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# Groundwater Arsenic Contamination and Associated Health Hazards in Kangra District of Himachal Pradesh, India

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## ABSTRACT

Groundwater arsenic contamination is a global problem. Many states of India are extensively polluted with arsenic, while a few have safe groundwater. Himachal Pradesh is still considered safe from groundwater arsenic contamination. Assessment of water quality and arsenic contamination in groundwater and its associated health hazards in the people of Kangra district. 300 samples were collected from all 17 blocks of Kangra and analyzed for arsenic, TDS, pH, and Depth of water source. Arsenic was analyzed through Merck's field test kits. TDS and pH were analyzed using standards protocol. A questionnaire-based health survey was carried out for health assessment. It was observed that the arsenic levels were above 10 ppb in 4.66% of samples. Khundian tehsil had a mean arsenic level of 25.83 ± 30.59 ppb, while Jawalamukhi had 10.38 ± 13.14 ppb. It was also observed that deep water sources have higher arsenic levels than shallow sources. TDS levels were also above the WHO limit in 24% of samples, and TDS was higher in deeper groundwater than in shallow sources. Groundwater pH levels were 5.78 in Bhawarna and 9.4 in Baijnath. 7.66% were reported with hypertension 3.32% with G.I. disorders, 3.66% with diabetes, 2.33% are showing skin diseases, and almost 7.66% are showing stone formation in the kidney and gall bladder. Kangra had groundwater arsenic contamination in 4.52% of samples. Cases of hypertension, diabetes, and stone formation were more than seven percent in people with chronic arsenic exposure.

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## INTRODUCTION

Arsenic (As) contamination in drinking water affects more than 107 countries worldwide. In Asia, almost 33 countries face this problem. Bangladesh, India, China, Nepal, Cambodia, Vietnam, Myanmar, Laos, and Indonesia are severely affected (Shaji et al., 2021). Significant groundwater arsenic contamination was reported in West Bengal, Assam, Bihar, Chhattisgarh, Haryana, Jharkhand, Karnataka, Punjab, and Uttar Pradesh in India (CGWB, 2023).

Arsenic exists in different metalloid forms like Arsenopyrite (FeAsS), Orpiment ($As_2S_3$), Niccolite (NiAs), and Realgar ($As_4S_4$). Arsenic is associated with hydroxide, sulfides, manganese, and iron within sedimentary rocks and soil (Rehman & Naranmandura, 2012). Arsenopyrite is a common mineral that is found in the deeper layer of the earth's crust (Smedley & Kinniburgh, 2002). Fertilizers, mine tailing, herbicides, and insecticides are some anthropogenic sources that play a vital role in groundwater contamination. Ghosh & Singh, (2009) have reported

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some common mechanisms for arsenic mobilization to groundwater, including oxidation of arsenic-bearing pyrite minerals, dissolution of arsenic-rich iron oxy-hydroxides (FeOOH) due to the onset of reducing conditions in the subsurface, and competitive exchange with phosphate ($H_2PO_4^-$) ions, due to which arsenic migrate into aquifers from subsurface soil contaminated with fertilizers. According to WHO (Gomez-Caminero et al., 2001), the permissible limit of arsenic in groundwater is 10 ppb. According to Lan et al., (2011), more than 30 million people drink arsenic-contaminated water above 50 ppb in Bangladesh and India. Arsenic interacts with oxygen, sulfur, and chlorine to generate inorganic compounds. Arsenic exists in the environment in arsenite and arsenate inorganic forms and interacts with oxygen, sulfur, and chloride. Trivalent inorganic forms are more toxic than other organic forms (Singh et al., 2013). Arsenite is 60–80 times more hazardous because its interaction with the thiol (-SH) portion of proteins is more thermodynamically stable than arsenate (Villaescusa & Bollinger, 2008). Arsenic reacts with sulfhydryl groups of cysteine residues, thereby damaging general protein metabolism (Shankar & Shanker, 2014).

According to the U.S. Environmental Protection Agency (EPA), inorganic arsenic is a "human carcinogen" with a maximum contaminated level of 10 ppb (Basu et al., 2015). Inorganic arsenic levels greater than 50 ppb can cause multi-site cancer in humans, having risk factors as high as 1% (Liu et al., 2013). Arsenic toxicity causes the inactivation of up to 200 enzymes involved in DNA repair and replication and other energy mechanisms in cells. It serves as a substitution for phosphate in high-energy molecules like ATP (Ratnaike, 2003). The negative health consequences of arsenic depend on two factors: the duration of exposure (Mukherjee et al., 2006). Acute arsenic poisoning can cause severe nausea and vomiting, muscle pain, weakness, abdominal pain, diarrhea, convulsions, coma, and death.

In contrast, the systems of the skin, liver, lungs, and blood are affected by chronic arsenic poisoning, which is more complicated and harder to diagnose than acute toxicity (Flora, 2020). The earliest sign of arsenic toxicity is typically shown in the skin, which is thought to be the most vulnerable organ (Rahman et al., 2009). Keratosis, melanosis (hyper-pigmentation), and leukomelanosis (hypo-pigmentation) are arsenic-associated clinical manifestations that result from chronic arsenic exposure (Figure 1) (Shrivastava et al., 2015).

Only a certain percentage (17–66%) of people exposed to arsenic develop these cutaneous

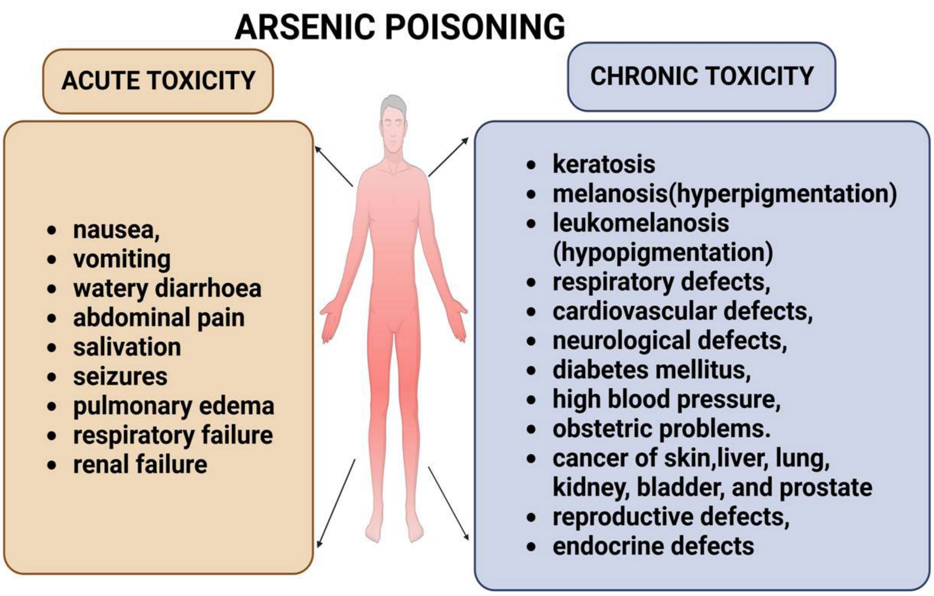



**Fig. 1.** Health effects of acute and chronic arsenic toxicity

illnesses (Huang et al., 2019). Some other health manifestations include diseases of the respiratory system, cardiovascular system, neurological effects, diabetes mellitus, high blood pressure, and obstetric problems (Abdul et al., 2015). The endocrine disruptor potential of arsenic could alter the gene expression of the steroidogenic pathway, leading to steroidogenic dysfunction (Reddy et al., 2011). Arsenic competes with retinoic acid receptors, thyroid hormone receptors, and estrogen receptors, thereby affecting hormonal regulations (Rahman et al., 2009). It is responsible for the generation of reactive oxygen species (ROS), which causes an imbalance between endogenous antioxidants and pro-oxidants, triggers signaling pathways, leads a cell towards apoptosis (Flora, 2011), and overall reduces the antioxidant defense system mechanism of the body (Manna et al., 2008). Arsenic is a known carcinogen that may induce cancer by both gastrointestinal and respiratory exposure (Centeno et al., 2006).

Since arsenic is associated with the tectonic movement of the Himalayan plateau (Guillot & Charlet, 2007), it is observed to be very high in the Ganga-Meghna-Brahmaputra region, due to which arsenic levels are found in these regions. The largest and most arsenic-rich area in the world, the Himalayan orogenic belt, was created as a result of the Eastern Tethys Sea's closing during the Cenozoic period when it took the shape of the Gulf of Eastern Pangaea (Stanger, 2005). Combining geochemical and biological processes results in arsenic-enriched groundwater linked to crustal evolution (Mukherjee et al., 2014). The Siwalik Group is regarded to be the arsenic's probable reservoir and serves as the direct source of the Himalayan deposits (Stanger, 2005). Because Himachal is so close to this belt, the status of the arsenic mobilization must be studied in detail.

The present study was designed to find groundwater quality assessment with particular emphasis on arsenic contamination in the Kangra District of Himachal Pradesh and its associated health hazards.

## MATERIALS AND METHODS

*Sampling site*

Kangra district is in the western region of the Indian state of Himachal Pradesh. The district has a geographical area of 5739 km$^2$, which accounts for 10.31 percent of the total land area of Himachal Pradesh. The district's geographic coordinates are 31°21' to 32°59' N latitude and 75°47'55" to 77° 45' E longitude (Ganguly et al., 2015). (Figure - 2)

All 17 blocks of district Kangra of Himachal Pradesh were chosen for this study. The samples were taken from a distance of 2 km between two sample sites. A grid on a map was prepared for sample collection and ensuring proper distance. The GPS was recorded for every sample, and sample collection sites were located through an Arc GIS map plotted for pH, TDS, and arsenic levels. All samples were collected from September 2021 to December 2021.

*Sample size*

Griding was done, and points were spotted at a 2 cm distance using a ruler on a map of Kangra. Sampling was done from every block at a uniform distance except Bara Bhangal, as people from that region use surface water for their daily needs. Three hundred water samples were collected from 142 hand pumps, 148 electrically powered pumps, one tube well, and nine taps from 15 different blocks of Kangra. A total of 300 samples were collected for this study.

*Sample collection*

50 mL reagent bottles were used to take the groundwater sample, and each bottle was thoroughly washed and rinsed with distilled water. Each hand pump and the electrically powered pump were purged for 10-15 min to ensure that fresh aquifer water was collected. Each sample was tested for arsenic level in the field using FTK (Merck KGaA, Germany, catalog no. 1.17927.0001, measuring range 0.005 - 0.010 - 0.025 - 0.05 - 0.10 - 0.25 - 0.50 mg/L)

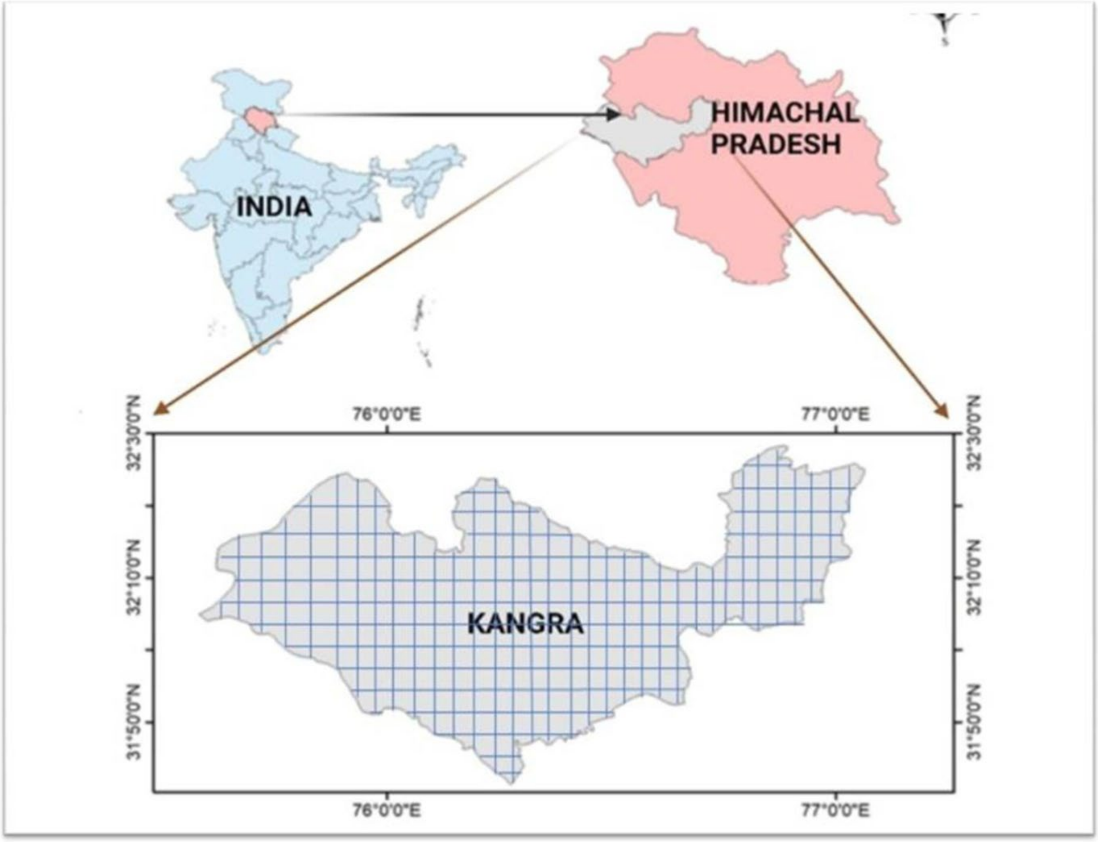



**Fig. 2.** Study area of District Kangra of Himachal Pradesh used for groundwater sample collection and questionnaire-based survey

arsenic. Physical parameters like pH and TDS were analyzed using digital pH and TDS meters (OAKION). Before determining the sample's pH, the pH meter was calibrated using buffer solutions with pH values of 4.0, 7.0, and 9.2. A questionnaire-based survey was conducted to find out the Depth of the water source and the socioeconomic and health status of people.

*Mapping of water sources and quality parameters through geographic information system*

Geographic Information System (GIS) consists of spatial datasets and data collection, management, and analysis software. In our research, ArcGIS software (10.1) was utilized. The geographical coordinates of the sampling site were noted using E dip Ahmet Taskin UTM location and GPS camera version 2.3 mobile applications.

*Statistical analysis*

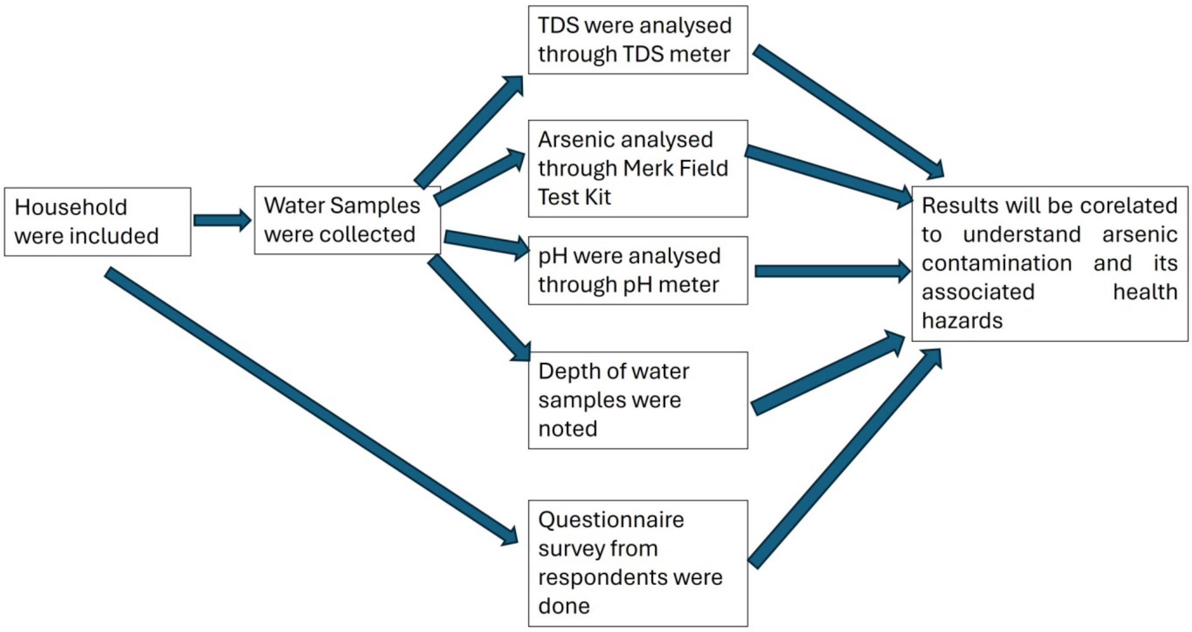
Graph Pad Prism version 5.03 was used for the statistical analysis. T-test and correlation analysis were applied to the collected data.

## RESULTS AND DISCUSSIONS

In District Kangra, a total of 300 samples were collected from all 16 blocks and tested for arsenic, TDS, Depth, and pH levels, and the results were analyzed (Table -1) using a graph pad prism and plotted on Arc GIS (Figure - 3 and Figure - 4).

*Total dissolved solids of groundwater samples of Kangra*

Total dissolved solids (TDS) levels in water samples of the Kangra district ranged from 30




**Flow diagram showing water sample collection and analysis, a questionnaire survey was also conducted**

**Table 1.** Showing Arsenic, Ph, TDS and depth of groundwater in the studied samples

| Parameters | No. of samples | Max. value | Min. value | Mean | Standard deviation |
|---|---|---|---|---|---|
| Arsenic (ppb) | 300 | 100 | 0 | 6.783 | 8.727 |
| Ph | 300 | 9.400 | 5.780 | 7.243 | 0.6450 |
| TDS (ppm) | 300 | 1100 | 30.00 | 333.9 | 13.32 |
| Depth (feet) | 300 | 440 | 20 | 180.6 | 82.67 |

**Table 2.** TDS level in collected samples from district Kangra

| S No. | TDS range | % of sample |
|---|---|---|
| 1. | < 300 ppm | 52% |
| 2. | 300-600 ppm | 32% |
| 3. | 600-900 ppm | 14% |
| 4. | 900-1200 ppm | 2% |

ppm to 1100 ppm. The average TDS in the Kangra district was 333.9 ± 230.7 ppm. Harchakian tehsil has the highest mean value of TDS (710 ± 160.90 ppm), followed by Jawalamukhi (647.7 ± 218.5 ppm) and Dehra (616 ± 211.8 ppm) (Figure-3A). Palampur tehsil had the lowest mean (89.86 ± 25.38 ppm), followed by Dharamshala (137.5 ± 61.33 ppm) and Nagrota Bagwan (168.8 ± 76.56 ppm). TDS levels exceeded the WHO-acceptable drinking limit in 24% of samples (Table - 2).

*pH of groundwater samples of Kangra*

The pH of district Kangra was between 9.4 and 5.78. The mean pH in district Kangra was 7.243± 0.6450. In contrast, the maximum mean value of pH was observed in Nagrota Surian tehsil at 7.967 ± 0.472, followed by Chadiar 7.72 ± 0.164 and Harchakian 7.667 ± 0.533 (Figure 3B) and the minimum mean pH value was observed in Palampur tehsil 6.266 ± 0.3409, followed

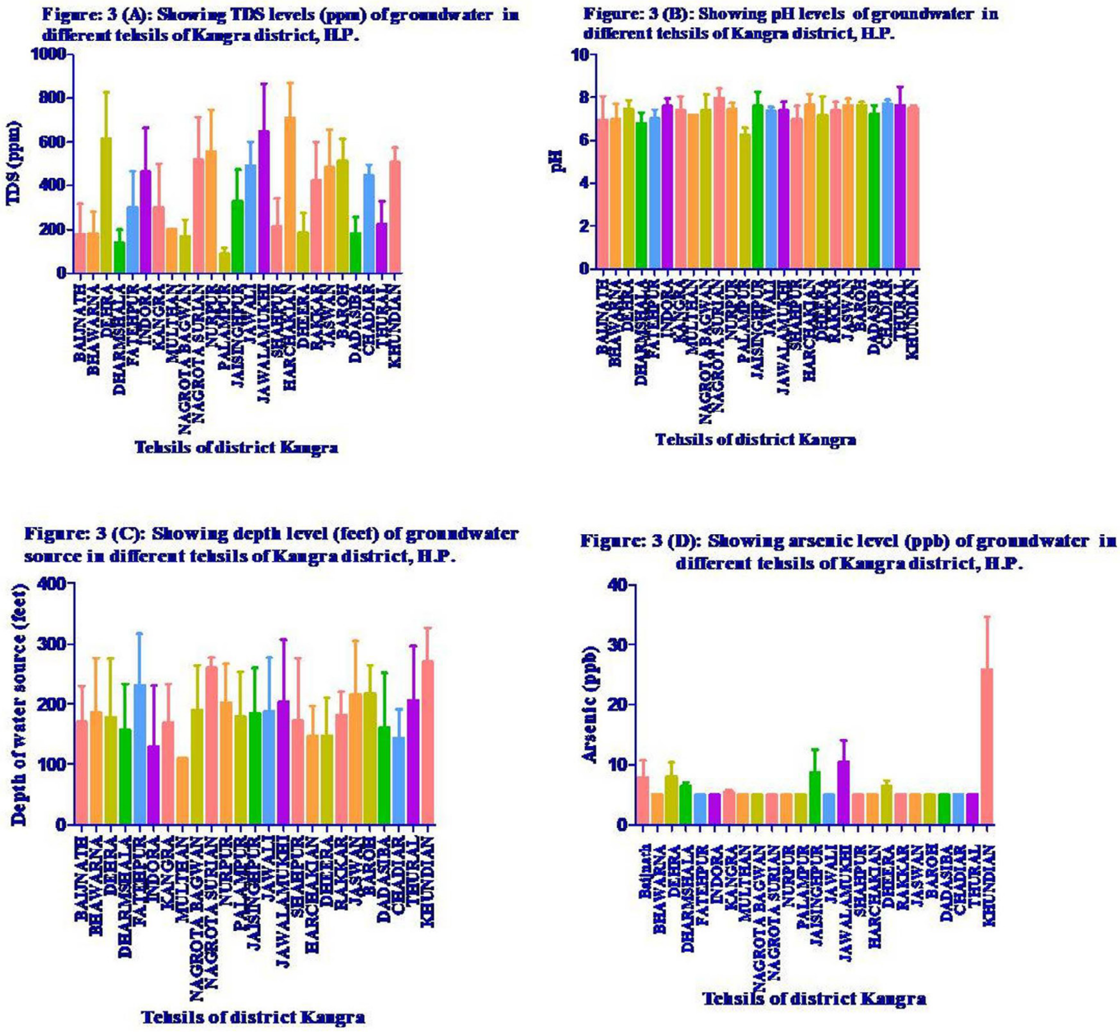



**Fig. 3.** Site-wise variation in TDS (ppm), pH, Depth (feet), and arsenic (ppb) levels in the groundwater of district Kangra. 3(A): Site-wise variation in TDS (ppm) showing Harchakian and Jawalamukhi having the highest mean value of TDS, whereas Palampur and Dharamshala have the lowest mean values. 3(B): Site-wise variation in pH showing Nagrota Surian, Chadiar, and Harchakian having the highest mean value of pH, whereas Palampur and Dharamshala have the lowest mean values. 3(C): Site-wise variation in Depth (feet) where the highest mean Depth for the well was found in Khundian and Nagrota Surian, whereas Multhan and lowest was found in Indore. 3(D): Site-wise variation in arsenic (ppb) showing Khundian, Jawalamukhi, and Jaisinghpur having the highest mean value of arsenic

by Dharamshala 6.809 ± 0.4967 and Baijnath 6.957 ±1.121. 2.66% of samples had a pH above the desirable drinking limit, and 13.33% had a pH below the desirable drinking limit.

*Depth of groundwater sources*

The Depth of groundwater sources in district Kangra was 20 to 440 feet. The mean depth of the groundwater source in district Kangra was 180.6 ± 82.67 feet. The maximum mean Depth was observed at Khundian tehsil at 270.0 ± 56.57 feet (Figure - 3C), followed by Nagrota Surian at 260.0 ± 17.32 feet and Fatehpur at 232.2 ± 84.23 feet, and the minimum mean was observed in Multhan tehsil 110.0 feet followed by Indora 129.5 ± 101.2 feet and Chadiar 144.0 ± 47.75 feet.

*Arsenic level in water samples of Kangra*

The observed range of arsenic was 0 to 100 ppb, whereas the mean arsenic in district Kangra was 6.783± 8.727 ppb. The highest level of arsenic was 100 ppb found in tehsil Khundian of district Kangra (Figure - 3D). The maximum mean value of arsenic was observed in Khundian Tehsil at 25.83 ± 30.59 ppb, followed by Jawalamukhi Tehsil at 10.38 ± 13.14 ppb. 4.66% of samples contain arsenic above the acceptable limit (Table - 3).

Arsenic, TDS, Depth, and pH results were plotted on a map using Arc GIS software (Figure - 4). The Depth of sources of the water sample and pH of the water sample is shown in Figure - 5.

**Table 3.** Arsenic contamination level in samples collected from Kangra district

| S. no. | Range in ppb | % of samples |
|---|---|---|
| 1. | 0-10 | 95.33% |
| 2. | 11-50 | 4.33% |
| 3. | 51-100 | 0.33% |
| 4. | Above 100 | -- |

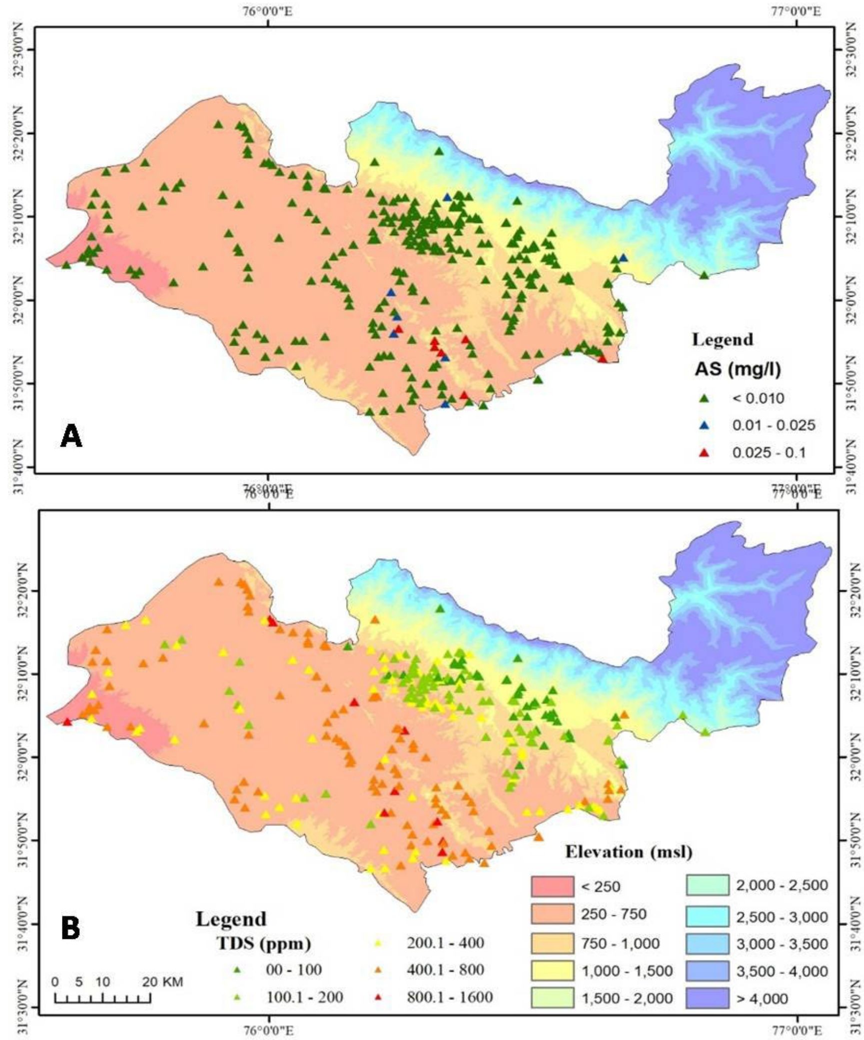



**Fig. 4.** Maps showing details about levels of arsenic (mg/L) and TDS (ppm) levels in the water sample collected from district Kangra. 4(A): The map shows details about levels of arsenic (mg/L) (1mg/L=1000ppb) of the water sample and the location where such findings were detected. The high levels of arsenic were found at an elevation of 250-750 msl from the Khundian and Jawalaji tehsils. 4(B): The map details TDS (ppm) levels in the water sample and the location where such findings were detected. Maximum samples containing TDS levels ranging between 400-800 ppm. Maximum TDS recorded in Jawalamukhi and Dehra tehsils. The TDS levels of water samples exceeding the permissible limit were found at an elevation of 250-750 msl

*Health outcomes*

The questionnaire-based survey was conducted to relate health symptoms and arsenic contamination status. Out of a study of 300 people, 25.66% were suffering from some disease (Table - 4). 7.66% were suffering from hypertension, 7.33% from diabetes, 5.33% from renal stones, 2.33% from gall bladder stones, and the rest were suffering from skin-related problems, gastric, and thyroid problems.

*Correlation analysis*

The correlation was applied between mean values of arsenic, pH, TDS, and Depth. The correlation coefficient between arsenic and pH was found to be 0.1674, with a p-value of 0.0036. The correlation coefficient between arsenic and TDS was 0.1237, with a p-value of 0.0322. The correlation coefficient between pH and TDS was 0.4547, with a p-value of <0.0001.

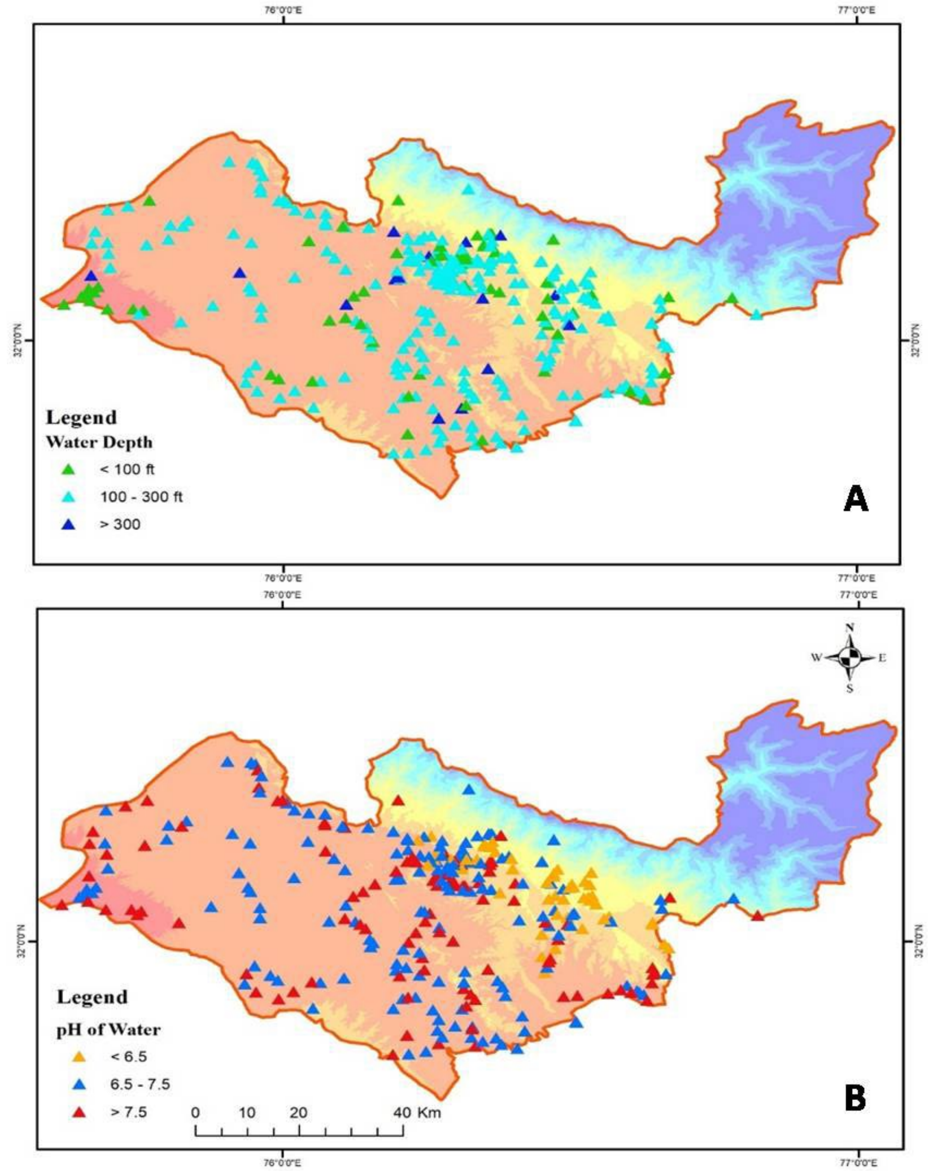



**Fig. 5.** Maps showing details about Depth of the water source and pH levels in the water sample collected from district Kangra. 5(A): The map details the Depth of the water source and the location where such findings were detected. The Depth of the water source of most collected samples was between 100-300 feet. Few water sources had a depth of >300 and <100 feet. 5(B): The map details the water source's pH and the location where such findings were detected. Few samples show pH<6.5, and maximum samples show a pH range between 6.5-7.5

The correlation coefficient between arsenic and Depth was 0.1492 with a p-value of 0.097. The correlation coefficient between pH and Depth was 0.04602 with a p-value of 0.4271. The correlation coefficient between TDS and Depth was 0.09424 with a p-value of 0.1033. pH and TDS showed a low positive correlation, whereas all other applied correlation tests have poor or negligible correlations (Figure - 6).

**Table 4.** Showing primary health symptoms in the people residing in surveyed area

| S. no. | Disease | No. cases | % of disease |
|---|---|---|---|
| 1. | Diabetes | 11 | 3.66 |
| 2. | Hypertension | 11 | 3.66 |
| 3. | Hypertension and diabetes both | 10 | 3.66 |
| 4. | Renal stones | 16 | 5.33 |
| 5. | Gall bladder stones | 7 | 2.33 |
| 6. | Skin allergy | 7 | 2.33 |
| 7. | GI disorders | 3 | 3.32 |

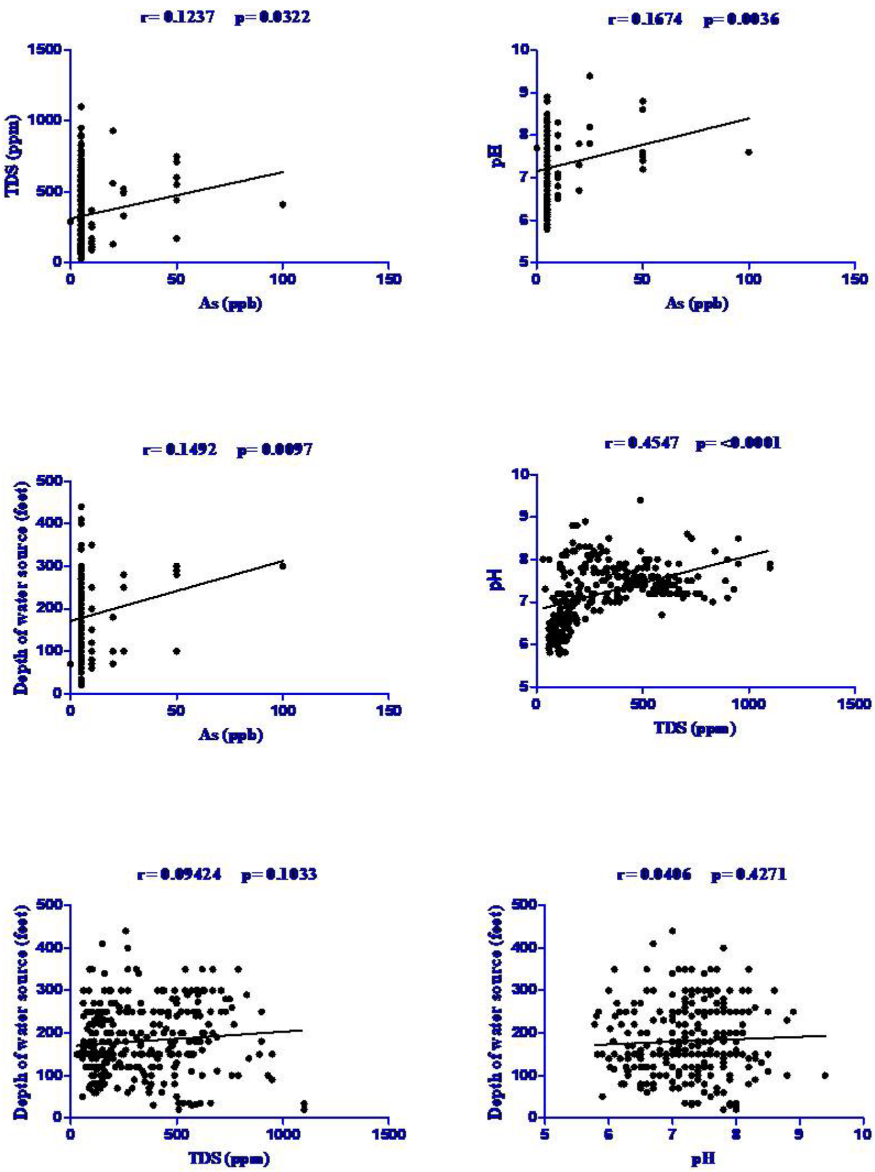



**Figure 6.** Correlation between various studied groundwater parameters

A T-test was applied to the mean values of arsenic, pH, TDS, and Depth. The p-value of arsenic and pH was 0.3659 ($R^2$=0.001368), with the difference between means being -0.4572 ± 0.5052. The p-value of arsenic and TDS was <0.0001 ($R^2$=0.5017), with a difference between means of -327.1 ± 13.33. The p-value of pH and TDS was <0.0001 ($R^2$=0.5014), and the difference between means was 326.7 ± 13.32. The p-value of arsenic and Depth was < 0.0001 ($R^2$=0.6878), with a difference between means of -174.0 ± 4.793. The p-value of pH and Depth was <0.0001 ($R^2$= 0.6890), with a difference between means of 173.5 ± 4.767. The p-value of TDS and Depth was <0.0001 ($R^2$= 0.1638), and the difference between means was 153.1 ± 14.15.

Groundwater has been used as the primary source of drinking water. According to WHO, the permissible limit for TDS is 500 mg/L. Water with TDS levels exceeding 500 mg/L causes gastrointestinal upset in consumers (WHO, 2003)(Organization, 2003). A high TDS is also associated with the odor and corrosiveness of water (Srilatha et al., 2014). Taste, hardness, and corrosion are all correlated with high TDS. High TDS water generally contains high concentrations of Na, K, Ca, Cl, $SO_4$, and many other elements that play a crucial role in digestive upset (Sarfraz et al., 2018). In our study, 24 % of samples from the Kangra district showed that the TDS concentration exceeded the acceptable threshold of 500 mg/L, which is associated with gastrointestinal abnormalities in people residing there. Renal as well as gallbladder stone cases were very high there.

According to WHO, (2011), water with a pH between 6.5 and 8.5 is considered safe. Water's pH plays a vital role in regulating homeostatic balance and is a lifesaving fluid. Ramesh & Vennila, (2012) reported that pH values below 6.5 or above 8.5 can damage the mucous membrane lining of tissues and eyes, and skin problems are reported through acidic pH water (Popoola et al., 2019). Highly alkaline pH is associated with the reduced reproductive rate in both males and females, the decline in biodiversity slowed growth, and the effects on the olfactory and optic systems. Ammonia and hydrogen sulfide become more poisonous at high pH, leading to higher human acidity levels (Dirisu et al., 2016). In our study, the values of pH range from more acidic to slightly alkaline, while at Baijnath, the pH is very high (9.4), and the lowest pH was 5.78 from Bhawarna. At Baijnath, various digestive problems along with diabetes are reported, and at Bhawarna, gastrointestinal upset cases are there.

Groundwater arsenic concentration is a common concern associated with human health. One study from Pakistan revealed that the arsenic concentration was less in shallow water sources; it was 25 ppb up to 100 feet and 125 ppb at greater than 110 feet depth (Nickson et al., 2005), while a study run by Shahid et al., (2018) showing a reverse trend of the above findings and reported that arsenic level was 32 ppb above 140 feet. It was just double below 140 feet. We studied arsenic levels from a 20- 440-foot water source and found that the arsenic level increased with an increase in Depth; the finding was different from the Bihar and West Bengal incidences.

Arsenic exposure is associated with many health effects, including skin changes, hyper-pigmentation, and gastrointestinal and neurological disorders (Chakraborti et al., 2017). In our study, 25.66% of people have signs of different diseases, out of which 7.66% were reported with hypertension 3.32% with G.I. disorders, 3.66% with diabetes, 2.33% are showing skin diseases, and almost 7.66% are showing stone formation in the kidney and gall bladder. In our study, 4.52% of the samples had arsenic levels above ten ppb. Kumar et al. (Magalona et al., 2019) reported that the risk of gall bladder cancer caused by arsenic-contaminated water in Bihar populations. The ROS and RNS formed due to pesticide and arsenic exposure contribute to gall bladder carcinogenicity (Hubaux et al., 2013).

## CONCLUSION

The study concluded that deep aquifers showed more arsenic contamination. 4.52 % of samples showed arsenic levels above ten ppb. Initial symptoms of raindrop pigmentation and hyperkeratosis were evident in residents consuming arsenic-contaminated water. Low pH and high TDS with arsenic toxicity were associated with diabetes and hypertension. 7.66% of cases of stone formation were reported from residents with very high TDS and arsenic toxicity. Still, Himachal Pradesh is considered safe from arsenic toxicity, but this study showed the prevalence of groundwater arsenic contamination with the initial signs of arsenic toxicity in residents.

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The present research did not receive any financial support.

## CONFLICT OF INTEREST

The authors declare that there is not any conflict of interests regarding the publication of this manuscript. In addition, the ethical issues, including plagiarism, informed consent, misconduct, data fabrication and/ or falsification, double publication and/or submission, and redundancy has been completely observed by the authors.

## LIFE SCIENCE REPORTING

The study has been approved by the Institutional Ethics Committee (IEC) of Central University of Himachal Pradesh, Dharamshala, Himachal Pradesh, India.

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