of fluoride cannot be ignored in drinking water (Huang *et al.* 2017; Fallahzadeh *et al.* 2018; Ghaderpoori *et al.* 2019; Kabir *et al.* 2020). Excessive and insufficient fluoride intake can cause adverse effects, including dental caries, dental, and skeletal fluorosis. The precise age when the teeth are most vulnerable is quite uncertain, with views ranging from the prenatal stage to the age of 2-5 when mineralization is optimum. However, it is widely recognized that teeth are no longer susceptible to harmful fluoride effects after 5-8 years (Adimalla and Qian 2019). Skeletal fluorosis is an advanced stage of bone fluorosis that results in localized cracking and bone weakening in both children and adults. Nevertheless, signs can only be visualized when the disease has reached an advanced stage. Adimalla and Li (2019) in their research stated that skeletal fluorosis begins at 10 years of age and reaches 100 percent at 20 years of age. As a result, children are usually not vulnerable to bone and skeletal fluorosis. To enhance the quality of healthy life, careful attention should be paid immediately to the NCHR assessment of residents' in the affected regions.

Sensitivity analysis

Sensitivity analyses were carried out to classify the main significant parameters that contribute to the performance values of the model's risk estimates. Results of the NCHR sensitivity study (oral and dermal absorption) are shown [Table 4S]. For oral ingestion exposure, F_C and D_WIR were found to be the most influencing parameters in all of the three age groups with correlation coefficients ranging from (0.96-0.98) and (0.16-0.23), respectively. For dermal exposure, F_W and S_{ET} posed the greatest influence on the output risks for the three age groups [Figure 4S]. Thus, a decrease in fluoride concentration, ingestion rate, shower exposure time, and fractional skin contact with water in all exposed groups will reduce the risk of oral intake and dermal absorption of fluoride. It was also found that sensitivity was inversely related to B_W. These results indicate that higher B_W is related to lower sensitivity.

Conclusion

In this study, fluoride concentration, and its NCHR were investigated in the study region using long-term data sets. The trend in fluoride parameter was identified and quantified using Mann–Kendall and Sen's slope tests. The highest fluoride concentrations were detected in the northern and central parts of the study area. GIS-based WQI was computed to demarcate groundwater quality zones. The computed WQI values range from 55.44-139.40 with 85% of the sampling locations falling under the “good” water class and the rest of them under the “poor” water class. Based on HRA, the children were found to be more prone to NCHR than the female and male due to oral ingestion of fluoride. The estimated levels of HQ_{oral} were greater in magnitude than those estimated from HQ_{dermal}. Sensitivity analysis suggested that the most important parameters in the model for minimizing the possible health impact were fluoride concentration in drinking water, ingestion rate, fractional surface skin contacts with water, and time of exposure to the shower. Thus, an increase in anthropogenic activities in the region is the main reason for the increase in fluoride concentration in the region apart from rock-weathering. Therefore, waste from the rural, domestic, and industrial sectors should be properly handled and disposed of. A suitable system for the continuous monitoring of water supplies for its hydrochemistry and hydrology be created so that a policy and action plan for the conservation and restoration of this valuable resource is formed. Defluoridation of drinking water in fluorotic areas should be implemented.

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Conflict of Interest

The authors declare no conflict of interest.

Contributions

AKC and PS conceived and designed the methodology; AKC and PS analyzed the results; AKC and PS wrote the paper; AKC and PS revised the paper.

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Figures

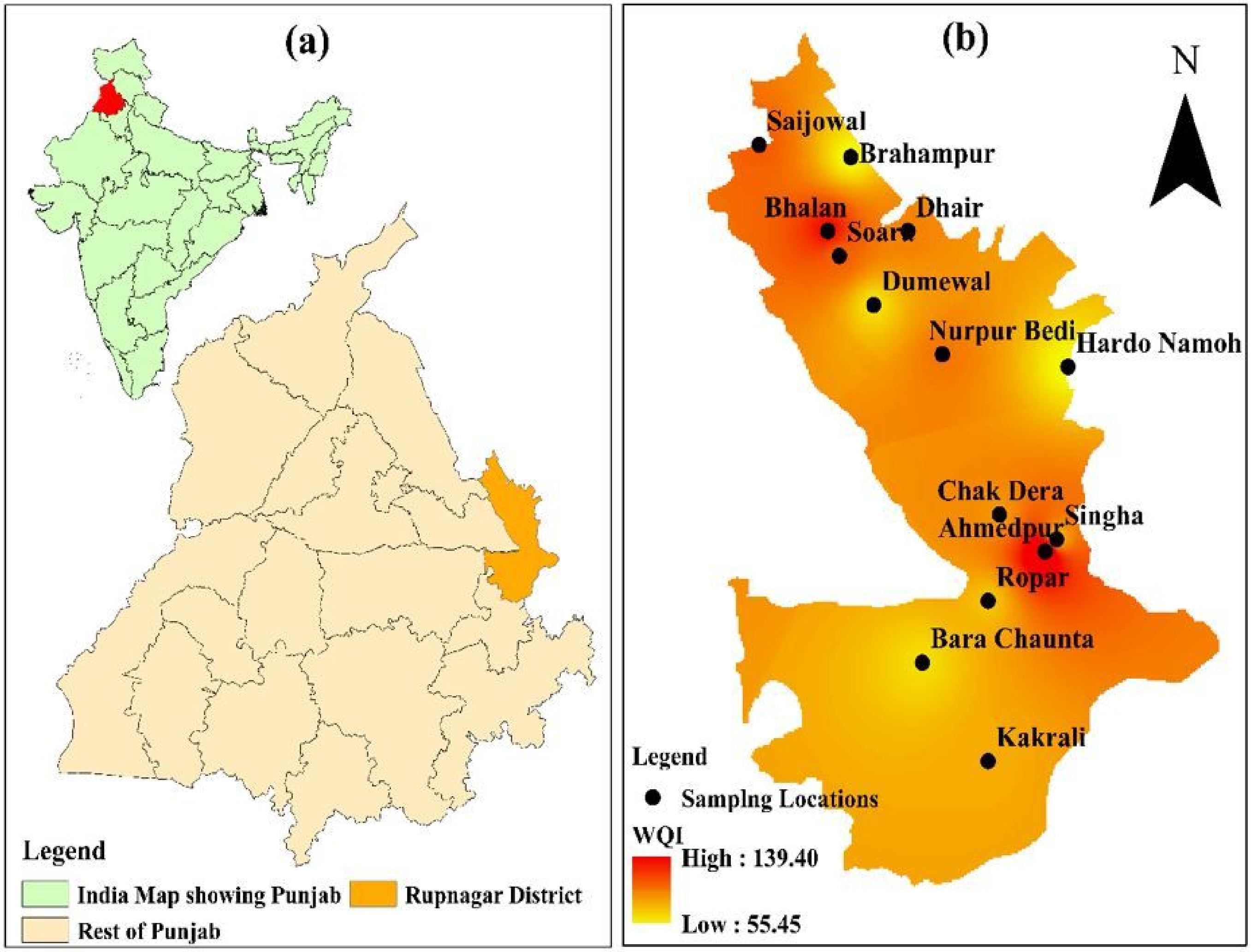

Figure 1

(a-b). (a) Location map of the study region; (b) Sampling locations and WQI distribution in the study region Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

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