Ground Water Contamination with Hexavalent Chromium and its Health Effects: Debatable and Unsettled Issue of Regulation

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ABSTRACT

Background & objectives: Ground water contamination with hexavalent chromium (Cr VI) is largely associated with the improper disposal of chromium-waste by the various industries. Due to the high mobility of Cr (VI) it penetrate soil and contaminate ground water. The data-base related to Cr (VI) toxicity in drinking is lacking in our country, however, high levels were reported in sewage water and factory effluent. Cr -toxicity is an emerging global health concern that can induce health problems including cancer. Keeping above facts in view, we have carried out a general survey by examining Cr (VI) in ground water obtained from Municipal Corporation and personal tube well.

Methods: A total number of 415 drinking water samples (AMC, n=125; Tube well water n=290) were analysed for Cr (VI) by ionchromatography using US EPA Method 218.6.

Results: A wide variation in the level of Cr (VI) in drinking water was observed in tube-well bore water. The average value was 14.57 µg/l (range 0.0 - 322.64 µg/l) with a median value of 6.39

µg/l. Cr (VI) was not detected in about 19% of the bore-well water (detection limit 0.3 ppb). On the contrary, AMC water samples had a considerably lower level of Cr (VI) (average 3.58 µg/l, range 0.0 – 13.78 µg/l, median 2.58 µg/l) and it was not detected in 39.5% of the samples. Cr (VI) in 7 bore water samples exceeded the maximum allowable limit of WHO (50 µg/l). Higher levels of Cr (VI) were several hundred to thousands fold-high than the non-enforceable proposed limit of Cr (VI) (0.02µg/l) as suggested by Public Health Goal (PHG). Relatively higher levels of Cr (VI) in water samples were detected from industrial areas when compared to non-industrial areas of the city.

Conclusions: Few samples exceeded the prescribed limit of WHO (50 µg/l) while all samples detected with Cr (VI) were exponentially high than the proposed limit of PHG. Higher exposure to (Cr VI) might be a risk factor to induce Cr - related health effects.

Keywords: Drinking water-hexavalent chromium- ionchromatography - public health goal, PHG-trivalent chromium

1. INTRODUCTION

Chromium (Cr) is an important industrial metal and two common forms of it Cr (III) and Cr (VI) are widely used in industries engaged in the manufacturing of steel and alloys, leather tanning, pigment manufacturing and processing, electroplating, textile trade etc[1, 2,] . The metal is also used as anti-corrosive, wood preservative agent and in cooling tower[2]. Thus enormous amount of toxic chromium-wastes is released into the environment that results in ground water pollution. Contribution from natural sources i.e. erosion of chromium rich rock (serpentine rocks) in certain specific region of the globe may also be significant[3]. Reports are also available that indicate the conversion of Cr (III) in chromite ore [Fe. Mg (CrO_3)] to Cr (VI) by ecological processes. The role of manganese oxide specifically pyroluscite has been reported for the oxidation of Cr (III) to Cr (VI)[4]. The Cr (VI) so formed leaches out from the soil by and large and contaminate drinking water ubiquitously. In 1993, Erin Brockovich has first highlighted ground water contamination with Cr (VI) to public attention in Hinkely, CA. In 2010, the Environmental Working Group[5] (EWG) reported trace levels of Cr (VI) in 31 of 35 US cities tap waters tested, but it was not a peer-reviewed scientific study. Natural contamination of drinking water has been reported from various countries and has been summarized by water Research Foundation[6]. Unlike arsenic toxicity, where millions people worldwide have been exposed to the elevated levels of arsenic in drinking water, epidemiological studies on oral exposure to Cr (VI) are very limited[6]. Zhang and Li[7] (1987) has reported elevated levels of all type of cancer including stomach cancer in Liaoning Province of China who have ingested higher levels of Cr (VI) through drinking water. However, this work was criticized later and became controversial and retrieved by the author. Similar finding was recently

presented by Linos *et.al*.[8] (2011) who reported significant increases in primary liver cancer mortality in citizens exposed to Cr (VI) through drinking water. However, reports from India are still lacking, as there are no consensus data on the exposure to Cr (VI) through drinking water. Although, several reports did also appear in literature, that indicate contamination of aquifer from the indiscriminate disposal of chromium- waste by various Cr- processing industries. According to a report, Orissa and Gujarat cities' are among world's 10 most polluted cities that observed ground water contamination. In Sukindia[9] (Orissa) there are about 97% of India's chromite ore deposits and here workers experience infertility, birth defects and still-birth. Ground water contamination with hexavalent chromium and its health effect has also been reported from Kanpur city of U.P. where chromium-waste from lather tanning industries is improperly disposed in to the environment[10]. Ahmedabad city of Gujarat State is known for textile work and a number of Cr-based dyes are used in small-, and large- scale-textile industries. Ground water contamination from this city remains poorly reported.

Currently, Cr- toxicity is regulated under 50 µg/L (ppb) by WHO[11] and traditionally it is assumed as the sum of Cr (III) and Cr (VI). As Cr (III) is relatively less toxic and insoluble in water, the toxic contribution from it in drinking water appears negligible. Cr (VI) seems to be the sole agent causing most of the toxicity in ground water that is used an important source of public utility. Moreover, Cr (VI) has no individual representation in water standard regulation. EPA is encouraging regular monitoring of Cr (VI) in drinking water and it had initiated several programs; California's Office of Environmental Health Hazard Assessment (OEHHA) plays important role to establish maximum contamination limits (MCL) and with the help of Public Health Goal (PHG). It has proposed an MCL for Cr (VI) as low as 0.02 µg/L. Till recently, it was difficult to analyse Cr (VI) at such a low level, EPA[12] has improved the existing method to detect Cr (VI) at the proposed level of PHG (0.02 µg/L). But commercially water standards at such a low level appear difficult to achieve due to the unfamiliar and non-cost effective technology. Thus, the present water standard regulation does not seem to comply in true sense and it needs further revision or a new MCL specifically for Cr (VI) is required. EPA is reviewing the available literature and by the end of 2014 it will hopefully decide whether the current limit 50 µg/L is to be revised or not.

We designed the present study to examine ground water contamination by measuring the level of Cr (VI) in drinking water obtained from Ahmedabad Municipal Corporation (AMC) and from personal bore water collected from industrial and non industrial area of Ahmedabad city.

2. MATERIALS AND METHODS

Chemicals: *All the chemicals and solvents used in this study were of high purity grade (99.9 % purity Analytical grade) and they were all free from chromium content. Reference standards,*

chromium III, chromium VI was purchased from Fluka (Switzerland). Milli Q water of 18 mega-ohms was used throughout the studies.

Sample collection and study location: Drinking water samples (about 100 mL) were collected from Municipal Corporation supply and personal bore water in fresh vials (sample containers) previously washed with 0.1 M HNO_3 followed by R.O water and then finally with ultra pure water (Milli Q, 18 mega ohms). A total number of 415 drinking water samples (Ahmedabad Municipal Corporation, AMC, N=125: Personal tube well bore water, N=290) were collected from the five different zones. Generally water samples were cleaned and did not require any special preparation. Total dissolved solute (TDS) of water samples were measured by Eutech Instrument (OAKTON®) and their pH was adjusted to 6.6 to 6.8 with sodium hydroxide. All the samples were filtered through 0.45µm Acrodisc® syringe filter before injection on to the ionchromatograph.

Assay method: Dionex IC (2500) was used to measure Cr (VI) according to the standard procedure described by EPA[13] (Method 218.6) with post column derivatization of Cr (VI) with diphenylcarbazide and measurement of absorption at 530 nm by UV- VISIBLE detection. Chromelion software 6.7 version was used for the acquisition and processing of data.

Method validation: Standard calibration curve for Cr (VI) at varying concentrations as mentioned below was prepared to obtain a five-points calibration curve. Solutions of Cr(VI) were prepared fresh each time at concentrations 0.5, 1.0, 2.0, 5.0, 10 and 25 ppb. One ml of the above standard mixture of solution was injected in duplicate and the average area was taken while preparing calibration graph. The instrument received 200 µL as injecting volume of sample/standard. Calibration curve, R^2 and relative standard deviation (RSD) were obtained using standard Chromelion Software (6.7 version programme) for automated data processing and quantifying Cr (VI).

Reproducibility, precision and accuracy: Ion chromatography is a highly sensitive technique to quantify hexavalent chromium and it can detect Cr (VI) below 1 ppb. The linearity was, performing multiple run of standard Cr (VI) starting from a very low concentration (1ppb) to a high concentration up to 25 ppb tested in this study checked reproducibility and sensitivity. Co-efficient determination (R2) and % RSD were determined with the help of Chromelion software. .

Statistical analysis: All statistical analysis was performed using SPSS 15 software. Analysis of variance (ANOVA) was used for comparison among groups. Linear regression was used for studying various relationships among continuous variables. Two- tailed tests were used for all comparisons with level of significance as 5% ($p<0.05$).

3. RESULTS

Analytical method (US EPA 218.6) employed in this study is highly sensitive (minimum detection limit 0.3 ppb) and reproducible. Calibration curve (Fig 1) was found linear up to 30 ppb for Cr (VI) as tested and co-efficient of determination (r^2) was 0.998 with a RSD 3.21%. Accuracy of the method was further proved by maintaining accurate retention time (about 5.08 min) with respect to elution profile of Cr (VI) that is shown in overlay of chromatograms and recovery in spiked sample (Fig 2 & Table 1).

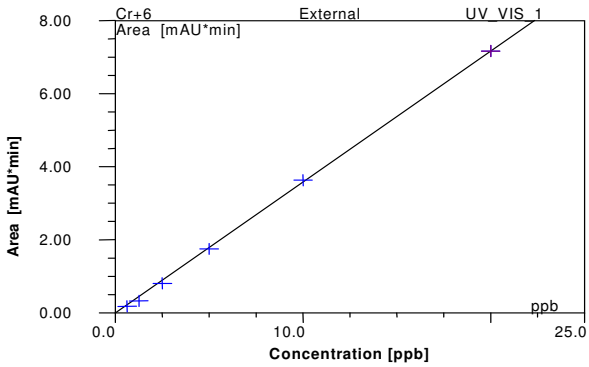

Fig 1. Calibration curve of Cr (VI)

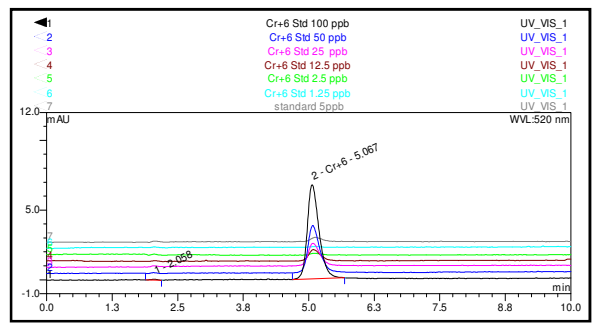

Fig 2. Overlay of chromatogram showing accuracy

Determination of Cr (VI) in un-spiked and spiked sample with the recovery is shown in Table 1. Water sample was first analysed to obtain background level of Cr (VI). It was found to be 8.18 ppb. This sample was spiked with known amount of standard Cr (VI) at two different concentrations, 10 and 30 ppb (Table1). The sample was mixed well and allowed to stand at room temperature for about 30 min. and then injected on to the column. Standard Cr (VI) 25 ppb was also injected in separate run to see its recovery. More than 96% recovery in all samples was obtained.

Table 1. Determination of Cr (VI) in spiked and un-spiked water samples showing recovery

Sr.No.	Sample (Water)	Spiked with Standard Cr (VI), ppb	Amount of Cr (VI) in (ppb)	% Recovery
1.	Blank	-	0	
2.	Un-spiked	-	8.18	
3.	Spiked (level 1)	10	17.58	96.7
4.	Spiked (level 2)	30	37.04	97.01
5.	25 ppb std.	-	24.08	96.32

Levels of Cr (VI) in drinking water

Levels of Cr (VI) in Municipal Corporation water samples and personal tube well water is summarized in Table 2. There was a wide variation in the level of Cr (VI) in drinking water obtained from bore wells as shown in Table 2.

Table2 Levels of Cr VI in drinking water from AMC and persona bore well water

Sr. No	Source	Total No. of samples	Samples detected With Cr (VI)	Samples not detected With Cr (VI)	Parameters	Cr (VI) (ppb or µg/l)
1	Personal tube bore water	290	234 (80.6%)	56 (19.4%)	Average	14.57
					Minimum	0.83
					Maximum	322.64
					Median	6.39
					GM	8.39
					SD	27.69
2	AMC water Supply	125	74 (60.5%)	51 (39.5%)	Average	3.58
					Minimum	0.32
					Maximum	13.78
					Median	2.58
					GM	2.43
					SD	2.90

The average value of Cr (VI) in personal tube well water was found to be 14.57 (range 0.83 - 322.64 µg/L) with a median value of 6.39 µg/L. Cr (VI) in one of the bore water was detected as high as 322.64µg/L. Cr (VI) was not detected in ~ about 19% of the bore well water samples (detection limit 0.3 ppb). On the contrary, AMC water supply had a considerably low level of Cr (VI) (mean 3.58 µg/L, range 0.32 – 13.78 µg/L, median 2.58 µg/L) and about 40 % of Municipal Corporation water samples showed no detection of Cr (VI).

In only three tube well water samples Cr (VI) concentration exceeded the prescribed limit of US EPA (100 µg/L). The 7 of the 290 samples were above the maximum contamination limit (MCL) of WHO and US office of California that regulates Cr (VI) as 50 µg/L. None of the sample in AMC water supply exceeded the prescribed (Table 3).

Percentage representation of hexavalent chromium in bore water of Ahmedabad city is shown in Table 3. It shows that the level of Cr (VI) in bore water was considerably elevated over the AMC water supply. Three of the 290 sample (1.03%) of the tube well water exceeded the prescribed limit of US EPA (100 µg/L), while 4 of the 290 samples (1.37%) were above the maximum contamination limit (MCL) of US office of California and WHO that regulates it as 50 µg/L, but these samples were below EPA limit (100 µg/L). None of the samples in AMC water supply exceeded MCL (Table 3).

Table 3: Distribution pattern of hexavalent chromium in drinking water supply of Ahmedabad city

Tube well personal bore water			Municipal corporation water supply		
Cr (VI) (ppb)	Number of samples	Percentage representation	Cr (VI) (ppb)	Number of samples	Percentage representation
> 100	3	1.03	-	-	-
> 50	4	1.37	-	-	-
< 50-30	11	3.79	> 10	2	1.6
< 30-10	77	26.55	> 5	16	12.8
< 10 -0.30	139	47.9	< 5	57	45.6
BDL/ND	56	19.3	ND	50	40
Total	290	99.5		125	100

Table 4 shows occurrence of Cr (VI) in the various zones of Ahmedabad city. A Kruskal-Wallis test was conducted to evaluate the effect of different zones of Ahmedabad city (Central, East, West, North and South) on the levels of Cr (VI) in AMC water and bore water. This test indicated that there is a significant difference in the mean rank among the levels of Cr (VI) in AMC water of different zones ($\chi 2$ (4, N = 74) = 6.92, from this data it can be concluded that there is a statistically significant difference between the AMC water and bore water group's median levels of Cr (VI) in both type of water (U = 6561, p = 0.001). It can be further concluded that the Cr (VI) levels in bore

water elicited statistically significant higher concentration than the AMC Water group (P =0.016). A Kruskal-Wallis test was conducted to evaluate the effect of different zone of Ahmedabad (Central, East, West, North and South) on the levels of Cr (VI) in AMC water and Bore Water.

Table 4. Distribution of Cr (VI) levels in various Zones of Ahmedabad

Zone	Municipal Corporation water supply							Bore water supply						
		Percentile representation			Mean Rank				Percentile representation			Mean Rank		
	N	50th	25th	75th		Min	Max	N	50th	25th	75th		Min	Max.
Central	15	5.8	5.7	6.1	52.4	5.3	6.1	27	6.8	4.5	9.2	101.7	1.2	28.0
East	14	5.4	5.4	5.6	30.5	5.2	6.8	23	13.0	8.8	16.3	149.4	1.3	51.2
West	14	5.5	5.4	5.6	27.7	5.3	5.7	75	6.5	3.2	13.4	109.3	1.0	59.0
North	14	5.6	5.3	5.8	35.1	5.3	7.0	62	9.4	5.6	15.4	134.5	0.28	28.0
South	17	5.6	5.5	5.9	40.0	4.5	6.1	54	6.8	3.2	13.3	109.5	0.83	322.6
Total	74	5.5	5.4	5.8	126.1	4.5	7.0	241	8.3	3.8	14.3	167.8	0.28	322.6

The results of Kruskal Wallies test indicated that there is a significant difference in the mean rank among the levels of Cr (VI) in AMC water of different zones ($\chi 2$ (4, N = 74) = 6.92, p =0.03) and difference is also significant in mean rank Cr (VI) levels in bore water of different zones $\chi 2$ (4, N = 74) = 10.40, p = .004). Thus, the different zones are effective in causing difference in Cr (VI) levels of AMC and Bore Water. Follow-up tests were conducted to evaluate pair-wise differences among the five different zones by controlling for Type I error across tests by using the Bonferroni approach. A post-hoc test using Mann-Whitney tests with Bonferroni correction showed the Cr (VI) levels in AMC water of Central zone is significantly higher than Eastern Zone (p=0.002) and Western Zone (p=0.004). Cr (VI) levels in bore water of Central Zone is significantly lower than Eastern Zone water (p=0.004).

Some residential areas of Ahmedabad city also observed small-scale industries including textile and chemical industries and they use some chemical - dye compounds containing chromium salts. They have their personnel bore and used them as a utility source to obtained water. Therefore, we have examined Cr (VI) level in industrial areas and compared our results from non -industrial areas. Table 5 indicates that relatively higher levels of Cr (VI) in water samples were detected from *Dhanilimda* area as compared to other non- industrial area such as *Kankarya* (105±92.1 Vs 5.54

±0.56, *mean ± GM*). Other industrial areas have also shown higher levels of Cr (VI) in their drinking water obtained from bore.

Table 5. Levels of Cr (VI) (µg/L) in industrial and non-industrial areas of Ahmedabad city

Zone	Industrial Area	Cr (VI) (ppb)	Non industrial Area	Cr (VI) (ppb)
South	a) Dani limda	105±92.1 N=9 (30.34-322)	1.Kankarya, 2.Maninagar	5.54 ±0.56 N=21 (0-6.15)
North	b) Naroda 1. GIDC Away from GIDC	13.24±4.06 N=12 (4.9-20.27) 5.87 ±2.3 N=11 (3.12-9.51)	3. Asarwa, 4.Meghaninagar	5.67 ±0.57 N=21 (0.0-7.01)
East	c) Nicol d) Odhav e) Vatva	18.2±10.14 N=17 (8.8-51.25)	5.Bapunagar, 6.Rakhiyal, 7.Hatkeshwar	5.68 ±0.49 N=13 (5.27-6.84)
West	f) Chandkheda, 1) IOC. Road	24.81±13.62 N=14 (8.36-59.04)	8.Navrangpura, 9.Juhapura, 10 Ranip	4.56± 1.21 N=18 (1.38-5.66)

(All values represent the mean ± GM; Values in parenthesis indicate the range)

A *Pearson product-moment correlation coefficient* was computed to assess the relationship between the Cr (VI), TDS and pH levels of AMC water and bore water of study area (Table 6). In AMC water, there is strong positive correlation between TDS and pH levels of water (r=0.991, n=74, p=0.001). But in bore water we found significant positive correlation between Cr (VI) and TDS levels (r=0.507 n= 105, p=0.001) and also positive correlation with pH levels (r=0.304, n=105, p=0.002).

Table 6: Effects of TDS and pH on the level of Cr (VI) and correlation ship among them

Location	Parameters	TDS (ppm)	pH
AMC	Cr (VI) (ppb)	0.072 (p=0.543, 74)	0.018 (p=0.880, 74)
	TDS	1	0.991 (p=0.001, 74)
Bore	Cr (VI)	0.507** (p=0.001)	0.304 (p=0.002, 105)**
	TDS	1	0.150 (p=0.128, 105)

4. DISCUSSION

Chromium compounds particularly Cr (VI) has high mobility in the environment and has potential to contaminate ground water. The process in soil environment is accompanied by adsorption, dissolution, precipitation etc. that play important role in leaching out Cr (VI) into drinking water. Natural weathering also plays a significant role in these processes. Though concentration of Cr (VI) in drinking water generally ranges from 0.4 to 8.0 µg/L, 10 to 20-folds of it have also been reported in spring water as summarized by WRF [6]. Data obtained in the present study for Cr (VI) concentration indicate that majority of the samples obtained from personal bore water (about 48%) fall in the line up to the level of 10 µg/L, a concentration that has been reported from USA in drinking water from natural sources[2] (ATSDR 2008). Spring water from various regions of the globe had higher levels[6]. It was noted that about 26% of the samples from bore water as well had shown relatively higher levels (10 to 30 µg/L). This level was quite higher when compared to Municipal water supply. Bore water may be considered as the raw water which is not usually filtered, while Corporation water is treated and filtered and required to meet certain norms of WHO standards. Only 7 bore samples out of 290 (about 6%) showed higher levels of Cr (VI) and they exceeded the prescribed limit of WHO (50 µg/L). None of the samples from Municipal Corporation exceeded WHO limit and were well below from the allowable limit of WHO (50µg/L).

Unlike arsenic toxicity, much of the epidemiological studies from chromium exposure are not available. And due to the paucity of database agreeable concessions regarding exposure limit could not be drawn at present. EPA is to set the exposure limit (MCL) by the end of this year, (2014). The current limit of chromium toxicity regulation (50 µg/L) also seems un-appropriate as it has no individual representation of Cr (VI) in directive, which is likely the sole component in drinking water as one of the contaminants. The situation would becomes more worse, if PHG proposed limit (0.02 µg/L) is taken into consideration, which indicates that exposure above 0.02 µg/L may

represent a risk of cancerous and non-cancerous effects. Globally, the advanced technology-updates are not sufficient enough to meet the required demand commercially.

5. CONCLUSION

There have been uncertainties about the ill- health effects of Cr (VI) exposure. The population may experience Cr (VI) - induced effects if continued to the excessive exposure. Preventive measures such as the use of filtered-water should be encouraged and the regulation norms be strictly enforced.

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