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Metal Contents in the Ground Waters of Tharparkar District, Sindh, Pakistan, with Special Focus on Arsenic

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Abstract. An integrated study was carried out to assess the ground water quality of Thar Desert, with exclusive focus on arsenic contamination, as ground water is the main source of drinking water in that area. Overall ninety nine (99) water samples were randomly collected from thirty three (33) different locations of District Tharparker and surveyed for arsenic (As) and other metals of potential concern in drinking water samples like iron (Fe), calcium (Ca), copper (Cu) and zinc(Zn). Ground water samples of two villages namely, Murid Khan Umarani and Khan Khanjar Reham have been found contaminated with As concentration present above 10 ppb while in the rest of the samples, As concentration is within the maximum permissible limit of all existing standards and has been found within narrow variations. This high profile of As in ground water is due to geochemical deposition of As in the vicinity. The quality of 76% of samples have been found to be extremely deteriorated due to the presence of high dissolved solids and hardness, making water unfit for drinking purposes.

Keywords: arsenic, ground water pollution, metal contents, Tharparkar

Introduction

Ground water contamination by As of non-anthropogenic origin has been found not only in developing countries but also in developed countries (Watanabe *et al*., 2004). It has been estimated that approximately one third of the world's population uses ground water for drinking purpose (Nickson *et al*., 2005).

Arsenic (As) is a metalloid and occurs naturally, being the 20th most abundant element in the earth's crust and component of more than 245 minerals. These are mostly ores containing sulphide, along with copper, nickle, lead, cobalt or other metals. Arsenic and many arsenic containing compounds are mobile in the environment. Weathering of rocks converts arsenic sulphide to arsenic trioxide, which enters the As cycle as dust or by dissolution in rain, rivers or ground water, thus making groundwater contamination by As is a serious threat to mankind all over the world. The terrestrial abundance of As is around 1.5-3 mg/kg and sources of As in the environment are natural as well as anthropogenic. Long before man's activities had any effect on the balance of nature, As was distributed ubiquitously throughout the earth's crust, soil, sediments, water, air and living organisms. Arsenic comprises about five hundred thousand of 1% (0.0005%) of the earth's crust (Mandal and Suzuki, 2002). It is used not only in medicines but also in agriculture, live stock feed, electronics industry and metallurgy (Gulledge and O'Connor, 1973).

As far as biochemical and physiological consequences of As are concerned, an adult human body contains about 18 mg of As. Arsenic is not considered essential but it is stimulatory and about 80% of the absorbed As is widely distributed in the tissues including the liver, abdominal viscera, bone, skin and particularly hair and nails from where it can be detected (Nriagu and Azcue, 1990). Chronic effects of As may appear from its accumulation in the body due to ingestion at low intake levels for prolonged periods of time. Strong evidences have been available to confirm that As in drinking water is proactive in causing skin, lung, kidney and bladder cancers (Ahmed, 2003).

Water pollution is one of the major threats to public health in Pakistan. Drinking water quality is poorly managed and monitored. Pakistan ranks 80th among 122 nations in drinking water quality. Drinking water sources, both (surface and ground water) are contaminated with coliforms, toxic metals and pesticides throughout the country (Azizullah *et al*., 2011). A study was conducted on the Indus River of Kohistan region

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along with the springs and streams to investigate As concentrations (As^{3+}, As^{5+} and total As) and other physico-chemical parameters. The results revealed that in some samples, As concentrations exceeded the permissible limits set by Pakistan Environmental Protection Agency and World Health Organization (2004); (Muhammad *et al*., 2010).

In the light of previous survey of ground water testing for As levels in Pakistan, significant proportion of underground water sources particularly in Sindh and Punjab provinces are contaminated and unsafe for human consumption (UNICEF & PCRWR, 1999-2000). Department of Community Health Sciences, Aga Khan University Karachi conducted a study to assess the health burden of As contamination of ground water of District Khairpur with the sponsorship of UNICEF. According to their findings, hyperpigmentation was the most common manifestation followed by keratosis of soles. Arsenicosis increases slightly with increasing age. Females were more commonly affected than males. Skin manifestations are considered the "tip of iceberg" for adverse affects of As (CHS-AKU Technical Report, 2006).

In the province of Sindh, ground water As concentration has reached up to 1100 ppb which far exceeds the World Health Organization (WHO) limit of 10 ppb (Haqi *et al*., 2007). Thar is the largest district of Sindh province having land area spread over approximately 22,000 sq. km and is the largest desert of Pakistan (Fig. 1). It has a population of more than one million. The study area has a tropical desert climate. Due to the arid and semi arid climate and insufficient surface water resources,

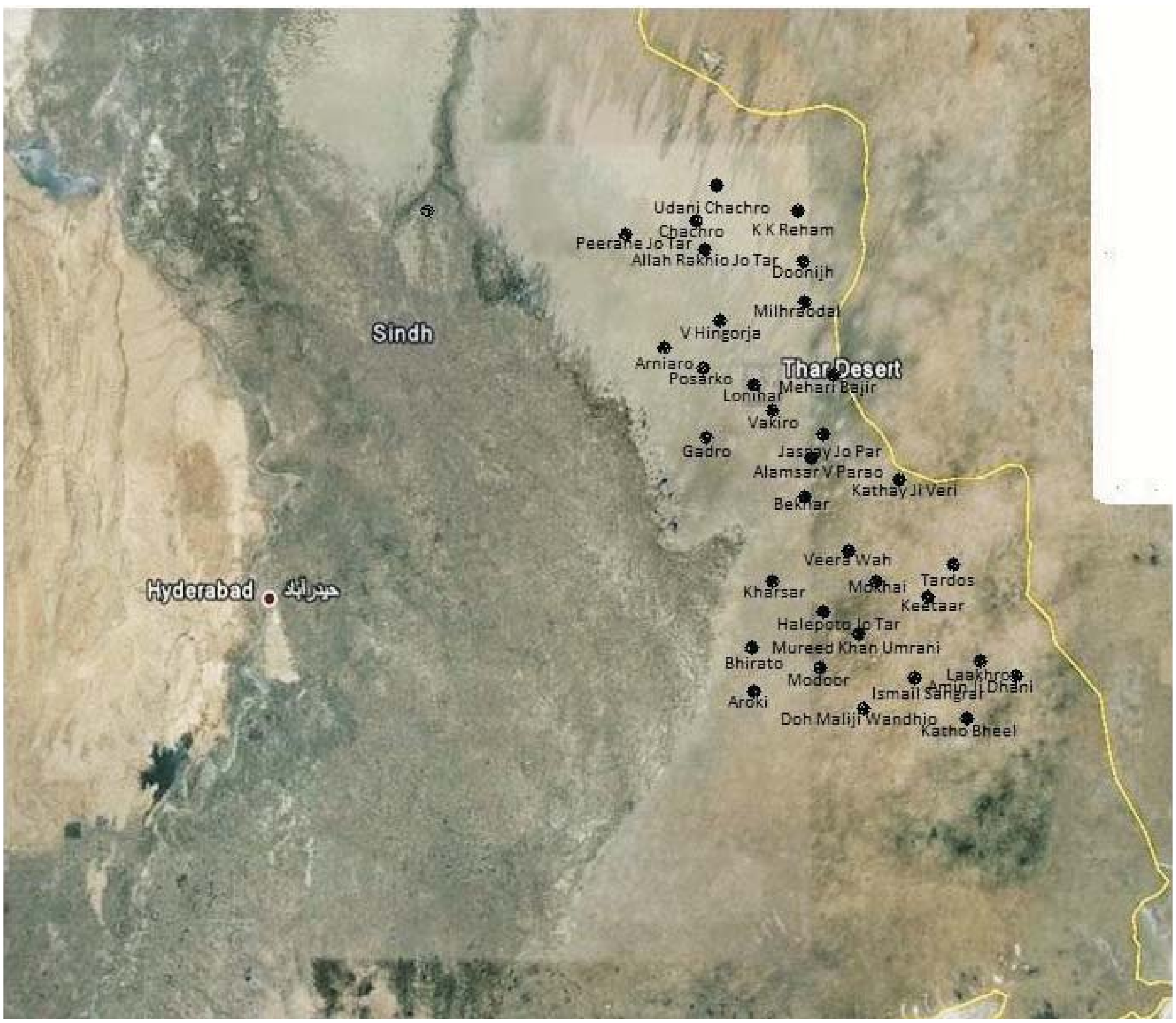

Fig. 1. Sampling locations with map of the study area.

ground water is the only source of water in almost all parts of Thar Desert and is used to meet the drinking and other domestic water needs of the inhabitants (Rafique *et al*., 2008). The geographical and geological set-up of Thar Desert suggests the deterioration of water quality in terms of trace and heavy metals (Ploethner, 1992). If ground water has to remain a mainstay of public water supply world wide, then there is an urgent need for better understanding of the mechanisms of enrichment of natural heavy metals in ground water (Watanabe *et al*., 2004).

Several studies have shown that As concentration in body tissues and fluid increases with an increase of As concentration in drinking water (Mandal *et al*., 1998; Valentine, 1994). In order to have a better understanding of the magnitude of As contamination in ground water and its effects on human beings, an integrated study was carried out in Tharparkar District of Sindh province. A detailed scenario as regards occurrence of different metal contents in the ground water of Thar Desert with special focus on As is presented in the study.

Materials and Methods

Sampling. Samples were collected in one liter polyethylene screw capped bottles, pre-cleaned with detergent, washed with plenty of tap water, soaked in 1% HNO_3 for 24 h and finally rinsed with de-ionized water. After cleaning operation blanks ($\sqrt{n}$=12) bottles were selected at random, were prepared in de-ionized water having 1% analytical grade HNO_3 and were analyzed for As and Pb in order to reject the sampling bottles having detectable levels of these trace metals. None of the sampling bottles were found to have detectable levels of metals. Sample bottles were then dried at 100 °C for 1 h, cooled to room temperature, recapped and labeled. 10 mL HNO_3 was added to the samples which were used for the estimation of trace metals. Ground water samples were collected in triplicate from thirty three (33) locations of District Tharparkar and were brought to the laboratory and immediately stored at 4 °C and analyzed as soon as possible. All necessary precautions were observed during their sampling, transportation and storage (APHA, 1998).

Analytical procedure. Analytical reagent grade chemicals were used in the preparation of reagents and standards. Physico-chemical analysis was performed for each sample in triplicate and the average values were recorded. The pH, TDS and electrical conductance of each sample were measured immediately using portable digital pH Meter and conductivity/TDS Meter (JENWAY/E.U/430 pH/cad./ portable/02162). Ca was determined by complexometric titration with EDTA. All other analytical estimations were performed within 48 h of sampling.

Trace metals analysis. For the analysis of trace metals (As, Fe, Pb, Cu, Zn) reference standards were prepared from Merck AAS standards (1000 ppm). Analyses were performed on a Hitachi Z-5000 Spectrophotometer with Zeeman background correction, equipped with graphite furnace, ZAA (Zeeman Atomic Absorption Spectrophotometer) software for electronic processing of the results, was used. Determinations of Pb and Cu were carried out by flameless (ETAAS) Electrothermal Atomic Absorption Spectrophotometer, Fe and Zn by Flame Atomic Spectrophotometer (FAAS) and As by hydride formation system.

In the determination of total As, the samples were treated with 20 % KI in order to reduce all the arsenate (As^{+5}) to arsenite (As^{+3}). For the acid channel 1.2 N HCl was used while for the reducing channel, $NaBH_4$ (1%) in NaOH (0.4%) were used. Blanks were treated in the same manner. Analysis of each metal was carried out in triplicate to get representative results. Indirect measure of the accuracy was obtained from recovery studies on ten water samples spiked with each metal. Recoveries for the spiked samples were 100±6%.

Results and Discussion

Table 1 summarizes the selective physico-chemical analysis of ground water samples, collected from Tharparkar District and analyzed in order to have an idea about the quality of water. The results indicate that most of the samples do not meet the WHO maximum permissible limits of drinking water with reference to Total Dissolved Solids (TDS) and thereby, deteriorating the ground water quality. As mentioned in Table 1, serial numbers 1, 7, 11, 21, 22, 23 and 32 samples were found within the permissible limit of 1000 ppm TDS. Almost 76% of villages were found to be effected due to the presence of high dissolved contents ranged up to 10280 ppm.

The pH of the analyzed samples, ranges from 6.9-8.6 indicating narrow variation and that most of the water samples are weakly alkaline. Due to the undesirable saline taste of these water samples, many of these wells are rendered non-operational. In areas where this problem

Table 1. Sampling locations, physical properties and metal levels in selected ground water samples in Tharparker District

Location	No. of Samples	pH	TDS ppm	Conductance mS/cm	As ppb	Pb ppb	Cu ppb	Zn ppm	Fe ppm	Ca ppm
Vakiro	03	7.5	580	1.16	0.77	2.75	BDL	0.70	0.01	64
Mehari Bajir	04	8.2	1628	3.26	1.04	1.54	BDL	0.58	0.01	108
Lonihar	03	6.9	3424	6.85	0.20	3.45	BDL	0.14	0.04	212
Posarko	05	7.0	2422	4.84	0.37	3.72	BDL	0.03	0.05	114
Arniaro	04	7.2	2112	4.22	3.34	1.14	BDL	0.27	0.01	220
Veehingorga	04	8.0	1008	2.02	0.16	3.01	BDL	0.80	0.01	89
Milhraodal	03	8.1	656	1.31	3.15	2.05	BDL	0.20	0.09	52
Doonijh	06	7.9	1432	2.86	5.53	4.36	BDL	0.04	0.02	78
Khan Khanjar Reham	05	8.2	2316	4.63	10.56	1.45	BDL	0.22	0.03	190
Udani Chachro	03	8.5	4172	8.34	0.18	3.42	BDL	0.23	0.02	138
Chachro	03	7.4	421	0.84	0.21	2.42	BDL	0.11	0.02	39
Allah Rakhio Jo Tar	04	8.3	8030	16.07	0.59	4.96	8.0	0.95	0.01	280
Peerane Jo Tar	05	8.4	7800	15.57	3.39	16.61	13.0	0.55	0.03	272
Gadro	03	7.6	1261	2.50	0.66	2.33	9.0	0.10	0.02	60
Jesse e Par	04	7.8	7670	15.29	1.89	16.33	8.0	2.99	0.01	208
Karanghi/Kathejiveri	04	8.5	4200	8.41	1.04	1.02	8.0	0.19	0.09	180
Beknar	04	8.6	10280	20.56	0.62	15.70	10.0	0.09	0.83	288
Almsar V Parao	06	8.3	6050	12.4	6.51	3.57	13.0	0.18	0.01	194
Tardos	05	7.7	1318	2.60	0.48	1.48	12.0	0.05	0.02	79
Keetar	04	8.1	2460	4.92	4.10	2.95	6.0	0.12	0.01	141
Mokhai	05	7.8	500	1.01	1.73	13.5	5.0	BDL	3.06	49
Hali Pota Poring No. 1	05	7.4	460	9.14	1.31	3.56	9.0	0.76	0.02	62
Virawah	05	7.2	240	0.49	2.10	2.39	4.0	BDL	0.15	20
Murid Khan Umarani	05	8.4	1534	2.98	29.88	3.26	9.0	0.57	0.42	161
Bhirato	05	7.4	250	0.51	0.92	BDL	1.0	BDL	0.02	22
Modoor	03	8.3	7230	14.45	0.55	15.02	4.0	1.27	0.02	301
Layakhro	06	8.1	4320	8.66	1.31	1.70	145	2.08	0.02	140
Amin Ji Dhani	04	7.9	1490	2.96	1.94	1.91	4.0	0.05	0.02	84
Ismail Sangrase	04	8.1	2070	4.14	0.95	1.94	9.0	0.27	0.10	88
Aroki	06	8.2	4340	8.63	0.90	2.32	117	2.0	0.02	158
Doh Maliji Jo Wandhio	04	8.4	2680	5.05	2.22	1.04	6.0	0.01	0.01	86
Katho Bhel	05	7.5	835	1.65	5.36	BDL	10	0.02	0.02	142
Kharsar	05	7.9	1330	2.60	3.37	4.97	6.0	0.01	BDL	200

BDL = below detection limit of (As: 0.15 ppb, Pb: 0.05 ppb, Cu: 0.05 ppb, Zn: 0.005 ppm and Fe: 0.01 ppm)
Note: mean values of 03 samples per location are presented.

is acute, people fetch water for drinking purpose from far flung places, where water is of somewhat better quality (EC, 1982).

Table 2 presents a comparison of different international drinking water norms with respect to As, Fe, Cu, Zn, Ca and Pb (CDWQ, 2010; USEPA, 2005; WHO, 2001). WHO (1971) has proposed two norms for As, namely highest desirable and maximum permissible, based on which three levels have been defined for As. Any parameter below highest desirable levels is marked as 'Safe', between highest desirable and maximum permissible levels as "Alert" and above permissible level marked as "Toxic". Based on their criteria, all the drinking water samples of Tharkparker collected in the study have been categorized as Safe, Alert or Toxic with respect to each of the parameters analyzed. A summary of the results obtained has been presented in Table 3. It shows that all the drinking water samples, collected from District Tharparker have been found to be safe with respect to As except two samples collected from 'Murid Khan Umarani'(30 ppb As) and Khan Khanjar Reham (11 ppb As) villages, which by the existing scale of WHO (1971), falls under the 'Alert range'. These observations are of concern, as As concentration in ground water is prone to sharp fluctuation depending upon geochemical conditions. It may further be seen from Fig. 2 that, all water samples have As concentration within the maximum permissible limits as regards existing drinking water guidelines except European Economic Community maximum guidelines (EEC, 1985) .

To look into the distribution pattern of As occurrence, analytical data were subjected to several statistical treatments and are shown in Table 4. As was analyzed in the collected samples in the range of 0.16 to 29.88 ppb. Small differences between the mean and median in each

Table 2. Comparative drinking water international standards*

Parameters	USEPA[a] Maximum contamination level (ppm)	CDWQ[b] Maimum acceptable concentration (ppm)	EEC[c] Maximum admissible concentration (ppm)	Japan Maximum admissible concentration (ppm)	WHO[d] Guideline (ppm)
As	0.01	0.01	0.05	0.01	0.01
Cu	1.0	2.0	2	1.0	1-2
Fe	0.3	0.2	0.2	0.3	0.3
Zn	5	5	-	-	3
Ca	-	200	-	-	200
Pb	0.015	0.01	0.01	0.05	0.01

*[a]USEPA, United State Environmental Protection Agency; [b]Canadian drinking water quality guidelines; [c]European Economic Community; [d]World Health Organization.

Table 3. Summary of safe (S) alert (A) and toxic (T) limits of drinking water according to WHO guidelines

Parameters	WHO guideline*		S (%)	A (%)	T (%)
	Highest Desirable Limit (HDL) (HDL)(ppm)	Maximum Permissible Limit (MPL) (ppm)	Below HDL	Between HDL and MPL	Above MPL
As	0.01	0.05	93.94	6.06	0
Cu	0.05	1.0	93.94	6.06	0
Fe	0.1	1.0	90.91	6.06	3.03
Zn	5	15	100	0	0
Ca	40	200	9.09	66.67	24.24
Pb	0.01	0.05	88	12	0

*WHO (1971)

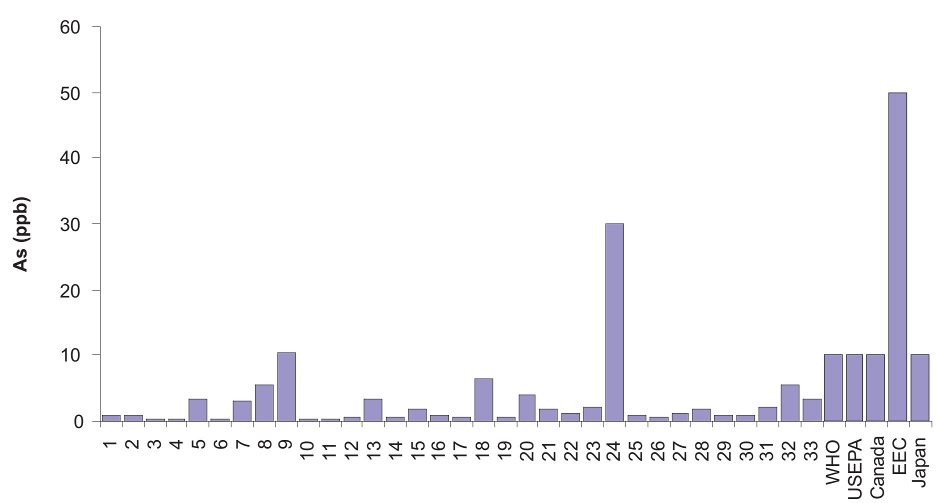

Fig. 2. Arsenic levels with comparison to different existing standards.

case and high standard deviation indicate that the distribution is 'normal'. Wide range of data indicates the presence of extreme values, which are likely to bias the normal distribution statistics. Occurrence of As is very sporadic and marked differences in concentration occur across very short distances. The geochemical setup of Thar Desert suggests that the natural source of As in ground water is the As bearing minerals associated with sedimentary rocks and the granitic basement of Thar (Rafique *et al*., 2008).

Pb is a general toxicant and a cumulative poison which is present in water to some extent as a result of dissolution from natural sources. Pb has increased in the environment during the last century. However, Pb is not easily mobilized from soils (EC, 1982). Ground water samples of Thar Desert contained 1.02-16.61 ppb of Pb with a mean value of 4.69 ppb.

Table 3 shows that 88% of analyzed samples were found in the 'Safe' range with respect to Pb, as per WHO limits (1971). Large differences between the mean and median and high standard deviation indicate that the distribution is widely off normal. 12% of the samples were categorized in 'Alert' zone for Pb, indicating that the samples are above the highest desirable limit of 10 ppb by WHO and are still within the maximum permissible limit of 50 ppb. It could be possible that these analyzed levels may lead to Pb toxicity in the water bodies. Being a toxic element, Pb causes neurological, physiological and behavioral problems, even at relatively low levels of exposure. Raised hearing threshold and decrease intelligence quotient (IQ), may lead to memory loss and death in case of long exposure (WHO, 1973).

Zn is an essential trace metal, found in all ground waters. It is obvious from Table 1 that ground waters of Thar are quite safe enough as regards their Zn content. Higher values for Zn accumulation were observed in the range of 0.01-2.99 ppm with a mean and median of 0.52 ppm and 0.21 ppm, respectively. Distribution analysis reveals that Zn is normally distributed due to minimum difference between mean and median. It is known that Zn in natural waters is low and does not appreciably contribute to human daily intake. However, Zn from galvanization process may reach a few ppb in soft acid water. In such cases drinking water may provide a considerable portion of the daily Zn intake (Monitor, 1981).

Fe is one of the most abundant metals in earth's crust and is found in natural waters in the range of 0.5 to 50 ppb. Higher concentration of Fe imparts taste and colour to water (WHO, 1993). This is underscored by the figures in Table 3 where combined value (A% & T%) 6.06+3.03 (9.09%) of ground water belongs to the 'Alert' or 'Toxic' categories. Normal distribution analysis on Fe values as shown in Table 4, reveal large difference between mean and median values indicating asymmetric distribution pattern and presence of extreme values in the form of outliers ranging from (0.01-3.06) ppm with high standard deviation. Although Fe is an essential element for human nutrition, drinking water is not considered to be a good source of this particular element due to its role in impairing physical/ aesthetic qualities of water (WHO, 1993).

Table 2 shows that most of the water samples appear to be reasonably hard with respect to Ca. The distribution of Ca; however appears to be normal, due to the closeness of mean and median values. Similarly analyzed Ca was found in the range of 20-301 ppm in the collected water samples (Table 4). Ca occurs in great abundance in all natural waters and is an important micronutrient. It is

Table 4. Normal distribution statistics for As in the groundwater of District Tharparkar*

Statistics	As	Pb	Zn	Cu	Fe	Ca
Mean	2.96	4.69	0.52	18.09	0.16	136.94
Median	1.31	2.95	0.21	7.96	0.02	138
Standard Deviation	5.33	4.91	0.714	36.04	0.55	80.15
Sample Variance	28.45	24.15	0.510	298.56	0.30	6424.06
Minimum	0.16	1.02	0.01	4.0	0.01	20
Maximum	29.88	16.61	2.99	145	3.06	301
Mode	1.04	-	0.27	9	0.02	-
**Sample nos.	-	25, 31	21, 23, 25	1 to 11	33	-

*only those water samples have been considered for statistical treatment in which the metals analyzed were found to be within detection limits; **samples No. as provided in Table 2, where mean values found below the LOD.

an important contributor to hardness in water and its excess may reduce the utility of waters for domestic purpose (Elin, 1988).

Cu is an essential element in human metabolism and is generally considered to be non-toxic for human at the levels encountered in drinking water but elevated concentration of Cu may affect the taste and colour of water (WHO, 1993). The distribution of Cu is widely off normal due to wide difference between the mean and median (Table 4). The extreme values were found to be 4.0 - 145 ppb, leading to high standard deviation. Water samples were therefore found to be safe according to all existing drinking water guidelines as may be seen in Table 3.

Pearson correlation coefficient analysis was obtained between metal pairs in the water samples. The product of the correlation coefficient (r) was evaluated as the range given in Table 5. According to the analysis, As shows almost no correlation with the other metals involved in the study. In case of Pb, low correlation was observed with Zn and Fe, medium correlation was however found between Cu and Zn.

Table 5. Pearson Correlation Coefficient Analysis

	As	Pb	Cu	Zn	Fe
As	1				
Pb	-0.081	1			
Cu	-0.062	-0.10	1		
Zn	-0.056	0.315	0.585	1	
Fe	0.053	0.414	-0.053	-0.152	1

0.0-0.3 = no correlation; 0.3-0.5 = low correlation; 0.5-0.7 = medium correlation

Conclusion

To look into the trend and distribution pattern of metal contents, analytical data has also been subjected to normal statistical analysis. A comprehensive statistical analysis of metal contents in ground water of District Tharparker has revealed that two locations namely, Murid Khan Umarani and Khan Khanjar Reham contain As above the permissible level of 10 ppb. One more location Alamsar V Parao is more likely, where the prolong use of As contaminated water may cause adverse effects to the consumer's health. As mobilization in two identified locations may follow reductive dissolution of oxy-hydroxides of Fe through oxidation of sedimentary rocks present in the vicinity (Zaigham, 2003).

Nine percent of samples were out of 'Safe' zone in case of Fe. While 3% were found to be in the 'Toxic' range of Fe. Twelve percent samples were out of 'Safe' zone with respect to Pb. There is no explicit correlation found between occurrence of As and the other analyzed metals in these samples. By and large, these water samples are characterized as hard water making it unfit for domestic use. In general the presence of As in drinking water at several locations within District Tharparkar is a great potential threat. An appropriate study should be designed that includes comprehensive risk assessment and elucidation of As mobilization mechanism. A mitigation plan should be developed on priority basis to address this situation of health health concern.

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