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Arsenic-contaminated groundwater in Holocene sediments from parts of Middle Ganga Plain, Uttar Pradesh, India

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Groundwater survey in Ghazipur, Varanasi and Mirzapur districts of Uttar Pradesh (UP) in Middle Ganga Plain shows that people in these areas are drinking arsenic-contaminated groundwater. Moreover, 60% of tubewells have arsenic concentration above the WHO guideline value of 10 μ g/l and 20% is above 50 μ g/l, the Indian standards for arsenic in drinking water. Depth information of 68 tubewells indicates that 85% tubewells are from shallow aquifers (10–42 m). Most of the arsenic-contaminated tubewells have a depth of 25–45 m. The iron content in tubewells varies from 0.1 to as much as 7.6 mg/l and 83% tubewells have iron more than 1 mg/l. The correlation between dissolved arsenic and iron in tubewell waters is positive.

Arsenic-contaminated tubewells in UP are located in Holocene Newer Alluvium entrenched channels and floodplains which are characterized by grey to black coloured organic-rich argillaceous sediments. Saidpur, Varanasi, Chunar and Mirzapur towns have arsenicsafe groundwater because their positions on the Pleistocene Older Alluvium upland surfaces are made up of oxidized yellowish-brown coloured sediments with calcareous and ferrugenous concretions. However, deeper tubewells (>60 m) in Pleistocene Older Alluvium aquifers in arsenic-contaminated areas would be a better option for arsenic-safe groundwater. Dugwells would be another source for arsenic-safe groundwater in arsenic-affected areas. The arsenic in groundwater in Middle Ganga Plain, UP is getting released from associated sediments which were mainly deposited from the Himalayan hill ranges and minor inputs from peninsular India.

Keywords: Arsenic, groundwater, Holocene Newer Alluvium, Pleistocene Older Alluvium, Middle Ganga Plain.

BEFORE the onset of the 21st century, 20 groundwaterarsenic contamination incidents had been reported in different parts of the world including five from Asia¹. Severe arsenic contamination in groundwater has been reported from Bangladesh¹⁻⁴ and West Bengal⁵⁻¹². Arsenic pollution in groundwater is also reported from parts of Indus delta in Pakistan¹³, inner Mongolia, the Xin-Xiang province in the People's Republic of China¹⁴ and Taiwan¹⁵. Arsenic pollution in groundwater is known from many fluvio-deltaic tracts of the world like Hanoi City and the upper end of the Red River delta¹⁶, as well as, from flood- and delta-plains of the Mekong River in Laos and Cambodia¹⁷. Groundwater-arsenic contamination was also reported in the Tarai belt of Nepal¹⁸. Thus arsenic problem is common to several alluvial plains in south and east Asia.

Arsenic contamination in groundwater in India was first reported from Chandigarh and different villages of Panjab, Haryana and Himachal Pradesh¹⁹. Another example of arsenic poisoning was reported from West Bengal²⁰. Groundwater-arsenic contamination was also reported from Middle Ganga Plain in Jharkhand, Bihar and Uttar Pradesh^{10,21,22}. Groundwater-arsenic contamination in tubewell was also reported from the states of north-east India^{23–26}. It appears that a good portion of all states and countries in the Ganga-Meghna-Brahmaputra (GMB) plain comprising an area over 500,000 km² and a population of over 450 million are at risk from groundwaterarsenic contamination. The upper permissible limit of arsenic in drinking water is 10 µg/l as per WHO guidelines²⁷, which has been endorsed by the Bureau of Indian Standards²⁸.

Uttar Pradesh (UP) in Middle Ganga Plain (Figure 1) is densely populated fertile land. The people of Ghazipur, Varanasi and Mirzapur districts are drinking arseniccontaminated groundwater. The objective of this study is to elucidate the role of geomorphology and Quaternary morphostratigraphy (geology control) on groundwaterarsenic contamination in entrenched channels and floodplains from parts of Middle Ganga Plain in UP (Figure 2).

Geomorphologic and Quaternary morphostratigraphic map of the region between Saidpur and Mirzapur towns of Uttar Pradesh along Ganga River is prepared based on the Survey of India topographic sheets of 1 : 50,000 scale,

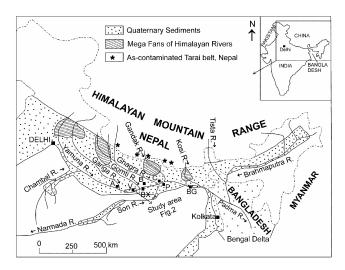


Figure 1. Quaternary sediments in Indo-Ganga foredeep and Bengal Basin. The study area (Figure 2) from parts of UP is shown in Middle Ganga Plain. A, Allahabad; V, Varanasi; BX, Buxar; B, Ballia; C, Chhapra; P, Patna, and BG, Bhagalpur.

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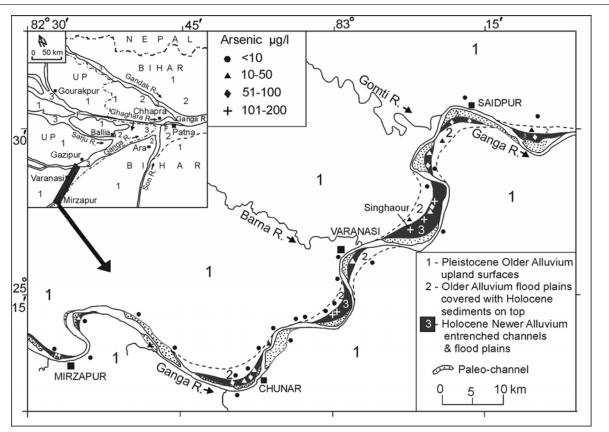


Figure 2. Geomorphologic and Quaternary morpho-stratigraphic map from parts of UP showing arsenic-affected areas.

with field checks to identify fluvial landforms and soil characters. The geomorphic features of the Ganga Plain show differences in their elevations, spatial distribution, and nature of sediments, indicating that their formation must have occurred at different times, under different climates, water budget and sediment supply during Pleistocene–Holocene period. The study area is subdivided into three zones, i.e. Pleistocene Older Alluvium upland inter-fluve surface (47 ± 12 ka), Holocene Newer Alluvium river valley terrace surface (3 ± 1 ka) and Holocene to Recent active channels and floodplains^{29,30}.

The Pleistocene Older Alluvium surface was recognized by yellow-brown coloured sediments with profuse calcareous and ferruginous concretions. The Holocene Newer Alluvium surface was recognized by grey to black coloured organic-rich argillaceous sediments in entrenched channels and floodplains of Ganga River (Figure 3). In this study, about 3000 km² has been mapped to delineate groundwater arsenic-contaminated and arsenic-safe areas from parts of UP (Figure 2). Groundwater arsenic in Ghazipur, Varanasi and Mirzapur districts has been tested in tubewells within Holocene Newer Alluvium aquifers and Pleistocene Older Alluvium aquifers.

Arsenic analysis of 245 water samples, mainly from tubewells and some from dugwells, was made from parts of UP using EZ Arsenic Test Kit, Hach, USA (cat. no. 28228-00). Kit data was used as a guide to delineate arsenic-contaminated area. The minimum determination limit of the kit is $10 \mu g/l$. Kit test of 245 tubewell water samples helped to delinate the arsenic-contaminated and arsenic-uncontaminated areas from parts of UP in Middle Ganga Plain. Out of 245 water samples, 68 tubewells water samples were taken for laboratory confirmation test and laboratory tested values of arsenic and iron are shown in Table 1.

Hand tubewell water samples were collected in acid prewashed 10 ml polythene bottles. The bottles were kept overnight in dilute laboratory grade nitric acid (1:1) and finally washed with distilled water. Immediately after collection, 1 drop of dilute nitric acid (1:1) GR Grade was added as preservative. Arsenic and iron were determined from these samples. All reagents are of Analar grade. A solution of 1.25% NaBH₄ (Merck, Schuchardt, Germany) was prepared in 0.5% NaoH (Merck, Mumbai, India). A 5.0 M solution of HCl ((Merck, Mumbai, India) was used. All these solutions were prepared using distilled de-ionized water. The flow rate for both tetrahydroborate and hydrochloride acid was 1 ml/min. Details of the reagents and glassware are given in refs 5 and 6.

The tubewell water samples were analysed for As and Fe at the School of Environmental Studies, Jadavpur University, Kolkata, following a flow injection hydride generation atomic absorption spectrometry (FI-HG-AAS) system. A Perkin-Elmer Model 3100 atomic absorption spectrometer equipped with a Hewlett-Packard Vectra Computer with GEM software, Perkin-Elmer EDL system-2,

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Figure 3. (*a*) Holocene Newer Alluvium grey to black coloured argilleaceous sediments with palaeo-channel (scale is geological hammer) and (*b*) Plesitocene Older Alluvium yellow-brown coloured oxidized sediments with calcareous and ferrugenous concretion (scale is pen) in Varanasi areas.

Table 1. Arsenic and iron concentration in tubewells water from parts	of UP
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Latitude	Longitude		Depth	As	Fe	Latitude	Longitude		Depth	As	Fe
(N)	(E)	Village	(m)	$(\mu g/l)$	$(\mu g/l)$	(N)	(E)	Village	(m)	(µg/l)	$(\mu g/l)$
25°15.26′	83°01.35′	Malihiya	42	6	3199	25°13.67′	83°00.28′	Ramnu	36	8	1750
25°19.55′	83°09.31'	Kaile	10	8	1891	25°18.06'	83°00.16′	Varanasi town	40	3	1432
25°20.49′	83°10.11′	Mapauli	14	3	1368	25°15.08′	83°01.12′	Malahai	36	3	160
25°26.09′	83°10.19′	Manhar kalan	50	6	784	25°12.73′	82°58.18′	Mora Deora	32	125	6619
25°27.28′	83°10.19′	Hardan Jura	50	35	2776	25°12.60′	82°59.02′	Mora Deora	30	110	6678
25°26.94′	83°08.65′	Hardan Jura	50	60	3805	25°12.71′	83°00.53′	Mora Deora	30	115	7654
25°28.41′	83°10.15′	Mathia	42	20	4004	25°12.70'	82°56.44′	Chittanaun	54	3	2615
25°28.31'	83°08.76′	Mathia	42	25	2340	25°12.56′	82°55.96′	Gausinpur	40	8	1750
25°28.59′	83°10.12′	Thanda Kalan	38	30	985	25°11.44′	82°53.84′	Maehhara	36	15	1954
25°30.95'	83°11.26′	Jamalpur	36	20	1256	25°10.45'	82°53.49′	Adalpur	40	3	804
25°31.16′	83°11.30′	Jamalpur	36	55	4567	25°08.45'	82°52.38′	Bedauli	27	16	1482
25°31.11′	83°13.87′	Marufpur	30	60	5504	25°08.32'	82°52.62′	Bedauli	36	25	2341
25°31.11′	83°13.62′	Marufpur	30	20	4578	25°07.93′	82°52.26′	Junar	36	3	1871
25°21.20'	83°05.13′	Singhaour	42	180	6619	25°07.07'	82°50.17′	Shilpi	30	65	2764
25°20.57'	83°07.19′	Singhaour	42	155	6789	25°06.99′	82°50.06′	Shilpi	30	25	3452
25°21.20'	83°06.45′	Chandpur	27	25	1523	25°07.03'	82°49.34′	Bhualpur	30	55	3342
25°21.59′	83°09.19′	Gobartha	36	65	1589	25°07.07'	82°49.64′	Bhualpur	30	25	2442
25°21.84'	83°09.42′	Gobartha	36	110	3429	25°07.28'	82°48.63′	Sikhar	36	30	2977
25°22.21′	83°09.91′	Mokolpur	48	55	925	25°07.17'	82°48.45′	Sikhar	36	50	2345
25°22.22′	83°10.02′	Mokolpur	48	100	4432	25°06.98′	82°51.26′	Madhia	26	30	4531
25°21.28′	83°09.82'	Rampur	40	120	3451	25°07.88'	82°47.15′	Ramgarh	27	3	2313
25°23.77'	83°10.05′	Chhatauni	42	6	725	25°08.20'	82°46.41′	Pemapur	27	10	1428
25°31.10′	83°06.29′	Tekuri	35	8	1086	25°11.54'	82°55.81′	Shivpur	33	6	1245
25°30.49′	83°06.98′	Dodhoua	21	3	744	25°10.36'	82°55.81′	Chitraha	33	3	1026
25°30.55'	83°06.98′	Khaurana	40	8	885	25°06.83'	82°48.50′	Shamaspur	42	3	201
25°29.16′	83°9.25′	Dubaitha	30	55	4321	25°10.51'	82°44.12′	Dhanukpur	27	5	1428
25°29.81'	83°19.41′	Chhapra	36	25	1684	25°10.39'	82°47.22′	Domanpur	33	15	4668
25°29.09′	83°20.23′	Chhapra	36	65	2345	25°13.29′	82°40.39′	Kaotabir	48	3	543
25°30.25'	83°20.49′	Rampur Manjha	ı 36	20	1784	25°13.06'	82°34.45′	Dingurpatti	27	22	3722
25°32.91′	83°21.25′	Deokoli bazar	35	6	2233	25°12.64′	82°35.36′	Dingurpatti	27	6	2130
25°32.46′	83°13.54′	Saidpur bazar	21	8	2213	25°09.76'	82°32.66′	Majhgawan	36	25	100
25°13.20′	82°59.09′	Tarapur	54	4	4044	25°09.66′	82°32.59′	Majhgawan	36	55	5621
25°13.17′	82°59.09′	Tarapur	48	6	2434	25°09.55′	82°32.62′	Majhgawan	36	60	2347
25°13.18′	82°59.71′	Tikari	45	6	1106	25°10.20'	82°32.57′	Majhgawan	36	6	1650

2, arsenic lamp (lamp current 400 mA) and Varian AAS Model Spectra AA-20 with hollow cathode arsenic lamp (lamp current 10 mA) were used. The minimum detection limit with 95% confidence level was $3 \mu g/l$ arsenic^{5,6}. Dissolved Fe of acidified water samples was analysed by phenanthroline method by using UV spectrophotometer. The accuracy of analytical method using FI-HG-AAS was verified by analysing Standard Reference Materials CRM (BND 301) NPL, Indian water (certified value 990 \pm 200 µg/l; observed 960 \pm 40 µg/l); Water SRM (quality

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control sample for trace metal analysis) from USEPA Environmental Monitoring and Support Laboratory, Cincinnati, Ohio, USA (certified value $17.6 \pm 2.21 \,\mu g/l$; observed $16 \pm 3.5 \,\mu g/l$). The different mineral phases of sediments are identified using an Automated Powder X-ray Diffractometer (Model Philips APD-15). Latitude and longitude of hand tubewells were recorded by a GPS (Garmin eTrex Vista).

Information of 68 tubewells along with sample ID, GPS East and North, depth, villages, arsenic concentration, iron concentration has been shown in Table 1.

Table 1 shows that 60% of tubewells have arsenic concentration above the WHO guideline value of 10 μ g/l and 20% is above 50 μ g/l, the Indian standard for arsenic in drinking water. Moreover, 23 villages have arsenic > 10 μ g/l and 12 villages have arsenic > 50 μ g/l in tubewell water. Maximum arsenic is 180 μ g/l in Singhaour village.

Most of the arsenic-contaminated tubewells are located in entrenched channels and floodplains of Holocene Newer Alluvium sediments between Saidpur and Mirzapur towns (Figure 2). Table 1 shows maximum arsenic concentration in tubewell water as 180 µg/l in Singhaour village, which is located in Holocene Newer Alluvium entrenched channels and flood plains of Ganga River. However, tubewells on its opposite side have arsenic-safe groundwater (<10 μ g/l) due to their position on Pleistocene Older Alluvium sediments (Figure 2). Few arseniccontaminated tubewells (arsenic $\leq 50 \,\mu g/l$) are located in Older Alluvium floodplain. These areas have a shallow cover of Holocene sediments on Older Alluvium floodplains. Tubewells located in Saidpur, Varanasi, Chunar and Mirzapur towns have arsenic-safe groundwater due to their positions on Older Alluvium surfaces. Historically, Varanasi is one of the oldest towns of Indian culture and civilization. Groundwater in Varanasi town is virtually arsenic-safe due to its position in Older Alluvial upland surfaces whereas villages located in Holocene Newer Alluvium sediments in entrenched channels and floodplains of Ganga River have arsenic-contaminated groundwater (Figure 4).

The population density of arsenic in tubewell water in Varanasi areas is higher compared to that of Mirzapur areas. Entrenched channels and floodplains of Ganga River near Varanasi town have a wide cover of Holocene sediments in comparison to that of the Mirzapur areas (Figure 2).

Table 1 shows that 83% of tubewell water samples have higher concentration of iron beyond its permissible limit of 1 mg/l. The iron content varies from 0.1 to as much as 7.6 mg/l. Even arsenic-safe tubewell water has high amount of iron (arsenic 4 μ g/l and iron 4 mg/l).

The correlation of dissolved arsenic and iron in tubewell water is positive. The higher value of correlation co-efficient (R) = 0.73 than that of PE(R) = 0.1301 (probable error of R) justifies significance of correlation.

However, lower values of arsenic correspond with the higher values of iron (Figure 5).

Depth information of 68 tubewells indicates that 85% of tubewells are from shallow depth (10–42 m). 51 tubewells have arsenic $\leq 40 \ \mu g/l$ within the depth of 10–54 m and 17 tubewells have arsenic $\geq 50 \ \mu g/l$ within the depth of 30–50 m. However, most of the arsenic-contaminated tubewells occur within the depth of 25–45 m. Maximum value of arsenic (180 $\ \mu g/l$) corresponds to a depth of 42 m (Figure 6).

Chemical analysis of Newer Alluvium soil (near Varanasi 25°14.50': 83°01.73') shows arsenic and iron concentrations of 8.7 mg/kg and 6.13 g/kg respectively and Older Alluvium soil (Varanasi town 25°18.75':

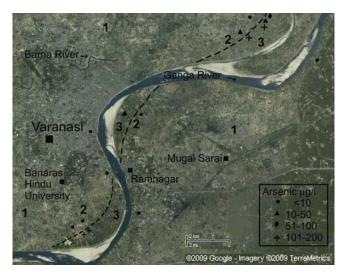


Figure 4. Google satellite image in and around Varanasi town showing spatial distribution of arsenic-contaminated tubewells in Holocene Newer Alluvium entrenched channels and floodplains. Geomorphologically, the area is subdivided into three zones, viz. 1, Pleistocene Older Alluvial upland surfaces; 2, Older Alluvium floodplains covered with Holocene sediments on top and 3, Holocene Newer Alluvium entrenched channels and floodplains.

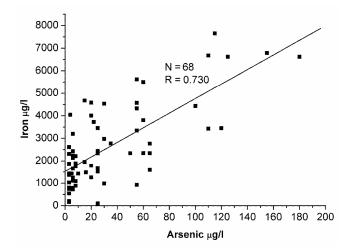


Figure 5. Correlation between dissolved arsenic and iron concentration in tubewells water from parts of UP.

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83°00.17') shows arsenic and iron concentrations of 5.65 mg/kg and iron 3.75 g/kg respectively.

X-ray diffraction (XRD) studies are done on arsenicsafe (Varanasi town 25°18.75': 83°00.17') Older Alluvium sediments and arsenic-contaminated (near Varanasi 25°14.50': 83°01.73') Newer Alluvium sediments. Mineralogical assemblages of those sediments are quartz, muscovite, chlorite, montmorillonite, kaolinite, feldspar, amphibole and goethite (Figure 7).

Ganga Plain is a shallow asymmetrical depression with a gentle eastern slope. Most of the rivers of northern

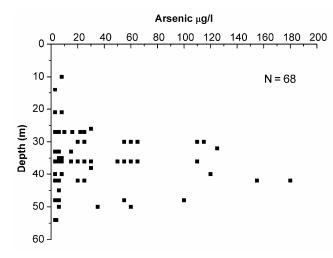


Figure 6. Correlation between arsenic and depth in tubewells water from parts of UP.

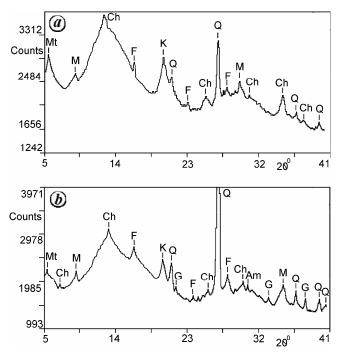


Figure 7. XRD studies on Pleistocene Older Alluvial sediments (*a*), and Holocene Newer Alluvium sediments (*b*), in Varanasi areas. Q, Quartz; Ch, Chlorite; M, Muscovite; Mt, Montmorillonite; F, Feldspar; K, Kaolinite; Am, Amphibole, and G, Goethite.

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Ganga Plain follow a southerly trend; those of southern part follow north-easterly trend; only those of the axial zone follow the eastern slope. Ganga River is the main river in Gangetic plain flowing west to east and forming Bengal delta before joining the Bay of Bengal. A large number of small drainages, dense networks of channel and numerous lakes and swamps have developed on the Gangetic plain³¹.

Geomorphologically, the Middle Ganga Plain is subdivided into three zones, viz. 1, Pleistocene Older Alluvium upland surfaces, 2, Older Alluvium floodplains covered with Holocene sediments on top and 3, Holocene Newer Alluvium entrenched channels and floodplains (Figure 8).

The major parts of the Gangetic plain consist of interfluve upland surface of Older Alluvium. Pleistocene Older Alluvium inter-fluve upland surface is characterized by yellow-brown coloured sediments with profuse calcareous and ferruginous concretions and is either exposed or occurs under shallow cover of Holocene sediments. Tubewells in Older Alluvium aquifers have virtually arsenic-safe groundwater. Major towns on the bank of Ganga River (Saidpur, Varanasi, Chunar and Mirzapur) are located on Older Alluvium upland surfaces and have arsenic-safe groundwater. However, tubewells in Holocene Newer Alluvium entrenched channels and floodplains characterized by grey to black coloured organic-rich argillaceous sediments have arseniccontaminated groundwater (Figure 2).

In Gangetic plain, the Pleistocene Older Alluvium yellow-brown coloured surface was subjected to erosion and oxidation. This was dissected by channels and floodplains and buried under Younger Newer Alluvium sediments. These oxidizing Pleistocene upland surfaces are well flushed by groundwater flow due to high-hydraulic head and sediments are low in arsenic and devoid of organic



Older flood plains covered with Holocene sediments on top

3 Holocene Newer Alluvium entrenched channels and flood plains

Figure 8. Schematic 3D diagram of Middle Ganga Plain in UP showing arsenic-contaminated areas in Holocene Newer Alluvium entrenched channels and floodplains and arsenic-safe areas in Pleistocene Older Alluvium upland surfaces. Older Alluvium floodplains covered with Holocene sediments on top are locally arsenic-contaminated in groundwater (modified after Singh³¹).

matter. The environment of the oxidized Pleistocene yellow-brown colour Older Alluvium sediments is not favourable to release sorbed arsenic to groundwater.

Schematic 3D diagram of Middle Ganga Plain shows arsenic-affected aquifers in Holocene entrenched channels and floodplains and arsenic-safe aquifers in Older Alluvium upland surfaces. Geomorphology and Quaternary morpho-stratigraphy play an important role in controlling groundwater arsenic from parts of UP in Middle Ganga Plain (Figure 8). Sediment characteristics (Pleistocene yellow-brown coloured oxidized sediments or Holocene grey to black coloured organic-rich argillaceous sediments) are important parameters for installation of tubewells for safe drinking water in UP. Tubewells located in Pleistocene Older Alluvium surfaces would be a better option for arsenic-safe groundwater in UP.

No specific sources of arsenic could be identified in Gangetic plain of UP but several potential minor sources have been identified both in the Himalayan belt as well as, in peninsular India. Gangetic plain was formed due to accumulation of bulk sediments from the Himalayan hill range whereas the input of peninsular India is minor. The southern belt of the Himalayas is subjected to high erosion and intense rainfall during the Holocene time^{32,33}. The possibility of erosion, oxidation and transportation of arsenic-bearing products in suspension and solution in the Gangetic plain is high.

Potential arsenic sources of arsenic from peninsular India include the following: (i) pyrite-bearing shale from the Proterozoic Vindhyan range with its Amjhore mine pyrite containing 260 mg/kg arsenic³⁴, (ii) As–Cu mineralization in the Bundelkhand Granite around Salaiya area, Chhatarpur district, UP³⁵, and (iii) gold belt of the Son valley with arsenic content in bedrock locally reaching 2.8–10 g/kg³⁶.

Groundwater-arsenic contamination is also recorded from Terai belt in Nepal¹⁸. The provenance of the Terai sediments is from the Himalayan hill range. Most of the rivers in northern parts of Gangetic plain are from the Himalayan hill range.

Abandoned channels, swamps and active channels in Middle Ganga Plain are perennially or seasonally water filled where the aquifers are presumed to be enriched in biomass and thereby organic carbon. Chemical data on arsenic-affected aquifer in Bangladesh reveals that arsenic mobilization was associated with recent inflow of carbon driven by recent biogeochemical processes³⁷. Reduction of hydrated iron oxide (HFO) is common and high concentration of dissolved iron (7.65 mg/l) indicates strong reducing condition.

Mobilization of arsenic in UP tubewell waters is expected to follow a similar course as observed in the Gangetic delta of West Bengal, India and Bangladesh. Arsenic sorbed on discrete phases of hydrated Fe–Mn oxide coated sediments grains were preferentially entrapped in grey to black coloured organic-rich argillaceous sediments in entrenched channels and floodplains. Biomediated reductive dissolution of HFO by anaerobic heterotrophic Fe^{3+} reducing bacteria (IRB) plays an important role in releasing sorbed arsenic to groundwater^{2,38–40}.

Even within arsenic-affected areas in UP, dugwells are found to be arsenic safe because of their oxygenated nature. It has been observed that often arsenic-contaminated tubewells in UP are being used for drinking water, whereas well constructed dugwells with arsenic-safe water are poorly maintained and used unhygienically for other domestic purposes. Thus properly maintained dugwells represent an important source of arsenic-safe water in arsenic-contaminated areas.

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Defining optimum spectral narrow bands and bandwidths for agricultural applications

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In this study, an attempt was made to define the optimum set of spectral narrowbands and the required bandwidth for agricultural applications. Spectral observations were collected using ASD handheld spectroradiometer (325-1075 nm) from major kharif (rainy) and rabi (winter) season crops, two different soil types and crops under different agronomic treatments. To identify best bands, a stepwise discriminant analysis (SDA) was carried out for each data set. Aggregating the bands selected from individual SDA, 13 optimum narrow bands were identified for crop and soil assessment. To find optimum bandwidth, the measured reflectance was aggregated to different bandwidths (3, 5, 10, 15, 20, 25 and 30 nm). The reflectance values at different wavelength regions were compared with the original spectra using root mean square error. It was found that the optimum bandwidth required for crop discrimination differed for different wavelength regions.

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