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Fluoride Contamination of Groundwater from Semi-Arid Regions of Western India

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ABSTRACT

A study on fluoride risk assessment was carried out in the semi-arid region of North Gujarat, India. The intricate link between groundwater fluoride and human health, lack of awareness, limited access to fluoride treatment facilities, and poor socio-economic conditions of the ~5.0 million rural population in the studied region make them vulnerable to fluoride. This study aimed to evaluate non-carcinogenic health risk, its severity, and the total population at risk in these regions due to chronic fluoride exposure. Fluoride in our samples (n=132) exhibits large spatial variability, and it ranges from ~0.13-8.64 $mg.L^{-1}$ (average: 1.64 ± 1.50 $mg.L^{-1}$) and 43% of them are more than the WHO limit of 1.5 $mg.L^{-1}$. Hazard Quotient (HQ) was used to assess health risks through the ingestion exposure route. The comparison of the range (0.1-8.55 versus 0.06-4.11), average (1.63 ± 1.49 versus 0.78 ± 0.72), and median (1.26 versus 0.60) of HQ between children and adults highlights that the former are at more risk compared to latter. Our conservative estimates suggest that ~0.45 million children and ~1.06 million adult population, ~55% and ~20% of the respective population classes, of the region, are potentially at risk. The empirical Bayesian Kriging model was used to produce risk-assessments maps. These can help policymakers in prioritizing the application of mitigation funding and resources, and in increasing testing efforts in high-risk areas. We believe this study should guide policymakers to adopt strategies in ensuring the public health safety of the rural population, children in particular, of the studied region.

INTRODUCTION

Human health risk associated with high fluoride in drinking water continues to draw considerable attention even in recent years (Ahada & Suthar 2019, Zhang et al. 2020) as it used to draw about a few decades back (Dissanayake 1991, Ozsvath 2009). The intricate link between dissolved fluoride and human health necessitates a critical understanding of the problem and requires approaches (e.g., modeling and simulation, direct measurements, etc.) to determine the associated human health risk. Geogenic sources of fluoride include minerals like fluorite, amphiboles, apatite, and amphiboles (Handa 1975, Hem 1985) while anthropogenic sources include phosphatic fertilizers (Kundu & Mandal 2009), brick kiln industries (Datta et al.1996), industries like aluminum smelting industries and coal-based power plants (Ali et al. 2016). Chronic consumption of drinking water with fluoride >1.5 $mg.L^{-1}$ is known to cause different types forms of fluorosis such as degradation of dental enamel, skeletal deformities, ligament-calcification, osteosclerosis, and crippling deformities of joints and spine (Dissanayake et al. 1991, Biglari et al. 2016). Recent studies have also indicated that excess fluoride consumption may even be the cause of neurological effects (Jiang et al. 2019); genetic effects (Cao et al. 2016); insulin resistance (Dey & Giri 2016); urinary tract diseases (Jha et al. 2011); thyroid hormone issues (Kravchenko et al. 2014); respiratory problems (Follin-Arbelet & Moum 2016) and may even cause cancer in bone, lungs, bladder, and uterus (Yang et al. 2000).

India accounts for ~50% of the total fluorosis-affected world population (Vithanage & Bhattacharya 2015, Podgorski et al. 2018). With a ~60% rural population, the Western state of Gujarat is one of the most severely affected states. The rural regions of Gujarat are mostly semi-arid and surface-water-scarce resulting in high reliance of residents on groundwater. Due to poor literacy rates and socio-economic conditions, these rural residents lack awareness about fluoride hazards, and with limited access to water/fluoride treatment facilities, they are coerced to consume untreated groundwater, resulting in their exposure to fluoride (and other contaminants). A few recent articles have highlighted the problem of high fluoride in Gujarat; for example, in Mehsana district Mandal et al. (2021) estimated that ~0.073 million children and ~0.467 million adults are at risk of fluorosis. Similarly (Shirke et al. 2020) reported >1.5 $mg.L^{-1}$ in 40%

of the wells in groundwater of the Amba Dongar region. Both these studies inferred that children are at higher risk compared to adults. Additionally, it is also socially pertinent to assess the number of people potentially affected by fluoride-related morbidities in these rural regions. To the best of our knowledge, such studies on Gujarat are at best very sparse (Mandal et al. 2021) despite predictive modeling assessment (Podgorski et al. 2018) indicating that ~11 million population of the state are potentially at risk.

We hypothesize that given the similar level of groundwater contamination, rural populations are likely to be impacted more compared to urban counterparts as the former lack awareness about contamination/hazards, and rural regions lack infrastructure facilities such as fluoride treatment plants. To examine the same, as a case study we carried out a first-time investigation on groundwater fluoride contamination from two northern districts of Gujarat which covers a combined area of ~16000 km^2 with a present-day cumulative population of ~5 million. The hazard quotient is used to estimate the non-carcinogenic risk and its severity, and subsequently, the total population at risk in these regions due to chronic fluoride exposure in groundwater was evaluated. Furthermore, the Empirical Bayesian Kriging model was used to identify the high and low-risk zones in these districts.

MATERIALS AND METHODS

Study Area

Banaskantha district: Banaskantha district (23°33' N-24°25' N; 71°07'E- 73°02' E) consists of twelve sub-districts (viz. Danta, Amirgadh, Dantiwada, Tharad, Vav, Bhabhar, Vadgam, Deodar, Kankrej, Deesa, Dhanera, and Palanpur; (Fig. 1(a)) and have a geographical area of 10, 303 km^2. It consists of 1249 villages with a population of ~3.12 million as per the 2011 census (https://www.censusindia.co.in/district/banas-kantha-district-gujarat-469). Long-term data suggest that average maximum and minimum temperatures were ~34°C and ~19°C respectively. The climate of the district is semi-arid, with an average yearly rainfall of 580 mm. Ephemeral rivers Banas and Saraswati constitute the drainage network of the district (CGWB 2011).

Patan district: Patan district is positioned between 23°24' and 24°09' N latitudes and 71°01' and 72°30' E longitudes in the northern part of Gujarat (Fig 1(b)). It occupies a geographical area of 5740 km^2. The district is divided into seven sub-districts and consists of 517 villages with a total population of ~1.34 million as per the 2011 census (https://www.censusindia.co.in/district/patan-district-gujarat-470). The average maximum temperature is 34.4°C during summer and the minimum temperature is 19.5°C during winter. The district witnesses very low average annual rainfall (403

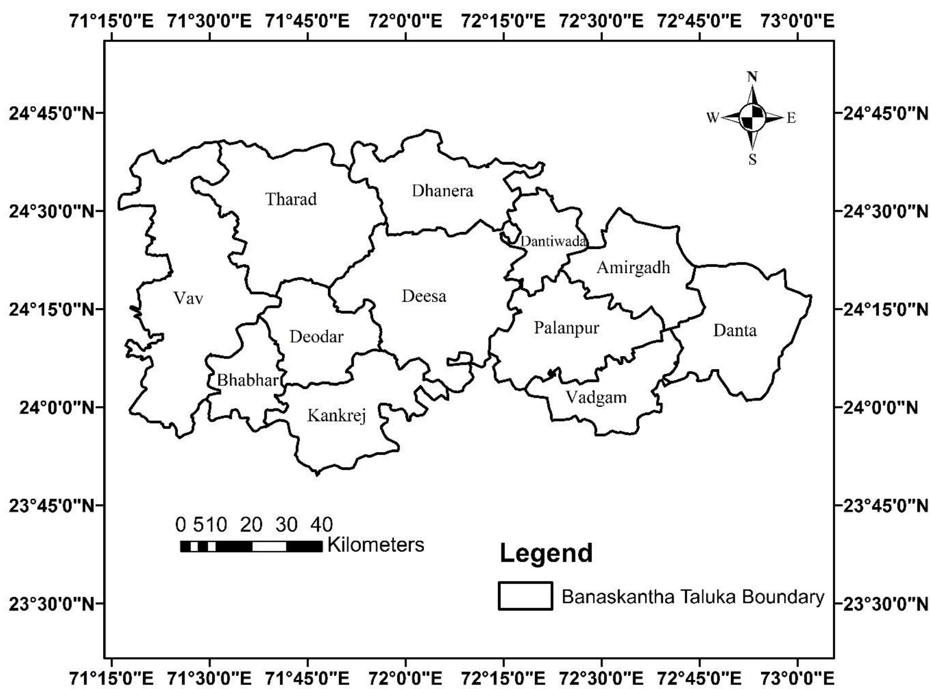

Fig. 1(a): Sub-district map of Banaskantha district.

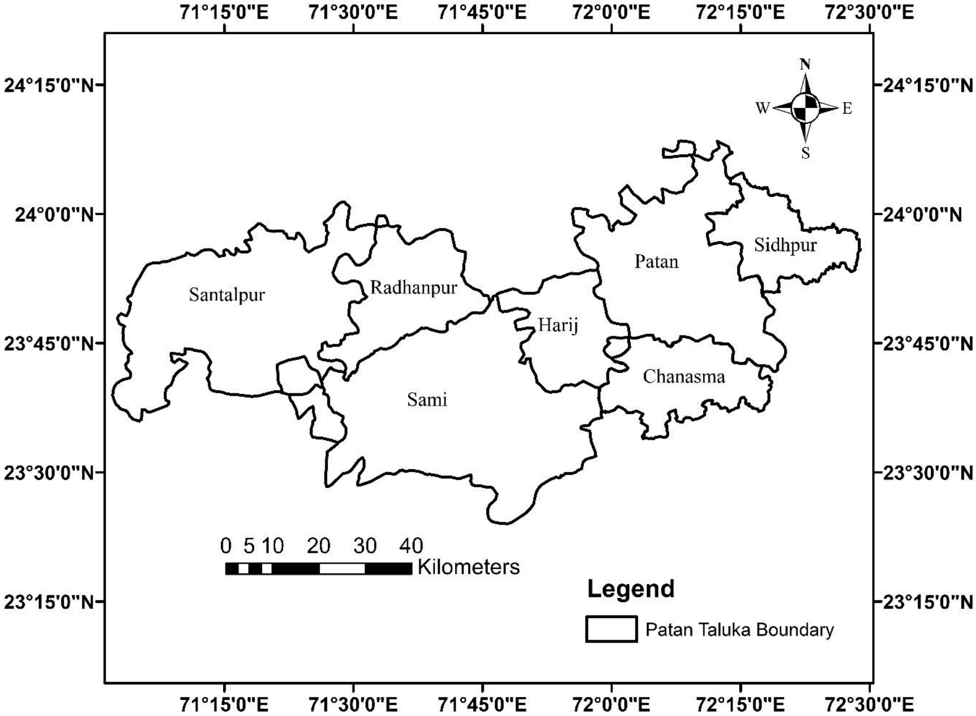

Fig. 1(b): Sub-district map of Patan district.

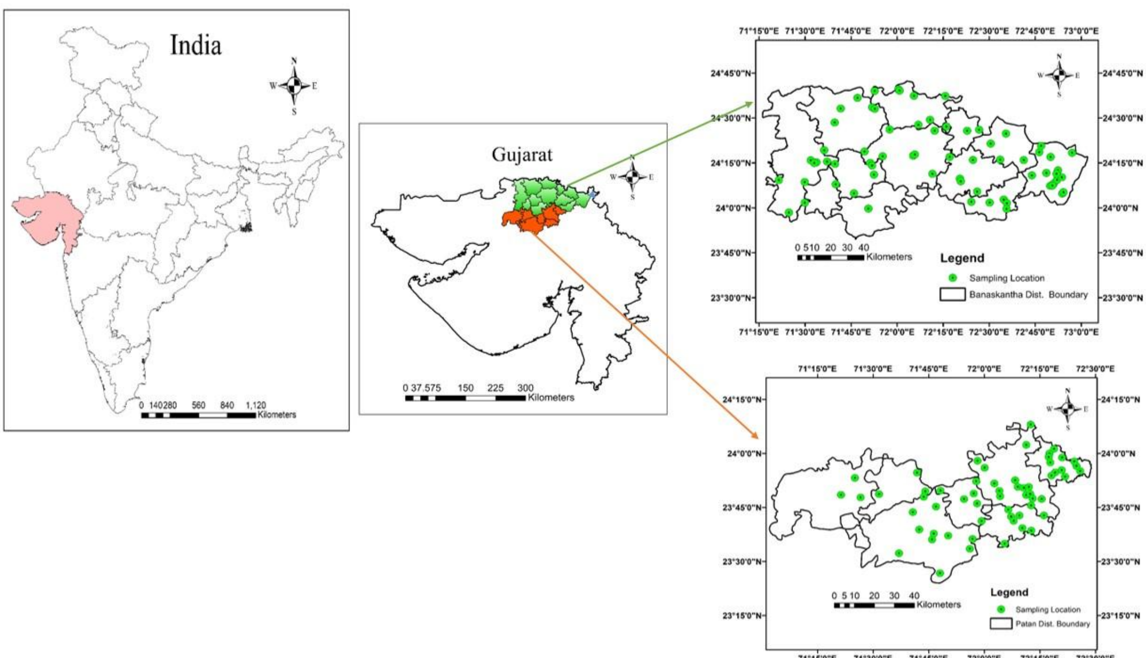

Fig. 1(c): Study area, and location map of the sampling sites in the Banaskantha and Patan districts.

mm) with Khari, Banas, and Umardasi being the three rivers basins (CGWB Report 2014). The district is predominantly covered by alluvium. It has a multi-layer aquifer system that comprises semi-consolidated Mesozoic and Tertiary formations and unconsolidated quaternary alluvial deposits.

Sampling and Analyses

Groundwater samples were collected either from hand pumps, dug wells, or open wells of the study area. After sufficient purging, bottles were rinsed thoroughly and samples were collected without bubbles into separate bottles each for measurements of alkalinity and major ions, water isotopes, carbon isotopes, and trace elements. In total samples were collected from 132 locations - 72 from the Banaskantha district and 60 from the Patan district (Fig. 1(c)). In some areas of the western part of the Banaskantha district, groundwater samples were not collected as the local population is dependent on the treated river water source for their daily consumption. Random sampling was done from different villages of each sub-district.

In-situ parameters such as pH, temperature, and conductivity measurements were made using portable meters (Eutech PCD 650) with respective precisions better than 0.002 units, 0.1°C, 1 $\mu S.cm^{-1}$, and 1 $mg.L^{-1}$. After collection, samples were stored at 4°C in the laboratory until the analysis of anions. Fluoride and other anions (Cl, SO_4, and NO_3) were analyzed in (0.45 µm) syringe filtered samples by Ion−chromatography (Thermofisher Scientific). AG23 and AS23 columns were used for the separation of the anions, then eluted with a mixture of 0.8 mM $NaHCO_3$ and 4.5 mM $NaCO_3$ solution, and finally detected with a suppressed electrical conductivity detector. The detector response was calibrated using laboratory-made mixed standards and the consistency of the instrument performance was ensured by monitoring the detector sensitivity throughout the analysis. Overall reproducibility ascertained by a coefficient of variation in repeat measurements is within ±5% for fluoride.

Health Risk-Assessment

Non-carcinogenic risks due to fluoride can arise from both oral and dermal exposure routes. In this study, the dermal risk is not taken into consideration as it was found to be very insignificant. Estimation of daily intake (EDI) and Hazard Quotient (HQ) due to fluoride has been made for the oral pathway following USEPA (1989).

$$EDI_{ORAL} = (C \times IR \times EF \times ED) / (BW \times AT) \quad \ldots(1)$$

$$HQ_{ORAL} = EDI_{ORAL} / RfD_{ORAL} \quad \ldots(2)$$

In the above equations, C is the measured concentration of fluoride in groundwater ($mg.L^{-1}$); IR is daily water intake rate ($L.day^{-1}$); EF is exposure frequency ($day.y^{-1}$); ED is exposure duration (y); AT is averaging time (day) and BW is average body weight (kg) of the consumer (e.g., adult or children). HQ_{ORAL} has been calculated separately for adults and children. Values of 350 days per year (EF), 30 years (ED), 10500 days (AT); 2 L day^{-1} (IR), and 70 kg (BW) are used for adults while the corresponding values used for children are 350 days per year (EF), 6 years (ED), 2100 days (AT); 0.89 $L.day^{-1}$ (IR) and 15 kg (BW). RfD_{ORAL}, the reference oral dose, has been taken as 0.06 mg $F.kg^{-1}$-body weight. day^{-1} (USEPA 1989, Ali et al. 2019). Using these values it is found that the product of EF and ED is equal to that of AT, thus equation 2 can further be simplified to:

$$HQ_{ORAL} = C \times IR / (RfD_{ORAL} \times BW) \quad \ldots(2a)$$

The magnitude of HQ determines the probabilistic non-carcinogenic risk with HQ values <1 indicating that the water is safe for consumption while HQ_{ORAL}>1 indicates potential risks of fluorosis.

Geostatistical Modeling

In our study, ESRI ArcGIS 10.7 software (License no - EFL000908614 ArcGIS Pro Geostatistical Analyst) was used to perform the geostatistical modeling. Empirical Bayesian Kriging (EBK) was applied to calculate HQ values to map its spatial variation and to identify the high-risk zones within the study area. The EBK model was preferred as it was reported to be more realistic and comprehensive than other interpolation techniques (Krivoruchko 2012, Mukherjee et al. 2019). It differs from the classical Kriging model as it automates the critical aspect of building a valid Kriging model through subsetting and simulation. In this interpolation technique, errors are estimated based on semivariograms obtained through repeated simulation (Emenike et al. 2018) whereas the other techniques need manual adjustment for accuracy. The accuracy and robustness of the EBK model were gauzed by the calculated error parameters such as root-mean-square predicted (RMSP), mean standardized error (MS), root-mean-square-standardized (RMSS), and average standard (AS) error.

RESULTS AND DISCUSSION

Fluoride Distribution

Fluoride concentrations in our groundwater samples are reported in Table 1. The fluoride in our samples (n=132) ranged from 0.13 to 8.64 $mg.L^{-1}$ with an average of 1.65 ± 1.50 $mg.L^{-1}$ and a median value of 1.27 $mg.L^{-1}$. The observed range and average for the Banaskantha region are 0.13-6.03 $mg.L^{-1}$ and 1.68 ± 1.27 $mg.L^{-1}$ while for the Patan region these values are 0.18-8.64 $mg.L^{-1}$ and 1.61 ± 1.75 $mg.L^{-1}$

respectively. About 44% (32 of the 72) and 42% (25 of the 60) of the samples collected in Banaskantha and Patan districts respectively have fluoride above the WHO's maximum permissible limit (MPL) of 1.5 $mg.L^{-1}$ for safe drinking. The distribution pattern of fluoride levels across the study area indicates that ~20% of the samples had <0.5 $mg.L^{-1}$; 22% within 0.5-1.0 $mg.L^{-1}$; 15% fall within the regulatory limits of 1.0-1.5 $mg.L^{-1}$ while 43% of the samples having fluoride higher than MPL.

Fluoride Risk-Assessment

In our study area, fluoride >1.5 $mg.L^{-1}$ in 43% of the samples (n=132) underscores the significance of assessing the risk due to the consumption of groundwater. The dermal risk appears to be insignificant compared to oral intake. Health risk assessment (HQ; equation 2) depends on three parameters, of which, the concentration of fluoride is measured, while IR and BW are average values recommended by environmental/health/medical agencies (e.g., USEPA, and Indian Council of Medical Research and National Institute of Nutrition (ICMR-NIN); Health Canada).

USEPA recommends the use of IR value of 2 $L.day^{-1}$ and 0.89 $L.day^{-1}$ for adults and children respectively, whereas the corresponding values for BW are 70 kg and 15 kg. For India, where most of the regions are hot (semi)-arid or humid, average water consumption by working rural resident adults can be higher than 2 $L.day^{-1}$ and thus the fluoride exposure may even be higher than calculated using USEPA parameters. Recently ICMR-NIN has reported values of water intake for Indian adult males (32-58 $mL.kg^{-1}$ BW), adult females (27-52 $mL.kg^{-1}$ BW), and children (60 $mL.kg^{-1}$ BW) —the lower limit and upper limit of the range are associated with sedentary-working and high-working persons (ICMR-NIN report, 2020). Similarly, the recommended average BW of adult males and adult females are 65 kg and 55 kg respectively. Based on these average BW values, the IR range translates to 2.1-3.7 $L.day^{-1}$ (adult male), 1.5-2.9 $L.day^{-1}$ (adult female) and 0.9 $L.day^{-1}$ (children). Calculated HQs using USEPA, and the range of ICMR-NIN parameters yield variable results (summarized in Table 1) showing that choice of average IR and BW values are critical. We have however used the lower-bound estimate of HQ (i.e., USEPA parameters) for evaluating risk and geospatial HQ mapping of the region. The distinction between males and females has not been made while calculating HQ values for adults though females are prone to more risk because of their lower BW (Table 1).

Calculated HQ values range from 0.06-4.11 for adults (average: 0.78 ± 0.72; median: 0.60) and 0.13-8.55 for children (average: 1.63 ± 1.49; median: 1.26) highlighting that children are more prone to risk because of their lower body weight though somewhat offset by their lower daily water consumption compared to adults. This inference is similar to the finding in previous studies (Adimalla et al. 2018, Ahada & Suthar 2019). To get better insight into the

Table 1: Calculated HQ_{Oral} values for adults and children using USEPA and ICMR-NIN parameters. The HQ values show high variability.

Reference	HQ								
	Population class	IR [$L.day^{-1}$]	BW [kg]	Min.	Max.	Average	Standard deviation	Median	% samples with HQ>1
USEPA	Adult	2	70	0.06	4.11	0.78	0.72	0.6	28
USEPA	Children	0.89	15	0.13	8.55	1.63	1.49	1.26	58
ICMR-NIN, India	Adult (male)	2.1	65	0.07	4.65	0.88	0.87	0.68	35
	Adult (male)	2.9	65	0.1	6.43	1.22	1.2	0.94	45
	Adult (male)	3.7	65	0.12	8.2	1.55	1.53	1.2	56
ICMR-NIN, India	Adult (female)	1.5	55	0.06	3.93	0.74	0.73	0.58	26
	Adult (female)	2.2	55	0.09	5.76	1.09	1.08	0.85	43
	Adult (female)	2.9	55	0.11	7.59	1.44	1.42	1.12	52
ICMR-NIN, India	Children	0.9	15	0.13	8.64	1.65	1.5	1.27	58

potential risk to the population, HQ values were analyzed sub-district-wise. This approach is particularly beneficial to undertake mitigation measures at smaller space-domain and targeting smaller populations by the local authorities. Significant percentages of samples having HQ_{adult} >1 are observed for Danta (67%), Dhanera (43%), Vadgam (40%), Dantiwada (33%), Palanpur (25%), Amirgarh (25%), Sidhpur (44%), Patan (43%) and Harij (33%). Importantly, the fact that in the remaining 11 sub-districts less than 20% of the collected samples is having HQ_{adult} >1 underscores the lesser risk associated with the local population in them.

The oral exposure risk turns out to be more serious for children with 76 of 133 samples (58%) having HQ_{child} >1. In our study area sub-districts with ≥50% of the collected samples having HQ_{child} >1 are Danta (100%), Dhanera (86%), Vadgam (60%), Dantiwada (100%), Palanpur (75%), Deesa (75%), Tharad (50%), Sidhpur (69%), Patan (57%), Radhanpur (67%) and Chanasma (55%). There are only three sub-districts (viz. Bhabhar, Sami, and Santalpur) in the entire region wherein the calculated exposure risk to children is nominal with no samples having HQ_{child} >1.

Population at Potential Risk

Translating the facts above (discussed in section 3.2) to the total adult and total child population potentially at risk in each of the sub-districts rests on the premise that the groundwater sampling is representative, and it requires population data in each sub-district and the % of risk-prone samples. High resolution and seasonal sampling address biases and uncertainties in representative sampling. Reliable estimates of sub-district level population data are available from the census of 2011. Two approaches can be adopted for determining the % of risk-prone samples in any given sub-district: either it can be taken as the same as that of % of samples with fluoride >1.5 $mg.L^{-1}$ (e.g., Podgorski et al. 2018) or equal to the % of samples having HQ>1 (e.g., Mandal et al. 2021). Our calculations demonstrate that the latter approach provides a lower-bound estimate of the adult population at risk. An independent substantiation of the latter approach is made by calculating the total no of risk-prone villages in the Patan district and further matching it with reported data. We estimated the number of risk-prone villages in a sub-district by multiplying the % of samples with HQ_{adult} >1 in the sub-district by the total number of villages in it. Such exercise shows that there are 103 risk-prone villages in the Patan district- similar to/slightly lower than the 126 reported by Central Groundwater Board (CGWB 2014). Such could not be verified for the Banaskantha district due to a lack of available information on the number of risk-prone villages in it.

Estimates of adult and child populations at risk in each sub-districts are presented in Table 2. Calculations show that an estimated ~0.92 million of the adult population and ~0.39 million of the child population in the study area are potentially at risk due to the consumption of groundwater fluoride. With a projected increase of ~15% in population during 2011-2020 (http://www.population.u.com/in/gujarat-population), currently, these values would translate to ~1.06 million adults and ~0.45 million children respectively. These values respectively are ~20% of the adult population and ~55% of the children population of the region. Our current estimate of ~1.51 million reveals that ~30% of the total (adult +children) population in the study area is potentially at risk.

An assessment of the potential risk severity was made based on Dissanayke's classification (1991). Results show that while 57% of the collected samples are conducive to tooth development and prevent decay, the probabilistic occurrence of dental fluorosis and dental-and-skeletal fluorosis is associated with ~37% and ~6% of the samples. With no samples having fluoride concentration >10 $mg.L^{-1}$ the chances of crippling fluorosis are almost negligible. Furthermore, field photographs of children suffering from dental fluorosis (Fig. 2a and 2b) from the Danta region (within the Banaskantha district) are a testament to a real depiction of the fluoride problem in the study area.

Geostatistical Modeling

Geostatistical HQ maps are depicted in Figs. 3(a-d). The validity, accuracy, and reliability of the geospatial modeling are assessed by parameters RMSS, MSE, RMSP, and ASE. In our case, the RMSS values obtained are 0.99 (Fig. 3a), 0.98 (Fig. 3b), 1.01 (Fig. 3c), and 1.02 (Fig. 3d); all values close to 1 are indicative of the significant accuracy in prediction estimates (Mukherjee et al. 2019). Furthermore, corresponding MSE values for these Figs. being close to 0 (0.02, 0.01, 0.02, 0.03) indicate valid predictions. Finally, the close values of RMSP and ASE with an average RMSP/ASE ratio of 1.00 ± 0.02 signify the validity of the model output.

HQ_{adult} mapping is shown for ranges (0.0 to 3.0) for Banaskantha (Fig. 3a) and (0.0 to 4.5) for Patan (Fig. 3c). It appears from Fig. 3a that adults are prone to the fluoride risk in Danta and eastern margins of the Amirgadh sub-districts (within Banaskantha region), and in parts of Sidhpur and Patan sub-districts (within Patan region; Fig. 3c). A significant yet concerning observation made from the comparisons of Fig. 3 (a, c) versus Fig. 3 (b, d) is that the children are more prone to risk compared to adults in both the sub-districts. In the Banaskantha region, very high $HQ_{children}$ values (>2.5 to 6) were observed in the Danta sub-district (Fig. 3b) while others such as Dhanera, Dantiwada, Amirgadh, Palanpur, and

Table 2: Sub-district-wise statistical data of HQ_{oral}, and the adult and children population at risk in Banaskantha and Patan districts.

	Sr.No	Sub-district	Total samples	% of samples in F>1.5	% of samples in HQ>1	No of vil-lages in the sub-district		Population (Adult/ Children)	Population at risk based on HQ
Banaskantha	1	Danta	15	86	60	184	A	224839	134903
					100		C	46737	46737
	2	Palanpur	4	50	25	116	A	438773	109693
					75		C	56326	42244
	3	Vav	8	12.5	12.5	121	A	246156	30769
					25		C	41637	10409
	4	Tharad	6	16.6	0	134	A	327289	0
					50		C	56373	28186
	5	Dantiwada	3	100	33.3	57	A	115221	38407
					100		C	19051	19051
	6	Dhanera	7	71	42.9	77	A	230741	98889
					85.7		C	40759	34936
	7	Deesa	12	33	8.3	148	A	588123	49010
					75		C	94341	70755
	8	Deodhar	4	0	0	70	A	177919	0
					25		C	29161	7290
	9	Vadgam	5	40	40	109	A	240326	96130
					60		C	32382	19429
	10	Amirgadh	4	25	25	69	A	132354	33088
					25		C	28030	7007
	11	Kankrej	2	0	0	101	A	275613	0
					50		C	44364	22182
	12	Bhabhar	2	0	0	51	A	123152	0
					0		C	21149	0
Patan	13	Sidhpur	16	10	37.5	53	A	213087	79908
					68.8		C	27375	18820
	14	Patan	14	7	42.9	139	A	449480	192634
					57.1		C	58359	33348
	15	Harij	6	2	33.3	39	A	94562	31521
					33.3		C	13127	4376
	16	Santalpur	3	0	0	73	A	128791	0
					0		C	22074	0
	17	Radhanpur	3	1	0	56	A	144266	0
					66.7		C	22416	14944
	18	Sami	7	0	0	98	A	182805	0
					0		C	26560	0
	19	Chanasma	11	5	18.2	59	A	130743	23771
					54.5		C	14868	8110

Fig. 2a: Field photograph of a child suffering from dental fluorosis. The child is a resident of the Danta sub-district in the Banaskantha district.

Fig. 2b: Field photograph of another child (resident of Danta) suffering from dental fluorosis.

Vadgam are all associated with high fluoride risk, $HQ_{children}$ values > 1.5.

CONCLUSIONS

Fluoride contamination was investigated in groundwater from Banaskantha and Patan district, Gujarat, Western India. The five million residents of the studied region are vulnerable to fluoride exposure as they are compelled to consume untreated groundwater due to the limited availability of surface water resources in the region, and their sparse access to water treatment facilities.

Health risks were assessed through the ingestion exposure route only while dermal contribution was found to be very insignificant. HQ_{adult} ranges from 0.06-4.11 (average:

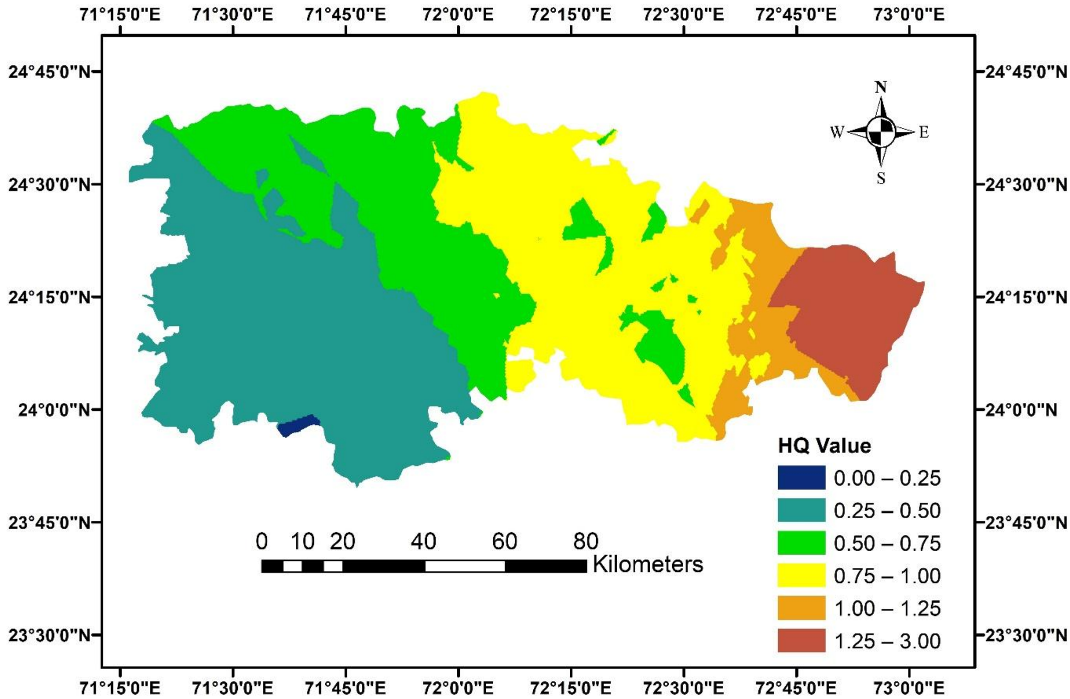

Fig. 3a: Health risk assessment (HQ_{oral}) for adults in the Banaskantha district. The map was produced using Empirical Bayesian Kriging (EBK) model.

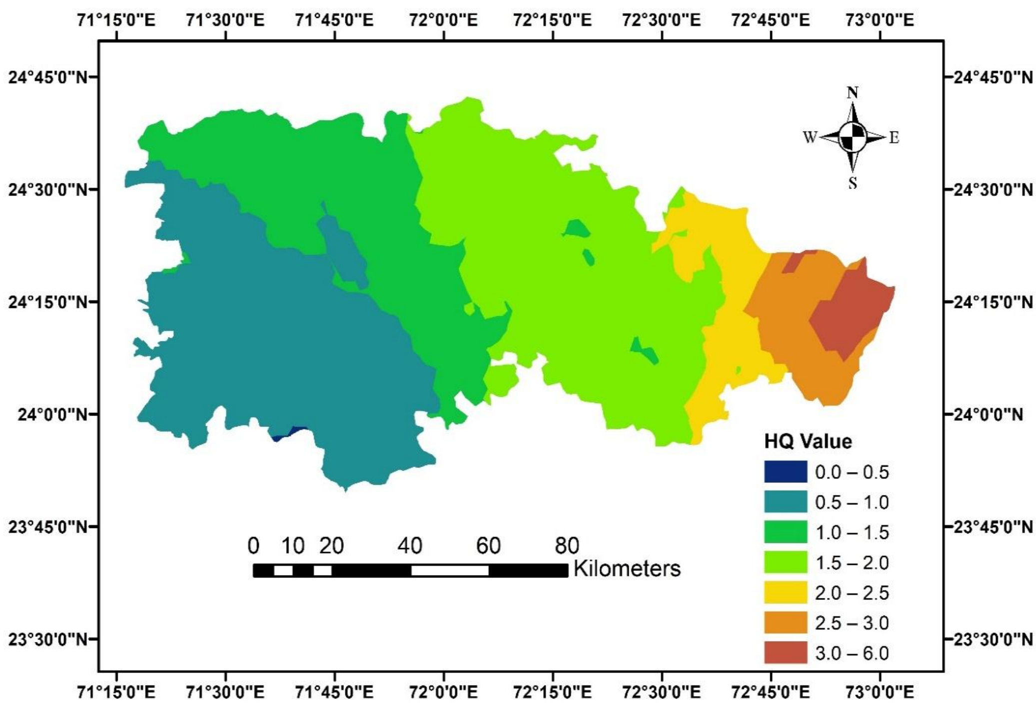

Fig. 3b: Health risk assessment (HQ_{oral}) for children in the Banaskantha district. (Map produced by EBK model).

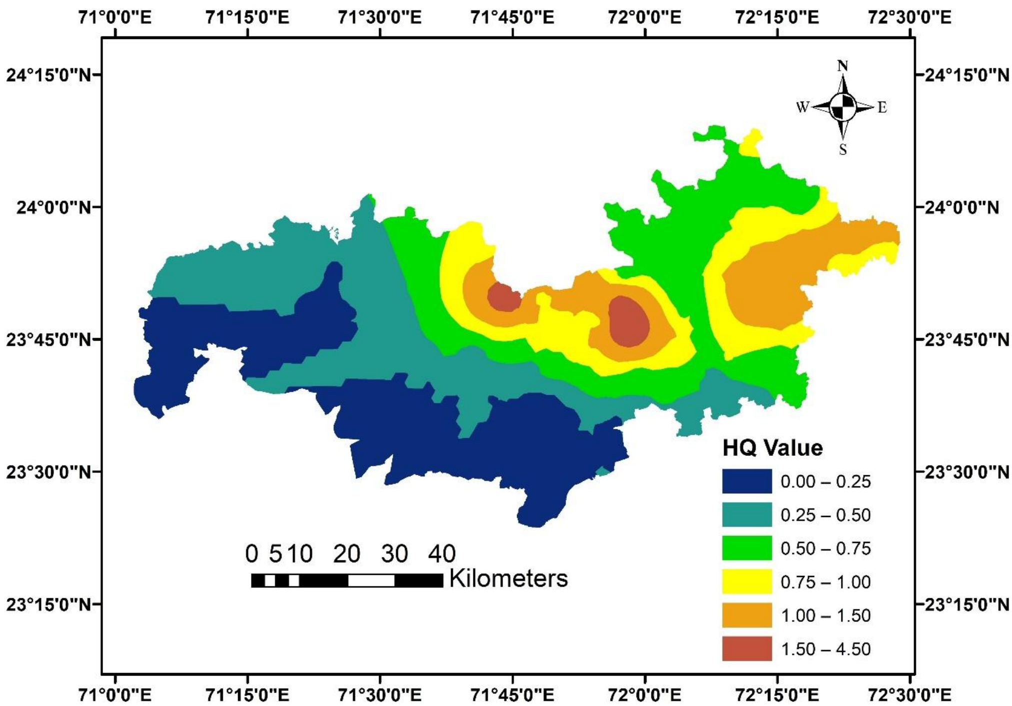

Fig. 3c: Health risk assessment (HQ_{oral}) for adults in the Patan district. (Map produced by EBK model).

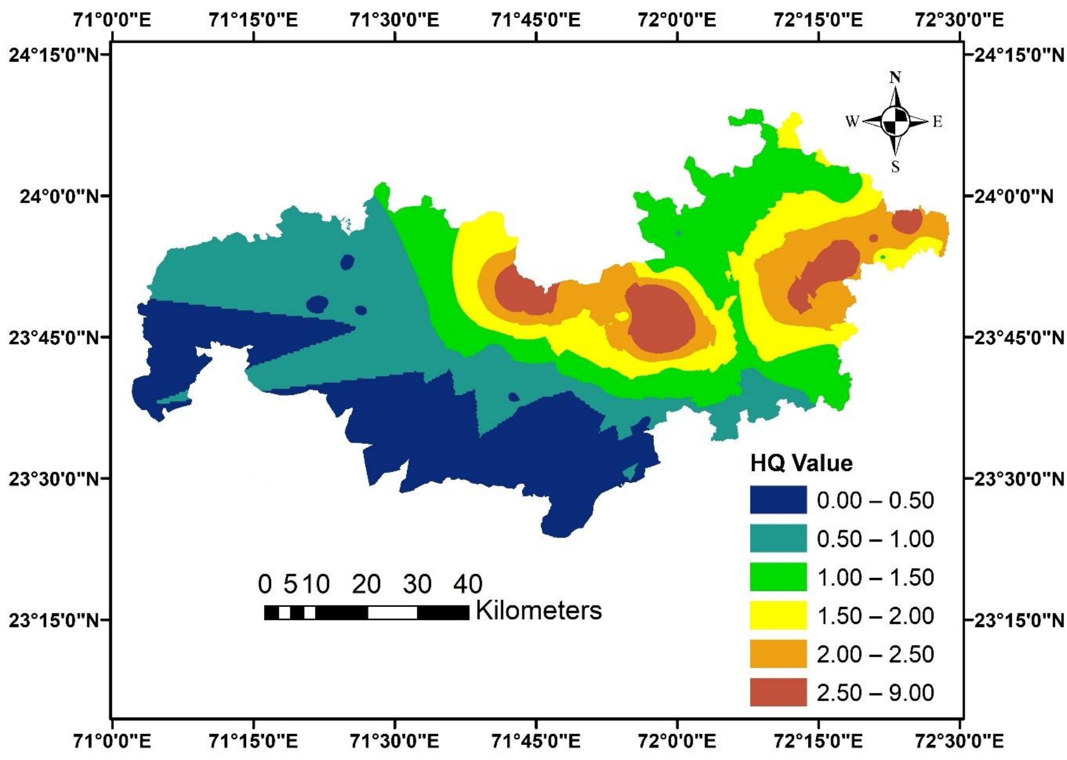

Fig.3d: Health risk assessment (HQ_{oral}) for children in the Patan district. (Map produced by EBK model)

0.78 ± 0.72; median: 0.60) and values >1 were found in 28% of samples. In comparison, the range (0.1-8.55), average (1.63 ± 1.49), median (1.26), and 58% samples having $HQ_{children}$ >1 all highlight that children are much more susceptible to fluoride risk compared to adults. Our conservative estimates suggest that ~20% of the adult population and ~55% of the child population of the region are potentially at risk. Photographs taken of children suffering from dental fluorosis are a testament to the real depiction of the hazard.

HQ maps of the endemic regions were produced by the Empirical Bayesian Kriging model. This mapping would help policymakers in channelizing mitigation funding and resources, and in increasing testing efforts in high-risk areas in Danta, Dantiwada, Dhanera, Vadgam, Sidhpur, and Patan sub-districts. Installation of fluoride treatment plants is suggested in these areas while rainwater harvesting is also suggested as a viable alternative. We hope that this study can help policymakers to adopt strategies to manage the regional groundwater resources better, in ensuring public health safety of the local population of the region, children in particular, from fluoride-related morbidities. We also believe that this article should stimulate further research concentrating on rural regions of India and other such countries, to estimate accurately the population impacted by fluoride contamination.

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REFERENCES

Adimalla, N., Li, P. and Qian, H. 2018. Evaluation of groundwater contamination for fluoride and nitrate in a semi-arid region of Nirmal Province, South India: A special emphasis on human health risk assessment (HHRA). Human Ecol. Risk Assess. Int. J., 14: 579 https://doi.org/10.1080/10807039.2018.1460579

Ahada, C.P and Suthar, S. 2019. Assessment of human health risk associated with high groundwater fluoride intake in southern districts of Punjab, India. Expos. Health, 11(4): 267-275.

Ali, S., Thakur, S.K., Sarkar, A. and Shekhar, S. 2016. Worldwide contamination of water by fluoride. Environ. Chem. Lett., 14(3): 291-315.

Biglari, H., Chavoshani, A., Javan, N. and Hossein Mahvi, A. 2016. Geochemical study of groundwater conditions with special emphasis on fluoride concentration, Iran. Desalin. Water Treat., 57(47): 22392-22399.

Cao, J., Chen, Y., Chen, J., Yan, H., Li, M. and Wang, J. 2016. Fluoride exposure changed the structure and the expressions of Y chromosome-related genes in the testes of mice. Chemosphere, 161: 292-299.

CGWB Report 2014. Aquifer Mapping and Management of Groundwater Resources Banaskantha District, Gujarat. Central Ground Water Board Department of Water Resources, River Development and Ganga Rejuvenation, Ministry of Jal Shakti Government of India.

CGWB. 2011. District Groundwater Brochure, Banaskantha District, Gujarat. Government of India, Ministry of Water Resources, Central Ground Water Board, West Central Region, Ahmedabad.

CGWB. 2014. District Groundwater Brochure, Patan District, Gujarat. Government of India, Ministry of Water Resources, Central Ground Water Board, West Central Region, Ahmedabad.

Datta, P.S., Deb, D.L. and Tyagi, S.K. 1996. Stable isotope (18O) investigations on the processes controlling fluoride contamination of groundwater. J. Contam. Hydrol., 24(1): 85-96. DOI: 10.1016/0169-7722(96)00004-6

Dey, S. and Giri, B. 2016. Fluoride fact on human health and health problems: A review. Med Clin Rev., 2(1): 11.

Dissanayake, C.B. 1991. The fluoride problem in the groundwater of Sri Lanka: Environmental management and health. Int. J. Environ. Stud., 38(2-3): 137-155.

Emenike, C.P., Tenebe, I.T and Jarvis, P. 2018. Fluoride contamination in groundwater sources in Southwestern Nigeria: Assessment using multivariate statistical approach and human health risk. Ecotoxicol. Environ. Saf., 156: 391-402.

Follin-Arbelet, B. and Moum, B. 2016. Fluoride: A risk factor for inflammatory bowel disease? Scand. J. Gastroenterol. 51: 1019e1024. https://doi.org/10.1080/ 00365521.2016.1177855.

Handa, B.K. 1975. Geochemistry and genesis of fluoride-containing ground waters in India. Groundwater, 13(3): 275-281.

Hem, J.D. 1985. Study and interpretation of the chemical characteristics of natural water (Vol. 2254). Department of the Interior, US Geological Survey, Virginia, US.

ICMR-NIN. 2020. Nutrient requirements for Indians recommended dietary allowances and estimated average requirements.

Jha, S.K., Mishra, V.K., Sharma, D.K. and Damodaran, T. 2011. Fluoride in the environment and its metabolism in humans. In: Whitacre, D.M. (ed.), Environmental Contamination and Toxicology, Springer, New York, NY, pp. 121e142. https://doi.org/10.1007/978-1-4419-8011-3_4

Jiang, P., Li, G., Zhou, X., Wang, C., Qiao, Y., Liao, D. and Shi, D. 2019. Chronic fluoride exposure induces neuronal apoptosis and impairs neurogenesis and synaptic plasticity: Role of GSK-3β/β-catenin pathway. Chemosphere, 214: 430-435.

Kravchenko, J., Rango, T., Akushevich, I., Atlaw, B., McCornick, P.G., Merola, R.B., Paul, C., Weinthal, E., Harrison, C., Vengosh, A. and Jeuland, M. 2014. The effect of non-fluoride factors on risk of dental fluorosis: evidence from rural populations of the Main Ethiopian Rift. Sci. Total Environ., 87: 595-606. https:// doi.org/10.1016/j.scitotenv.2013.12.087

Krivoruchko, K. 2012. Empirical Bayesian kriging. ArcUser Fall, 6(10): 71.

Kundu, M.C. and Mandal, B. 2009. Assessment of potential hazards of fluoride contamination in drinking groundwater of an intensively cultivated district in West Bengal, India. Environ. Monit. Assess., 152(1-4): 97. DOI: 10.1007/s10661-008-0299-1

Mandal, R., Das, A., Sudheer, A.K., Kumar, S., Verma, S., Gaddam, M. and Deshpande, R.D. 2021. Sources, controls, and probabilistic health risk assessment of fluoride contamination in groundwater from a semi-arid region in Gujarat, Western India: An isotope–hydrogeochemical perspective. Environ. Geochem. Health, 11: 1-17.

Mukherjee, I., Singh, U.K. and Patra, P.K. 2019. Exploring a multi-exposure-pathway approach to assess human health risks associated with groundwater fluoride exposure in the semi-arid region of eastern India. Chemosphere, 233: 164-173.

Ozsvath, D.L. 2009. Fluoride and environmental health: a review. Rev. Environ. Sci. Biotechnol., 8(1): 59-79.

Podgorski, J.E., Labhasetwar, P., Saha, D. and Berg, M. 2018. Prediction modeling and mapping of groundwater fluoride contamination throughout India. Environ. Sci. Technol., 52(17): 9889-9898.

Shirke, K.D., Kadam, A.K. and Pawar, N.J. 2020. Temporal variations in hydro-geochemistry and potential health risk assessment of groundwater from lithological diversity of the semi-arid region, Western Gujarat, India. Appl. Water Sci., 10(6): 1-20.

USEPA. 1989. Risk Assessment Guidance for Superfund Volume I: Human

Health Evaluation Manual (Part A). U.S. Environmental Protection Agency, Washington, DC.
Vithanage, M. and Bhattacharya, P. 2015. Fluoride in the environment: sources, distribution, and defluoridation. Environ. Chem. Lett., 13(2): 131-147.
Yang, C.Y., Cheng, M.F., Tsai, S.S. and Hung, C.F. 2000. Fluoride in drinking water and cancer mortality in Taiwan. Environ. Res., 82: 189e193. https://doi.org/10.1006/ enrs.1999.4018.
Zhang, L., Zhao, L., Zeng, Q., Fu, G., Feng, B., Lin, X., Liu Z., Wang, Y. and Hou, C. 2020. Spatial distribution of fluoride in drinking water and health risk assessment of children in typical fluorosis areas in north China. Chemosphere, 239: 124811.