

Supplementary material

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Fluoride contamination of groundwater resources in Ghana: Country-wide hazard modeling and estimated population at risk

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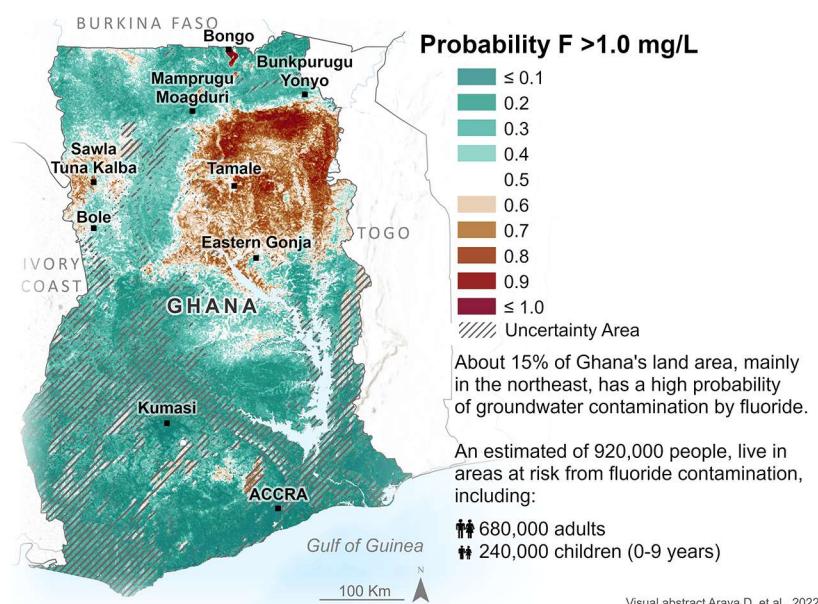
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Highlights

- Groundwater fluoride risk map ($> 1.0 \text{ mg/L}$) created with random forest modeling identifies risk areas in Ghana
- Geology and climate are major drivers of geogenic fluoride contamination
- Most affected areas are in the northeast of the country
- About 920,000 people, including 240,000 children (0–9 years), live in at-risk areas

Graphical abstract



Supplementary material

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Tables

S1. Fluoride data

Table S1. Groundwater fluoride measurement data collected for this study (n=3234) as well as compiled from existing sources .

Region	Source	No. of samples	% N F> 1.0 mg/L	% N F> 1.5 mg/L
Ashanti	(Boakye Opoku, 2013)	10	0%	0%
	Authors' data	2	0%	0%
	(Smedley, 1996)	4	0%	0%
	Total	16	0%	0%
Brong Ahafo	Authors' data	46	15%	2%
	UNHCR 2020	2	50%	0%
	Total	48	17%	0%
Central	(Agyemang, 2020)	14	0%	0%
	Authors' data	28	4%	4%
	(Mensah-Essilifie, 2013)	7	0%	0%
	UNHCR 2020	1	0%	0%
	Total	50	0%	0%
Eastern	Authors' data	177	15%	1%
	(Kulinkina et al., 2017)	172	5%	0%
	Total	349	0%	0%
Greater Accra	(Arko W. et al., 2019)	10	0%	0%
	Authors' data	6	17%	0%
	(Egbi et al., 2019)	17	0%	0%
	(Nkansah et al., 2019)	17	0%	0%
	Total	50	0%	0%
Northern	(Affam et al., 2012)	32	22%	19%
	(CIDA, 2011)	1039	26%	14%
	Authors' data	335	17%	6%
	(Gastineau, 2015)	32	31%	16%
	(Zango et al., 2019)	88	81%	69%
	Total	1526	0%	0%
Upper East	(Affam et al., 2012)	129	45%	41%
	(Anornu et al., 2017)	82	30%	13%
	(Chegbeleh et al., 2020)	39	13%	0%
	(CIDA, 2011)	420	18%	14%
	Authors' data	10	0%	0%
	(Smedley, 1996)	10	40%	40%
	(Smedley et al., 2002)	184	29%	23%
Upper West	Total	874	0%	0%
	(CIDA, 2011)	233	14%	8%
	(Smedley, 1996)	2	0%	0%
Volta	Total	235	0%	0%
	(Abusa et al., 2018)	34	0%	0%
	(Addo et al., 2011)	6	0%	0%
	(Avi et al., 2019)	9	11%	0%
	(CIDA, 2011)	9	22%	11%

	Authors' data (Egbi et al., 2019)	7	0%	0%
	Total	21	0%	0%
		86	0%	0%
	Total	3234	22%	13%

S2. Predictor variables

Table S2. List of variables tested for the modelling of groundwater contamination by fluoride. The variables finally used in the model are shown in grey.

Class	Predictor variable	Resolution	Reference
Soil	Clay fraction (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Sand fraction (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Silt fraction (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Cation exchange capacity of the soil (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Water capacity until wilting point (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Coarse fragments (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Soil pH (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Soil Nitrogen (2m depth)	250 meters	ISRIC (Hengl et al., 2015)
	Ustalfs	250 meters	ISRIC (Hengl et al., 2017)
	Lixisols	250 meters	ISRIC (Hengl et al., 2017)
Geology	Haplic Gleysols	250 meters	ISRIC (Hengl et al., 2017)
	Gskf - K-feldspar-rich granitoid, mainly granite and monzonite	polygon	GSD (GSD, 2009)
	Vba - Sandstone, mudstone, siltstone, and carbonate	polygon	GSD (GSD, 2009)
	VolSK - Sandstone	polygon	GSD (GSD, 2009)
	vos	polygon	GSD (GSD, 2009)
	bmrg	polygon	GSD (GSD, 2009)
	bsa	polygon	GSD (GSD, 2009)
	gvhr	polygon	GSD (GSD, 2009)
	bmbt	polygon	GSD (GSD, 2009)
	gsb	polygon	GSD (GSD, 2009)
	bvm	polygon	GSD (GSD, 2009)
	tmht	polygon	GSD (GSD, 2009)
	gvbm	polygon	GSD (GSD, 2009)
	vaf	polygon	GSD (GSD, 2009)
	bs	polygon	GSD (GSD, 2009)
	bvc	polygon	GSD (GSD, 2009)
	tmbt	polygon	GSD (GSD, 2009)
	tmbz	polygon	GSD (GSD, 2009)
	tmmg	polygon	GSD (GSD, 2009)
	vde	polygon	GSD (GSD, 2009)
	gsmg	polygon	GSD (GSD, 2009)
	qa	polygon	GSD (GSD, 2009)
	vdb	polygon	GSD (GSD, 2009)
	vcp	polygon	GSD (GSD, 2009)
	msd	polygon	GSD (GSD, 2009)
	gsbg	polygon	GSD (GSD, 2009)

	gshd	polygon	GSD (GSD, 2009)
	vts	polygon	GSD (GSD, 2009)
	vyb	polygon	GSD (GSD, 2009)
	dmgf	polygon	GSD (GSD, 2009)
	tcq	polygon	GSD (GSD, 2009)
	bmss	polygon	GSD (GSD, 2009)
	bv	polygon	GSD (GSD, 2009)
	gsht	polygon	GSD (GSD, 2009)
	bmb	polygon	GSD (GSD, 2009)
	bsw	polygon	GSD (GSD, 2009)
	tect	polygon	GSD (GSD, 2009)
	dmgi	polygon	GSD (GSD, 2009)
	acks	polygon	GSD (GSD, 2009)
	tpms	polygon	GSD (GSD, 2009)
	gvht	polygon	GSD (GSD, 2009)
Climate	Temperature	1 Kilometer	WorldClim (Fick and Hijmans, 2017)
	Priestley Taylor alpha coefficient (AET/PET)	1 Kilometer	CGIAR (Trabucco and Zomer, 2010)
	Potential evapotranspiration (PET)	1 Kilometer	CGIAR (Trabucco and Zomer, 2019)
	Actual evapotranspiration (AET)	1 Kilometer	CGIAR (Trabucco and Zomer, 2019)
	Mean annual precipitation	1 Kilometer	CGIAR (Trabucco and Zomer, 2019)
	Aridity (MAP/MAE)*	1 Kilometer	CGIAR (Trabucco and Zomer, 2019)
Topography	Slope	90 meters	HydroSHEDS/WWF(Lehner et al., 2008)
	Elevation	90 meters	HydroSHEDS/WWF(Lehner et al., 2008)
	Terrain forms	250 meters	OpenLandMap (Amatulli et al., 2018)
	Drainage basins	250 meters	
	Drainage density	250 meters	
	Topographic wetness index (TWI)	1 Kilometer	OpenLandMap (Hengl, 2018)
Others	Water table depth (WTD)	1 Kilometer	(Fan et al., 2013)
	Crop land	1 Kilometer	(ESA, 2017)

*Aridity = Mean Annual Precipitation (MAP)/Mean Annual Potential Evaporation (MAE)

S3. List of previous groundwater fluoride studies in Ghana

Table S9. List of previous fluoride studies by district and region. Included are studies that have provided direct or indirect reports of fluoride concentrations in Ghana. These studies were assigned to approximate locations within the corresponding districts. This facilitated the comparison of model results with results from previous studies.

District	Region	Studies
North Tongu	Volta	(Abusa et al., 2018; Ansa-Asare et al., 2009)
Awutu Senya	Central	(Agyemang, 2020; Amoako et al., 2011)
Agona East	Central	(Agyemang, 2020; Amoako et al., 2011)
New Juaben Municipal	Eastern	(Danquah et al., 2019)
Birim South	Eastern	(Kulinkina et al., 2017)
Birim Municipal	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017)
West Akim	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017)
Atiwa	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017)
Ayensuano	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017; Sunkari et al., 2019)
Upper West Akim	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017; Sunkari et al., 2019)
Suhum Municipal	Eastern	(Amoako et al., 2011; Kulinkina et al., 2017)
Kwaebibirem	Eastern	(Amoako et al., 2011; Kortatsi et al., 2008; Kulinkina et al., 2017)
Akyem Mansa	Eastern	(Amoako et al., 2011; Kortatsi et al., 2008; Kulinkina et al., 2017)
Wa Municipal	Upper West	(CIDA, 2011; Loh et al., 2012)
Nadowli-Kaleo	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi, 2009; Loh et al., 2012)
Mamprugu Moagduri	Northern	(Anku et al., 2009; CIDA, 2011; Gastineau, 2015; Kortatsi, 2009; Loh et al., 2012)
Nabdam	Upper East	(Anku et al., 2009; CIDA, 2011; Loh et al., 2012)
Lawra	Upper West	(Anku et al., 2009; CIDA, 2011; Loh et al., 2012)
Nandom	Upper West	(Anku et al., 2009; CIDA, 2011; Loh et al., 2012)
Builsa North	Upper East	(Anku et al., 2009; Anornu et al., 2017; CIDA, 2011; Loh et al., 2012)
Kasena Nankana East	Upper East	(Anku et al., 2009; Anornu et al., 2017; CIDA, 2011; Loh et al., 2012)
Kasena Nankana West	Upper East	(Anku et al., 2009; Anornu et al., 2017; CIDA, 2011; Loh et al., 2012)
Jirapa	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi et al., 2009; Loh et al., 2012)
Lambussie Karni	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi et al., 2009; Loh et al., 2012)
Daffiamma Bussie	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi et al., 2009; Loh et al., 2012)
Sissala West	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi et al., 2009; Loh et al., 2012)
Sissala East	Upper West	(Anku et al., 2009; CIDA, 2011; Kortatsi et al., 2009; Loh et al., 2012)
Bawku West	Upper East	(Anornu et al., 2017; CIDA, 2011; Loh et al., 2012)
Builsa South	Upper East	(Anornu et al., 2017; CIDA, 2011; Loh et al., 2012)
Wa East	Upper West	(CIDA, 2011; Kortatsi, 2009; Loh et al., 2012)
Cape Coast Metro	Central	(Mensah-Essilifie, 2013)
Ejura Sekye Dumase	Ashanti	(Osei-Nuamah, 2016)

Asante Akim North	Ashanti	(Osei-Nuamah, 2016)
Bunkpurugu Yonyo	Northern	(CIDA, 2011; Salifu et al., 2012)
Zabzugu	Northern	(CIDA, 2011; Salifu et al., 2012)
Tatale	Northern	(CIDA, 2011; Salifu et al., 2012)
Chereponi	Northern	(CIDA, 2011; Salifu et al., 2012)
Saboba	Northern	(CIDA, 2011; Salifu et al., 2012)
Yendi Municipal	Northern	(CIDA, 2011; Salifu et al., 2012)
Nanumba North	Northern	(CIDA, 2011; Salifu et al., 2012)
Nanumba South	Northern	(CIDA, 2011; Salifu et al., 2012)
West Mamprusi	Northern	(CIDA, 2011; Gastineau, 2015; Salifu et al., 2012)
Karaga	Northern	(CIDA, 2011; Salifu et al., 2012; Sunkari et al., 2020; Zango et al., 2019)
Mamprusi East	Northern	(CIDA, 2011; Salifu et al., 2012; Sunkari et al., 2020; Zango et al., 2019)
Akatsi North	Volta	(Abusa et al., 2018)
Ketu North	Volta	(Abusa et al., 2018)
Ketu South	Volta	Adoo 2011
Berekum	Brong Ahafo	(Akoto and Adiyah, 2007)
Jaman South	Brong Ahafo	(Akoto and Adiyah, 2007)
Sene West	Brong Ahafo	(Akoto and Adiyah, 2007)
Atebubu Amantin	Brong Ahafo	(Akoto and Adiyah, 2007; Tekpor Michael, 2012)
Upper Manya	Eastern	(Amevenku et al., 2012)
Ajumako-Enyan-Esiam	Central	(Amoako et al., 2011)
Gomoa West	Central	(Amoako et al., 2011)
Gomoa East	Central	(Amoako et al., 2011)
Agona West	Central	(Amoako et al., 2011)
Asikuma / Odoben / Brakwa	Central	(Amoako et al., 2011)
East Akim	Eastern	(Amoako et al., 2011)
Denkyembeh	Eastern	(Amoako et al., 2011)
Bolgatanga Municipal	Upper East	(Anku et al., 2009; Anornu et al., 2017; Apambire et al., 1997; CIDA, 2011; Loh et al., 2012; Zango et al., 2021)
Shai Osu Doku	Greater Accra	(Arko W. et al., 2019)
Kpone Katamanso	Greater Accra	(Arko W. et al., 2019)
Akwapem North	Eastern	(Arko W. et al., 2019)
Kma	Ashanti	(Nkansah et al., 2010)
Tamale North Sub Metro	Northern	(CIDA, 2011)
Tolon	Northern	(CIDA, 2011)
Savelugu Nanton	Northern	(CIDA, 2011)
Kumbumgu	Northern	(CIDA, 2011)
Sagnarigu	Northern	(CIDA, 2011)
Mion	Northern	(CIDA, 2011)
Nkwanta North	Volta	(CIDA, 2011)
Kpandai	Northern	(CIDA, 2011)
Krachi East	Volta	(CIDA, 2011)
Krachi Nchumuru	Volta	(CIDA, 2011)
Nkwanta South	Volta	(CIDA, 2011)
East Gonja	Northern	(CIDA, 2011)

Bole	Northern	(CIDA, 2011)
Wa West	Upper West	(CIDA, 2011; Loh et al., 2012)
Sawla/Tuna/Kalba	Northern	(CIDA, 2011; Loh et al., 2012, 2020)
Bongo	Upper East	(Affam et al., 2012; Alfredo et al., 2014; Anku et al., 2009; Anornu et al., 2017; Apambire et al., 1997; CIDA, 2011; Craig et al., 2018; Firempong et al., 2013; Loh et al., 2012; Smedley et al., 2002; Zango et al., 2021)
Pusiga	Upper East	(Anornu et al., 2017; CIDA, 2011; Zango et al., 2014)
West Gonja	Northern	(Arhin and Affam, 2010; CIDA, 2011)
Gonja Central	Northern	(Arhin and Affam, 2010; CIDA, 2011)
North Gonja	Northern	(Arhin and Affam, 2010; CIDA, 2011)
Gushiegu	Northern	(Anim-gyampo et al., 2012; CIDA, 2011; Salifu et al., 2012, 2013; Sunkari et al., 2020; Zango et al., 2019)
Garu Tempane	Upper East	(Anku et al., 2009; Anornu et al., 2017; CIDA, 2011; Zango et al., 2014)
Talensi	Upper East	(Anku et al., 2009; Chegbeleh et al., 2020; CIDA, 2011)
Bawku Municipal	Upper East	(Anku et al., 2009; CIDA, 2011; Zango et al., 2014)
Binduri	Upper East	(Anku et al., 2009; CIDA, 2011; Zango et al., 2014)
South Tongu	Volta	(Egbi et al., 2019)
Ada East	Greater Accra	(Egbi et al., 2019)
Birim North	Eastern	(Kortatsi et al., 2008)
Adansi South	Ashanti	(Kortatsi et al., 2008)
Adansi North	Ashanti	(Kortatsi et al., 2008)
Bosome Freho	Ashanti	(Kortatsi et al., 2008)
Asante Akim South	Ashanti	(Kortatsi et al., 2008)
Asante Akim Central Municipal	Ashanti	(Kortatsi et al., 2008)
Ejis Juaben	Ashanti	(Kortatsi et al., 2008)
Bosomtwe /Atwima / Kwanwoma	Ashanti	(Kortatsi et al., 2008)
Tarkwa Nsuaem	Western	(Kortatsi, 2009)
Prestea / Huni Valley	Western	(Kortatsi, 2009)
Wassa Amenfi East	Western	(Kortatsi, 2009)
Wassa Amenfi West	Western	(Kortatsi, 2009)
Wassa Amenfi Central	Western	(Kortatsi, 2009)
ingo Prampram	Greater Accra	(Nkansah et al., 2019)
Accra Metropolis	Greater Accra	(Nkansah et al., 2019)
La Dade Kotopon	Greater Accra	(Nkansah et al., 2019)
Ga Central Municipal	Greater Accra	(Nkansah et al., 2019)
Ga South	Greater Accra	(Sunkari et al., 2019)
Ga East	Greater Accra	(Sunkari et al., 2019)
Ga West	Greater Accra	(Sunkari et al., 2019)

Figures

S1. Geology of Ghana

The country's geology can be broadly subdivided into four major distinct lithostratigraphic/lithorectonic complexes (Geological Survey Department, GSD, 2009). The first is constituted by the Paleoproterozoic supracrustal and intrusive rocks (i.e. Birimian Supergroup, Tarkwaian Group, Tamnean Plutonic Suite and Eburnean Plutonic Suite), which formed between 2195 Ma and 2072 Ma. The second complex unit, formed between 1000 and 950 Ma, is the Neoproterozoic to early Cambrian, lithologically diverse platform of sediments (Voltaian Supergroup). The main stratigraphic units under the Voltaic Supergroup include the Kwahu Morago Group (also called Bombouaka) at the base, followed by the Oti-Pendjari Group, which was deposited after 600 Ma, and for the subsequent deposition of the Obosum Group in the late Neoproterozoic to early Cambrian. The third distinct complex is the rocks associated with the Panafrican Dahomeyide orogenic belt, listed in order of the degree of metamorphism and deformation: the Buem Structural Unit, the Togo structural Unit, a variety of gneisses associated with the Dahomeyan Supergroup (peak metamorphism at c 600 Ma) and some interleaved Eburnean protoliths. The fourth complex unit is the Coastal Sedimentary basin of Ordovician to Cretaceous age, mostly related to the opening of the Atlantic or proto-Atlantic Ocean. Major groups associated with this formation include Sekondian, Accraian, Amisian and the Apollonian Group).

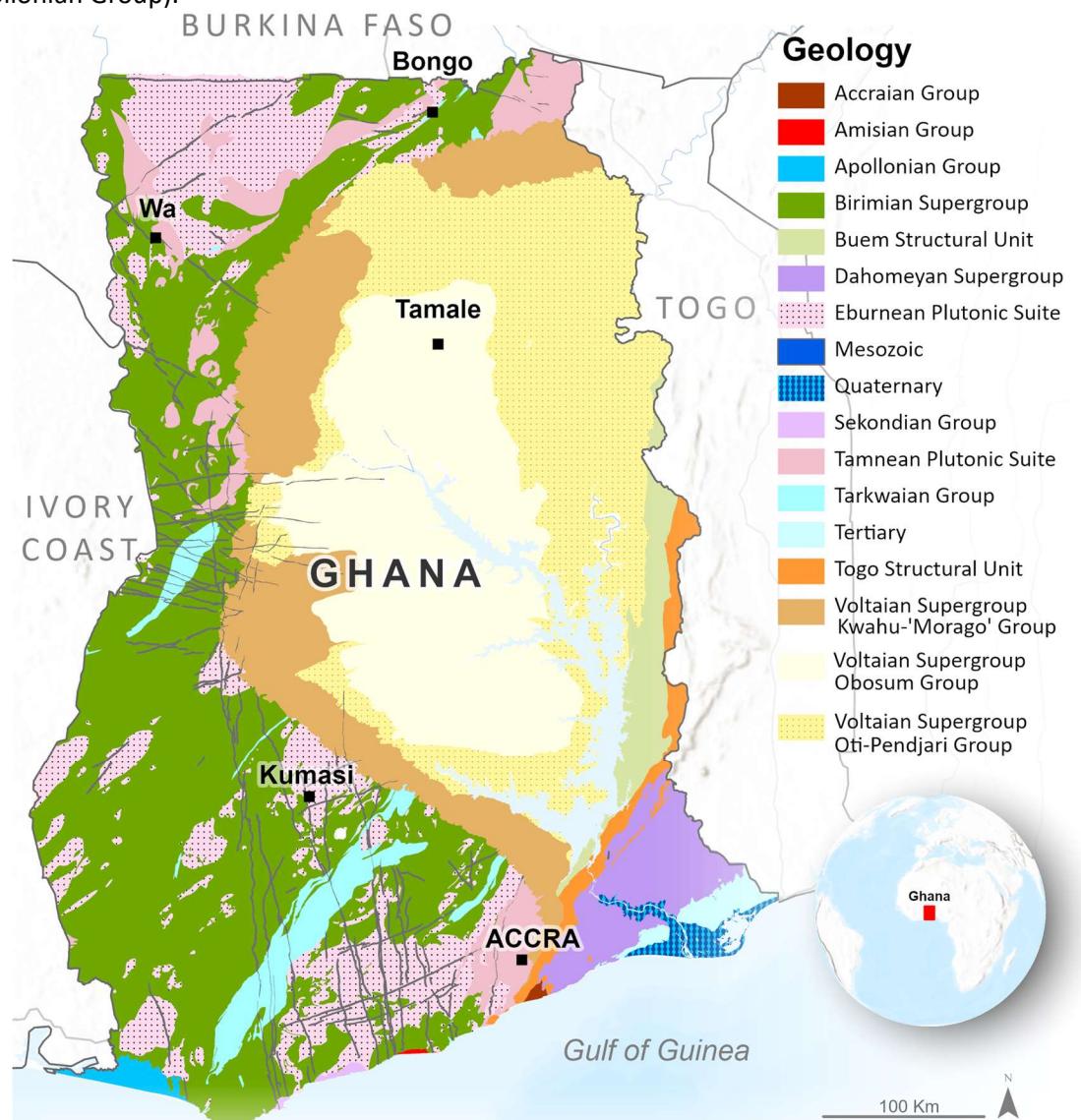


Fig. S1. Geology map of Ghana.

S2. Predictor variable maps

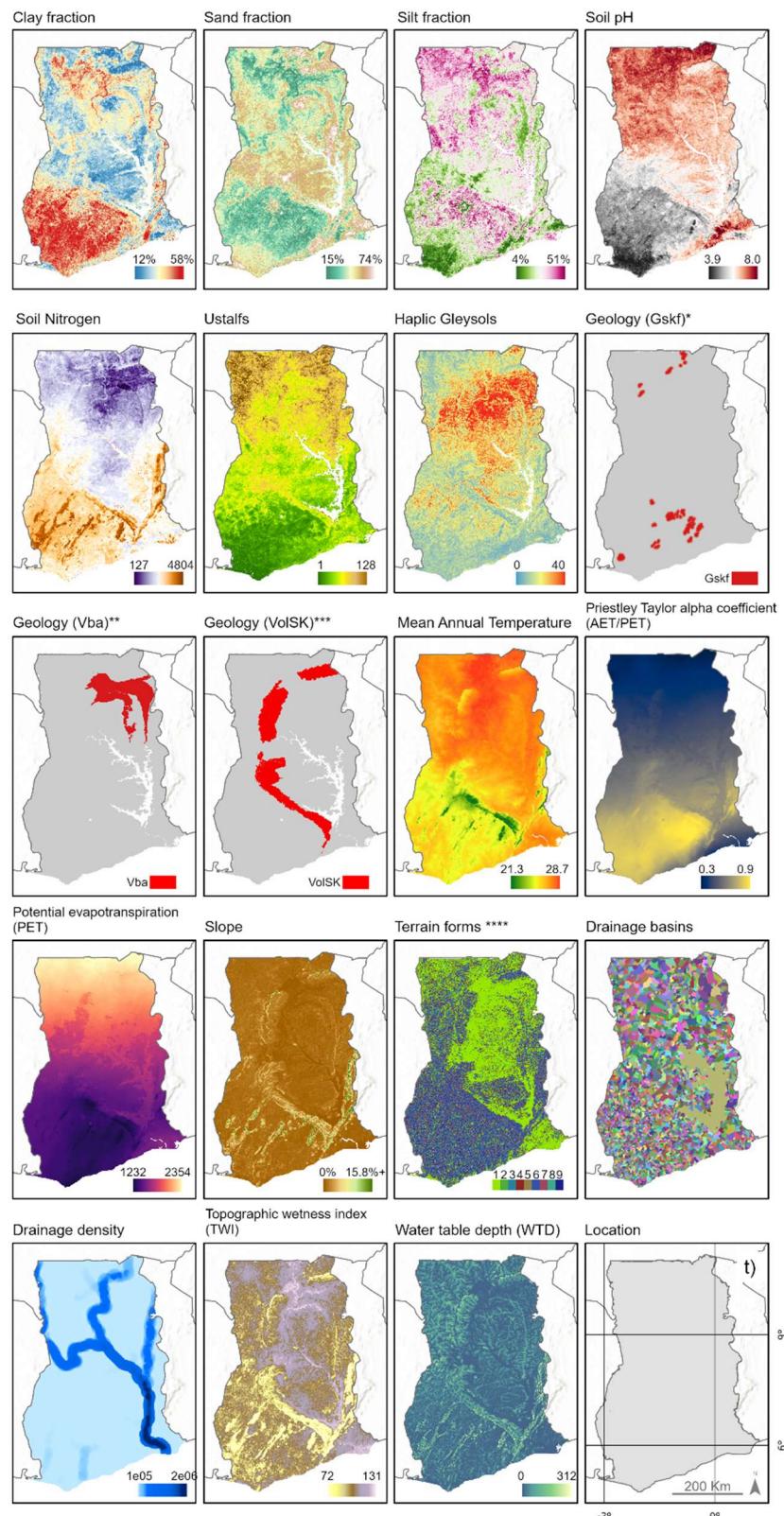


Fig. S2. Predictor variables. *Gskf – Eburnean Plutonic Suite – K-feldspar-rich granitoid, mainly granite and monzonite, **Vba – Voltaian Supergroup, Oti-Pendjari Group – Sandstone, mudstone, siltstone, and carbonate, ***VolSK – Voltaian Supergroup, Kwahu-Morago – Sandstone, ****Terrain forms (1 flat, 2 summit, 3 ridge, 4 shoulder, 5 spur, 6 slope, 7 hollow, 8 footslope, 9 valley).

S3. Spatial cross-validation

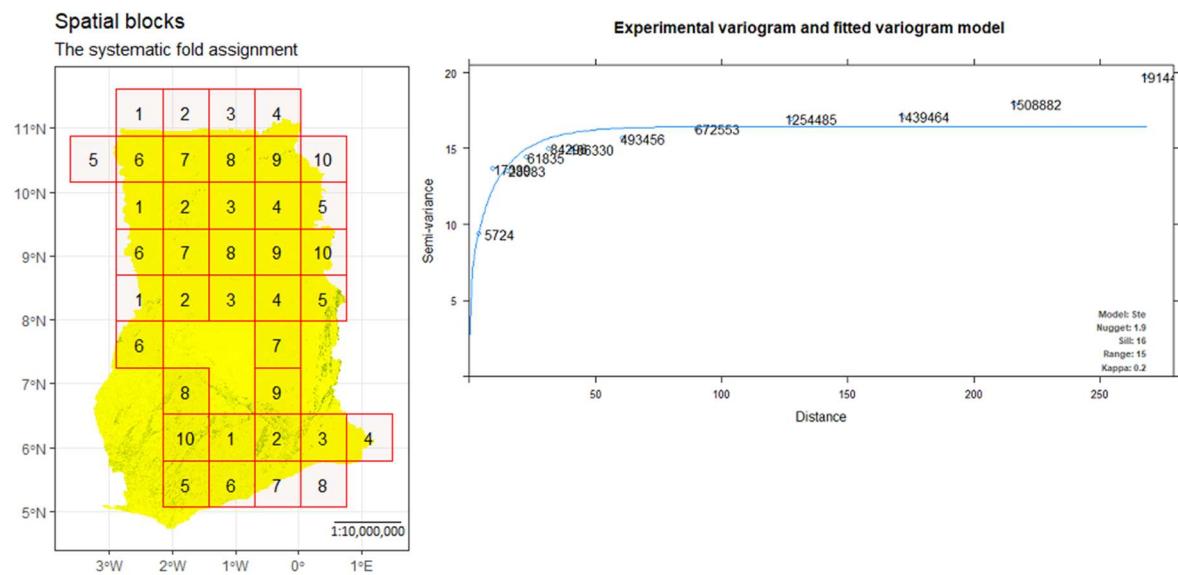


Fig. S3. Spatial cross validation. (a) Fold assignment, and (b) semivariogram. The semivariogram shows a sharp loss of spatial autocorrelation between the predictor variables at distance over 76 km.

S4. Partial dependence plots for the predictor variables

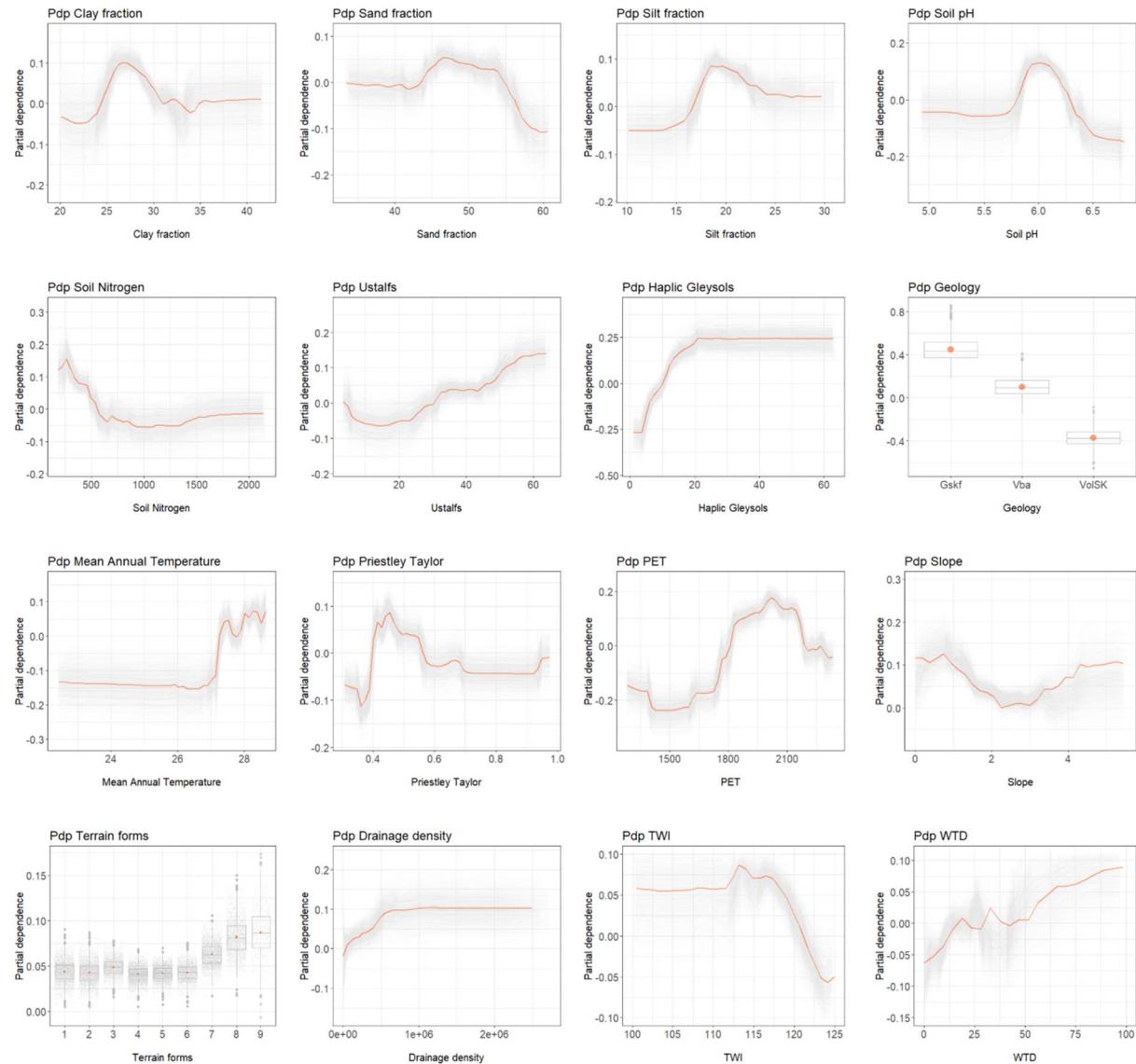


Fig. S4. Partial dependence plots for the predictor variables. The x-axis shows the distribution of the data for the explanatory variable, and the y-axis reports the impact of the variable on the prediction of fluoride in concentrations greater than 1.0 mg/L. The geology and PET graphs have a different scale on the y-axis than those shown in Figure 3b; this is because in Figure 3b they are presented in terms of probability. Terrain forms (1 flat, 2 summit, 3 ridge, 4 shoulder, 5 spur, 6 slope, 7 hollow, 8 footslope, 9 valley).

S5. Maps indicating the accuracy of the predictions overlaid on the map of Ghana

We identified regions in Ghana with groundwater fluoride levels exceeding 1.0 mg/L by averaging the 1000 probability maps of the country. We then classified the fluoride hazard in groundwater as high or low using the average of the probability cut-offs of the 1000 runs. The cut-off was chosen at the point at which the sensitivity and specificity of the analysis are equal; in a balanced sample between low and high values, this usually corresponds to the highest overall accuracy over all cut-off points. This was used to identify the high hazard areas ($\geq 49\%$ probability), which are shown in gray in Fig. S6a. This area represents 17.6% (~42,100 km²) of the country and is mainly located in the northeastern region of the country. As shown in Fig. S6b, this predicted area strongly agrees with the distribution of the ground truth sampling points with fluoride > 1.0 mg/L. A standard deviation ($SD \leq 0.53$), represented in dark pink in Fig. S6c, indicates that no significant discrepancies exist between the 1000 high hazard area maps (see methods 2.5). The pixels with a standard deviation equal to or greater than 0.53 make up only 0.7% of the fluoride risk area and are primarily located at the boundary of the risk area. Finally, Fig. S6d shows that, in 72.6% of the study area, the model can learn from the training data; therefore, the estimated cross-validation performance is reliable. This area covers most of the northern part of the country, where most of the high fluoride areas are located. The areas outside the area of applicability represent uncertain quality of prediction and are mainly located in the southwestern part of the country and represent 27.4% of the study area. Overall, the effect of excluding pixels with a standard deviation above a threshold and the area outside the AOA from the map of high hazard areas of fluoride concentration is minimal; therefore, the final map of the high hazard areas represents 15.6% (~37,300 km²) of the area of the country.

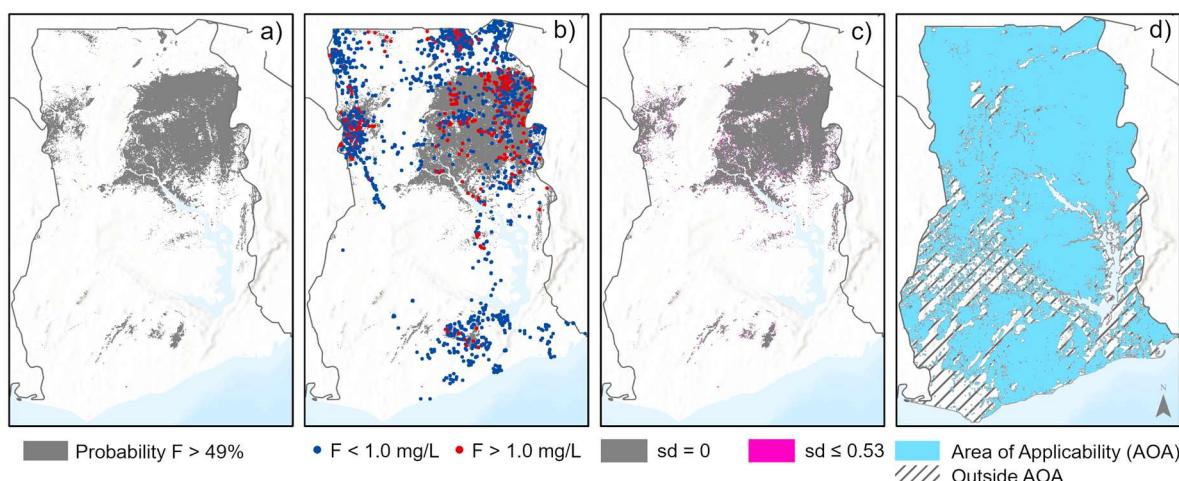


Fig. S5. Maps indicating the accuracy of the predictions overlaid on the map of Ghana. (a) Map of high hazard areas ($\geq 49\%$ probability) and (b) plotted against the sampling points supplied by field testing. Colors indicate fluoride concentration above (red) and below (blue) 1.0 mg/L. (c) The standard deviation of the high-hazard maps for the minimum, average, and maximum probability cut-off points of the 1000 iterations, with dark pink pixels highlighting disagreements ($SD \leq 0.53$) on the classification of the high-probability areas across the 1000 maps. (d) Area of applicability map indicating the pixels where the prediction is reliable (light blue) and where the prediction is uncertain (hatched gray shading). All the maps were created by the authors using ArcGIS Pro v.2.7.2 software. Base maps are from Esri within ArcGIS Pro v.2.7.2 and are credited to: Sources: Esri, Airbus DS, USGS, NGA, NASA, CGIAR, N Robinson, NCEAS, NLS, OS, NMA, Geodatastyrelsen, Rijkswaterstaat, GSA, Geoland, FEMA, Intermap, and the GIS user community.

S6. Proportion of children aged 0–9 years in relation to the total population for each district

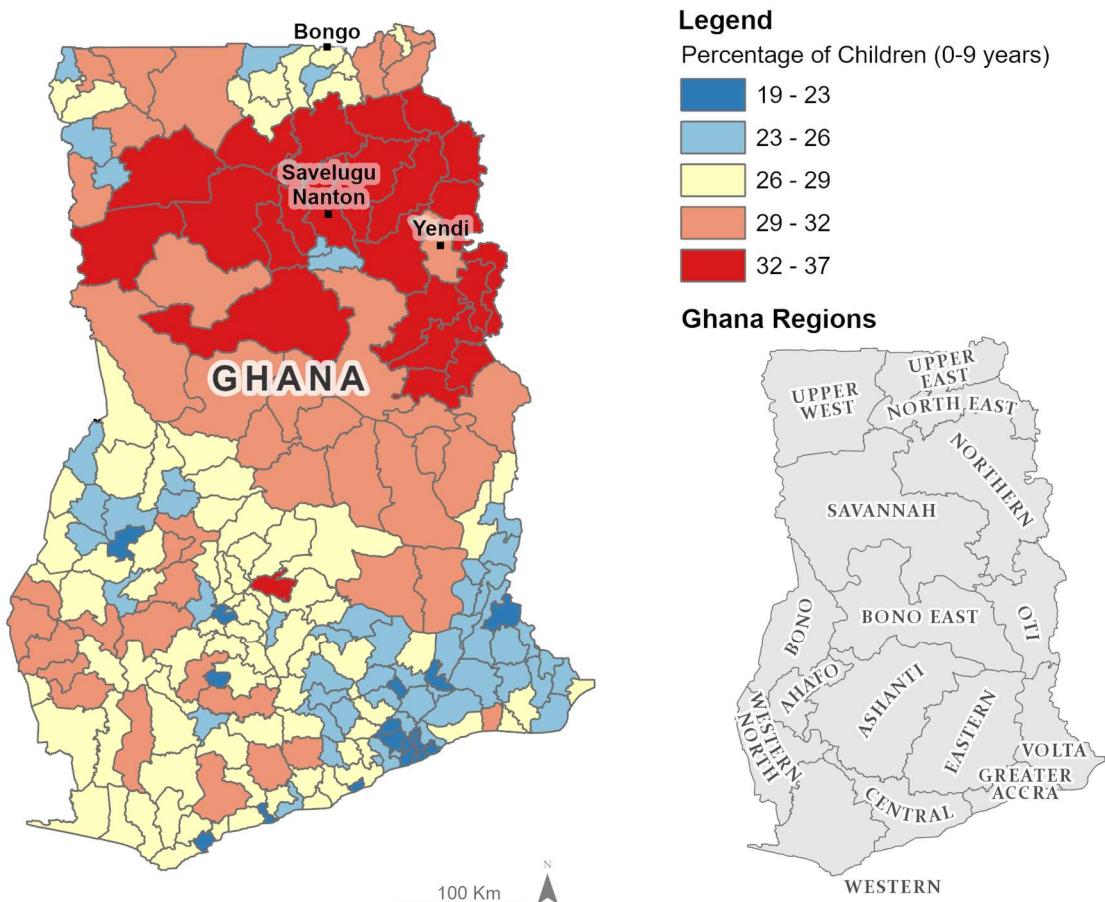


Fig. S6. Proportion of children aged 0–9 years at district level in relation to the total population. The Northern and Savannah regions have the highest proportions of children (GSS, 2020). All the maps were created by the authors using ArcGIS Pro v.2.7.2 software. Base maps are from Esri within ArcGIS Pro v.2.7.2 and are credited to: Sources: Esri, Airbus DS, USGS, NGA, NASA, CGIAR, N Robinson, NCEAS, NLS, OS, NMA, Geodatastyrelsen, Rijkswaterstaat, GSA, Geoland, FEMA, Intermap, and the GIS user community.

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