

Health risk assessment of high concentration of fluoride and nitrate in the groundwater – A study of central India

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Abstract

This study analyses spatio-temporal variation in hydrochemical parameters of groundwater with special reference to Fluoride (F) and Nitrate (NO₃) contamination and prepares a village-level health risk map for contaminated parts of the Kelapur and Maregaon tehsils, Yavatmal District, Maharashtra, India. The study found prolonged rock-water interaction resulting in high F contamination responsible for rising instances of acute dental and skeletal fluorosis in the region. Weathering and leaching of fluoride rich rocks were found to be the primary reason behind such problems. Variation in farming practices, moderate rainfall, excessive use of fertilizers for growing multiple crops in a year, domestic animal wastes, and open sewage systems have resulted in high NO₃ contamination increasing the risk of methemoglobinemia among the infants. The health risk map for F shows that very high F values during pre-monsoon season have been recorded in Kumbha and Kopamandavi villages, possibly responsible for skeletal fluorosis. In contrast, high fluoride contamination recorded in some of the villages of Kelapur and Maregaon is probably responsible for dental fluorosis. Higher than permissible level of NO₃ is a serious concern in both the seasons particularly in the Kelapur tehsil. The present study concludes that the presence of higher level of F and NO₃ in groundwaters are due to natural processes and anthropogenic activities respectively in the tribal region of central India.

Keywords: Fluoride, Nitrate, groundwater, Dental Fluorosis, Skeletal Fluorosis, Methemoglobinemia, Yavatmal.

Introduction

Groundwater is a valuable resource that mankind relies on it for a variety of uses. However, fluctuations in the level of ionic concentration such as fluoride (F) and nitrate (NO₃) in the groundwater render it unsuitable for human use due to its adverse health impacts, a problem that has been reported in many parts of the world. The situation is far more serious in areas inhabited by tribal people who are generally perceived to be socially,

economically and educationally backward. The present study therefore considers the problem of ground water pollution in a largely tribal inhabited area where the inhabitants are vulnerable to F and NO₃ pollution. This research aims at identifying sources of the F and NO₃ and its impact on human health by conducting a risk assessment analysis of Kelapur and Maregaon tehsils (sub-district) of Yavatmal district in Maharashtra.

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Contamination of groundwater resulting from both geogenic and anthropogenic causes have resulted in a series of problems in many regions of the world and in India too (Reyes-Toscano et al., 2020; Reddy and Sunitha, 2020; Haji et al., 2021; Raja and Neelakantan, 2021). Fluoride and nitrate are the two ions, if consumed above the permissible limit, cause serious health problems in humans (Raja and Neelakantan, 2021). The fluoride contamination in groundwater has adversely impacted the health of millions of people worldwide (Rao et al., 2016; World Health Organization, 2011; Keesari et al., 2021). Its intake within the permissible limit is beneficial, but excess consumption causes dental and skeletal fluorosis (Gaikwad et al., 2019; Marghade et al., 2020; Keesari et al., 2021). The natural sources of F include weathering F-rich minerals like apatite, fluorite, biotite, hornblende, and rainwater infiltration which increase its concentration in groundwater (Karunanidhi et al., 2019; Rao et al., 2016). Agricultural fertilizers and coal combustion are the anthropogenic sources of Fluoride. (Sharma et al., 2016).

Nonpoint sources such as leaching of chemical fertilizers, animal manure, and sewage discharges are sources of nitrate in groundwater (Karunanidhi et al., 2020; Gaikwad et al., 2020a; Gaikwad et al., 2020b). The most prevalent contaminant of groundwater is dissolved nitrogen in the form of NO_3 .

As per the Bureau of Indian Standards (2003) and World Health Organization (2011), the permissible limit for F and NO_3 in groundwater are 1.5 mg/L and 45 mg/L respectively. Above this limit, ingestion of F and NO_3 will adversely affect human health. Lithologies of Archean, Vindhyan,

Gondwana and Deccan trap basalt dominate the area under investigation (Central Ground Water Board, 2013; Madhnure et al. (2007); Madhnure et al., 2016). Madhnure and Malpe (2007) reported the highest F concentration 16.6 mg/L of groundwater in rural areas of the Yavatmal district. Madhnure et al. (2007); Madhnure and Malpe (2007) and Madhnure et al. (2016) have identified the sources of F in groundwaters of the Yavatmal district of Maharashtra. However, health risk assessment is rarely addressed in most of these studies. The study area is largely inhabited by tribal communities who are vulnerable to F and NO_3 pollution.

Study area

The study area comprises tribal-dominated Kelapur and Maregaon tehsils in the northeastern parts of Yavatmal district, Maharashtra, India (Figure 1). The total area of the Kelapur is 835 km² with 141 villages, whereas Maregaon is spread over 607 km² containing 115 villages. The area lies between 19°26' and 20°42' north latitudes and 77°18' and 79°9' east longitudes. The relief of the area is characterised by a complex interplay of hilly, rugged terrain, broad valleys, and plains of eastern parts of the Deccan plateau. Cotton (*Gossypium hirsutum*), pigeon pea (*Cajanus cajan*), soybean (*Glycine max*), and sorghum (Sorghum bicolor) are the main crops grown during the monsoon season. While Wheat (*Triticum aestivum*) and chickpea (*Cicer arietinum*) are frequently farmed during the winter season (Jangir et al., 2019). The water divide between the tehsils is responsible for groundwater fluctuation in the study area. Purna is the main river, and other important rivers are Man, Murna, Kate, etc., flowing through the district.

Deccan flood Basalt is the most

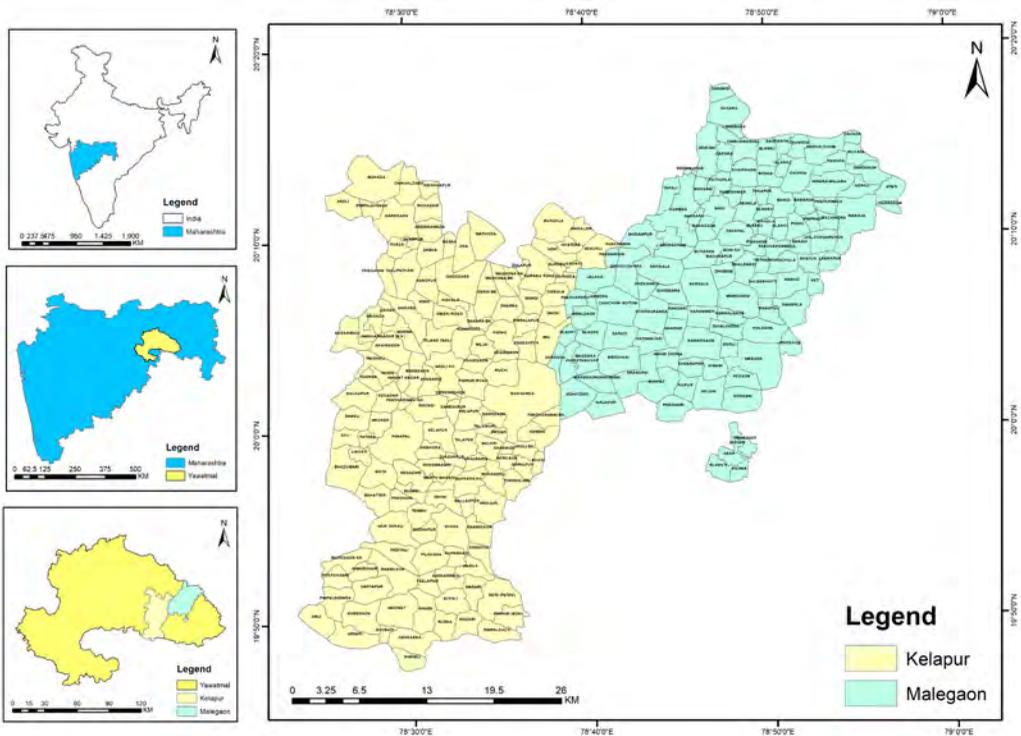


Fig. 1: Location of the study area (Kelapur and Malegaon)

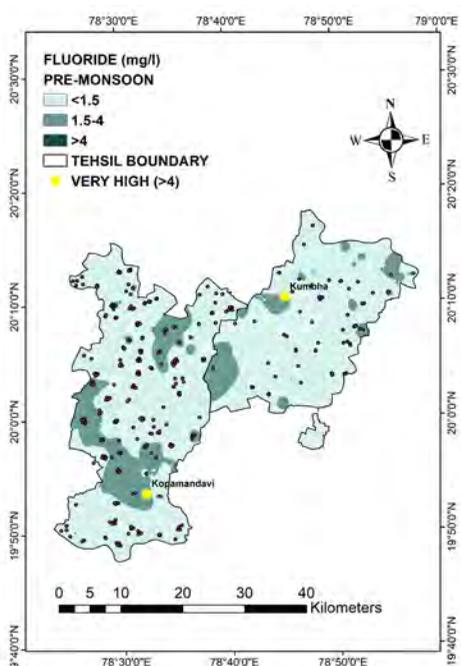


Fig. 2: Spatial distribution of Fluoride (PRMS)

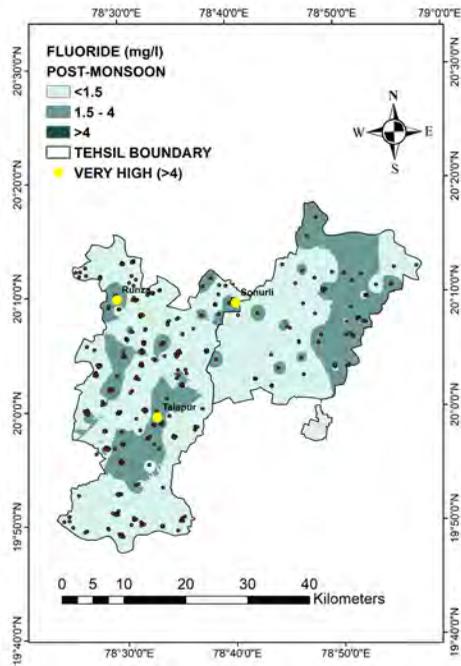


Fig. 3: Spatial distribution of Fluoride (POMS)

Table 1: Physio-chemical data of significant elements in groundwater (in mg/L except EC in $\mu\text{S}/\text{cm}$ and pH)

Parameters	Pre-monsoon (PRMS)			Post-monsoon (POMS)		
	Min	Max	Mean	Min	Max	Mean
pH	6.49	9.71	7.62	5.7	8.7	7.32
EC	182	5360	1173.47	101	3260	1036.06
TDS	183	2680	631.54	77	1630	553.06
TH	31	1660	397.37	48	1036	390.01
Cl	23	1128	171.67	16	636	140.39
F	0.008	4.73	1.09	0.1	4.4	1.25
NO ₃	0.19	234	87.51	0	215	78.89
SO ₄	0.24	560	89.27	3	324	52.97

common water-bearing formation, which occupies almost the entire area, followed by the Gondwana formation, which includes sandstone and shale sequences (Madhnure and Malpe, 2007; Central Ground Water Board, 2013). The Gondwana formation is made up of Kamthi and Barakar sandstone, with shale occupying the north-south direction in areas of Maregaon and Wani tehsils. Patches of alluvium can be found along the banks of the Wardha and Penganga rivers, as well as their major tributaries. It comprises clay and silt with lenticular sand and gravel bodies. (Madhnure and Malpe, 2007; Central Ground Water Board, 2013).

Yavatmal's climate is characterized by hot summers and general dryness throughout the year; the south-west and retreating monsoon seasons are an exception. During May, the mean daily maximum temperature reaches 41.8°C, whereas the mean daily minimum temperature is as low as 15.1°C during December. The annual rainfall is 850 to 1150 mm increasing from northwest to southeast. The mean annual rainfall is 1052 mm, with 56 average rainy days during the southwest monsoon. The population of the

tehsils mainly consists of Gond and Kolam tribes suffering from acute poverty as most of them counted below the poverty line (Madhnure and Malpe, 2007).

Database and methodology

Maregaon and Kelapur tehsils were chosen for the study due to comparatively high F and NO₃ contaminated sites in Yavatmal district. The present study uses hydrochemical data of PRMS (Pre-monsoon season) and POMS (Post-monsoon season) for the year 2018. The district's highest rainfall, 1109.5 mm, was recorded in 2018 (higher than average 780 mm) (IMD data for 2010 to 2019) even though the high F and NO₃ contamination were recorded in 2018 for both the seasons and in these two tehsils. Large concentration of the poverty ridden tribal communities in these tehsils only adds to the vulnerability of these groups to health problems.

The study area is covered in the Survey of India toposheet nos. 55I/5, 55I/9, 55L/8, 55L/12, 55L/15, 55L/16 and 56I/13. Geological map of the Yavatmal district was acquired from the Geological Survey of India. The hydrochemistry data used in the

present study is obtained from Groundwater Surveys and Development Agency (2018) under Hydrological Data User's Group membership, especially for pre-monsoon season and post-monsoon season. The present study is obtained Bureau of Indian Standards (2003) and World Health Organization (2011) drinking water standards for health risk assessments. The data was disaggregated for the villages based on the permissible limit as recommended by Bureau of Indian Standards (2003) and World Health Organization (2011). Village level F and NO₃ health risk maps for PRMS and POMS were prepared using the interpolation method in the ArcGIS (ver.10.1) software.

Results

The present study used significant elements of the physio-chemical data of groundwater (Table 1).

Spatio-temporal distribution of Fluoride in groundwater

In the study area, F concentration in groundwater ranges between 0.008 and 4.73 mg/L in PRMS and from 0.1 to 4.4 mg/L in POMS (Table 1).

In the PRMS, excessive F concentration is observed in the Kumbha village of Maregaon tehsil (4.73 mg/L), and in Kopamandavi village of Kelapur tehsil (4.02 mg/L) represented by yellow dots in figure 2, which can cause skeletal fluorosis. In contrast, the northern, eastern, and western parts of Kelapur tehsil and a few patches of Maregaon tehsil fall under high fluoride concentration (1.5 to 4 mg/L), which causes dental Fluorosis (Fig. 2).

In the POMS (Fig. 3), the maximum

concentration of F is reported the number in the Runza, Sonurli, and Talapur villages of Kelapur tehsil with 4.00 mg/L, 4.02 mg/L, and 4.37 mg/L concentration respectively. These villages have reported the number of cases of dental and skeletal fluorosis. Some patches in Kelapur and Maregaon tehsils show a high fluoride concentration of 1.5 to 4 mg/L, resulting in dental fluorosis (Figure 3).

Earlier studies have shown that rock and mineral weathering result in high F in the groundwater (Madhnure and Malpe, 2007; Wagh et al., 2016; Gaikwad et al., 2019; Gaikwad et al., 2020a; Raja and Neelakantan, 2021). The lowest concentration of F is observed at the Padha village in Kelapur tehsil (0.008 mg/L) and in Khandani village of Maregaon tehsil (0.1 mg/L).

Spatio-temporal distribution of Nitrate in groundwater

The NO₃ concentration in groundwater ranges from 0.19 to 234 mg/L in PRMS and from 0 to 215 mg/L in POMS (Table 1). More than 90 percent of the Maregaon tehsil shows NO₃ concentration below the permissible limit <45 mg/L in PRMS and POMS (Fig. 4 and 5).

In Kelapur tehsil, NO₃ concentration >100 mg/L is observed in some samples where the cases of Methemoglobinemia are reported. High NO₃ is due to fertilizer use in agriculture, sewage water used for irrigation, humans, and livestock waste (Pawar et al., 2008; Gaikwad et al., 2020c; Rahman et al., 2021).

In PRMS, the highest concentration of NO₃ of 234 mg/L is reported at Wagholi, while the lowest of 0.19 mg/L is observed at Gondwakadi of Kelapur tehsil. High NO₃ (45

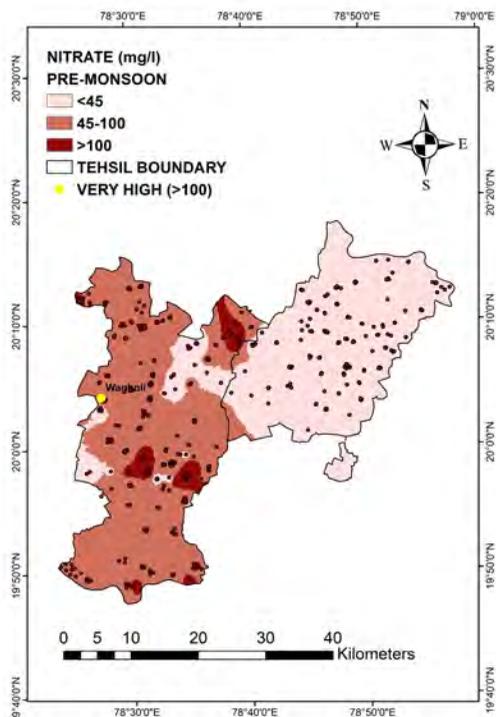


Fig. 4: Spatial distribution of Nitrate (PRMS)

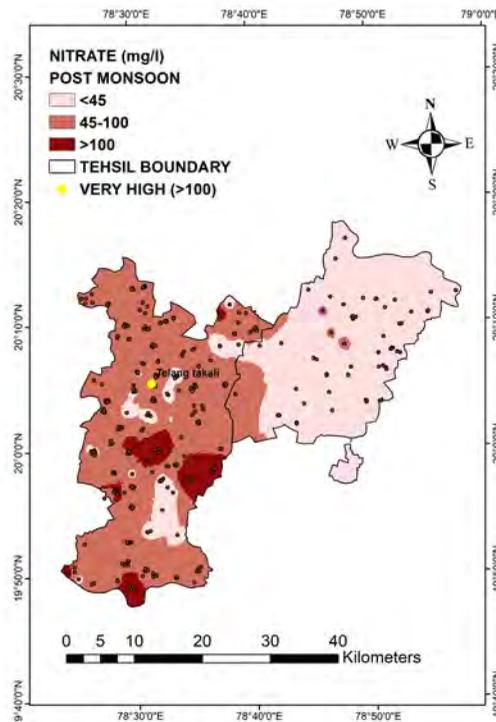


Fig. 5: Spatial distribution of Nitrate (POMS)

to 100 mg/L) was found in most parts over the Kelapur and a few patches of Maregaon (Fig. 4).

In POMS, the highest concentration (215 mg/l) of NO_3 is reported at Telang Takali, Kelapur. Some villages of the central eastern and southern parts of Kelapur tehsil have high NO_3 contamination (>100 mg/L) (Figure 5) whereas Chopan village has the lowest concentration of NO_3 .

Correlation analysis

The Pearson's correlation coefficients are presented in Table 2 and 3 for PRMS and POMS respectively. In an acidic medium, fluoride is absorbed by clays; however, it is desorbed in an alkaline medium, hence an alkaline pH is more favourable for fluoride dissolving activity

(Gaikwad et al., 2019). During PRMS, the average pH is alkaline (7.61), while in POMS, the average pH is almost acidic, suggesting the highest fluoride mobility in weakly alkaline conditions (Saxena and Ahmed, 2003; Sharma et al., 2016). The correlation between pH and F reflects a low positive correlation ($r = 0.215$ and 0.178) for PRMS and POMS respectively. Similar observations were reported by Brindha and Elango (2011). Fluoride shows a strong negative correlation with Nitrate, ($r = -0.253$) and ($r = -0.250$) in PRMS and POMS respectively. Similar results were reported by Sakram et al. (2019) and Sharma et al. (2016). It implies that fluoride sources are mostly geogenic, enriching the groundwater (Madhnure and Malpe, 2007; Gaikwad, 2012a; Central Ground Water Board, 2013; Kumar

Table 2: Hydrochemical parameters correlation matrix for Pre-Monsoon Data (n=502)

Variables	pH	EC	TDS	TH	Cl	F	NO ₃	SO ₄
pH	1.0							
EC	-0.199*	1.0						
TDS	-0.200*	0.832*	1.0					
TH	-0.263*	0.679*	0.845*	1.0				
Cl	-0.112	0.787*	0.877*	0.670*	1.0			
F	0.215*	-0.092	-0.140	-0.234*	-0.064	1.0		
NO ₃	-0.183*	-0.201*	-0.089	0.019	-0.218*	-0.253*	1.0	
SO ₄	-0.020	0.422*	0.292*	0.208*	0.387*	-0.015	-0.502*	1.0

(*Correlation is significant at the 0.05 level; non-significant correlation is without sign*)

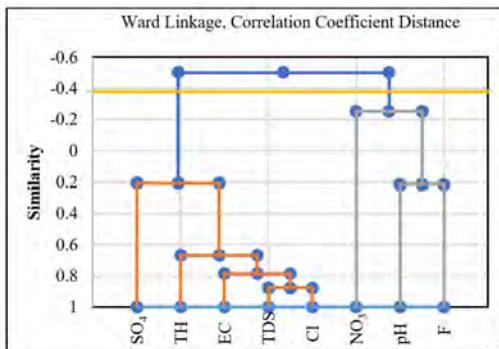


Fig. 6: Pre-monsoon (PRMS) clustering variables

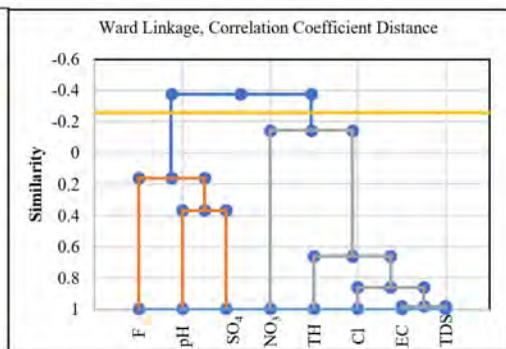


Fig. 7: Post-monsoon (POMS) clustering variables

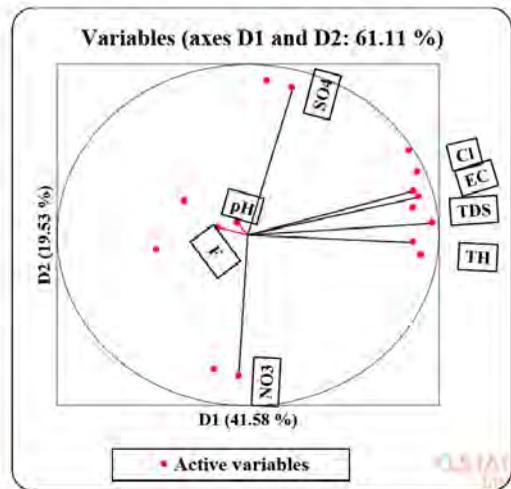
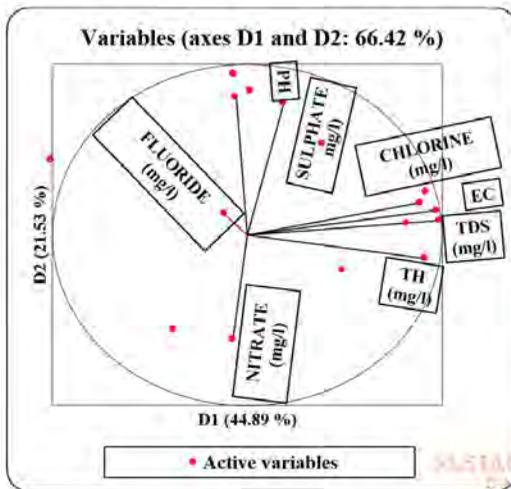


Fig. 8: Varimax rotated factor loadings and factor scores for the first two factors of PRMA and POMS, respectively.

Table 3: Hydrochemical parameters correlation matrix for Post-Monsoon Data (n=369)

Variables	pH	EC	TDS	TH	Cl	F	NO ₃	SO ₄
pH	1.0							
EC	0.075	1.0						
TDS	0.035	0.982*	1.0					
TH	-0.107	0.859*	0.897*	1.0				
Cl	0.107	0.861*	0.866*	0.658*	1.0			
F	0.178*	-0.154	-0.160	-0.227*	-0.013	1.0		
NO ₃	-0.306*	-0.116	-0.101	0.020	-0.143	-0.250*	1.0	
SO ₄	0.367*	0.279*	0.220*	0.016	0.301*	0.162	-0.377*	1.0

(*Correlation is significant at the 0.05 level; non-significant correlation is without sign*)

Table 4: PCA for pre-monsoon season

Post-monsoon	Factor 1	Factor 2	Factor 3
pH	-0.058	0.808	-0.048
EC	0.971	0.143	-0.071
TDS	0.987	0.084	-0.063
TH	0.911	-0.138	-0.123
Cl	0.887	0.188	0.076
F	-0.119	0.125	0.945
NO ₃	-0.075	-0.617	-0.377
SO ₄	0.185	0.770	0.087
Eigen value	3.679	1.875	0.833
Variability (%)	44.889	21.526	13.425
Cumulative %	44.889	66.415	79.840

et al., 2017). High F and NO₃ at a few places are due to weathering of F-rich minerals from rock and animal waste, wastewater disposal, use of nitrate fertilizers, etc. (World Health Organization, 2011; Adimalla et al., 2021).

Hierarchical cluster analysis (HCA)

The HCA can check similar groups of ions across various sampling sites (Yidana et al., 2012; Taoufik et al., 2017; Sakram et al., 2019). In the present study, Q-mode HCA is performed using XLSTAT 2021 software. Dendrograms obtained represent

the hierarchical relationship between objects and allocated objects to clusters. The HCA has been performed on the standardized variables using correlation coefficient and Ward's linkage method with two clusters. For PRMS and POMS samples, cluster analysis is used to assess the geochemical behaviour of groundwater parameters, particularly F and NO₃. The two colours on the dendrogram indicate the two clusters for PRMS and POMS (Fig. 6 and 7). The F comes under cluster 2 in PRMS (Fig. 6 and 7), but it is in cluster 1 during POMS, indicating a primary

Table 5: PCA for post-monsoon season

Pre-monsoon	Factor 1	Factor 2	Factor 3
pH	-0.150	0.042	0.716
EC	0.867	0.257	-0.095
TDS	0.968	0.068	-0.097
TH	0.866	-0.045	-0.233
Cl	0.897	0.221	0.008
F	-0.050	0.065	0.801
NO ₃	-0.049	-0.831	-0.304
SO ₄	0.236	0.863	-0.124
Eigen value	3.644	1.655	0.925
Variability (%)	41.578	19.531	16.687
Cumulative %	41.578	61.109	77.795

geogenic source with mainly carbonate weathering and the silicate weathering acting as the dominant factor. The rainwater may dilute the proportion of F during the POMS. The NO₃ in PRMS and POMS is present in cluster 2, suggesting anthropogenic inputs (Gaikwad et al., 2020b).

Principal Component Analysis (PCA)

As per PCA the first three factors explain 79.84 percent of the variation in PRMS and 77.80 percent of the variation in POMS (Table 4 & 5). The same variation is explained by the first three factors when varimax rotation is carried out (Fig. 8). In PRMS and POMS, the variables Electrical conductivity (EC), Total dissolved solids (TDS), Total Hardness (TH), and Chloride (Cl) have the greatest impact on the first factor, showing carbonate weathering and reverse ion exchange processes. (Sonkamble et al., 2012; Gaikwad et al., 2020b) (Table 4 & 5; Figure 8). Factor 2 is highly affected by variables like NO₃ and SO₄ having mixed anthropogenic sources such as detergents, human waste, and agricultural runoff in both PRMS and POMS (Table 4 &

5; Figure 8). The factor 3 in both the seasons is highly affected by the F variable mainly because it is controlled by the geogenic process.

Health risk assessment

The quantitative estimation of fluoride and nitrate reveals that a few water samples in the study area exceed the drinking-water standards for F and NO₃.

Fluoride (F)

Village-wise health risk map of F is prepared using the criteria adopted by Adimalla et al. (2018, 2021), which accepts <1.5 mg/L for low health risk, 1.5 to 4 mg/L for high health risk, and >4 mg/L for very high health risk (Table 6 and 7). Out of 502 samples in PRMS and 369 in POMS groundwater, <1.5 mg/L concentration of F was found in 76.49 percent (384 samples) and 68.02 percent (251 samples) respectively that are below the maximum acceptable limit. This means that about 70 to 80 percent of PRMS and POMS samples show that the water is potable at these locations posing little health risk to the

Table 6: Groundwater classification based on F concentration (PRMS)

F (mg/L)	Classification of health risks	Number of samples (n=502)	% of Samples
< 1.5	No risk	384	76.49
1.5 - 4	High risk	116	23.11
> 4	Very high risk	2	0.40

Table 7: Groundwater classification based on F concentration (POMS)

F (mg/L)	Classification of health risks	Number of samples (n=369)	% of Samples
< 1.5	No risk	251	68.02
1.5 - 4	High risk	118	31.98
> 4	Very high risk	0	0

humans. In contrast 23.51 percent samples (116) in PRMS and 31.98 percent (118) samples in POMS show health risks (High risk 23.11% and very high risk 0.40%) where the maximum permissible limit (1.5 to 4 mg/L) is exceeded which may lead to skeletal and dental fluorosis (Table 6 and 7; Figure 9). Weathering of rocks and minerals is the probable reason for the high F in groundwater of the study area. In POMS however none of the villages fell in the very high health risk category (Table 7).

The study establishes that F contamination is restricted to the areas having F-bearing minerals like amphiboles and biotite in host rock formations. In such rock formations, F mobilization in groundwater is facilitated by low rainfall, high temperature, and high alkalinity. In addition, excess of ions such as sodium, bicarbonate, and chloride give rise to such contamination (Madhnure and Malpe, 2007; Gaikwad and Pawar, 2012b; Gaikwad et al., 2020a).

Nitrate (NO₃)

With regard to NO₃, Adimalla et al. (2018, 2021) suggested <45 mg/L for low health risk, 46 to 100 mg/L denotes high health risk, and >100 mg/L stands for very high

health risk. PRMS and POMS groundwater samples in Table 8 and 9 show that 15.34 percent (77) and 29.27 percent (108) samples respectively have NO₃ below the maximum acceptable limit (<45 mg/L) meaning thereby that the groundwater is potable with little health risk to the people. While, most of the samples are contaminated and unsuitable for human consumption. Groundwater in as much as 84.66 percent (425) samples in PRMS (High risk 38.45% and very high risk 46.21%) and 70.73 percent (261) samples in POMS (High risk 35.77% and very high risk 34.96%) were found unsuitable for human consumption as far as NO₃ is concerned. It means that the wells of Kelapur tehsil are by and large unsuitable for drinking purposes and can lead to methemoglobinemia among infants if consumed (Table 8 and 9; Figure 9). The animal waste, open wastewater disposal facility, and higher use of nitrate fertilizers for growing cotton crops are the primary sources of high nitrate concentration in the groundwater of the study area.

Conclusion

The present study has revealed that both natural processes and anthropogenic activities pollute the groundwater of Kelapur and Maregaon,

Table 8: Groundwater classification based on NO₃ concentration (PRMS)

NO ₃ (mg/L)	Classification of health risks	Number of samples (n=502)	% of Samples
<45	No risk	77	15.34
46 - 100	High risk	193	38.45
>100	Very high risk	232	46.21

Table 9: Groundwater classification based on NO₃ concentration (POMS)

NO ₃ (mg/L)	Classification of health risks	Number of samples (n=369)	% of Samples
<45	No risk	108	29.27
46 - 100	High risk	132	35.77
>100	Very high risk	129	34.96

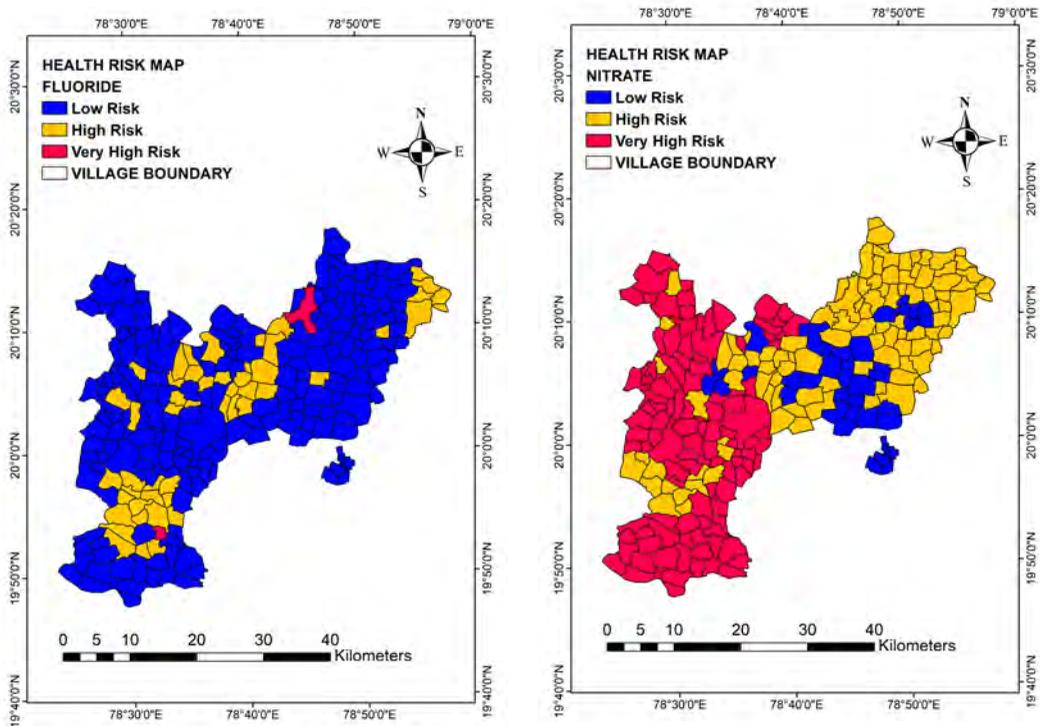


Fig. 9: Health risk based on F and NO₃ concentration

especially in the cases of F and NO₃. Fluoride with very high contamination is observed in two villages namely Kopamandavi in Kelapur and Kumbha in Maregaon during PRMS, responsible for dental and skeletal Fluorosis, whereas fluoride high contamination is experienced in 30 villages in Kelapur and 20

in Maregaon resulting into dental Fluorosis.

Very high to high nitrate contamination is observed in 125 and 38 villages respectively in Kelapur largely due to excessive use of fertilizers for the cotton crops, manures, domestic animal waste and open sewage

system. The farmers cultivate 2 to 3 crops in a year using the water of the Khuni River and the Saikheda dam. About 90 percent of the sewage is open sewage system. On the other hand, low nitrate contamination is observed in Maregaon due to less use of fertilizers and farmers practice only Kharif crops (monsoon season) and dryland farming.

In both the seasons, a negative correlation found between F and NO_3 indicates that the significant source of F in groundwater is from lithologies but cannot be from the fertilizers. High F and NO_3 at a few places are due to weathering of F-rich minerals from rock and animal waste, wastewater disposal use of nitrate fertilizers. The cluster analysis demonstrates that F in PRMS and POMS, indicating a major geogenic source dominated by carbonate weathering and silicate weathering. The presence of NO_3 in PRMS and POMS indicates that the studied area has a high level of NO_3 as a result of anthropogenic inputs. Principal Component Analysis shows that the first factor is mainly affected by Electrical conductivity, Total dissolved solids, Total Hardness, Chloride in POMS and PRMS, indicating carbonate weathering and reverse ion exchange processes. In both seasons, variables like NO_3 and SO_4 with anthropogenic sources such as detergents, human waste, and agricultural runoff have a significant impact on Factor 2. The factor 3 in both the seasons is highly affected by the F variable mainly because of the geogenic process controlling it. The maximum permissible limit of F (1.5 to 4 mg/L) observed in 23.51 percent cases during PRMS and 31.98 percent in POMS samples show a high health risk, particularly skeletal and dental fluorosis for the tribal people of the study area. This groundwater is undoubtedly

not suitable for drinking purposes and even for agriculture. The F contamination is restricted to these areas due to prolonged rock-water interaction of F-bearing minerals like amphiboles and biotite and the excess Sodium, Bicarbonate, and Chloride in host rock formations. The NO_3 of more than 45 mg/L proportion is observed in 84.66 percent PRMS and 70.73 percent POMS groundwater samples which show high health risks, specifically methemoglobinemia among the infants, particularly in the Kelapur than the Maregaon tehsil. Due to high NO_3 , such groundwater is not suitable for drinking and farming purposes. In the study area, variation in farming practices, moderate rainfall, excessive use of fertilizers for growing multiple crops in a year, domestic animal wastes, and open sewage systems have resulted in high nitrate concentration in the groundwater.

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