STM JOURNALS

Research & Reviews: A Journal of Toxicology
ISSN: 2231-3834(Online), ISSN: 2349-1264(Print)
Volume 10, Issue 1
www.stmjournals.com

Uranium and Heavy Metal Contamination of Sirhind Canal Water and Groundwater in the Malwa Belt of Punjab and its Mitigation Strategies

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Abstract
Inductively coupled plasma mass spectroscopy (ICPMS) has been used to measure the Uranium and heavy metals content of the Sirhind canal water and ground water samples of adjoining villages of the collection points along the canal. The aim of this study is to propose mitigation strategies for Uranium in the groundwater of Malwa belt. The uranium and heavy metal content of the water samples collected at the five different locations downstream along the canal are below the detection limit (BDL), except for the Iron content which is below the acceptable limit in three samples. According to Central Groundwater Board report, 18 out of 22 districts (80% area) of Punjab are affected by Uranium contamination of groundwater. Mitigation strategies for Arsenic, Fluoride and Uranium were planned under the World Bank Project sanctioned to Punjab in 2015.Our investigations suggest that the best solution for the mitigation of Uranium in the groundwater of Malwa belt is to opt for the supply of canal water as a substitute for underground water being supplied through tubewells.

Keywords: *BDL, cancer risk, heavy metals, Sirhind canal, underground water, Uranium content*

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INTRODUCTION

The World Health Organization (WHO) [1] recommended a reference level of 30 μg l^{-1} (ppb) as the permissible limit of Uranium (U) in drinking water. However, the Atomic Energy Regulatory Board (AERB) has recommended 30 ppb as acceptable limit and 60 ppb as permissible limit for India, which is double than the WHO limit. It has been investigated that U accumulation inside the human body beyond the permissible limit set by WHO is a major health hazard due to its chemical and radioactive effects particularly for the kidneys and lungs as the major target organs [2–4]. The major source of supply of the Uranium to the human body is drinking water, which contributes about 85% of ingested Uranium and the rest 15% is contributed by the food intake [5]. The transient chemical damage to the kidneys is due to an exposure of about 0.1 mg/kg of body weight of soluble natural Uranium [6]. Uranium itself is a weak radioactive metal but it may be hazardous to human health if its contamination is high in the drinking water. The assessment of health hazards risk is important if the concentration of Uranium in water and its extent of getting ingested into the human body is higher than the safe limit provided by WHO [1].

Punjab is facing a crisis situation due to high levels of Uranium (U) contamination of groundwater in the Malwa belt of Punjab, Arsenic contamination in the Majha belt, and Selenium contamination in the Doaba belt of Punjab [7]. High toxicity of U in the groundwater of the Malwa belt of Punjab has been reported by Virk [8–15] and some other authors [16–18]. The groundwater contamination is so acute that Public Interest Litigation (PIL) had been filed in the Punjab and Haryana High Court in 2010 by BS Loomba. Directions have been issued by the Hizgh Court to the Union of India and Punjab to explore the possibility of deactivating the Uranium for provision of clean potable water to the public in the Malwa region [19].

Honourable Judges on the bench took suo moto notice [20] of our groundwater contamination report published in The Tribune [21].

The source of groundwater Uranium is yet to be ascertained in a foolproof manner. Some reports have been published to explain Uranium contamination of groundwater in the Malwa belt as follows:

- Guru Nanak Dev University, Amritsar group attributed high U content in water due to leaching of Uranium from basement granite rich rock formations with link to Tosham granite rocks of Haryana [18, 22].
- Punjab University, Chandigarh group reports that high U content in water can be attributed to high salinity of water and Phosphatic fertilizers being used in Malwa region of Punjab. Calcium bicarbonate acts as a leaching agent for U in soil and it gets concentrated in groundwater by geochemical processes [23].
- S. Singh and HS Virk reported U conc. of 612 ppm in Mussoorie phosphorite using f.t. technique [24]. Phosphate fertilizers seem to be plausible cause of U in the groundwater of Punjab.
- Patnaik et al. [25] report that mobilization of Uranium from Siwalik Himalayas is the cause of enhanced geogenic Uranium in the Malwa belt of Punjab.
- Duke University group suggests that the primary source of U is geogenic but anthropogenic factors such as groundwater table decline, and nitrate pollution may further enhance U mobilization [26].

The present report is based on the data collected by the authors from Malwa belt of Punjab and survey carried out by Punjab Water Supply and Sanitation Department (PWSSD), Mohali, Punjab, India [27]from seven districts, namely, Fazilka, Moga, Ferozepur, Barnala, Patiala, Sangrur, and Bathinda of the Malwa belt of Punjab (Figure 1).

THE STUDY AREA AND GROUNDWATER QUALITY

The State of Punjab is part of the Indus River System in the north and north-west of the Indian subcontinent. It is separated from the Ganga basin by the Ghaggar River which flows seasonally in the south-eastern parts of the state. As a consequence of diversity in the natural environment like climate, topography, parent rocks, drainage and vegetation cover spread over a span of time, the soils of Punjab developed largely on alluvium, vary widely and show difference in their nature, properties and profile development. The Punjab State is mainly underlain by quaternary alluvium of considerable thickness, which abuts against the rocks of Siwalik system towards North-East [28]. The British laid a network of canals to irrigate Punjab landmass in undivided India. After 1947, major canals were left in Pakistan and Indian Punjab dug up Bhakra canal to augment the supply of water being released in the Sirhind canal system. The Sirhind canal and its distribution network are spread over a length of 3215 km. The system's headworks, where it draws its water, are on the Sutlej River at Ropar. The main source of water in the Sutlej River originates in Himachal Pradesh (HP).

The State of Punjab was at the forefront of ushering in Green revolution in 1969. The target of achieving National Food Security also involved the use of large quantity of fertilizer, pesticides, insecticides, weedicides, use of hybrid varieties of seeds and excessive extraction of ground water. Thus, Punjab became the hub of agricultural activity. Over the years, the agricultural practices have changed the cropping pattern on an extensive scale which has made the agro-ecosystem of the State extremely fragile in the context of water depletion, soil health, weeds and pest, human health and overall living environment [29]. Due to overexploitation of groundwater and cumulative effects of fertilizers, pesticides, weedicides etc., Punjab's Malwa belt has become 'Cancer Capitol' of India.

METHODOLOGY USED FOR SAMPLE COLLECTION AND ANALYSIS

For collection of samples, 20 ml bottles of superior quality plastic are used. The bottles are washed first with soap solution and then with distilled water. These are rinsed with deionised water and dried. Canal water

***Fig. 1:** District Map of Punjab showing Seven Districts of Malwa Belt.*

samples were collected from different spots along the Sirhind canal and its main branches downstream from its origin. The depth of water collection varied from 2 to 6 feet from the surface. Groundwater samples were collected from handpumps or tubewells along the banks of the canal adjoining the spots from where canal water was collected.

Groundwater from the source is allowed to flow freely before collection in plastic bottles. 10–20 ml of water is collected from the running water source. For dissolved metal determinations, samples must be filtered through a 0.45-µm capsule filter at the field site. Nitric acid (0.5M HNO3) solubilization is required before the determination of total recoverable Uranium. The preservation and digestion of Uranium in acid is used in order to aid breakdown of complexes and to minimize interferences by poly-atoms.

The Uranium and heavy metal analysis of collected water samples has been done using Model 7700 Agilent Series ICP-MS following standard procedure in the Punjab State laboratory set up in Mohali. The method measures ions produced by a radiofrequency inductively coupled plasma. Analyte species originating in a liquid are nebulized and the resulting aerosol is transported by Argon gas into the plasma torch. The ions produced by high temperatures are entrained in the plasma gas and introduced, by means of an interface, into a mass spectrometer. The ions produced in the plasma are sorted according to their mass-to-charge ratios and quantified with a channel electron multiplier. Interferences must be assessed, and valid corrections applied. Interference correction must include compensation for background ions contributed by the plasma gas, reagents, and constituents of the sample matrix.

A mass spectrometer with inductively coupled plasma (ICP) suitable for multi-element and isotope analysis is required. The spectrometer should be capable of scanning a mass range from 5 m/z (AMU) to 240 m/z (AMU) with a resolution of at least 1 mr/z peak width at 5% of peak height (mr = relative mass of an atom species; z = charge number). The instrument may be fitted with a conventional or extended dynamic range detection system. Most quadrupole ICP-MS, high-resolution ICP-MS and collision cell ICP-MS instrumentation is fit for this purpose. Data analysis is done automatically by inbuilt system of ICP-MS. In addition to Uranium, data for 40 more trace elements can be retrieved using ICP-MS.

RESULTS AND DISCUSSION

Ingestion of the Uranium through drinking water results in both the radiological risk (carcinogenic) and chemical risk (noncarcinogenic). The methodology used for the assessment of the radiological and chemical risks due to Uranium concentrations in the water samples and the associated risk factors is described elsewhere [8]. The range of Uranium content in the groundwater of seven districts of the Malwa belt is given in Table 1. It is observed that groundwater of Malwa districts of Punjab is highly contaminated as compared with other districts of Punjab. The highest numbers of water quality affected habitations are found in Fazilka district followed by Moga, Ferozepur, Barnala, Patiala, Sangrur and Bathinda districts.

The Uranium content variation does not show very high fluctuations from district to district. Its highest value of 366.0 ppb has been recorded in the tubewell of village Kotha *alias* Lakhman Pura in Fazilka district. Excess cancer risk corresponding to the highest Uranium contamination is also listed in Table 1. It is alarming for inhabitants of village Kotha with a probability of 10 cancers per 10,000 population. The mean value of excess cancer risk in the surveyed habitations of this district comes out to be 5.01×10^{-4}, which works out to be five per 10,000 people [8].

Table 2 records the Uranium and heavy metals content in the surface water flowing in the Sirhind canal passing through the terrain of four districts of Malwa belt. The results are obtained using Model 7700 Agilent Series ICP-MS following standard procedure in the Punjab State laboratory set up in Mohali. The values of Uranium, Arsenic, Lead, Nickel, Cadmium and Chromium are below the detection limit. The acceptable limit (AL) for Iron is 1.0mg/l (1ppm) in India. Hence the Iron contamination recorded in three canal water samples is much below the AL.

Table 3 records the Uranium and heavy metals content in the groundwater of villages situated near the spots from where canal water was collected for this study. Surprisingly, the heavy metals contents are below the detection limit for Arsenic, Lead, Iron, Nickel, Cadmium and Chromium but Uranium contamination is found above the detection limit in seven samples out of eight. The highest U content of 85.48 ppb, which is above the permissible limit, is found in the groundwater of Bhure village of Barnala district in the Malwa belt. Comparison of Tables 2 and 3 shows that Sirhind canal water is safe for drinking purpose as compared with groundwater of Malwa belt.

Table 4 is a repeat survey of Sirhind canal water from five different spots. The analysis was done using ICP-MS facility of Punjab Agricultural University (PAU), Ludhiana. Overall, fifteen elements were analysed in the canal water but results for ten heavy metals are recorded in Table 4. It is observed that heavy metals, Manganese, Iron, Lead, Copper, Cobalt, Zinc, Nickel, Arsenic, Cadmium and Chromium, contents are either negligible or below the acceptable limit. These samples were not analysed for Uranium as our previous analysis (Table 2) of canal water has established that its presence is below 5 ppb, which is the detection limit of the instrument. We intend to extend this survey to the tributaries of the Sirhind canal in the Malwa belt.

Central Ground Water Board (CGWB) and Duke University reported [26] that 18 districts out of 22, covering 80 percent area of Punjab, are affected by groundwater contamination of Uranium, which is highest in India. It is most exhaustive survey of Uranium contamination of Groundwater Resources in India.

Table 1: *Uranium Content in Groundwater of Malwa Belt of Punjab.*

S.N.	Name of District	No. of Affected Habitations	Source of Groundwater	Uranium Range (ppb)	Excess Cancer Risk x 10^{-4}
1	Fazilka	217	Tubewell/Handpump	30.0–366.0	10.37
2	Moga	203	do	30.0–346.7	9.82
3	Ferozepur	139	do	30.0–331.4	9.39
4	Barnala	115	do	30.0–290.6	8.23
5	Patiala	96	do	30.0–267.0	7.56
6	Sangrur	93	Tubewell	30.0–230.3	6.52
7	Bathinda	52	Tubewell/Handpump	30.0–325.1	9.21

Table 2: *Uranium and Heavy Metals Contents in the Surface Water of Sirhind Canal.*

S.N.	District	Village near testing point	Uranium(ppb)	Lead	Iron(ppm)	Arsenic	Nickel	Chromium	Cadmium
1	Sangrur	Jahangir	BDL	BDL	BDL	BDL	BDL	BDL	BDL
2	Sangrur	Ballian	BDL	BDL	0.010	BDL	BDL	BDL	BDL
3	Barnala	Mander Khurd	BDL	BDL	BDL	BDL	BDL	BDL	BDL
4	Mansa	Rallan	BDL	BDL	0.020	BDL	BDL	BDL	BDL
5	Bathinda	Kotli Kalan	BDL	BDL	0.023	BDL	BDL	BDL	BDL

BDL- Below detection limit; MDL- Method detection limit (MDL of Iron is 0.010 ppm).

Table 3: *Uranium and Heavy Metals Contents in Groundwater Samples of Villages Situated along the Sirhind Canal in the Malwa Belt of Punjab.*

S.N.	District	Village	Uranium(ppb)	Lead	Iron	Arsenic	Nickel	Chromium	Cadmium
1	Sangrur	Jahangir	BDL	BDL	BDL	BDL	BDL	BDL	BDL
2	Sangrur	Dandiwal	6.87	BDL	BDL	BDL	BDL	BDL	BDL
3	Barnala	Bhure	85.48	BDL	BDL	BDL	BDL	BDL	BDL
4	Barnala	Mander Khurd	13.55	BDL	BDL	BDL	BDL	BDL	BDL
5	Barnala	Pandher	39.21	BDL	BDL	BDL	BDL	BDL	BDL
6	Mansa	Bhaini Bhaga	25.63	BDL	BDL	BDL	BDL	BDL	BDL
7	Mansa	Mohr Khurd	5.86	BDL	BDL	BDL	BDL	BDL	BDL
8	Bathinda	Pakka Khurd	10.56	BDL	BDL	BDL	BDL	BDL	BDL

BDL- Below detection limit; MDL- Method detection limit (MDL of Uranium is 5 ppb).

Table 4: *Heavy Metals Contents in the Surface Water of Sirhind Canal Downstream.*

S.N.	Heavy Element	Gurthali (Ludhiana)	Sirthala (Ludhiana)	Jabomajra (Sangrur)	Mahorana (Sangrur)	Babanpur (Sangrur)
1.	Chromium	0.000	0.000	0.001	0.000	0.000
2	Manganese	0.002	0.004	0.002	0.010	0.004
3	Iron	0.031	0.012	0.016	0.012	0.036
4	Cobalt	0.000	0.000	0.000	0.000	0.000
5	Nickel	0.001	0.001	0.001	0.001	0.001
6	Copper	0.004	0.002	0.002	0.002	0.001
7	Zinc	0.002	0.001	0.001	0.001	0.001
8	Arsenic	0.003	0.003	0.003	0.003	0.002
9	Cadmium	0.000	0.000	0.000	0.000	0.000
10	Lead	0.000	0.000	0.000	0.000	0.000

Uranium Mitigation Strategies in Punjab

Mitigation of Uranium in groundwater is a global problem. Uranium is toxic to humans, as well as animals, due to its radioactive and heavy metal nature. It is an emerging challenge to environmental scientists to prevent the spread of Uranium in the environment. Removal of Uranium (VI) by zerovalent iron has been suggested as a feasible pathway to control Uranium contaminations in seepage waters. Available information in the literature, however, presents discrepant evidence on the process responsible for the mitigation effect. On basis of an EH-pH diagram of Uranium and Iron, it is outlined that these discrepancies may be explained by the aqueous chemistry of Uranium and Iron [30].

Several methods are available for the removal of Uranium from drinking-water, although some of these methods have been tested at laboratory or pilot scale only. Coagulation using ferric sulphate or aluminium sulphate at optimal pH and coagulant dosages can achieve 80–95% removal of Uranium, whereas at least 99% removal can be achieved using lime softening, anion exchange resin or reverse osmosis (RO) processes.

Microbial reduction of Uranium is still a hot topic in the field of bioremediation. Novel bacteria have been isolated and characterized for U(VI) reduction, and investigations have been carried out to study the molecular genetic mechanisms involved in the microbial reduction of U(VI) [31]. Many microbes are proved to be capable of reducing highly soluble U(VI) to sparingly insoluble U(IV) form and precipitate the reduced Uranium as mineral uraninite. These microbial processes might be an ideal solution to clean-up Uranium contaminated environments, because of their relatively low cost and minimum environmental disruption [32], but most of these are at pilot study level.

Punjab state report published in The Tribune [33] mentions about supply of canal water for 340 villages affected by Arsenic and Fluoride under the World Bank Project of Rs. 378 crores. There is no mention of any such measures for mitigation of Uranium contamination of groundwater in the Malwa belt. The World Bank Report [34] "Towards Managing Rural Drinking Water Quality in the State of Punjab, India", approved in 2015 with a grant of 248 million US Dollars for improvement of water quality, classified contaminants in three categories:

Group 1 contaminants Arsenic, Uranium and Fluoride have geogenic origins, with over-abstraction of groundwater increasing the mobilization of these contaminants.

Group 2 contaminants are anthropogenic in origin, namely Lead, Selenium, Mercury, Nickel, Chromium, Aluminium, Iron, Nitrate, total dissolved solids, pesticides.

Group 3 are biological contaminants or pathogens, which continue to be a major health risk in the State.

This report states: "Arsenic and Fluoride present the most urgent risk, surpassing the risk of Uranium, with the duration of exposure increasing the health impacts". The fact of matter is that Department of Water Supply and Sanitation (DWSS), Punjab in its report [35] "Water Quality Mitigation Strategy" gives top priority to mitigation of mandatory parameters, namely, Arsenic, Fluoride, TDS, Nitrate and Iron. Uranium, Aluminium, Selenium and other heavy metals are classified under 'Emerging Parameters' and given lesser importance for purpose of mitigation.

DWSS report mentions that out of 1540 habitations, affected with Emerging Parameters, 494 habitations are covered/being covered with RO plants, 34 are being covered under surface water projects. And thus, a balance of 1012 habitations affected with Emerging Parameters is left to be covered. In the year 2019–20, 15 habitations affected with Mercury, 113 habitations affected with Uranium and 16 habitations affected with Selenium will be provided with treatment technologies [35]. Since the World Bank Project is coming to its end during December 2019, it is obvious that DWSS is lagging far behind its targets of mitigation of Uranium in the groundwater of Malwa belt.

CONCLUSIONS

1. Uranium contamination is highest in the Malwa belt of Punjab and it must be given top priority in mitigation programmes of DWSS of Punjab State.
2. Our investigations reveal that Uranium and heavy metals are either absent or present below the detection limit in the surface water flowing in the Sirhind canal in the Malwa region of Punjab state.
3. Supply of canal water for purposes of drinking and irrigation is the best option available to mitigate the Uranium contamination problem of groundwater in the Malwa belt.
4. Reverse Osmosis (RO) process may be continued as an alternative option in the Malwa belt.
5. DWSS proposal for digging deep submersible tubewells with depth >200feet to supply Uranium free water in the Malwa belt may be discarded as an option.

ACKNOWLEDGEMENTS

One of the authors, Savneet Kailley, is grateful to the authorities of Desh Bhagat University, Gobindgarh for permission to undertake research investigations in this area. The authors are thankful to Secretary, Punjab Water Supply and Sanitation Department, Govt. of Punjab and Director Water Quality Mohali, for supply of Uranium data of Malwa belt and copy of World Bank Project Report. They acknowledge help of technical staff of PAU, Ludhiana and DWSS, Mohali for analysis of water samples.

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Cite this Article
Savneet Kaur Kailley, Hardev Singh Virk. Uranium and Heavy Metal Contamination of Sirhind Canal Water and Groundwater in the Malwa Belt of Punjab and its Mitigation Strategies. *Research & Reviews: A Journal of Toxicology*. 2020; 10(1): 34–41p.