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Monitoring and prediction of high fluoride concentrations in groundwater in Pakistan

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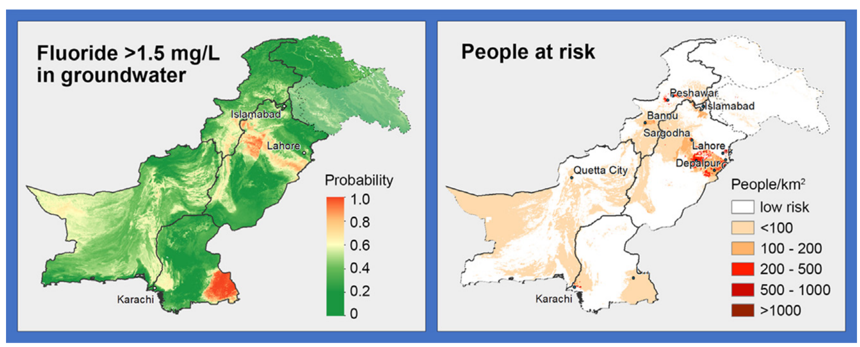
HIGHLIGHTS

- Groundwater fluoride risk maps (>1.5 mg/L) created with >5000 data and machine learning for all of Pakistan.
- Arid climate and soil composition are statistically important predictors of geogenic fluoride contamination.
- The high-resolution maps reveal the vulnerable areas and the number of people exposed.
- An estimated 13 million people (6% of the population) are at risk of fluorosis.
- Most affected areas are in the Thar Desert, the Thal Desert, and scattered along the Sulaiman Mountain Range.

GRAPHICAL ABSTRACT



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ABSTRACT

Concentrations of naturally occurring fluoride in groundwater exceeding the WHO guideline of 1.5 mg/L have been detected in many parts of Pakistan. This may lead to dental or skeletal fluorosis and thereby poses a potential threat to public health. Utilizing a total of 5483 fluoride concentrations, comprising 2160 new measurements as well as those from other sources, we have applied machine learning techniques to predict the probability of fluoride in groundwater in Pakistan exceeding 1.5 mg/L at a 250 m spatial resolution. Climate, soil, lithology, topography, and land cover parameters were identified as effective predictors of high fluoride concentrations in groundwater. Excellent model performance was observed in a random forest model that achieved an Area Under the Curve (AUC) of 0.92 on test data that were not used in modeling. The highest probabilities of high fluoride concentrations in groundwater are predicted in the Thar Desert, Sargodha Division, and scattered along the Sulaiman Mountains. Applying the model predictions to the population density and accounting for groundwater usage in both rural and urban areas, we estimate that about 13 million people may be at risk of fluorosis due to consuming groundwater with fluoride concentrations >1.5 mg/L in Pakistan, which corresponds to ~6% of the total population. Both the fluoride prediction map and the health risk map can be used as important decision-making tools for authorities and water resource managers in the identification and mitigation of groundwater fluoride contamination.

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1. Introduction

Fluorine is greatly abundant in Earth's crust (Amini et al., 2008; Edmunds and Smedley, 2013). It is usually present in the ionic form in the natural environment as a consequence of having the highest electronegativity among the chemical elements. Fluorine is prevalent in a wide variety of minerals but mainly in fluorite, fluorapatite, topaz, and to a moderate degree in biotite and muscovite (Farooqi, 2015; Kumar et al., 2020). Fluorites are commonly found as cements in carbonate rocks (García and Borgnino, 2015). Aside from natural sources, anthropogenic activities can introduce fluoride to the environment, for example, through fluoride phosphate fertilizer effluents (Ali et al., 2019) or fossil fuel combustion (García and Borgnino, 2015). These fluorine-bearing minerals gradually become enriched in groundwater primarily through dissolution and desorption from metal oxides (Kumar et al., 2020), the processes of which are promoted by high alkalinity, low calcium concentration, and the sodium bicarbonate water type in groundwater (Banerjee, 2015; Farooqi, 2015; Kumar et al., 2016; Kumar et al., 2020; Rafique et al., 2009; Singh and Mukherjee, 2015). Elevated fluoride concentrations are also tied to arid climatic conditions (Handa, 1975; Rasool et al., 2018), which are associated with processes such as a higher cation exchange rate, faster dissolution from fluoride-containing minerals and prolonged groundwater residence times and therefore water-rock interactions (Ali et al., 2019; Podgorski et al., 2018).

While lower concentrations of fluoride in drinking water (0.5–1.0 mg/L) are known to prevent tooth decay (World Health Organization, 1994), excessive exposure to fluoride (above 1.5 mg/L) can lead to dental and/or skeletal fluorosis (Wang et al., 2004). It has been estimated that more than 200 million people in the world are at risk of fluorosis (Ayoob and Gupta, 2006). Given that drinking water is one of the major sources of fluoride for humans, the World Health Organization (WHO) maintains a health-based guideline of 1.5 mg/L for fluoride in drinking water (World Health Organization, 2011), which is also the permissible limit in Pakistan (Khwaja and Aslam, 2018). However, some countries such as India (Podgorski et al., 2018; Shah and Bandekar, 1998) and China (Bo et al., 2003) have adopted the fluoride concentration limit of 1.0 mg/L to account for arid climates and other fluoride intake pathways, e.g. food (Ozsvath, 2009).

Groundwater is used extensively to serve the growing population of Pakistan, supplying around 39% of drinking water (World Health Organization and UNICEF, 2019) and 73% of irrigation water (Qureshi, 2020). Having a predominantly semi-arid to arid climate, the conditions in Pakistan are favorable for fluoride accumulation in groundwater. Furthermore, fluoride-bearing rocks such as granites are present in many parts of the country (Naseem et al., 2010), and many cities contain high levels of Na^+ and K^+ in groundwater (Raza et al., 2017), which promote calcite precipitation by cation exchange and reinforce fluoride release from minerals (Farooqi, 2015).

Elevated fluoride concentrations in groundwater (>1.5 mg/L) have been identified in many places in Pakistan, though mainly confined to the populous and flat-lying Punjab and Sindh Provinces (Ali et al., 2019; Brahman et al., 2013; Farooqi, 2015; Farooqi et al., 2007; Iwasaki, 2007; Khattak et al., 2022; Rafique, 2008; Rafique et al., 2008; Rafique et al., 2009). For instance, 27.2% of the samples in a study along the riverine systems in the Punjab and Sindh exceeded 1.5 mg/L (Ali et al., 2019). The aquifers of these two provinces are recharged primarily from rainwater along with infiltration from the five major rivers in Punjab: the Sutlej, Ravi, Jhelum, Chenab, and Indus Rivers, all of which come together as the Indus that flows further south through Sindh. Very high fluoride concentrations up to 30 mg/L have been measured in the Thar Desert (Iwasaki, 2007; Rafique et al., 2009), which is situated at the southeastern corner of the country where sand dunes and kaolin/granite abound. Widespread concerns have been raised about dental and skeletal fluorosis that has been detected, particularly among children, in the cities of Mianwali, Quetta, Lahore, Karachi, Peshawar, and the Thar Desert (Ahmad et al., 2020; Khan et al., 2004; Khan et al., 2015; Rafique et al., 2015; Sami et al., 2016).

To protect public health, it is essential to determine if wells and springs contain safe or hazardous levels of fluoride. As such, maps of affected areas can provide a key first step in determining the locations of safe and hazardous wells. The most comprehensive nationwide analysis previously conducted of the distribution of fluoride in groundwater in Pakistan consisted of some 1000 measurements that were summarized at the subtehsil scale (Khan et al., 2002). This corresponds to an average size of about 2100 km^2 per administrative unit, which provides only very limited spatial resolution of fluoride contamination. Furthermore, no attempt had been made to make predictions beyond the data collected.

Thanks to the availability of high-resolution data sets of environmental predictor variables, machine learning approaches have been used to create accurate geospatial prediction models of various groundwater as well as soil parameters (Chen et al., 2017; Erickson et al., 2021; Hengl et al., 2017; Podgorski and Berg, 2020; Podgorski et al., 2020; Reichstein et al., 2019; Winkel et al., 2008; Wu et al., 2021). In contrast to conventional geostatistical techniques based upon interpolation among observations, machine learning models have the potential to be applied with high accuracy to larger scales (regional to country scale) where sufficient data are available.

Machine-learning methods, such as random forest, have proved effective in modeling a binary target variable (e.g. fluoride concentrations above a threshold) that is effectively unchanging in time and predicting its occurrence on the basis of relevant predictors (e.g., climate, geology) (Podgorski et al., 2017; Podgorski et al., 2018; Winkel et al., 2008). Since machine learning models learn from data, relationships can be inferred between predictors and the target variable from modeling results and thus learn more about how hydro-geochemical conditions regulate fluoride concentrations in groundwater.

In this paper, we analyzed fluoride in over 2100 groundwater samples from all over Pakistan, including in the as yet poorly examined Thal Desert, and combine these with previously published data to produce with machine learning a first-ever high-resolution prediction map of high fluoride concentrations in Pakistan. This allows us to generate a health-risk map, estimating the locations and number of people at risk of excessive fluoride exposure. Furthermore, the statistically important predictor variables are also discussed for their insight into the environmental conditions associated with high fluoride concentrations. The main purpose of these hazard and risk maps is to identify areas with greater and lesser chances of containing high fluoride concentrations in groundwater. They can thereby provide valuable guidance for authorities and water resource managers in testing for and ultimately mitigating hazardously high concentrations of fluoride for the protection of health.

2. Materials and methods

2.1. Study area

Pakistan is characterized by a variable geomorphology, with the flat-lying Indus plain that comprises the provinces of Punjab and Sindh in the east; the Hindu Kush, Karakoram, and Himalaya ranges in the north; and the vast Baluchistan Plateau in the west. The climatic conditions are primarily arid to semi-arid, with temperate conditions in the northwest and arctic conditions in the northern mountain ranges. Desert areas cover around one-third of the country, with mountain areas, grasslands and agricultural regions covering the other parts (Greenman et al., 1967). Pakistan's geology is dominated by young (Quaternary age) alluvial and deltaic deposits (Sanaullah et al., 2019) that outcrop across much of the Indus plain (Punjab, Sindh) and Baluchistan basin, while older formations (granites, metamorphic rocks) are mainly restricted to the Khyber Pakhtunkhwa region (WAPDA/EUAD, 1989). Most of Pakistan's population is concentrated in Punjab and Sindh due to fertile soil and an abundant water supply. Over 100 million inhabitants of Punjab and Sindh rely on groundwater replenished by the Indus River and its tributaries (Jhelum, Chenab, Ravi, and Sutlej) for drinking water and agricultural uses (Bhowmik et al., 2015). There are more than one million private tube wells in the country, of which 3.8% are in Khyber-Pakhtunkhwa, 6.4% in Sindh, 4.8% in Baluchistan and more than 80% in Punjab, with a total groundwater abstraction of about 60 billion m^3 per year (Qureshi, 2020). Unregulated groundwater abstraction, unsustainable pumping, agricultural irrigation and increasing water demands in urban areas result in fluctuating groundwater levels and generally impact the water quality of the aquifers of the Indus plain (Rasheed et al., 2022; Ullah et al., 2022).

2.2. Groundwater samples

We sampled 2160 groundwater wells between 2013 and 2019 throughout Pakistan (Fig. S2) and measured fluoride with a portable photometer field test kit (FTK; HI96739, Hanna Instruments) and verified values by ion chromatography (IC). The majority of measurements was taken in well-populated Punjab and Sindh Provinces, especially in the Thal Desert where a comprehensive study of fluoride contamination has been lacking.

To test the accuracy of the FTK, water samples with known fluoride concentrations ranging from 0 to 8 mg/L were prepared and subsequently measured by IC and the FTK. This comparison showed that the FTK is generally able to recover the fluoride concentrations with high accuracy (Table S3), in particular in the range of 0 to 2 mg/L. In addition, 54 of the field samples were also verified by IC. These mostly plot along the 1:1 line (Fig. S3), with deviations generally being due to higher FTK measurements. With respect to the threshold of 1.5 mg/L, 83% of these samples were classified the same by both measurement methods.

The original 2160 fluoride measurements were combined with data from other sources to form a dataset of 5543 geolocated concentrations of fluoride in groundwater throughout Pakistan (see Fig. 1, Fig. S5, and Table S1), which were later used in modeling. Of these, 2814 samples were collected between October 2013 and March 2014 by the Rural Water Quality Monitoring Program (RWQMP) of the Pakistan Council of Research in Water Resources (PCRWR, 2015). Approximately 573 of the 2865 villages in Punjab and Sindh Provinces were covered in this program, with 4 to 5 samples collected in each village. In addition, we were able to include a few hundred data points from the Indus plain in Punjab and Sindh (Ali et al., 2019) as well as the Thar Desert (Rafique, 2008; Rafique et al., 2008; Rafique et al., 2009), where 79% of the samples exceeded 1.5 mg/L.

Overall, 911 of the 5543 groundwater samples (16%) are greater than 1.5 mg/L, and 1451 samples (26%) are above 1 mg/L. The clusters of samples reported at the same location in a village were averaged after removing outliers, the points of which are distinctly different from their neighboring data (e.g. in the RWQMP project). This reduced the number of the total data points from 5543 to 5483, with 16% of points having a high fluoride level after outlier removal and averaging. The fluoride concentrations were then converted into binary levels “high” (F > 1.5 mg/L) and “low” (F ≤ 1.5 mg/L).

2.3. Predictor data sets

A variety of environmental data sets dealing with climate, lithology, land cover and soil (n = 48, see Fig. S11, and Table S4) were considered for modeling based on known or possible links with fluoride contamination (García and Borgnino, 2015; Handa, 1975; Podgorski et al., 2017; Podgorski et al., 2018). All of the variables represent data at the surface, or up to 2 m depth in the case of soil parameters (Hengl et al., 2017). Each level of the categorical predictors of soil groups, geologic age, and lithology was converted into a binary data set to enable testing the significance of each category in relation to fluoride in subsequent variable selection procedures. Pearson correlations were determined between each continuous predictor variable and fluoride concentration, whereas the fraction of fluoride concentrations exceeding 1.5 mg/L was calculated for categorical variables.

Predictor values were extracted at the locations of the 5483 fluoride measurements and added to the fluoride data set. As most predictors were available in 250 m resolution, rasters with a coarser (1000 m) or finer (100 m) resolution were resampled by nearest neighbor and bilinear methods respectively, to 250 m for subsequent prediction procedures, allowing the resolution of the prediction maps to be 250 m.

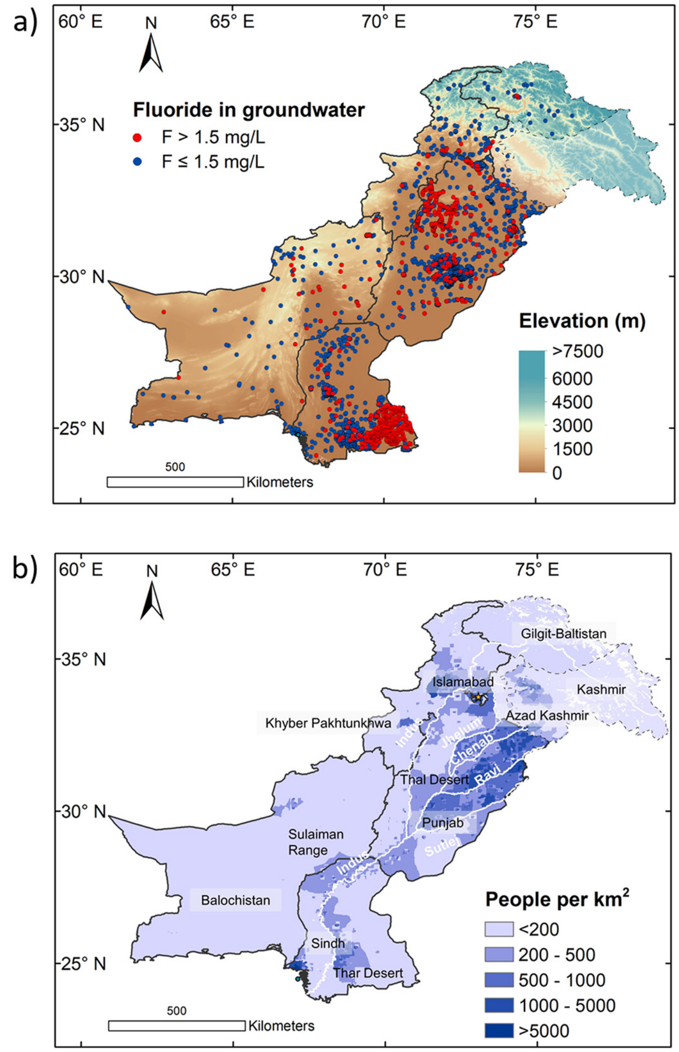

Fig. 1. Groundwater fluoride concentrations (n = 5543) from original and existing measurements. a) High (F > 1.5) and low (F ≤ 1.5) fluoride levels are plotted with topography. About 16% of the wells exceed the WHO drinking-water guideline of 1.5 mg/L. **b)** Population density map of Pakistan with labels of provinces and rivers.

2.4. Training and testing data sets

Training and test data sets were split at the ratio of 80% to 20%. Tightly clustered data points selected by random sampling may result in data rows that do not contain much variance in the predictor variables. Therefore, a method was developed that ensures greater predictor variability as follows:

i). Selection of continuous predictors with the highest correlations with fluoride (Table 1, Table S4): coarse fragments fraction, nitrogen fraction, organic carbon density, potential evapotranspiration (PET), aridity index, compound topographic index, and slope.
ii). The quantile interval (0–25%, 25–50%, 50–75%, 75–100%) of each predictor in step i) was determined for each of the 5483 fluoride measurements.
iii). The data points were then divided into groups that share the same predictor quantile interval combinations (1714 unique groups).
iv). Training dataset (80% of data) created by random sampling, ensuring that at least one data point is selected from each group. The remaining data points are assigned to the test dataset (20% of data).

Table 1
Correlations and significance of environmental parameters used in the model with fluoride concentrations.

Type	Variable	Resolution	Correlation (p)
Climate (continuous)	Aridity index (Zomer et al., 2007; Zomer et al., 2008)	1000 m	−0.1276 (3.09E-21)
	Actual evapotranspiration (AET) (mm/year) (Trabucco and Zomer, 2010)	1000 m	0.0276 (4.17E-02)
	Potential evapotranspiration (PET) (mm/year) (Zomer et al., 2007; Zomer et al., 2008)	1000 m	0.1550 (1.07E-30)
	Precipitation (mm/year) (Fick and Hijmans, 2017)	1000 m	0.0412 (2.31E-03)
	Temperature (°C) (Fick and Hijmans, 2017)	1000 m	0.0902 (2.53E-11)
Soil (continuous)	Silt fraction (g/kg) (Hengl et al., 2017)	250 m	−0.0319 (1.84E-2)
	Nitrogen fraction (cg/kg) (Hengl et al., 2017)	250 m	−0.1363 (4.83E-24)
	Coarse fragments fraction (cm^3/dm^3) (Hengl et al., 2017)	250 m	0.2515 (1.75E-79)
	Organic carbon density (g/dm^3) (Hengl et al., 2017)	250 m	0.1111 (1.91E-16)
Soil (categorical)	Arenosols (Hengl et al., 2017)	250 m	0.6677
	Calcisols (Hengl et al., 2017)	250 m	0.3010
	Cambisols (Hengl et al., 2017)	250 m	0.0993
	Gypsisols (Hengl et al., 2017)	250 m	0
	Solonchaks (Hengl et al., 2017)	250 m	0.0376
	Solonetz (Hengl et al., 2017)	250 m	0.0500
Lithology (categorical)	Carbonate sedimentary rocks (Hengl, 2018)	250 m	0.2468
	Metamorphic rocks (Hengl, 2018)	250 m	0.0566
	Mixed sedimentary rocks (Hengl, 2018)	250 m	0.2025
	Evaporite (Hengl, 2018)	250 m	0.1644
	Siliciclastic sedimentary rocks (Hengl, 2018)	250 m	0.0222
Topography (continuous)	Elevation (m) (Verdin, 2017)	100 m	−0.0275 (4.22E-2)
Land cover (categorical)	Shrubland (Buchhorn et al., 2020)	Polygon	0.5664
	Cropland (Buchhorn et al., 2020)	Polygon	0.0885
	Herbaceous vegetation (Buchhorn et al., 2020)	Polygon	0.3839

Applying this procedure, the proportion of “high” to “low” fluoride classes in the training and test sets remains approximately 16% (Table S5).

2.5. Random forest modeling

Random forest is a machine-learning algorithm that constructs an ensemble of decision trees, which recursively partition predictor variables to predict a dependent variable (Breiman, 2001). Individual decision trees consider a random subset of candidate predictors at each split, which reduces correlation between decision trees and helps avoid overfitting (James et al., 2013). Increasing the number of trees can further reduce overfitting (Breiman, 2001). Randomness is also introduced by growing trees with bootstrapped samples (sampling with replacement) of the training set, which results in approximately one-third of sample data being left out of each tree. The unselected, out-of-bag samples can also be used to estimate the generalization error of the random forest model (Breiman, 2001).

The R programming language (R Core Team, 2013) was used with the “randomForest” package (Liaw and Wiener, 2002) to create random forest classification models of groundwater fluoride. The output of the random forest is therefore the probability of the occurrence of high fluoride concentrations. As such, the WHO drinking water guideline of 1.5 mg/L was generally used to define the cut-off between high and low fluoride concentrations. Bootstrapped sampling was made with a balance between the two categories of high and low fluoride.

The performance of a model can be evaluated through the accuracy of the predictions for a given probability cut-off value. The Area Under the ROC (receiver operator characteristic) Curve (AUC) overcomes the subjectivity of choosing a threshold by summing up over true positive and false positive rates at all cut-off values (Huang and Ling, 2005). The AUC score is bounded between 0 and 1 (perfect classifiers); an uninformative classifier that uses random guessing may yield 0.5 (Tharwat, 2020).

Variance importance plots of a random forest model help evaluate the influence of individual variables. They are composed of two importance indices, namely, Mean Decrease Accuracy (MDA) and Mean Decease Impurity (MDI). MDA is calculated by averaging the change in out-of-bag error estimates after permuting an individual variable's values in out-of-bag observations (Biau and Scornet, 2016). In general, permuting the values of an important variable will lead to deterioration of model performance, thus a decrease in accuracy. Likewise, MDI is defined as the average decrease in node Gini impurity from splitting an individual variable over all grown decision trees (Biau and Scornet, 2016). Gini impurity indicates the homogeneity of a node, which is lower after splitting an important variable that divides observations roughly into the same class.

To simplify the model but retain its predictive power, variable selection by recursive feature elimination (RFE) was conducted with the caret package in R (Kuhn, 2009), which backwardly reduces the number of variables by removing the least important one at each step. Random forests were created starting with all 48 variables (Table S4) and working down to just one variable. The predictor subset ultimately chosen was that with which the corresponding model was simplest and obtained high test AUC score.

For all models, 1000 trees were grown, and the default number of variable candidates at each split (i.e. square root of the number of features) was used. Sampling for each tree was made with replacement and an even balance between high and low fluoride classes. The optimal number of samples from each class was determined by trying 70%, 80%, 90%, and 100% of the minority class (high fluoride) and selecting the sample size with the highest AUC score in 10-fold cross validation. The final random forest model was built with the RFE-selected variables and optimal sample size. A prediction map of the occurrence of fluoride contamination >1.5 mg/L was then created by applying the final random forest model to the predictor datasets using the raster package (Hijmans, 2021).

We then estimated the population in Pakistan at risk of exposure to high fluoride concentrations from groundwater used as drinking water by multiplying the population density in 2020 (Gao, 2017; Jones and O’Neill, 2016) by the probability of high fluoride concentrations and accounting for the average rate of domestic groundwater usage (39.1%) (World Health Organization and UNICEF, 2019). Only areas above the probability cut-off value at which the accuracy rates for the two classes (sensitivity and specificity) are equal were taken into consideration.

3. Results and discussion

3.1. Fluoride concentrations in groundwater

Summary statistics, box plots and spatial distributions of the new groundwater fluoride measurements ($n = 2160$) are shown in Table S2, Fig. S1-and Fig. S3, respectively. Whereas the data points from previously published studies (Ali et al., 2019; Brahman et al., 2013; Khattak et al., 2022; Naseem et al., 2010; Rafique, 2008; Rafique et al., 2008; Rafique et al., 2009) are confined almost exclusively to the Punjab and Sindh provinces, our data span the entire country (Fig. S5).

High fluoride concentrations were detected in all regions, including maximum values of 27.5 mg/L in Punjab and 33.3 mg/L in Sindh. However, the average concentrations by province are all less than 1.5 mg/L (Table S2). As seen in the inset of Fig. S3, high-fluoride areas were discovered in the Sargodha Division in northwest Punjab, which had an average measured fluoride concentration of 1.8 mg/L. Particularly affected are the upper-Thal Desert districts of Bhakkar, Khushab and Mianwali.

Correlations between fluoride and other geochemical indicators are shown in Fig. S2. The alkaline environment and the presence of bicarbonates create a favorable condition for high fluoride waters, while calcium ions suppress fluoride in groundwater.

3.2. Prediction modeling

3.2.1. Random forest model

Of the 48 variables considered, eight were selected by the RFE process: actual evapotranspiration (AET), aridity index, coarse fragments fraction, elevation, nitrogen fraction, PET, precipitation, and temperature. The RFE algorithm selects variables based on their importance, which is generally lower in categorical variables than in continuous ones due to only a fixed (generally small) number of possible values in the former. The following binary predictors were therefore manually added: arenosols, calcisols, cambisols, carbonate sedimentary rocks, cropland, evaporite, gypsisols, herbaceous vegetation, mixed sedimentary rocks, shrubland, siliciclastic (non-carbonate) rocks, solonchaks, solonetz, and herbaceous vegetation, which are considered to be important from a geochemical standpoint, for example, by acting as a sink or source of fluoride (Ali et al., 2019; García and Borgnino, 2015; Podgorski et al., 2018). The optimal sample size determined by tuning was 562, which is 80% of the number of the minority class (high fluoride) in the training dataset.

The final random forest model attained an AUC score of 0.92 as determined with the test dataset (Fig. 2b). The cut-off value of 0.47 was found at the point at which sensitivity equals specificity, that is, where the accuracy rates for the two classes are evenly balanced. Using this cut-off, the overall accuracy with the test set is 0.83, which is comparable to the out-of-bag accuracy of 0.82 (accuracy calculated with out-of-bag samples during training), confirming that the distribution of data in the training and testing datasets is generally similar.

Unsurprisingly, the measured importance of the continuous variables is higher than that of the binary ones (Fig. 2a). The climate predictors temperature, precipitation, PET, aridity index, and AET as well as nitrogen content, coarse fragments fraction, and elevation received the highest importance. Among the binary predictors, calcisols and cropland are the most important.

3.2.2. Prediction map

The prediction map derived from the random forest model (Fig. 3) indicates that approximately 30% of Pakistan is at-risk of fluoride concentrations in groundwater exceeding 1.5 mg/L. Two particularly high-hazard areas include the Thar Desert (Sindh Province) and the Bhakkar, Mianwali, and Khushab districts in the upper Thal Desert (Sargodha Division, Punjab Province). Both locations have an arid climate and are situated within the flat-lying Indus Plain. Higher probabilities are also found in the Sulaiman Mountains in eastern Balochistan. While most areas of the prediction map clearly show higher or lower hazard, the determination is more ambiguous in southwestern Balochistan, where the probabilities are around 0.50. This may result from a relative lack of measurements to adequately sample the distribution of fluoride in this region, whereas most of the data used in the model stem from the Indus Plain.

The fluoride prediction map was compared with a similar fluoride map of India (Podgorski et al., 2018) (Fig. S8). High probabilities of fluoride exceeding 1.5 mg/L in northern Punjab align very well with those across the border in Indian Punjab. Similarly in the Thar Desert, which forms a natural boundary between Pakistan and India, the predictions on either side of the border are compatible. Whereas these sections are well constrained by fluoride data, border areas in between match less well, which may be due in part to a lack of measurements in southern Pakistani Punjab.

Since much of Pakistan has a warm, dry climate and people consequently drink more water, a prediction model was also created for fluoride concentrations exceeding 1.0 mg/L (Fig. S7). However, with only 543 of the 5483 measurements falling in the range of 1.0–1.5 mg/L, the high-hazard regions of the prediction maps for 1.5 mg/L (Fig. 3) and 1.0 mg/L (Fig. S7) are largely similar. For example, higher probabilities for 1.0 mg/L were found for central Pakistan near the borders of Punjab, Khyber Pakhtunkhwa, and Balochistan. The selection of variables by RFE was also similar for both models (1.0 mg/L and 1.5 mg/L).

3.2.3. Predictor importance

Some of the most important predictors are the climate parameters (Fig. 2a). The correlations in Table S4 also confirm that drier climate conditions favor fluoride release. High importance was also found for the elevation variable, which is strongly negatively correlated with temperature.

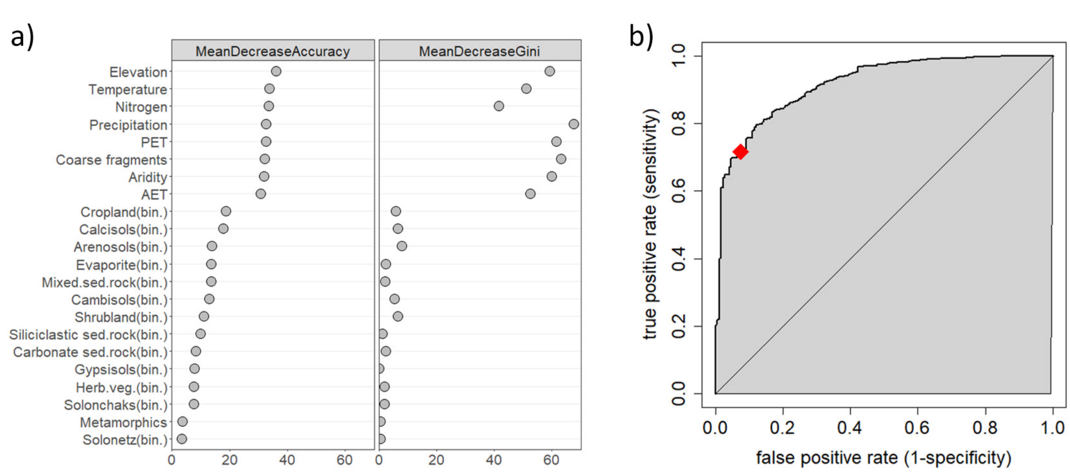

Fig. 2. Random forest modeling results of groundwater fluoride concentrations in Pakistan exceeding the WHO guideline of 1.5 mg/L. a) Variable importance plots of Mean Decrease Accuracy and Mean Decrease Gini Impurity for each variable in the random forest model. Due to having only two degrees of possible values, the binary variables (bin.) show lower importance scores relative to the other continuous variables. b) ROC curve (AUC score: 0.92) of the model response using the test dataset plotted with the true positive rate (sensitivity) against the false positive rate (1 – specificity). The red dot indicates the cut-off value of 0.47.

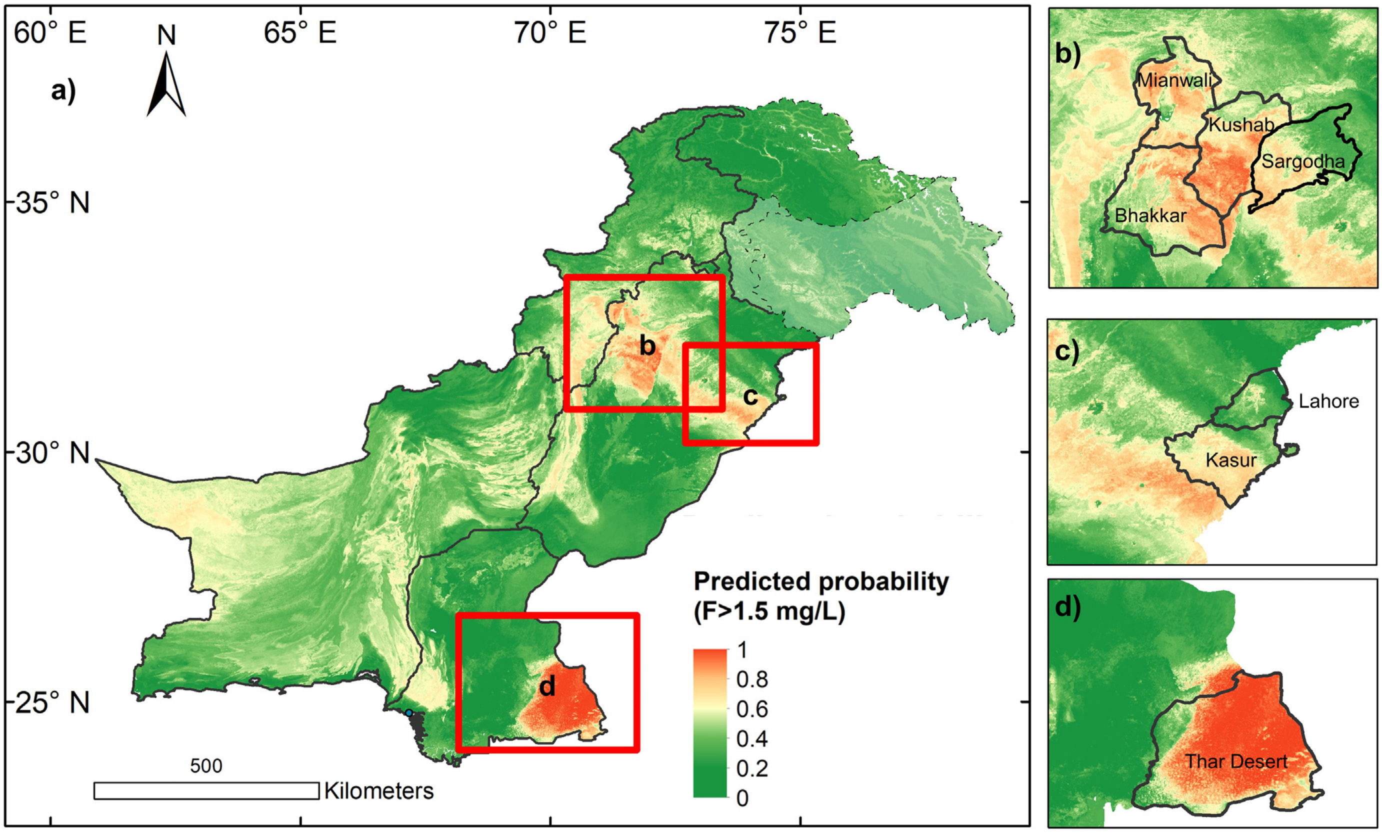

Fig. 3. Prediction map at a resolution of approximately 250 m of groundwater fluoride in Pakistan exceeding the WHO guideline of 1.5 mg/L. (AUC score is 0.92). **a)** Full map with locations of insets. **b)** Fluoride hotspot of the Sargodha Division in the upper Thal Desert (Bhakkar, Mianwali, Khushab districts). **c)** Hotspots in the Kasur and densely populated Lahore districts. **d)** Hotspot of the Thar Desert. The prediction map is also available for viewing at high-resolution on the GIS-based Groundwater Assessment Platform (GAP), www.gapmaps.org.

The two continuous variables of soil parameters, fraction of coarse soil fragments and nitrogen fraction, also have high importance measures. The fraction of coarse fragments is high in the Sulaiman Mountains of eastern Balochistan and western Punjab and Sindh and are composed of mixed or carbonate sedimentary rock that may contain fluoride-bearing minerals (Fig. 4). On the other hand, nitrogen fraction is associated closely with the presence of forested mountains in the north, which has lower temperatures, and where the sparse measurements generally show low fluoride concentrations. The “calcisols” binary soil predictor, which is associated with substantial accumulations of lime, is connected to the presence of high fluoride concentrations, as the precipitation of calcite removes calcium from dissolution and results in higher fluoride concentrations (Banerjee, 2015).

Carbonate sedimentary rocks can represent an important source of fluorite (García and Borgnino, 2015), and silicate minerals may contain small amounts of fluorine. Shrubland and herbaceous vegetation are the principal vegetation cover type in the highly contaminated Thar Desert, and thus assigned high importance in the model, though it is unclear if this relationship exists elsewhere.

In the Thal Desert (Punjab), calcisols likely control Ca^{2+} levels and enhance the dissolution of F-bearing minerals. Moreover, alkaline pH conditions can promote F^- leaching by ion exchange processes. This is particularly the case along the Jhelum River where water samples are dominated by the Na-Cl type of water (Ali et al., 2019). A rapid change in aquifer recharge in recent years has resulted in increased fluoride levels (Younas et al., 2019). High alkaline waters in the Lahore and Kasur areas, which experience excessive pumping, are also associated with high fluoride leaching (Farooqi et al., 2007).

While calcisols are also the dominant soil type in the Sulaiman Mountains (Fig. 2d), fluoride levels are not as high as in the Thal Desert, possibly due to a stable water table. In the south in the Thar Desert (Sindh), dolomite dissolution and arid climatic conditions promote evaporation process and the dissolution of evaporites, contributing to the formation of saline groundwater.

3.3. Health risk map

A health risk map was made using the optimal cut-off value of 0.47 (Fig. 5). It identifies numerous densely populated regions where people rely on groundwater associated with high fluoride. In total, over 13 million people are estimated to be potentially affected by fluoride contamination in groundwater, which is 6.0% of the total population in Pakistan. However, this number would be even larger if the population in regions with probabilities under the cut-off of 0.47 would be taken into consideration. With the increasing population in Pakistan (population growth rate around 2% in the year 2020) (World Bank, 2020), the problem may become more severe in the future if the reliance on groundwater remains high. Large at-risk populations are found in northern Punjab, Islamabad, and Khyber Pakhtunkhwa. Furthermore, the fluoride risk map (Fig. 5) indicates that residents in the cities of Lahore, Sargodha, Depalpur, Peshawar, Bannu, Karachi, Quetta, and others are at high risk, which is confirmed by the high prevalence of fluorosis in some of these cities (Ahmad et al., 2020; Mohsin et al., 2014; Rahman et al., 2018; Sami et al., 2016). Conversely, the total number of people at risk in the Thar Desert (SE Sindh), which has high probabilities of fluoride contamination, is far smaller owing to its sparse population, yet residents there are still under high risk.

The presented probability and health risk maps (Figs. 3 and 5) raise awareness about fluoride contamination and its adverse health impacts in Pakistan. Furthermore, they can help authorities in prioritizing areas

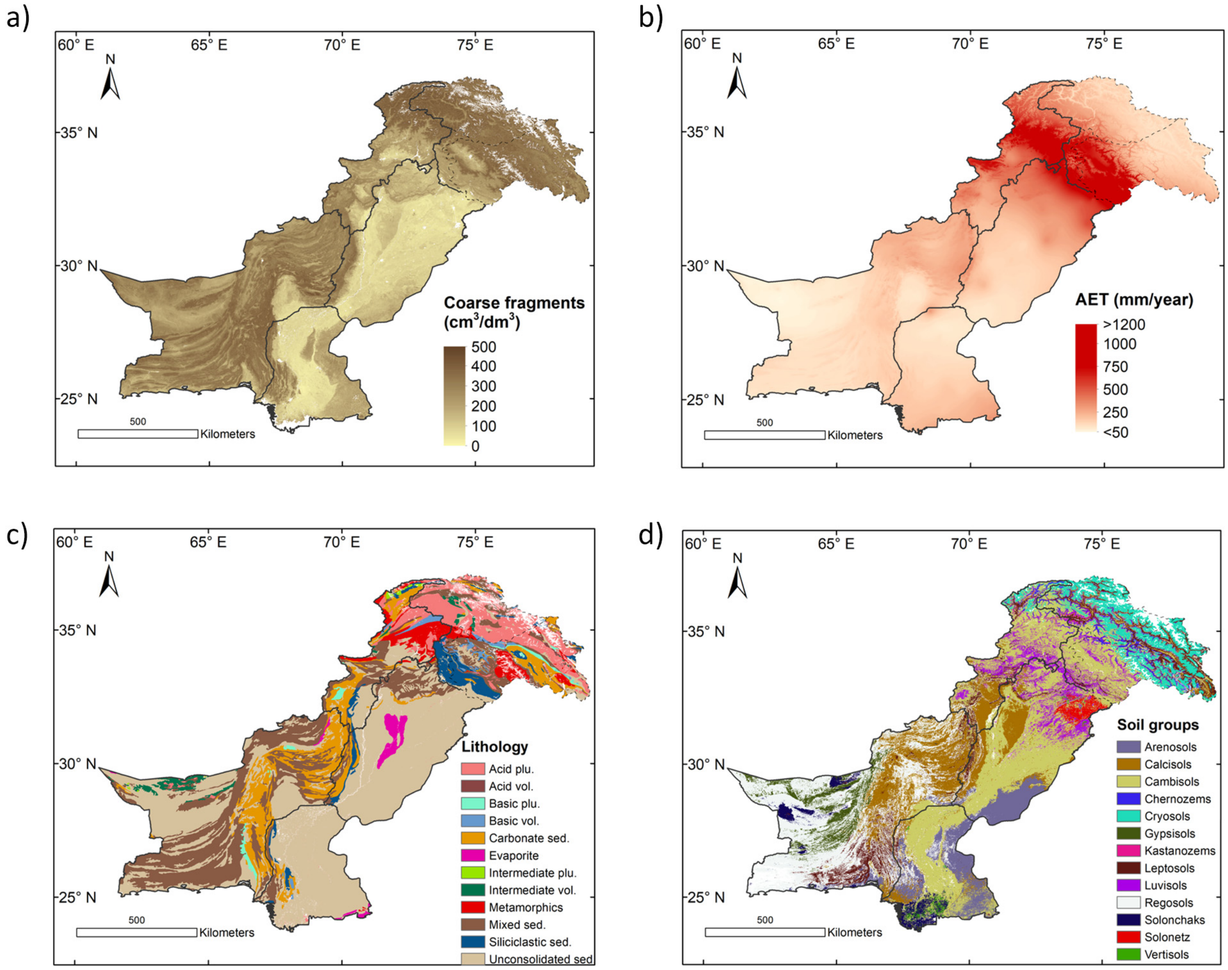

Fig. 4. Maps of selected predictor variables of a) coarse fragments fraction, b) AET, c) lithology, and d) soil groups.

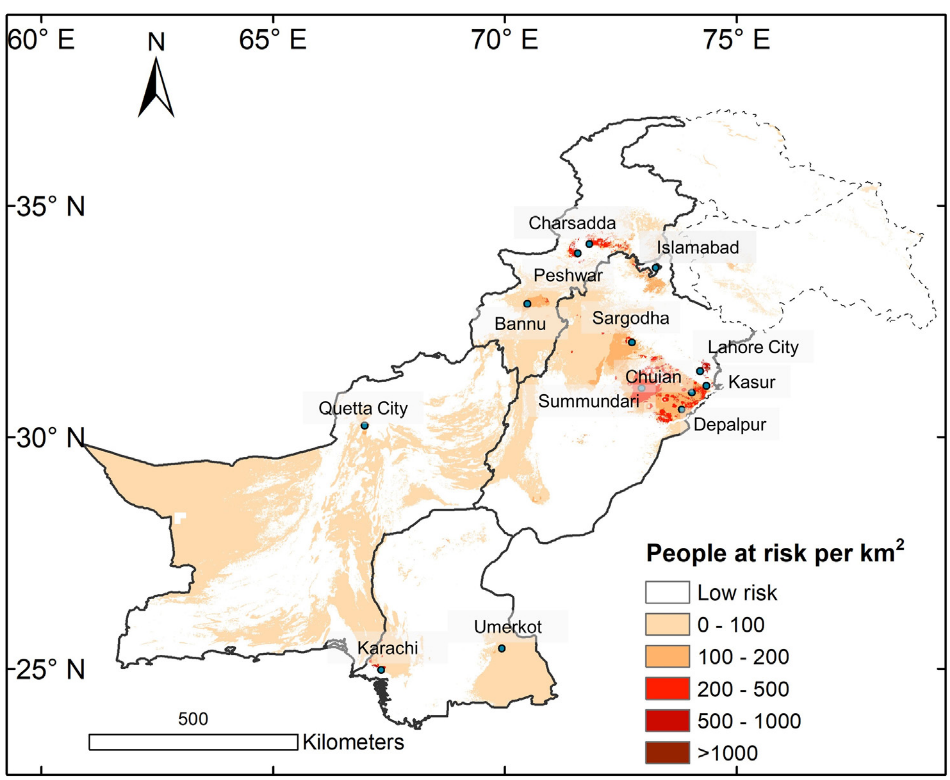

Fig. 5. Population at risk of exposure to fluoride concentrations in groundwater exceeding 1.5 mg/L.

for implementing mitigation measures. These could include monitoring programs for drinking water wells or fluoride removal, e.g. adsorption treatment (Bhatnagar et al., 2011) or membrane separation processes (Waghmare and Arfin, 2015), and improving health management systems. Compared to the previous nationwide representation of fluoride at the sub-tehsil-scale (Khan et al., 2002), the novel maps presented here have a 3–4 order of magnitude higher spatial resolution (250 m), are based on much larger new datasets, and predict the probability of high groundwater fluoride for areas where data are lacking. Also in relation to a recent study by Khattak et al. that contains clusters of many groundwater fluoride measurements across much of Punjab (Khattak et al., 2021), the new maps identify hotspots, e.g. in the Sargodha Division, that that study did not uncover.

4. Conclusions

This study, which presents a large new dataset of fluoride in groundwater across Pakistan, combined with geospatial modeling and risk mapping using various environmental predictors, highlights several regions where exposure to high fluoride levels pose a significant public health risk. Hot spots include the Thal Desert in Punjab (Sargodha Division), the Thar Desert in Sindh, and the Sulaiman Mountains in the western part of the country. Analysis of the importance of the predictor variables and their correlation with fluoride show that high fluoride concentrations in groundwater benefit from arid climatic conditions with high temperatures and evapotranspiration, the presence of fluoride-bearing minerals (e.g. carbonate sedimentary rock), and the presence of calcisols.

Knowing the countrywide groundwater fluoride risk and affected populations shall be helpful for authorities and water resource managers in identifying fluoride-contaminated wells and mitigating the risk for residents. All groundwater wells in areas with a high probability (e.g., above the cut-off value of 0.47) should be tested, for instance, in the Thar Desert and the Sargodha Division (especially the Bhakkar, Mianwali, and Khushab districts in the upper Thal Desert). Particular attention should also be paid to risk areas with a high population density such as Lahore, Sargodha, Depalpur, Peshawar, and Bannu. Mitigation measures include monitoring, provision of alternative sources of drinking water, fluoride removal treatment, and awareness-raising campaigns. These maps are not a replacement for actual groundwater testing but indicate hazard and risk for drinking water use.

Future work could consider additional groundwater contaminants, e.g. uranium, nitrate, pesticides or salinity in order to obtain a more comprehensive understanding of the safety of groundwater. Model accuracy could be further improved by incorporating additional data and other predictor variables, such as hydrological parameters, if available.

CRediT authorship contribution statement

Yuya Ling: Data curation, Formal analysis, Investigation, Methodology, Software, Validation, Visualization, Writing – original draft, Writing – review & editing. **Joel Podgorski:** Conceptualization, Data curation, Formal analysis, Funding acquisition, Investigation, Methodology, Project administration, Software, Validation, Visualization, Writing – review & editing. **Muhammad Sadiq:** Data curation, Formal analysis, Validation. **Hifza Rasheed:** Data curation, Formal analysis, Validation. **Syed Ali Musstjab Akber Shah Eqani:** Conceptualization, Funding acquisition, Project administration, Supervision, Data curation, Validation, Writing – review & editing. **Michael Berg:** Conceptualization, Funding acquisition, Investigation, Methodology, Project administration, Supervision, Visualization, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2022.156058.

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