Fluoride contamination in groundwaters of Sonbhadra District, Uttar Pradesh, India

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Fluoride (F^-) concentration over and above the permissible limits (1.5 mg/l) in drinking water leads to human health hazards, such as dental and skeletal fluorosis affecting millions of people in many parts of India. Preliminary investigations indicate that severe health disorders have been identified in parts of the Kachnarwa region, which is in the upper Panda river basin, Sonbhadra District, Uttar Pradesh, due to excess intake of fluoride through drinking water. The lithological units of the study area mainly consist of granite and gneissic complex rocks of the Chota-Nagpur Plateau. In order to understand the probable source of fluoride and its concentration, 17 groundwater samples mostly from granite and phyllite regions were collected for fluoride estimation during May 2006. The concentration of fluoride in the groundwater of the study area varies from 0.483 to 6.7 mg/l. Among the 17 samples analysed, 8 exceeded the maximum permissible limits of fluoride in the study area. The source of fluoride in the groundwater of the study area is mainly from geological occurrence (i.e. fluoride-bearing minerals, viz. apatite and biotitic mica). Microscopic analysis of the rock samples showed 5–10% apatite and 20–25% biotite, but other fluoride-bearing minerals like fluorite and hornblende were characteristically absent. X-ray diffraction studies showed that apatite mineral peak profile of the rock samples corroborated with the fluorapatite of the standard set by the JCPDS. The worst fluoride-affected villages are Rohiniyadamar, Madhuri, Neruiyadamar, Gobardaha and Kunrwa. Most people in these villages suffer from dental and skeletal fluorosis such as mottling of teeth, deformation of ligaments, bending of spinal column and ageing problem. An urgent need is to educate the people on the causes of fluorosis, encouraging rainwater harvesting and providing fluoride-free drinking water in the study area.

Keywords: Dental and skeletal fluorosis, fluoride, granite gneissic rock, groundwater.

FLUORIDE (F^-) occurs in almost all waters from trace to high concentrations[1]. Fluoride concentration in natural waters depends on various factors such as temperature, pH, solubility of fluorine-bearing minerals, anion exchange capacity of aquifer materials (OH^- for F^-), and the nature of geological formations drained by water and contact time of water with a particular formation. Minerals which have the greatest effect on the hydrogeochemistry of fluoride are fluorite, apatite, mica, amphiboles, certain clays and villiamite. Fluoride is among the substances for which there are both lower (0.6 mg/l) and upper (1.2 mg/l) limits of concentration in drinking water, with identified health effect and benefits for human beings[2]. Very low doses of fluoride (<0.6 mg/l) in water promote tooth decay. However, when consumed in higher doses (>1.5 mg/l), it leads to dental fluorosis or mottled enamel and excessively high concentration (>3.0 mg/l) of fluoride may lead to skeletal fluorosis.

In general, fluoride content in water between 1.5 and 2.0 mg/l may lead to dental mottling, which is characterized initially by opaque white patches on the teeth and in advanced stages leads to dental fluorosis (teeth display brown to black staining) followed by pitting of teeth surfaces. High manifestations of dental fluorosis are mostly found in children up to the age of 12 years, and skeletal fluorosis[3] may occur when fluoride concentrations in drinking water exceed 4–8 mg/l. The high fluoride concentration manifests as an increase in bone density leading to thickness of long bones and calcification of ligaments. The symptoms include mild rheumatic/arthritic pain in the joints and muscles to severe pain in the cervical spine region along with stiffness and rigidity of the joints. The disease may be present in an individual at subclinical, chronic or acute levels of manifestation[4]. Crippling skeletal fluorosis can occur when the water supply contains more than 10 mg/l of fluoride[5,6]. The severity of fluorosis depends on the concentration of fluoride in the drinking water, daily intake, continuity and duration of exposure, and climatic conditions.

In India, an estimated 62 million people, including 6 million children suffer from fluorosis because of consuming fluoride-contaminated water. Although fluorosis was identified[7] as early as 1937, a programme for controlling the disease through networking between State Rural Drinking Water Supply Implementing Agencies and Health Departments was launched[8] during 1986–87. Generally, high fluoride contamination in hard-rock terrain is common due to water quality variation and changes in shallow and deep aquifers zones. But, in alluvial plain groundwater, the variation and changes in fluoride levels are usually rare. A number of cases of fluorosis have been reported mostly from the granite and gneissic complex of different states such as Andhra Pradesh[9–12], Assam[13], Bihar[14], Delhi[15], Gujarat[16], Karnataka[17,18], Kerala[19], Madhya Pradesh[20,21], Maharashtra[22], Orissa[23,24], Rajasthan[25,26] and Tamil Nadu[27,28] (Table 1).

In the Ganga alluvial plain of Uttar Pradesh (UP), fluoride content has been reported by various researchers, and State and Central Governments in the districts of Varanasi[29], Unnao[30], Kanpur, Agra[31] and Mathura[32] (Table 1).

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Table 1. Lithology and fluoride concentration in the groundwater in different states of India

Sample location	Geological formation	Fluoride (mg/l)	Reference
Guntur, Andhra Pradesh	Archean granite and gneissic complex	0.6–2.5	9
Anantapur, Andhra Pradesh	Archean granite and gneissic complex	0.56–5.8	10
Hyderabad, Andhra Pradesh	Archean granite and gneissic complex	0.38–4.0	11
Ranga Reddy, Andhra Pradesh	Archean granite and gneissic complex	0.4–4.8	12
Karbi Anglong, Assam	Cretaceous sandstone	0.4–20.6	13
Bihar	Archean gneissic complex	0.1–2.5	14
Delhi	Amphibolites and quartzites	0.2–32.5	15
Gujarat	Basalt and quaternary sediments	0.1–40	16
Bellary, Karnataka	Archean granite and gneissic complex	0.33–7.8	17
Karnataka	Archean granite and gneissic complex	1–7.4	18
Kerala	Archean granite and gneissic complex	0.2–5.75	19
Chandidongri, Madhya Pradesh	Archean granite and gneissic complex	1.5–4	20
Shivpuri, Madhya Pradesh	Archean granite and gneissic complex	0.2–6.4	21
Chandrapur Maharastra	Archean granite and gneissic complex	–	22
Orissa	Archean granite and gneissic complex	0.3–10.1	23
Orissa	Archean granite and gneissic complex	0.1–16.4	24
Churu, Rajasthan	Granites and micaceous sands	0.1–14	25
Dungarpur, Rajasthan	Archean granite and gneissic complex	0.1–10	26
Tamil Nadu	Gneissic complex	0.51–4.0	27
Tamil Nadu	Charnokite and gneissic complex	0.5–4.0	28
Varanasi, Uttar Pradesh	Quaternary–Upper Tertiary deposits	0.2–2.1	29
Unnao, Uttar Pradesh	Quaternary–Upper Tertiary deposits	–	30
Agra, Uttar Pradesh	Quaternary–Upper Tertiary deposits	0.1–17.5	31
Mathura, Uttar Pradesh	Quaternary–Upper Tertiary deposits	0.6–2.5	32
Sonbhadra District, Uttar Pradesh	Archean granite and gneissic complex	0.483–6.7	Present study

Fluoride contamination in many parts of UP (Unnao, 2 mg/l; Debraspur, 2.1 mg/l; Janghai, 3.2 mg/l; Kulpahar, 3 mg/l; Babera, 3.3 mg/l; Karchhana, 2.8 mg/l; Jhansi, 2.8 mg/l, and Etah, 3 mg/l) has been reported mainly in the Quarternary–Upper Tertiary deposits[33]. Ray *et al.*[29] reported 24.91% dental fluorosis, but no case of skeletal fluorosis was reported in Ledhupur and Rustampur villages near Varanasi. Maximum prevalence of dental fluorosis was found in the 11–15 years age group; prevalence was higher in males than females. Misra *et al.*[32] have reported fluoride contamination of 0.6–2.5 mg/l in Saidabad Tahsil, Mathura District in the Ganga alluvial plain. A survey conducted by the local human rights organization (NGO) on the health hazards of the human population found that the dental and skeletal disorders are due to the fluoride content in groundwater in parts of Chopan, Dudhi and Myorpur blocks of Sonbhadra District, UP[34]. The main aim of present study is to understand the source of fluoride concentration in groundwater and the severity of fluorosis problems in the drought-prone area of Kachnarwa panchayat, Sonbhadra District.

The study area located in the Chopan block (Figure 1), experiences semi-arid and arid climate, with an average annual minimum and maximum temperatures 10°C and 47°C respectively. Rainfall is the main source of groundwater recharge in the study area and the region receives an average annual rainfall of ~750 mm. Most of the agricultural activities mainly depend on rainfall, since the availability of groundwater resources is scarce. The major litho-units of the study area comprise of granitic and phyllites rocks, which are overlaid by red, sandy soil cover and the drainage is dendritic to sub-dendritic. Secondary intrusive such as dykes, pegmatite and quartz veins, which occur to a limited extent, are present in the rocks. Groundwater occurs under phreatic conditions in the weathered and fractured zones. The depth to water table ranges from 5.6 to 12.1 m below the ground surface. The depth of dug wells ranges from 8 to 13 m and the diameter of the dug wells ranges from 2 to 7 m. The maximum depth of the hand pump for drinking water use is 40 m below ground level.

Groundwater samples were collected from 17 dug wells and hand pumps (where there is no dug well) located in Kachnarwa region of Sonbhadra District during May 2006 (Figure 1). They were analysed for major ion chemistry using the standard methods[35]. Electrical conductivity and pH were measured using EC and pH meters. Total dissolved solids were estimated by ionic calculation method. Total alkalinity (TA), CO_3 and HCO_3 were estimated by titrating with HCl. Total hardness (TH) and Ca were analysed titrimetrically using standard EDTA. Mg was computed, taking the difference between TH and Ca values. Na and K were measured by flame photometer, Cl was estimated by standard $AgNO_3$ titration and SO_4 was measured by the turbidimetric method. Fluoride was estimated by using an ion selective electrode (model 96-09) with 720 pH/ISE meter (Orion, USA). XRD was performed with a RIGAKU XRD instrument using CuKα, at the Department of Ceramic Engineering, Banaras Hindu University, Varanasi. The rocks were broken and hydrous mineral–apatite-rich portions were hand-picked and powdered using agate mortar. The instrument was calibrated with Si standard before and after the analyses.

The results of chemical analysis of the groundwater samples are presented in Table 2. The pH value of groundwater in the study area varies from 7.0 to 7.7, indicating an alkaline condition which favours the solubility of fluorine-bearing minerals. In acidic medium (acidic pH), fluoride is adsorbed in clay; however, in alkaline medium, it is desorbed, and thus alkaline pH is more favourable for fluoride dissolution activity[36]. Fluoride has a unique chemical behaviour towards most of the anions and can be easily replaced even under normal temperature and pressure conditions[37]. In general, apatite and fluorite, besides replacement of hydroxyl by fluoride ions in mica, hornblende and soil that mostly consists of clay minerals, are major sources of fluoride in circulating water[38]. The concentration of Ca, Na, hydroxyl ion and certain complexing ions can alter the concentration of fluoride in the groundwaters[39]. It has been observed that 88% of EC values of the groundwater samples were less than 1000 mg/l (Table 2). Total dissolved solids, a salinity indicator for the classification of groundwater, varies from 270 to 714 mg/l in the study area. The bicarbonate content varies from 152 to 396 mg/l, and this high value indicates intense chemical weathering of the parent granite rock. The concentration of fluoride in the groundwater of the area varies from 0.483 to 6.7 mg/l (Table 2). Due to the common ion effect, the dissolution of fluorite is suppressed when the concentration of Ca is above the limit of fluorite solubility. The calcium content in the groundwaters of the study area varies from 15 to 44 mg/l and sodium values of groundwaters vary from 27.6 to 230 mg/l. The calcium concentrations were lower than the sodium concentrations (Table 2), which indicates the higher fluoride content in the groundwater of the study area. A strong negative correlation between Ca and F in the groundwaters that contain Ca in excess of that required for the solubility of fluoride minerals, has been observed by many researchers[40,41]. Generally, high concentration of sodium will increase the solubility of fluoride-bearing minerals in the waters. This is the cause for the higher levels of fluoride in the groundwaters of the study area. Under the prevailing semi-arid climatic conditions, during weathering of granite gneissic rocks, fluorine is released from apatite and biotite to the circulating alkaline groundwater. In the study area fluoride contamination is mainly a natural process, i.e. leaching of fluorine-bearing minerals, since no man-made pollution has been noticed.

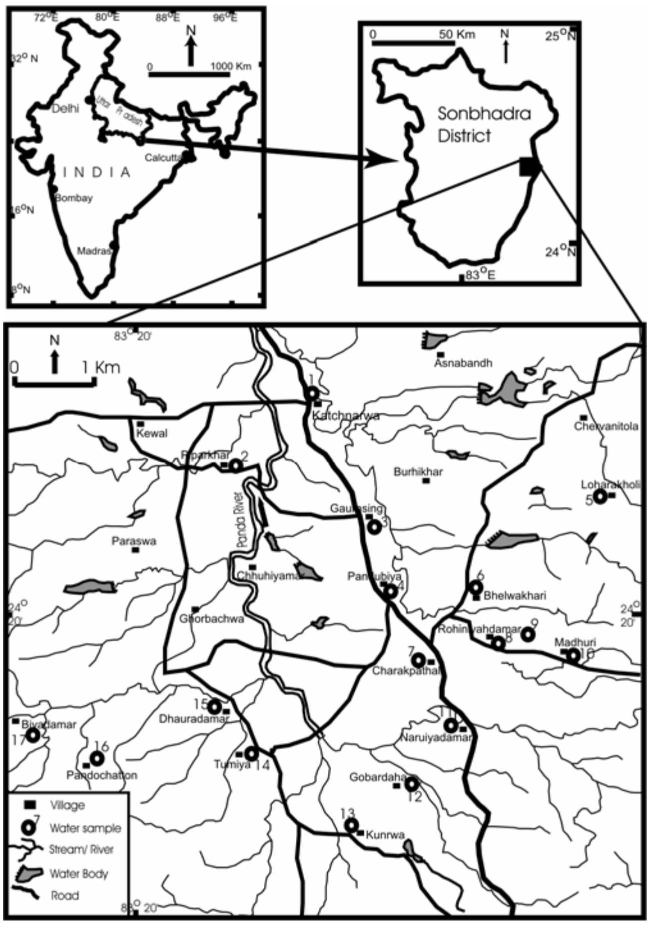

Figure 1. Location map of the study area. 1, Kachnarwa; 2, Piparkharmore; 3, Gaurasing; 4, Pandubiya; 5, Lohrakholi; 6, Bhelwakhari; 7, Charakpatli; 8, Rohiniyadamar (a); 9, Rohiniyadamar (b); 10, Madhuri; 11, Neruiyadamar; 12, Gobardaha; 13, Kunrwa; 14, Tumiya; 15, Dhauradamar; 16, Pandochatton; 17, Biyadamar.

Since fluorite, apatite, mica and various other minerals take part during rock–water interaction and liberate fluoride into the groundwater, it is imperative to know the presence of minerals in the rock specimen microscopically. Microscopic analysis of the rock samples shows quartz, microcline, biotite, apatite, sphene and minor chlorite, in which apatite and biotite are the fluorine-bearing minerals. Normally biotite in granite rocks may contain as high as 0.91% fluorine, hornblende contains 0.17% fluorine and fluorapatite has fluorine concentration[42] as high as 3.72%. The model analysis data show 20–30% quartz, 40–45% feldspar, 5–10% apatite, 20–25% biotite with minor chlorite and sphene. Feldspar is occasionally sericitized. Apatite grains are euhedral to subhedral with variable grain size (elongated grains are sometimes as long as 200 μm) and are closely associated with the hydrous minerals at the boundary of the coarse-grained feldspar and quartz (Figure 2). As far as immediate association is concerned, apatite grains are closely related to biotite and at places chlorite and myrmekites. The close association of apatite grains with the hydrous phases and myrmekites at the grain boundary of feldspar megacrysts is probably caused by the late fluid activity from crystallization of the granite body.

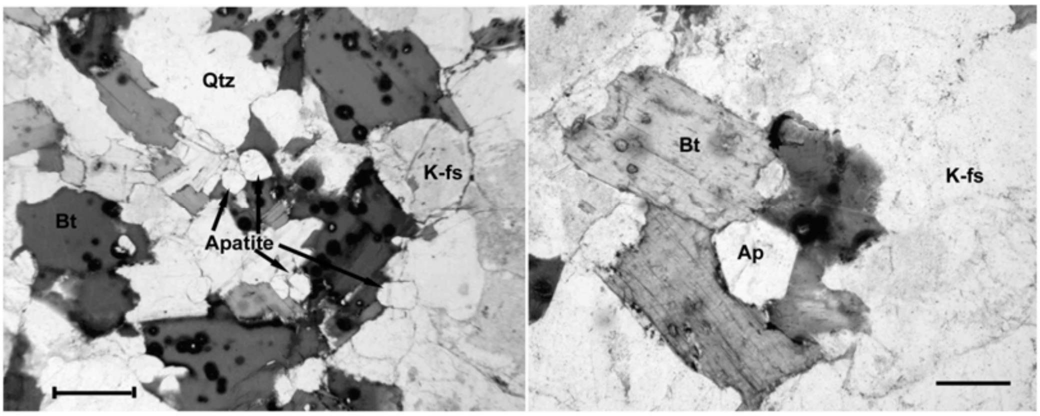

Figure 2. Microscopic identification of minerals in the rock samples.

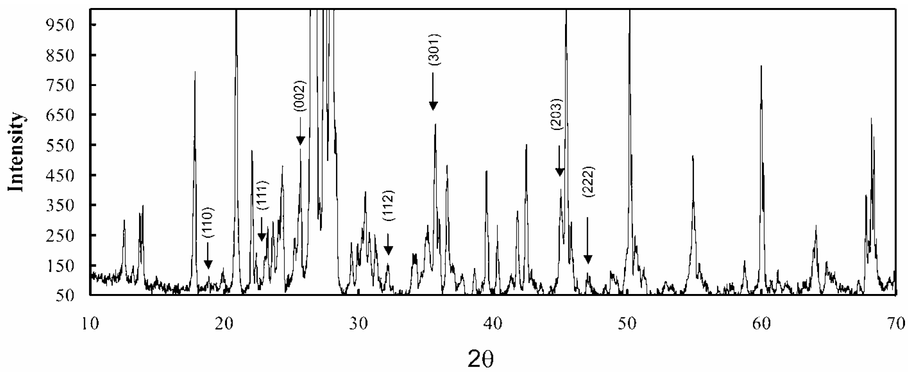

Figure 3. Profile peaks of apatite mineral in granite rock of the study area.

Figure 4. Fluorosis-related health disorders of the habitants in Kachnarwa area, Sonbhadra District.

XRD studies were conducted to know the variety of apatite occurrences in the granite rock by comparing with the standards. The peak profile (Figure 3) of this powdered granitic rock sample was indexed using the major known peaks of the mineral apatite. Lattice parameters were calculated using the program[43] CELLCALC 1.51. Lattice parameters calculated for a hexagonal system from the indexed peak profiles are presented in Table 3. These are comparable with the values of lattice parameters available for fluorapatite (standard JCPDS card number 15-876) using the same hexagonal crystal system, with space group of $P6_3/m$. This implies a close similarity with fluorapatite. Hence, fluorapatite along with biotite mica releases fluoride ion to the groundwaters of the study area.

Since there are no published data available on the incidence of fluoride in the groundwater and its health hazards in the Kachnarwa panchayat area, Sonbhadra District, we have carried out investigations on the fluoride content in groundwaters of the villages affected with dental and skeletal fluorosis and also the probable source of fluoride in groundwaters of the study area. High fluoride (>1.5 mg/l) distribution was found mainly in the villages of Lohrakholi, Charakpatli, Rohiniyadamar, Madhuri, Neruiyadamar, Gobardaha and Kunrwa (Table 2). The highest fluoride concentration (6.7 mg/l) was recorded from a dug well in Neruiyadamar village situated on the granite gneissic

Table 2. Location and geochemical analysis of groundwater in the study area

Serial no.	Location	pH	EC (μS/cm)	TDS (mg/l)	Ca^{2+} (mg/l)	Na^{+} (mg/l)	HCO_3^- (mg/l)	F^- (mg/l)	WT (m)
1	Kachnarwa$	7.5	550	355	44	27.6	180	0.483	9.5
2	Piparkharmore*	7.0	627	377	36	36.2	152	1.14	6.9
3	Gaurasing$	7.6	728	466	39	75.2	252	1.15	12.1
4	Pandubiya$	7.5	900	550	27	118.3	252	1.12	8.4
5	Lohrakholi$	7.7	930	571	16	98.9	316	1.99	7
6	Bhelwakhari$	7.5	640	414	34	45.6	216	1.02	9.5
7	Charakpatli*	7.3	760	457	35	75	248	2.06	9.3
8	Rohiniyadamar (a)$	7.5	820	522	28	124.3	216	5.6	6.7
9	Rohiniyadamar (b)$	7.4	880	520	19	132.2	268	5.38	7.5
10	Madhuri*	7.1	630	417	43	74.7	224	1.91	7.2
11	Neruiyadamar$	7.5	1190	724	15	230	396	6.7	10.7
12	Gobardaha*	7.2	825	504	28	93.9	276	3.33	9.5
13	Kunrwa$	7.5	1130	680	22	173	340	3.68	10.9
14	Tumiya$	7.0	740	480	32	96.8	196	1.39	9
15	Dhauradamar$	7.4	820	506	17	79.8	244	1.24	6.7
16	Pandochatton$	7.5	806	484	20	61.3	228	1.2	5.6
17	Biyadamar*	7.0	450	280	18	37.6	164	0.659	6.5

EC, Electrical conductivity; TDS, Total dissolved solids; WT, Water table; $Dug wells; *Bore wells.

Table 3. Lattice parameters for the apatite calculated for the hexagonal system from peak profile

	Mineral apatite in granite rock	Standard for fluorapatite
Lattice parameter	a = 9.34862 Å (2) c = 6.91427 Å (3) V = 523.326 Å^3 (2)	a = 9.3684 Å c = 6.8841 Å

complex. In the worst-affected villages of the study area, such as Rohiniyadamar, Madhuri, Neruiyadamar, Gobardaha and Kunrwa, the people suffer from dental and skeletal fluorosis (Figure 4). In the fluoride-affected villages, both children and adults suffer from health disorders like mottling of teeth, deformation of ligaments, bending of spinal column and ageing problem. In other villages such as Tumiya, Lohrakholi and Charakpatli, the fluoride concentration varies from 1.39 to 2.06 mg/l, where the people suffer mainly from dental fluorosis. The rest of the villages are not affected by fluoride disorders, because fluoride content in the groundwaters is within the permissible limit (<1.5 mg/l). About 47% of the groundwater samples analysed in the study area exceeds the maximum permissible limits of fluoride (1.5 mg/l) set by the ISI[2] and WHO[5] (Table 4). The weathering activity characterized by alternate wet and dry conditions of the semi-arid climate is responsible for leaching fluoride from the minerals present in the soils and rocks[44]. Easy accessibility of circulating water to the weathered product dissolves and leaches the minerals, contributing F^- to the groundwater[45].

The present study of groundwater quality with reference to fluoride concentration in the Kachnarwa region indicated that the groundwaters are alkaline in nature. Fluoride concentration in the groundwaters of the study area varied from 0.483 to 6.7 mg/l. High fluoride concentration was found in the villages covered with granitic gneissic complexes than the other rock formations. The highest fluoride concentration was found to corroborate with low calcium values and high sodium content in the groundwaters (Table 2). Weathering and leaching of fluorine-bearing minerals in rock formations under alkaline environment lead to the enrichment of fluoride in the groundwaters. Among the 17 groundwater samples analysed in the study area, 47% of samples had high fluoride content than the maximum permissible limit (1.5 mg/l). A high rate of evapo-transpiration, comparatively low rainfall, intensive irrigation and heavy use of fertilizers, alkaline environment, longer residence time of water in the weathered aquifer zone and low rate of dilution are favourable factors for the dissolution of fluorine-bearing minerals and thereby increase of fluoride concentration in the groundwater.

The present study was confined to a small area in the Kachnarwa region. A more detailed study is necessary for better understanding of the source and effects of fluoride problems in other parts of the Sonbhadra District. Local people ingesting the groundwater have not received medical attention in the study area till date. Since these people are dependent on the groundwater for domestic use, remedial measures such as importing of drinking water and rainwater harvesting are needed. Nutritional diet such as calcium and phosphorus-rich food should be recommended to those affected with fluorosis, as it decreases rate of accumulation of fluoride in the human body[22]. Environmental awareness programme for the health implications of fluoride should be emphasized through education of the public and community participation.

Table 4. Comparison of fluoride content in the groundwater of the study area with drinking water standards

	ISI standards		WHO standards			
Parameter	Highest desirable (mg/l)	Maximum permissible (mg/l)	Highest desirable (mg/l)	Maximum permissible (mg/l)	Number of samples exceeding permissible limits	Percentage of the total number of samples exceeding limits
Fluoride	0.6–1.2	1.5	0.5	1.5	8	47

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ACKNOWLEDGEMENTS. N.J.R. thanks the University Grants Commission, New Delhi, for financial support under a major research project during 2008–11. We thank P. Ram and Janmejoy Singh for help during the field work and laboratory chemical analysis.

Received 27 November 2007; revised accepted 13 January 2009

Late Holocene uplift in the lower Narmada basin, western India

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A late Holocene depositional surface has been identified in the lower Narmada basin of western India. This terraced surface occupies the valley formed by the incision of the mid–late Holocene surface. Radiocarbon dating of organic-rich clay horizons indicates that these terrace sediments were deposited between 1900 and 1200 yrs BP, well corresponding with the large-magnitude palaeoflood records of the central Narmada basin. These terraces occur as 2–4 m high uplifted surfaces and point to a latest phase of tectonic uplift in the lower Narmada during the historical times.

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Keywords: Late Holocene, radiocarbon dating, tectonics, terrace sediments.

IN western India, basement controlled tectonics have been extensively investigated[1–5]. Horst and graben structure in the subsurface is basically the manifestation of the reactivation of basement trends[3]. Having originated during the Mesozoic[2], the Cambay and the Narmada Mesozoic basins occupy a large part of mainland Gujarat. These basins have undergone several phases of subsidence and uplift since their formation. Repeated movements along faults in several phases throughout the Mesozoic and Cenozoic have shaped the landscape of mainland Gujarat. In the lower Narmada basin the reactivation of the basement trends since Precambrian is well established[4]. The tectonic uplift in phases during the Quaternary has exposed the Tertiary and Quaternary sediments along various rivers. The study area is located within the lower Narmada basin, between lat. 21°35′ to 21°55′N and long. 73°00′ to 73°25′E (Figure 1 *a* and *b*). It lies on the Narmada–Ankleshwar block of the Cambay rift basin, bounded by the ENE–WSW seismically active Narmada–Son Fault (NSF) in the north. The detailed tectonic and geomorphic history of the lower Narmada valley up to mid–late Holocene is well understood[4,5]. Here we report the occurrence and significance of a late Holocene depositional surface within the mid–late Holocene surface of the lower Narmada basin.

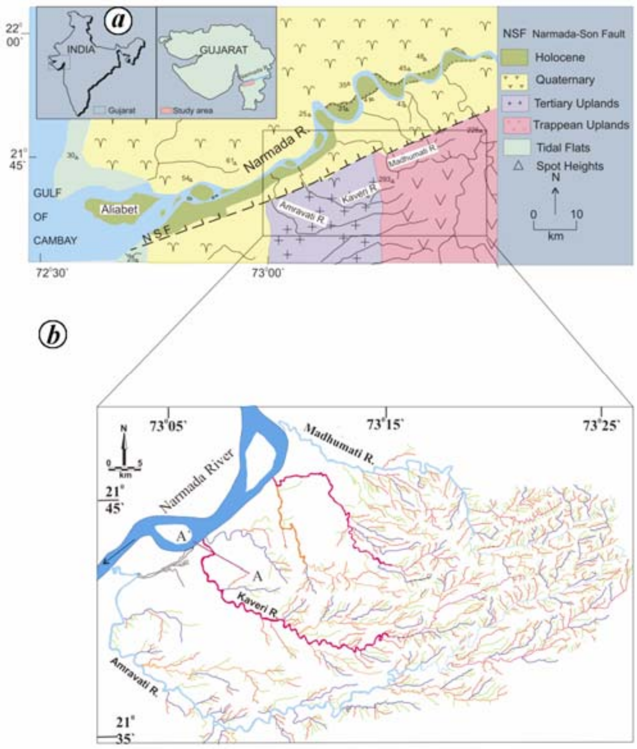

Figure 1. ***a***, Location map showing the general geology and geomorphology of the area[4]. ***b***, Area of study comprising Amravati, Kaveri and Madhumati rivers, meeting the Narmada river in its lower reach. Line *A–A′* marks the cross-section of Figure 3 *b*.