

Study of Groundwater Arsenic in Terai Belt of Eastern Uttar Pradesh, India

Anil K Dwivedi

PEARL, Department of Botany, DDU Gorakhpur University, Gorakhpur, Uttar Pradesh, India

ABSTRACT

The present study investigates the groundwater solute chemistry, hydrogeochemical behavior of arsenic (As) and the assessment of health risks through ingestion and dermal contact pathways to the adults and children of the Bahraich district, falling under the middle Gangetic plain, Ghaghara river sub-basin, Terai region of India. A Monte Carlo simulation and a sensitivity analysis were also performed to quantify the uncertainties and impact of various input variables in risk calculations, respectively. Concentrations major ions (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , SO_4^{2-} , PO_4^{3-} , NO_3^- , HCO_3^- , F^- and Cl^-), dissolved organic carbon (DOC), and trace metals (As, Fe, and Mn) as well as physical parameters (EC, pH, and Eh) were measured on the collected groundwater and river water samples. The concentration of As in the groundwater samples was found in range between 0.64 $\mu\text{g/L}$ to 104 $\mu\text{g/L}$ and ~37% of the samples had $\text{As} > 10 \mu\text{g/L}$ (WHO and BIS acceptable limit of 10 $\mu\text{g/L}$ for drinking water). River water samples also displayed high As concentrations (mean of 14 $\mu\text{g/L}$) with ~73% of samples having $\text{As} > 10 \mu\text{g/L}$, which could be attributed to infiltration from As-enriched shallow groundwater of the nearby regions. The groundwater of the study area was predominantly of Ca^{2+} - Mg^{2+} - HCO_3^- type. Bivariate weathering plots showed groundwater was influenced by silicate weathering and carbonate dissolution, along with the ion exchange and reverse ion exchange processes, with a minor contribution from evaporate dissolution. Most of the groundwater samples ($n = 57$) were anoxic and had low SO_4^{2-} and NO_3^- , and high Mn, Fe and DOC concentrations. The observed values and correlations between various measured parameters, including groundwater As concentrations and saturation indices calculations, indicated the reductive dissolution of iron oxyhydroxide as the major process for As mobilization in the study area. The results of total non-carcinogenic risk (HI) estimated by the deterministic and probabilistic techniques were nearly identical for both adults and children, but an overestimation was observed in the case of carcinogenic risk calculated by the deterministic approach. Health risk results also showed that children were more susceptible to non-carcinogenic risk, whereas adults were at a higher risk of cancer in the study area. Sensitivity analysis indicated that the concentration of As in groundwater and exposure duration (ED) were the most effective variables for non-carcinogenic (HI) and carcinogenic (TILCR) risk estimation in both adults and children.

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KEYWORDS: terai belt, arsenic, groundwater, eastern Uttar Pradesh, gangetic plain, India

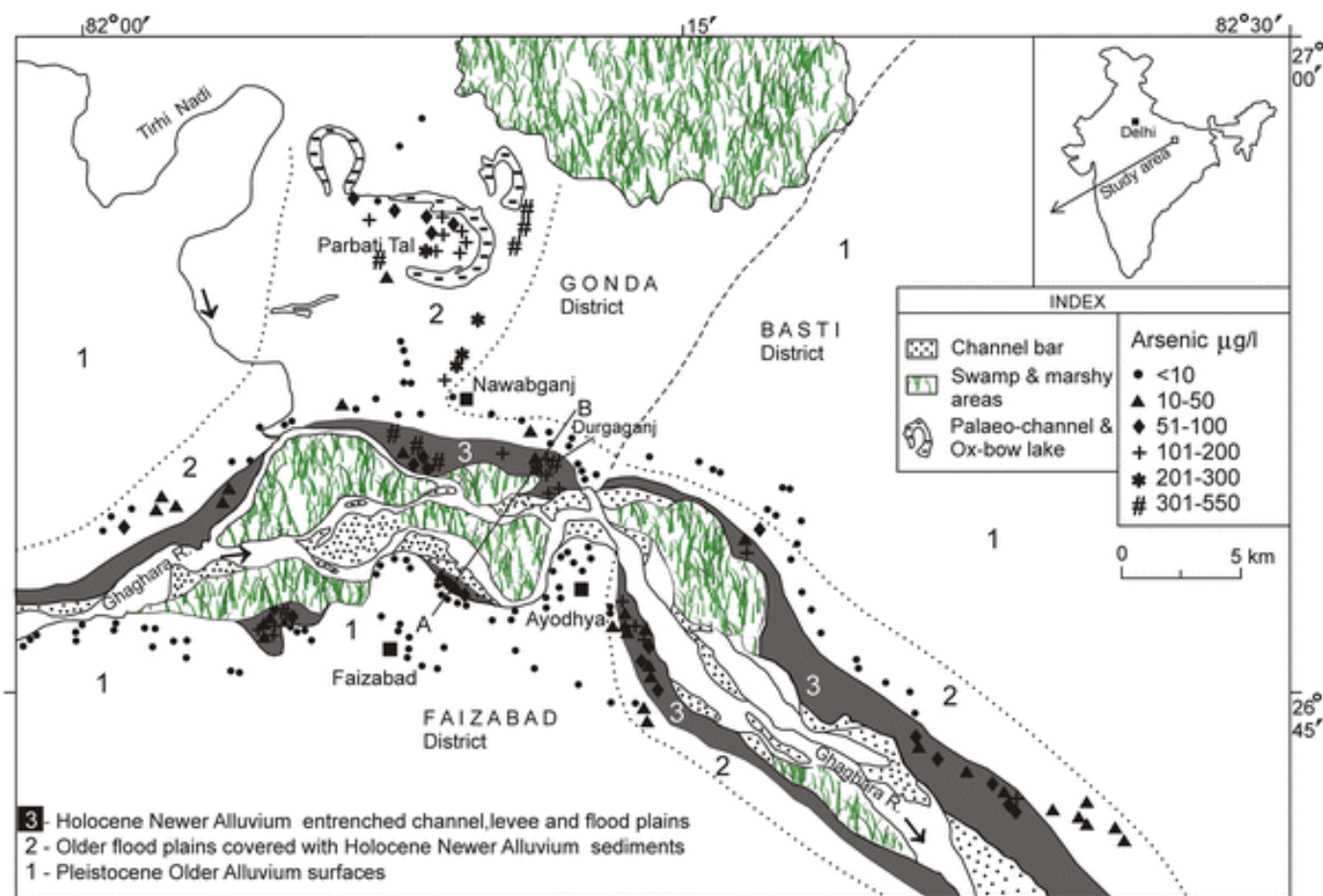
INTRODUCTION

Ingestion of Arsenic (As) through groundwater has become a severe public health concern around the world. In >70 countries groundwater with elevated concentrations of As has been reported. The present study estimates the As concentration as high as 399

$\mu\text{g/L}$ in Rapti River Basin which lies in Terai region of Uttar Pradesh, India. The villages on the left bank showed As contamination whereas villages on the right bank does not show As concentration above WHO standards of 10 $\mu\text{g/L}$. High concentration of

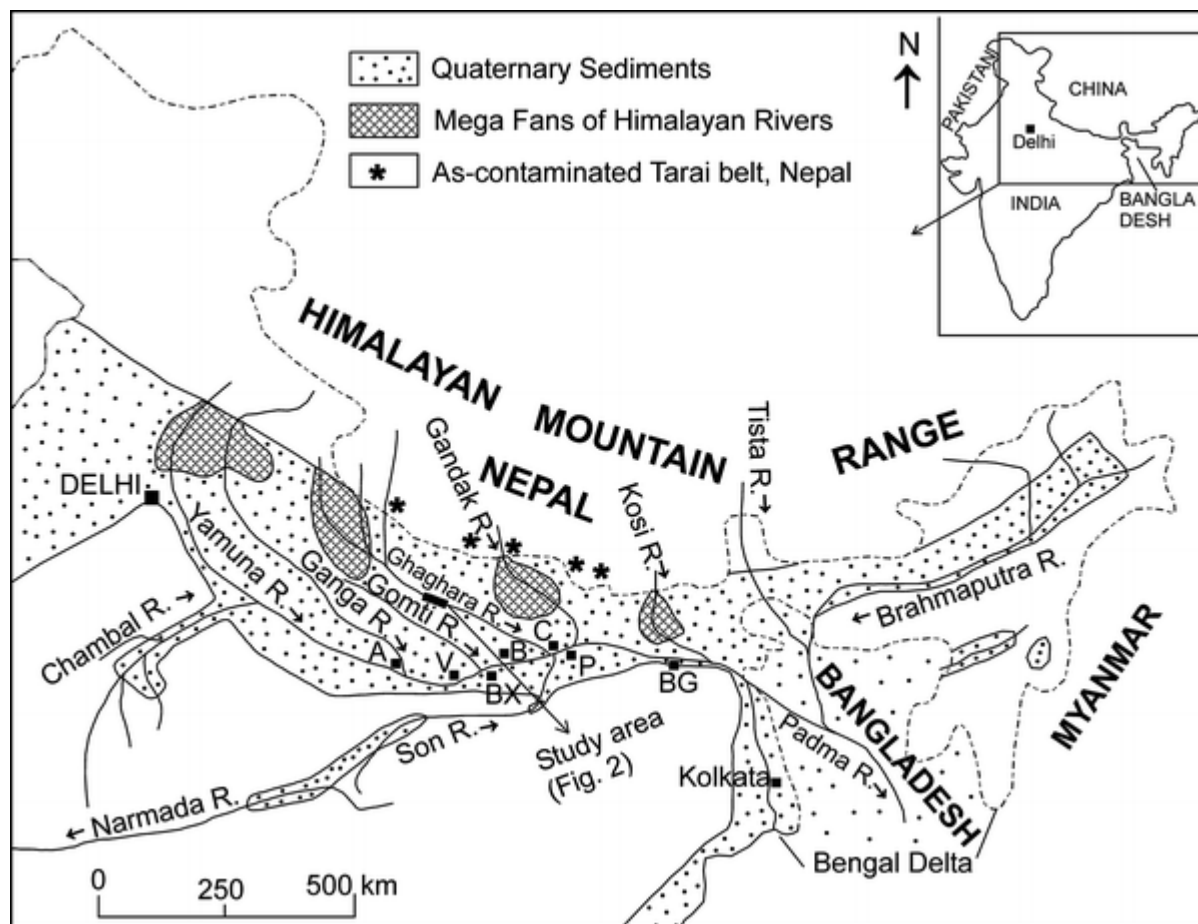
iron (Fe) along with bicarbonate (HCO_3^-) and As indicates reductive dissolution of FeOOH leading enrichment of As in groundwater. Highly reducing

conditions are also indicated by low values of SO_4/Cl ratio (ranges between 0 and 3.57). [1,2]



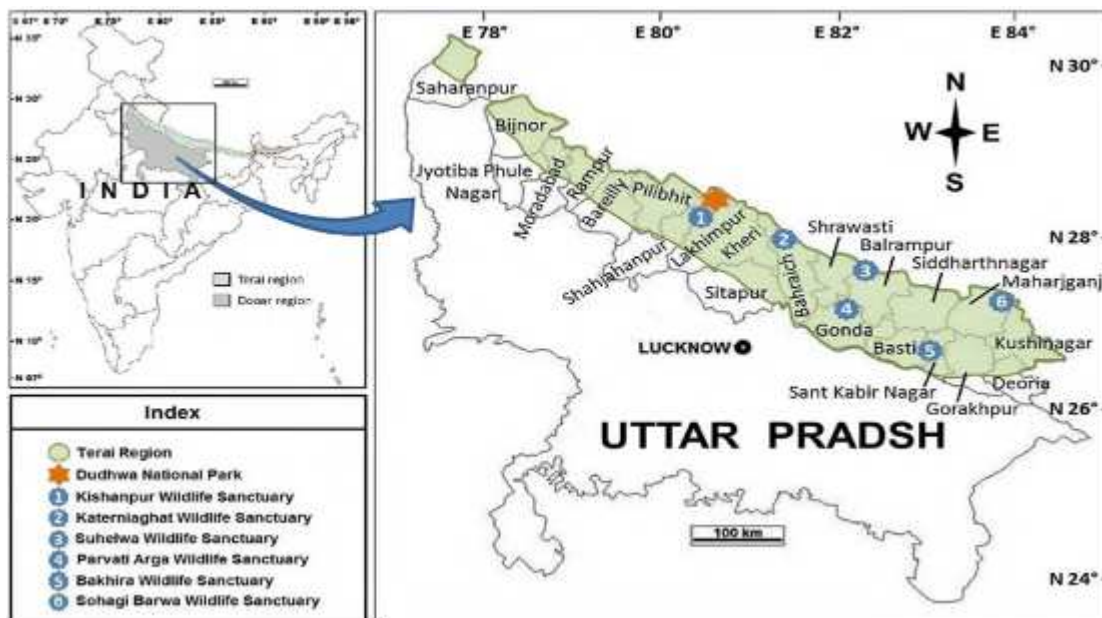
Topography, geology and river dynamics seem to govern the As contamination as depressions/low lying areas shows presence of high As in the aquifers. Nepal, a small land-locked South Asian country, lies between latitude $26^{\circ} 22' \text{ N}$ to $30^{\circ} 27' \text{ N}$ and longitude $80^{\circ} 4' \text{ E}$ to $88^{\circ} 12' \text{ E}$. Elevation ranges from 60 to 8,848 metres, including the highest peak in the world—the Mount Everest. It is bounded on the north by Tibet (China), on the east by Sikkim and West Bengal, on the south by Bihar and Uttar Pradesh, and on the west by Uttar Pradesh of India (Fig.). It has roughly a rectangular shape encompassing an area of 147,181 sq km with an average length of 885 km east to west and an average breadth of 193 km north to south. Topographical diversity provides a wide range of climates, differing according to variations in altitude and location. Likewise, annual rainfall differs with seasonal variations depending on the monsoon cycle. Nepal's landscape is broadly defined by three contrasting physiographic bands, which run in more or less parallel, east to west: lowland terai region in the south; Hill region in the centre and Mountain region in the north [3,4] The terai region, also called the 'grainary' of Nepal, is a low-lying tropical and sub-tropical belt of flat alluvial deposits stretching along the Nepal-India border that comprises 20 districts with an average width of 29 km. It is the northern extension of the Gangetic Plain and has an altitude ranging from 60 to 310 metre above the mean sea-level. Three major rivers—the Kosi, the Narayani (Gandak River), and the Karnali—feed the region. The terai makes up only 23% of the total land area but accommodates 50% of the total population of the country.[5,6]

Late Quaternary stratigraphy and sedimentation in the Middle Ganga Plain (MGP) (Uttar Pradesh–Bihar) have influenced groundwater arsenic contamination. Arsenic contaminated aquifers are pervasive within narrow entrenched channels and flood plains (T_0 -Surface) of fine-grained grey to black coloured argillaceous organic rich Holocene sediments (Newer Alluvium). Contaminated aquifers are often located close to distribution of abandoned or existing channels and swamps.[7,8]



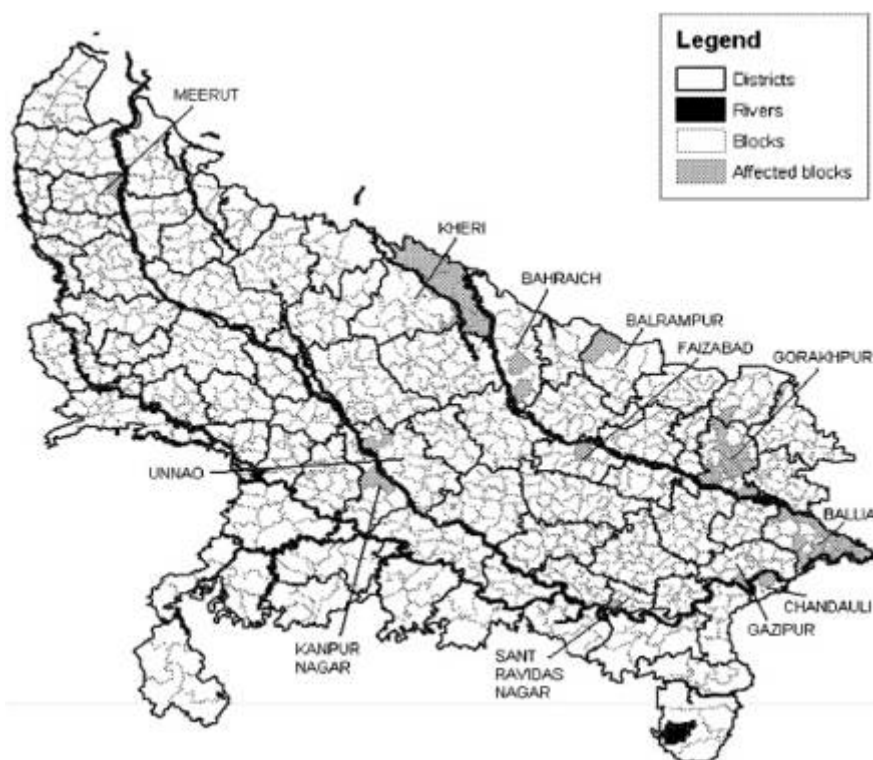
The Pleistocene Older Alluvium upland terraces (T_2 -Surface) made up of oxidized yellowish brown sediments with calcareous and ferruginous concretions and the aquifers within it are free of arsenic contamination. MGP sediments are mainly derived from the Himalaya with minor inputs from the Peninsular India. The potential source of arsenic in MGP is mainly from the Himalaya. The contaminated aquifers in the Terai belt of Nepal are closely comparable in nature and age to those of the MGP. Arsenic was transported from disseminated sources as adsorbed on dispersed phases of hydrated-iron-oxide and later on released to groundwater mainly by reductive dissolution of hydrated-iron-oxide and corresponding oxidation of organic matter in aquifer. Strong reducing nature of groundwater is indicated by high concentration of dissolved iron (11.06 mg/l). Even within the arsenic-affected areas, dugwells are found to be arsenic safe due to oxygenated nature.[9,10]

Exploratory studies carried out by ONGC in collaboration with CGWB in the Gangetic alluvium indicated existence of auto flowing fresh groundwater at more than 1000 m depth. Similarly, free flow of ground water has been reported at deeper depths from some areas like Terai and Sub-Terai belt of Uttar Pradesh. As no energy is required for extraction of ground water from such auto flowing aquifers, development of groundwater from these auto flow zones may be economically viable and eco-friendly. It is scientifically an established fact that the vast water resources of Uttar Pradesh including river systems have developed in the foreland of Himalayas that lap on Vindhya and Granite system of Indian Peninsular region, due to various depositional activities occurred during formation of Ganga basin. The studies revealed that this foreland is in dynamic state due to plate-tectonic activity and ONGC has seismic data of Ganga basin which can reveal the depth to basement as well as the changing dynamics of aquifers at different depths with their spatial and temporal distributions and the geological changes occurring overtime.[11,12]



The Terai or Tarai is a lowland region in northern India and southern Nepal that lies south of the outer foothills of the Himalayas, the Sivalik Hills, and north of the Indo-Gangetic Plain. This lowland belt is characterised by tall grasslands, scrub savannah, sal forests and clay rich swamps. In northern India, the Terai spreads from the Yamuna River eastward across Haryana, Uttarakhand, Uttar Pradesh, Bihar and West Bengal. The Terai is part of the Terai-Duar savanna and grasslands ecoregion. The corresponding lowland region in West Bengal, Bangladesh, Bhutan and Assam in the Brahmaputra River basin is called 'Dooars'. In Nepal, the term is applied to the part of the country situated north of the Indo-Gangetic Plain. Nepal's Terai stretches over 33,998.8 km² (13,127.0 sq mi), about 23.1% of Nepal's land area, and lies at an elevation of between 67 and 300 m (220 and 984 ft). The region comprises more than 50 wetlands. North of the Terai rises the Bhabar, a narrow but continuous belt of forest about 8–12 km (5.0–7.5 mi) wide.[13,14]

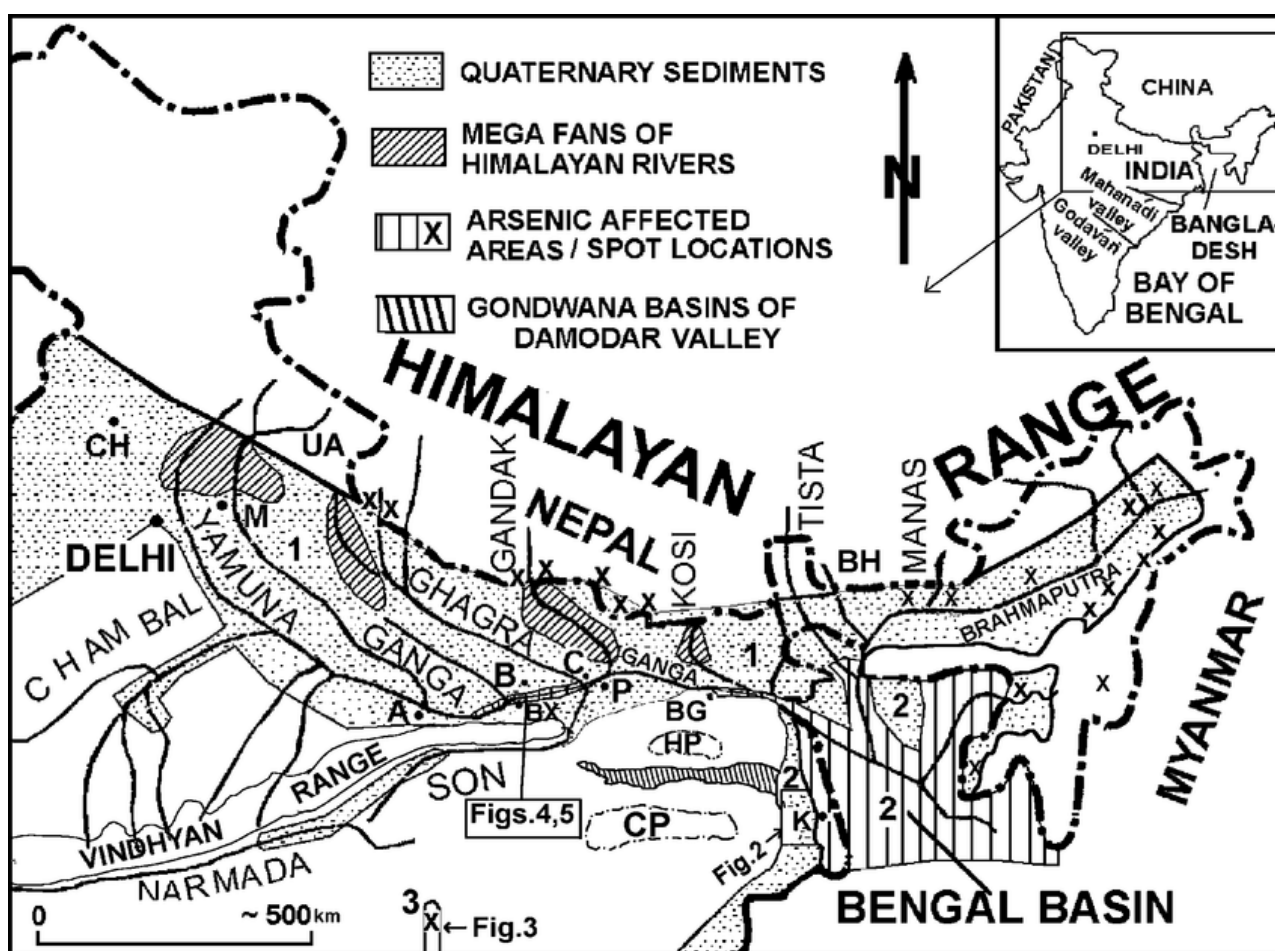
The Terai is crossed by the large perennial Himalayan rivers Yamuna, Ganges, Sarda, Karnali, Narayani and Kosi that have each built alluvial fans covering thousands of square kilometres below their exits from the hills. Medium rivers such as the Rapti rise in the Mahabharat Range. The geological structure of the region consists of old and new alluvium, both of which constitute alluvial deposits of mainly sand, clay, silt, gravels and coarse fragments. The new alluvium is renewed every year by fresh deposits brought down by active streams, which engage themselves in fluvial action. Old alluvium is found rather away from river courses, especially on uplands of the plain where silting is a rare phenomenon.[15]



A large number of small and usually seasonal rivers flow through the Terai, most of which originate in the Sivalik Hills. The soil in the Terai is alluvial and fine to medium textured. Forest cover in the Terai and hill areas has decreased at an annual rate of 1.3% between 1978 and 1979, and 2.3% between 1990 and 1991. With deforestation and cultivation increasing, a permeable mixture of gravel, boulders and sand evolves, which leads to a sinking water table. But where layers consist of clay and fine sediments, the groundwater rises to the surface and heavy sediment is washed out, thus enabling frequent and massive floods during monsoon, such as the 2008 Bihar flood. [16,17]

In India, the Terai extends over the states of Haryana, Uttarakhand, Uttar Pradesh, Bihar and West Bengal. These are mostly the districts of these states that are on the India–Nepal border:

- Haryana: Panchkula district
- Uttarakhand: Haridwar district, Udham Singh Nagar and Nainital districts
- Uttar Pradesh: Pilibhit district, Lakhimpur Kheri district, Bahraich district, Shravasti district, Balrampur district, Siddharthnagar district, Maharajganj district
- Bihar: West Champaran district, East Champaran district, Sitamarhi district, Madhubani district, Supaul district, Araria district, Kishanganj district
- West Bengal: Siliguri subdivision of Darjeeling district,^[15] Jalpaiguri Sadar subdivision of Jalpaiguri district



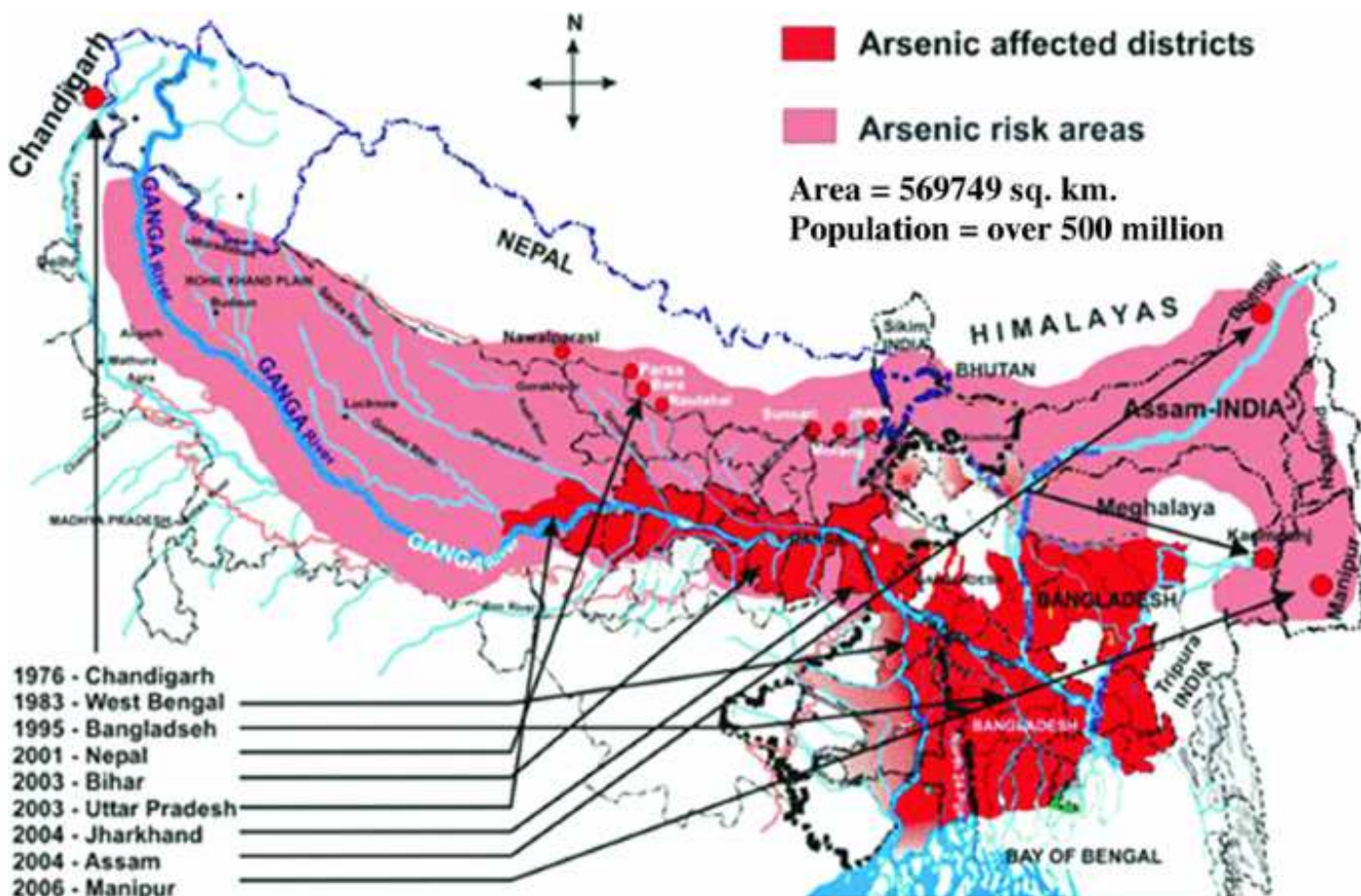
The Inner Terai consists of five elongated valleys located between the Mahabharat and Sivalik ranges. From north-west to south-east these valleys are:

- Surkhet Valley in the Surkhet district, north of the Kailali and Bardiya districts;
- Dang Valley in the Dang Deokhuri district;
- Deukhuri Valley located south of the Dang Valley,
- Chitwan Valley stretching across the Chitwan and Makwanpur districts;
- Kamala Valley, also called Udayapur Valley, in the Udayapur district north of the Siraha and Saptari districts.[18,19]

The Outer Terai begins south of the Sivalik Hills and extends to the Indo-Gangetic Plain. In the Far-Western Region, Nepal it comprises the Kanchanpur and Kailali districts, and in the Mid-Western Region, Nepal Bardiya and Banke districts. Farther east, the Outer Terai comprises the Kapilvastu, Rupandehi, Nawalparasi, Parsa, Bara, Rautahat, Sarlahi, Mahottari, Dhanusa, Siraha, Saptari, Sunsari, Morang and Jhapa districts.

Discussion

Lakhimpur Kheri, also known as mini-Punjab of Uttar Pradesh, is a new flashpoint in India's most populous state that goes to poll early next year. On October 3, eight people were killed in the district in violent clashes. Geographically, Kheri is UP's largest district and contributes 3.38% to the state's agriculture, forestry, and fisheries sector GDP. However, the district fares poor on several social and health indicators. A 2018 study titled 'Arsenic Occurrence in Ground Water and Soil of Uttar Pradesh, India and its Phytotoxic Impact on Crop Plants' shows that the groundwater in Lakhimpur Kheri and Unnao districts of Uttar Pradesh contain arsenic in the range of 23 to 140 microgram per litre ($\mu\text{g/l}$), which was far above the World Health Organization's permissible limit of $10 \mu\text{g/l}$. Microgram per litre is also referred to as parts per billion, or ppb. The BIS (Bureau of Indian Standards) acceptable limit of arsenic in drinking water is also 10 ppb.[20,21]



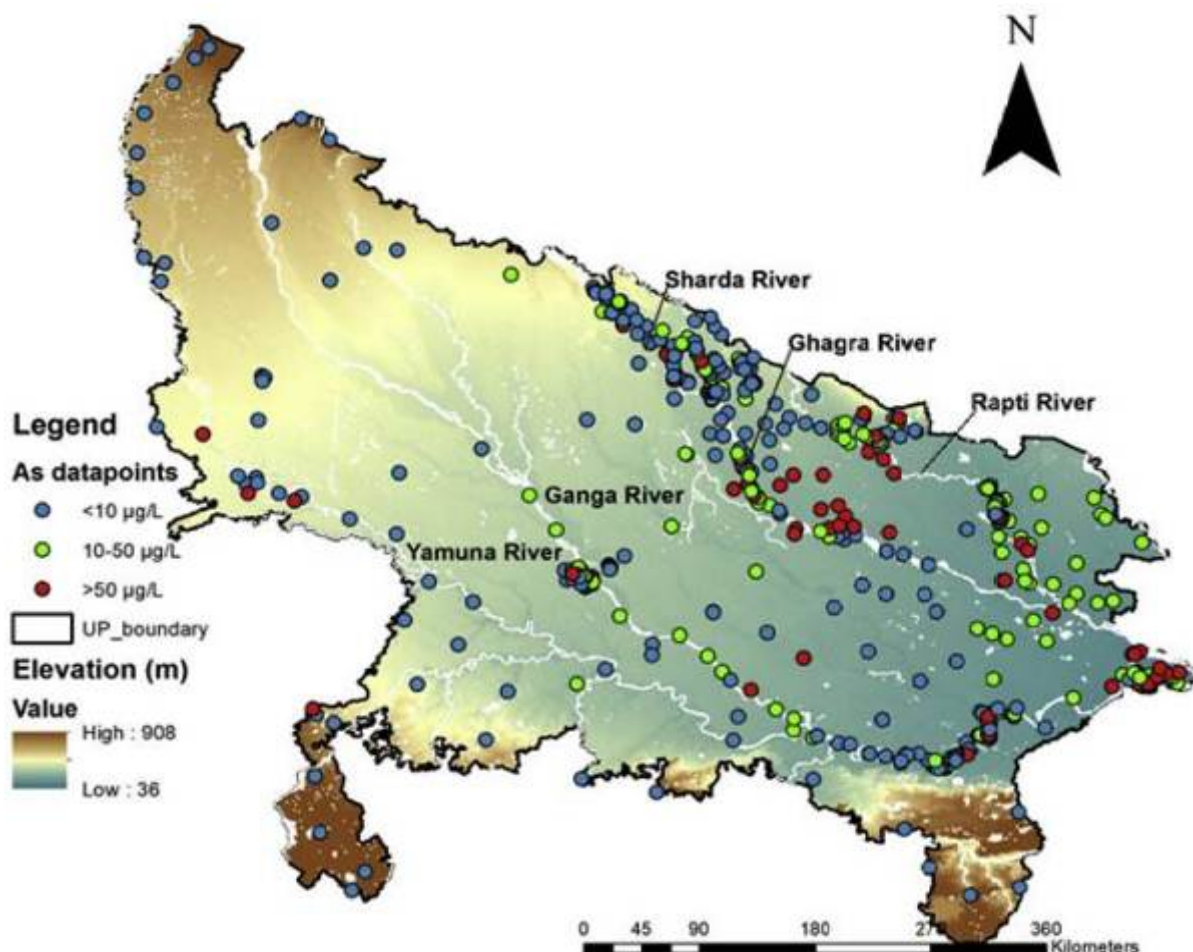
Surprisingly, the 2018 study found that the soil samples in the district contained almost 40-45 times more arsenic than that found in the groundwater samples of the same site.

This is worrisome as a large chunk of rural population in the district depends on groundwater to meet its drinking water needs and also irrigated the crops.[22]

Another study, titled 'Predicting groundwater arsenic contamination: Regions at risk in highest populated state of India', published in 2019 found that as many as 23.48 million people in rural areas of Uttar Pradesh are exposed to high levels of arsenic in groundwater. Lakhimpur Kheri is among the worst affected districts, noted the 2019 study.

"The blocks [in Lakhimpur Kheri] such as Pallia, Nighasan and Ishanagar are having arsenic poisoning. The water in these areas of the district is high in arsenic. Arsenic can cause a number of health problems such as skin, lung, liver cancer. It can cause cardiac arrest too," Chander Kumar Singh, author of the report, told Gaon Connection. Singh is working as an associate professor, Department of Energy and Environment, TERI School of Advanced Studies.

This is not all. "Mitauli block in the district has a fluoride problem. Fluoride is problematic because it causes bone related problems. The kids might suffer dental or skeletal fluorosis," the author and researcher added.[23,24]



Arsenic concentration in groundwater of Uttar Pradesh shown by red, green, and blue circles. This problem of poor groundwater quality has also been recorded by the Central Ground Water Board (CGWB), under the Ministry of Jal Shakti, Department of Water Resources. As per the CGWB, Nighasan in Lakhimpur Kheri is among the most affected blocks by arsenic content.

The 2012-13 report titled 'Ground Water Scenario of Lakhimpur Kheri district, UP' shows that the arsenic concentration of more than 10 ppb has been recorded in Palia, Nighasan, Ramia Bihar, Dhaurahara and Issanagar blocks of Lakhimpur Kheri.

Phosphate is nil in ground water of the area. "It is observed that 20 per cent of the samples analysed have high nitrate, which is most likely due to the use of fertilisers for agriculture and other improper waste disposal," read the CGWB's 2012-13 report.[25,26]

In a study, Of the 20 districts in the terai region where 18,635 tube wells were examined and, on average, arsenic contamination was detected in 7.4% of the tubewells as reported previously (4), comparatively six higher arsenic-contaminated districts were included in this study. The districts are: Nawalparasi, Bara, Parsa, Rautahat, Rupandehi, and Kapilvastu. Investigation of health effects due to chronic toxicity of arsenic was also carried out at village level on a total coverage basis in purposively-selected four vill-

ages, namely Goini, Thulo Kunwar, Sano Kunwar, and Patkhoul of Nawalparasi district, where water contamination was higher. Water samples from all 217 tubewells of all four study villages with a total population of 2,339, living in 358 households, were examined for arsenic contamination. Arsenicosis cases were identified by observing skin manifestations on the body and by organizing medical camps in different places of the studied villages. The day before organizing each medical camp, all the households of that particular area of the village were informed, and volunteers requested them to visit the camp the next day for clinical examinations. All the villagers visiting the medical camps were examined for arsenicosis, and there was no refusal. One of the authors carried out clinical examinations and diagnosis of arsenicosis. Of the total population ($n=2,339$), 1,864 (79.7%) were included in the study. [27,28] The villagers who were not exposed to arsenic-contaminated water and those who were not available at the time of the study period were not included in the study. Following the measurement of arsenic contamination level in tubewell water, targeted subjects were clinically examined to identify arsenicosis cases. Necessary information relating to arsenicosis was also collected. Any person showing pigmentary changes, such as hyperpigmentation (melanosis) or hypopigmentation (leucomelanosis) and/or hyperkeratinization (keratosis), with or without

other manifestations of chronic arsenic toxicity and having a history of arsenic exposure through water for more than six months, was diagnosed as an 'arsenicosis' case. The patients were categorized into three different stages based on clinical manifestations (9) observed on the body, which were as follows: First stage (mild stage): Melanosis, keratosis (mild), conjunctivitis (conjunctival congestion), and bronchitis. Second stage (moderate stage): Depigmentation (leucomelanosis), keratosis (moderate to severe), oedema (non-pitting), peripheral neuropathy, hepatopathy, and nephropathy (early stage). Third stage (severe stage): Hepatopathy and nephropathy (late stage), gangrene of the limbs, pre-cancerous skin lesions, and cancer. Arsenic concentrations in water samples were measured by atomic absorption spectrophotometer (AAS) equipped with a continuous hydride generator in the research laboratory of the Environment and Public Health Organization (ENPHO), Kathmandu, Nepal. Level of arsenic in water samples was determined, following pre-reduction with 5% (w/v) potassium iodide (KI) and 5% (w/v) ascorbic acid in 10% (v/v) HCl. The accuracy of determination of arsenic in water was ensured by measuring standard reference material (SRM), NIST SRM 1640 with arsenic of 26.67 ± 0.41 $\mu\text{g}/\text{kg}$. The detection limit (DL) of the HG-AAS was 3 $\mu\text{g}/\text{L}$ for arsenic in water. The variables, which were common, included geographical distribution, demographic characteristics, types of skin lesion, and stages of arsenicosis cases. Data of these common variables were summarized and analyzed to find out the prevalence of arsenicosis in relation to different demographic characteristics. Surveys for the determination of arsenic contamination in the study areas were organized by the Nepal Red Cross Society (NRCS), Department of Water Supply and Sanitation (DWSS), and Rural Water Supply and Sanitation Support Program (RWSSSP) during 2001-2004. The community-based health surveys were, however, carried out by the Department of Human Ecology, School of International Health of Tokyo University, Japan, Department of Dermatology of University of Miyazaki, Japan, and Department of Occupational and Environmental Health of National Institute of Preventive and Social Medicine (NIPSOM), Bangladesh, in collaboration and coordination with ENPHO and other local institutions during 2002-2004.

Results

An about 100 km² of the middle Ganga plain in Uttar Pradesh, experiencing intensive groundwater extraction. In order to recognize the arsenic contamination zones of the Varanasi environs, sixty eight groundwater samples have been collected and

analyzed for major ions, iron and arsenic. Twenty one sediment samples in the four boreholes were also collected to deduce the source of arsenic in the groundwater. The preliminary survey reports for the first time indicates that part of rural and urban population of Varanasi environs are drinking and using for irrigation arsenic contaminated water mostly from hand tube wells (<70 m). The study area is a part of middle Ganga plain which comprises of Quaternary alluvium consists of an alternating succession of clay, clayey silt and sand deposits. The high arsenic content in groundwater samples of the study area indicates that 14% of the samples are exceeding the 10 $\mu\text{g}/\text{l}$ and 5% of the samples are exceeding 50 $\mu\text{g}/\text{l}$. The high arsenic concentration is found in the villages such as Bahadurpur, Madhiya, Bhojpur, Ratanpur, Semra, Jalilpur, Kateswar, Bhakhara and Kodupur (eastern side of Ganga River in Varanasi), situated within the newer alluvium deposited during middle Holocene to Recent. The older alluvial aquifers situated in the western side of the Ganga River are arsenic safe (maximum As concentration of 9 $\mu\text{g}/\text{l}$) though the borehole sediments shows high arsenic (mean 5.2 mg/kg) and iron content (529 mg/kg) in shallow and medium depths. This may be due to lack of reducing conditions (i.e organic content) for releasing arsenic into the groundwater. Rainfall infiltration, organic matter from recently accumulated biomass from flood prone belt in the newer alluvium plays a critical role in releasing arsenic and iron present in sediments. The main mechanism for the release of As into groundwater in the Holocene sandy aquifer sediments of Varanasi environs may be due to the reductive dissolution of Fe oxyhydroxide present as coatings on sand grains as well as altered mica content. The high societal problems of this study will help to mitigate the severity of arsenic contamination by providing alternate drinking water resources to the people in middle Ganga plain and to arrange permanent arsenic safe drinking water source by the authorities.[29]

An evaluation of three arsenic removal technologies, the Three-Gagri System, the Jerry Can System, and the Arsenic Treatment Unit (ATU), was conducted. In addition, a comparison of two arsenic field tests, EM Quant® test strips and Arsenic Check™, was made. Finally, water source samples from Parasi, Nepal were analyzed to determine their arsenic content. The three arsenic removal technologies were evaluated for their effectiveness and appropriateness. Effectiveness is the measure of a technology's ability to remove arsenic to or below 10 $\mu\text{g}/\text{L}$ (micrograms per liter or parts per billion), the guideline set by the World Health Organization (WHO) for arsenic in drinking water. The WHO set this guideline because drinking

arsenic contaminated water above this limit can cause adverse health effects. Appropriate technologies are easy to assemble, simple to use in rural settings, and made with locally available, inexpensive materials. The Three-Gagri System was found to be both effective and appropriate, but the clogging problem and the question of whether or not this system promotes microbial growth in the water need to be addressed. The Jerry Can System was found to be ineffective and inappropriate with its current design. The ATU, while very effective, is inappropriate for implementation in Nepal due to its high cost. This study has also concluded that the Arsenic Check™ test is a safer and more accurate than the EM Quant® test strips. Finally, this study found that groundwater from some water sources in Parasi Nepal is contaminated with arsenic above the WHO guideline.

Conclusions

The ATU (Arsenic Treatment Unit) system was tested over ten consecutive days, and one influent and one effluent sample from each day were analyzed. Since we installed this system the day before we left Parasi, the author collected only the samples for the first day. For the nine days following, representatives from the Nepal Red Cross Society, who had been instructed on the proper method of sample collection, took samples and express mailed them to MIT so that the author could analyze them. A pump was installed for this system to replace the gravity pump. It can be seen that the influent concentrations are quite variable and this is due to the stronger suction applied to the aquifer by the new pump. The stronger suction may have allowed more particles or colloids, to which arsenic may have sorbed, to be introduced to the well; this effect was probably exaggerated by the fact that the filter in the pump was missing.

Besides being effective, it is also an appropriate technology. The system has a simple design and was easily constructed. It has a relatively low cost of about US\$10.50. The materials necessary to assemble the system were obtained locally, with the exception of the iron filings. As mentioned, it is expected that zero valent iron filings, or turnings, could be obtained at a local foundry but this was not confirmed during the field site visit to Nepal. It was shown that iron nails could be obtained locally, replace the filings, and successfully remove arsenic below the WHO guideline; although this approach was also shown to result in a very low flowrate. There are two drawbacks to this system. One of the drawbacks is that it has been known to become easily clogged. In Parasi, it was observed to take longer and longer for each batch of water to filter through. If the system becomes too clogged, it will not be able to provide an

adequate supply of water to its users. Also, it is not known if this system encourages bacterial growth due to the water's long residence time. It has been suggested that wood charcoal be added to the middle gagri for the purpose of removing organic impurities¹⁰¹, but wood charcoal was not found in Parasi so it was left out of the design.^[28]

The results of the Three-Gagri System compared well with results found in recent literature about a study on the Three-Kalshi System but there were some differences. The Three-Gagri System proved to be more effective at removing arsenic than the results from the Three-Kalshi. While the Three-Gagri System removed arsenic to an average concentration of 4 ug/L (with an average influent concentration of 215 ug/L) in field tests in Parasi, Nepal in January 2001, the Three-Kalshi System had an average removal of 17 ug/L (with an average influent concentration of 90 ug/L)¹⁰². Since the testing for the Three-Kalshi System was done in Bangladesh, where the drinking water standard for arsenic in drinking water is set at 50 ug/L, the results for that system were satisfactory. Nepal, on the other hand, does not have drinking water quality standards, so we were aiming to remove arsenic to the WHO guideline. Also, there were differing results for flowrate as the Three-Gagri System flowed at 4L/hour and the Three-Kalshi System is reported to have flowed at an average of 5L/hour

There are differences associated with building the system out of the aluminum gagrins as opposed to the Bangladeshi kalshis, also known as kolshis. The kalshis used in the above study were fired, unglazed clay pitchers. These cost less than the aluminum gagrins, US\$5-6 for three kalshis as opposed to US\$10.50 for three gagrins. The clay also allows for continuous diffusion of air and water vapor through the porous ceramic kalshi. This provides a more oxidizing environment to allow for the complete conversion of zero valent iron to hydrous ferric oxide, the active component for arsenic removal in the Three-Gagri/Kalshi System.^[29]

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