

Uranium Estimation in the Drinking Waters of Punjab State, India: A Survey

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Abstract: Uranium is a ubiquitous element and is present in rocks, soil, natural materials, and in small quantities in air, water and food. The biokinetics, metabolism, and chemical toxicity of uranium including the toxic effects of this metal on kidney functions are well established. The United States Environmental Protection Agency has classified uranium as a confirmed human carcinogen and has suggested that zero tolerance only is a safe acceptable limit for the carcinogenic risk. High uranium concentrations in some drinking waters of Punjab State, India were reported for the first time in 1995; since then many surveys have been carried out. These surveys indicate wide variation in the uranium concentration in drinking waters. At some locations very high values upto 644µg/l have been reported which crosses the guideline value of 30µg/l by the International Agencies. The results of some of these surveys of Punjab are presented in the paper. Also the results of a recent survey carried out in the Amritsar District of Punjab State are presented where the uranium concentration varies from a minimum of 5.5µg/l to 135.5µg/l.

Keywords: Uranium, Drinking Water, Punjab, India.

1. Introduction:

Uranium occurs naturally in the +2, +3, +4, +5 and +6 valence states, but is most commonly found in the hexavalent form. Naturally occurring uranium is a mixture of three radionuclides (^{234}U , ^{235}U and ^{238}U), all of which decay by both alpha and gamma emissions [1,2] and it consists almost entirely of the ^{238}U isotope, with the ^{235}U and ^{234}U isotopes respectively comprising about 0.72% and 0.0054% [3]. Uranium is widespread in nature; occurring in granites and various other mineral deposits [2,4].

Uranium is present in the environment as a result of leaching from natural deposits, release in mill tailings, emissions from the nuclear industry, the combustion of coal and other fuels and the use of phosphate fertilizers that contain uranium. Its presence in drinking water is most commonly from natural sources. Intake of uranium through air is extremely low, and it appears that intake through food is between 1 and 4µg/day. Intake through drinking water is normally low; however, in circumstances in which uranium is present in a drinking water source, the majority of intake can be through drinking water [5].

Although ubiquitous in the environment, uranium has no known metabolic function in animals and is currently regarded as non-essential [6]. The average human gastrointestinal absorption of uranium is 1-2% [7]. Following ingestion, uranium rapidly appears in the blood stream [8], where it is associated primarily

with the red cells [9]. Clearance from the bloodstream is also rapid, and the uranium subsequently accumulates in the kidneys and the skeleton, whereas little is found in the liver [7]; the uranyl ion replaces calcium in the hydroxyl apatite complex of the bone crystal [10].

Nephritis is the primary chemically induced effects of uranium in humans [11]. A survey on two groups of subjects with chronic exposure to uranium in drinking water, one exposed to less than 1µg/l and the other exposed to 2-781µg/l, was carried out in Canada [12]. This study found no correlation between exposure and albumin in urine, but it did find a correlation with alkaline phosphatase and β_2 -microglobulin in urine. This study concluded that the concentrations observed in the study affected kidney function at the proximal tubule. In a study carried out in Finland [13,14,15,16] on people who consumed drinking water with elevated levels of uranium concentrations has found that in addition to kidneys, bone may be another target of chemical toxicity of uranium in humans.

2. Guideline values for uranium concentration in drinking water:

The international and national agencies have given guideline values for uranium concentration in drinking water which have been revised from time to time as more evidence appears from the new research. The World Health Organization (WHO) [5] has given a provisional guideline value of 30µg/l as the tolerable daily intake (TDI). This value is derived on the

epidemiological studies on population exposed to high uranium concentration and is based on the consumption of 2 litres of drinking water per day which supersedes the earlier value of TDI as 15µg/l [17] and was based on animal experiments.

Health Canada [18] has given an Interim Maximum Acceptable Concentration (IMAC) as 20µg/l. The Environmental Protection Agency (EPA) of USA has given the Maximum Contamination Level (MCL) for naturally occurring uranium as 30µg/l [19]. The Atomic Energy Regulatory Board of the Department of Atomic Energy of India has given a TDI value of 60µg/l for drinking water [20].

3. Surveys of the World:

In most natural waters, uranium is present in concentrations between 0.1 and 10µg/l and concentrations greater than 1mg/l can occur in water associated with uranium ore deposits [21].

In one of the very early surveys carried out in the region of Helsinki of Finland on drilled wells,

uranium concentration as high as 14,870µg/l was observed [22]. Since then, many surveys for the measurement of uranium content in water and/or its health effects on animals/ humans are being carried out all over the world [12, 13, 14, 15, 16, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32 and 33].

4. Surveys of the Punjab State, India:

The district map of the Punjab State, India is shown in Fig. 1. Punjab is located in the northwest of India between latitude 29°30'N – 32°32'N and longitude 73°55' – 76°50'E at an average elevation of 300m above sea level and between the great river systems of the Indus and the Ganges. The soil of Punjab is loose, sandy and calcareous and alluvial, which is an admixture of gravel, sand, silt and clay in various proportions. The land is predominantly used for agriculture. Hydrogeology of this region is characterized by unconfined and semi-confined aquifers at a depth of 2.3 to 23.3m below ground level.



Fig. 1: District map of Punjab State, India

High uranium content (up to 113.7µg/l) in the drinking waters of some southern parts (Bathinda district) of Punjab were reported for the first time in 1995 [34]. A survey carried out by Mukesh et al., in 2003 [35] in this area has reported uranium concentration varying from 1.65 µg/l to 74.98µg/l. In another survey, Rani and Singh in 2006 [36] has reported uranium values from 1.39 µg/l to 98.25µg/l in Punjab. The problem further came into lime light when in March 2009, Ms. Carin Smit, a clinical metal toxicologist, associated with the UK-based non-government organization, Defeat Autism Now and Vera Dirr, a specialist alarmed the Indian government after observing a high incidence of abnormalities in local children at the Baba Farid Center for special children in Faridkot (Southern Punjab), a not for profit organization working with kids, ailing from autism, cerebral palsy and other neurological disorders. 149 hair samples were collected to evaluate the trace and toxic metal concentration from Microtrace Mineral Laboratory, Germany. The hair of the children tested showed high values for Ba, Cd, Mn, Pb and U signifying long-term exposure. Testing of six randomly selected water samples showed concentration above the European maximum contaminant level for uranium in three samples and lead in one [37]. 88% of the hair samples of the children below the age 12 years exceeded the uranium reference range [38].

After coming these reports in the media, Bhaba Atomic Research Centre (BARC), Mumbai signed a MOU with Physics Department of Guru Nanak Dev University, Amritsar to carry out a joint venture for collection and analysis of water samples for uranium

concentrations. In the initial studies total 235 samples were collected from the four districts; Bathinda, Mansa, Faridkot and Firozpur of Southern Punjab. One of the author has participated in these investigations and the results have been published [39]. Some of the results of this work are presented in Table 1.

The uranium concentration varied from < 2µg/l to 644µg/l with an average of 73.1µg/l. In Bathinda, Mansa, Faridkot and Firozpur 50%, 42.5%, 36.7% and 31.1% of the water samples crossed the guide line value of 60µg/l set by the Atomic Energy Regulatory Board of the Department of Atomic Energy, India [20]. These studies clearly demonstrate the need for further investigations.

A study carried out by Alrakabi et al. [40] in the Bathinda district has found uranium concentrations from < 2 to 212µg/l. The state government at its own level has also got analysed hundreds of water samples from BARC collected from different locations, many of which exceed the guideline value for uranium concentration as is evident from the news appearing from time to time in the print media. The TDS values are also high in many parts and the water is not potable. Taking into consideration a serious problem, the state government is going to install Reverse Osmosis (RO) systems at the village level. A specialized laboratory equipped to check contamination of groundwater by uranium and other heavy metals is also being established at Mohali in Punjab.

Table 1: Uranium concentration of subsurface water in four districts (Bathinda, Mansa, Faridkot and Firozpur) of Punjab State in India (all data in µg/l) [39]

District (No. of Samples)	Mean concentration of U (µg/l)	Minimum	Maximum
Bathinda (80)	80.83 ± 5.3	2	644
Mansa (80)	75.83 ± 6.7	< 0.2	498
Faridkot (30)	68.56 ± 8.3	16.5	473.5
Firozpur (45)	62.12 ± 7.3	2.8	184.4

5. A recent survey in the Amritsar District, Punjab State:

Recently we have carried out a survey in the Amritsar district of Punjab State for uranium estimation in the ground water. The experimental procedure, results and discussions are given below:

5.1 Materials and Methods

The water samples were collected with 1L-size plastic containers which have been previously washed and rinsed with acid. The collected samples were acidified with 0.01M of nitric acid to obtain a pH < 2 in order to avoid adsorption of the radionuclides on the walls of the container. In total we have collected 30 samples from different parts of the Amritsar district. The sources comprise handpumps and borewells used for drinking and agricultural purposes. The depth of the sources was from 50ft – 500ft.

5.2 Experimental

The uranium concentration was measured by Laser Fluorimeter. It is a compact analytical instrument to measure uranium in trace and ultra trace levels in aqueous environmental samples, since the concentration of uranium in aquatic environment is very low, may be in ppb or sub-ppb level. The method is based on luminescence of the uranyl ion in solution and fluorescence measurement technique is fabricated by Laser Instruments Sections of Raja Ramana Centre of Advanced Technology, Department of Atomic Energy, India. Laser fluorimetry is one of the quick, sensitive and reliable methods of estimating ultratrace concentrations, though having the difficulties of measurements in higher concentrations and devoid of isotopic determination. The instrument consists of a sealed-off molecular nitrogen laser tube as the excitation source lasing at 337.1 nm, along with sample compartment and a photomultiplier tube, as a radiation transducer. The detector is present at right angle to the radiation source, thus the background signal due to the source will be negligible and a lower detection level can be achieved than atomic absorption or emission techniques. The laser source emits the intense pulsed ultra violet radiation carrying energy of 20 micro joules

of pulse width of 7 nano seconds at a repetition rate of 10 pulses per second. The 337.1 nm output of the laser is focused onto the sample cell made up of suprasil quartz (9ml) that excites the uranium complex compounds, formed when sodium pyrophosphate (fluorescence enhancing agent) is added to the sample solution along with organic matter present in the samples. The fluorescence of uranyl ion is measured by a photomultiplier tube (PMT). The fluorescence of organic matters excited by the nitrogen laser have very short lifetime (< 100 ns) compared to that of uranyl complex (~ 200 μ s). By measuring the delayed fluorescence signal (only for a 100 μ s period starting from 30 μ s after the laser pulse) and fluorescence signals from the PMT are integrated for 4s and then displayed. Thus the fluorescence due to organic compounds dies down and only the fluorescence of uranium isotopes is collected by the time gated PMT. The fluorescence is proportional to concentration of uranium in aqueous medium. Otherwise the fluorescence peak of organic matters is around 400 nm and that of uranium in complexing media are 494, 516, 540 and 565 nm, maximum around 510 nm. Therefore, using an optical filter at 450nm, the contribution of organic matters is nullified. Thus the fluorescence interference due to organic matters is corrected by using proper optical filter, fixed electronic delay and gating technology.

5.3 Results and discussion

The results of the present investigations are presented in Table 2. We find that the uranium concentration varies from a minimum of 5.5 μ g/l to a maximum of 135.5 μ g/l with a mean value of 41.1 μ g/l. Out of 30 samples, 11 samples cross the maximum contamination level (MCL) of 30 μ g/l set by WHO [5] and EPA [19], and 7 samples cross the MCL of 60 μ g/l set by Department of Atomic Energy, India [20]. This distribution is shown as a pie chart in Fig 2. It is also seen from the table that in general water samples from a depth of more than 220ft are having lower concentration of uranium as compared to the water samples which are from a depth of upto 120ft. So it is concluded that the depth of the water source also plays an important role in the variation of uranium content in drinking water.

Table 2. Uranium concentration in various drinking water samples of Amritsar District, Punjab State, India.

S No.	Sample Code	Depth (ft)	Uranium Concentration (µg/l)
1.	AT-1	150	12.1
2.	AT-2	90	23.8
3.	AT-3	110	91.0
4.	AT-4	110	97.5
5.	AT-5	50	15.8
6.	AT-6	200	62.2
7.	AT-7	-	9.6
8.	AT-8	-	8.0
9.	AT-9	110	27.3
10.	AT-10	415	8.8
11.	AT-11	100	56.6
12.	AT-12	-	22.5
13.	AT-13	100	10.7
14.	AT-14	100	8.8
15.	AT-15	220	9.5
16.	AT-16	180	14.2
17.	AT-17	150	57.1
18.	AT-18	80	52.5
19.	AT-19	400	5.5
20.	AT-20	500	15.7
21.	AT-21	500	17.5
22.	AT-22	-	26.5
23.	TT-1	120	111.3
24.	TT-2	120	132.0
25.	TT-3	120	95.7
26.	TT-4	120	135.5
27.	TT-5	80	59.6
28.	TT-6	350	19.3
29.	UWS	450	12.0
30.	NMP	450	13.5

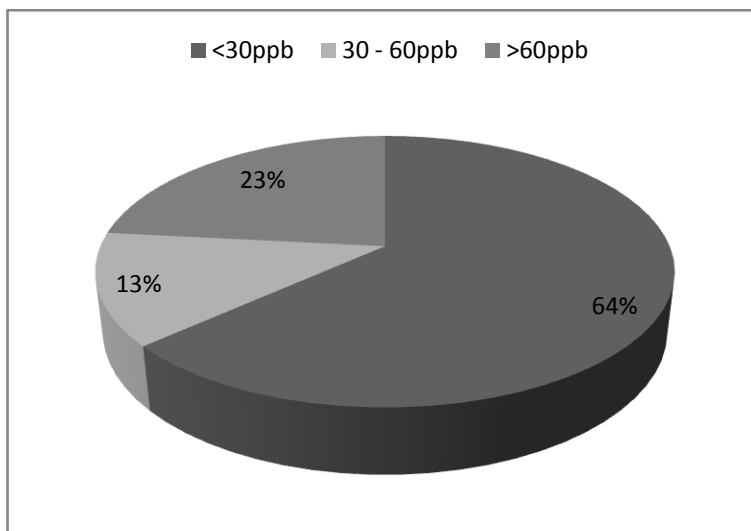


Fig 2. The percentage of water samples having uranium concentration comparable to the national and international guidelines.

6. Conclusions:

Uranium in drinking water may pose health effects if present in high concentrations. After the first reporting on high uranium concentrations in drinking water of Bathinda district of Punjab state in 1995, many surveys have been carried out by different research groups from the universities, by Bhaba Atomic Research Centre (BARC), Mumbai and by the state government. In a survey carried out jointly by BARC and the Guru Nanak Dev University, Amritsar in the malwa region of Punjab State (Bathinda, Mansa, Faridkot and Firozpur districts), again high uranium concentrations (< 2 to 644 $\mu\text{g/l}$) have been observed in many samples which confirm the earlier findings of this area. These values cross the guideline value of 30 $\mu\text{g/l}$ set by the World Health Organization (WHO) and the Environmental Protection Agency (EPA) of USA, as well as the guideline value of 60 $\mu\text{g/l}$ prescribed by the Atomic

Energy Regulatory Board of the Department of Atomic Energy, India (AERB, DAE). In a recent survey carried out in the Amritsar district of Punjab State, the uranium concentration varies from 5.5 $\mu\text{g/l}$ to 135.5 $\mu\text{g/l}$ with a mean value of 41.1 $\mu\text{g/l}$. Out of the 30 samples, 11 samples cross the guideline value of 30 $\mu\text{g/l}$. In this survey it is also observed that more the depth of the water source, lesser is the uranium content in the water samples of this area.

The state and the central governments are trying to provide clean drinking water to the public. More surveys are required in the areas which have not been covered in the previous surveys.

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References

- [1] C. R. Cothorn and W. L. Lappenbusch, "Occurrences of uranium in drinking water in the US", *Health Physics*, 45, 89-99, 1983.
- [2] D. R. Lide, (ed). *Handbook of chemistry and physics*. CRC Press, Boca Raton, FL., 1993.
- [3] N. N. Greenwood, A. Earnshaw. *Chemistry of the elements*. Oxford, Pergamon Press. 1984.
- [4] C. E. Roessler, Z. A. Smith, W. E. Bolch and R. J. Prince, "Uranium and radium-226 in Florida phosphate materials", *Health Physics*, 37, 267-269, 1979.
- [5] WHO, World Health Organization. "Uranium in drinking water, Background document for development of WHO guidelines for drinking-water quality", WHO/SDE/WSH/03.04/118/Rev/1, 2012.
- [6] M. Berlin and B. Rudell, "Uranium In: Handbook on the toxicology of metals. 2nd edition." L. Friberg, G. F. Nordberg and V. B. Vouk(eds). Elsevier Science Publishers, Amsterdam, 623-637, 1986.
- [7] M. E. Wrenn, P. W. Durbin, B. Howard, J. Lipsztein, J. Rundo, E. T. Still and D.L. Willis, "Metabolism of ingested U and Ra", *Health Physics*, 48(5), 601-633, 1985.
- [8] Y. D. La Touche, D. L. Willis, O. I. Dawydiak, "Absorption and biokinetics of U in rats following an oral administration of uranyl nitrate solution", *Health Physics*, 53(2), 147-162, 1987
- [9] I. M. Fisenne and P. M. Perry, "Isotopic U concentration in human blood from New York city donors", *Health Physics*, 49, 1272-1275, 1985.
- [10] M. A. Moss, "Chronic low level uranium exposure via drinking water- clinical investigations in Nova Scotia. Halifax, Nova Scotia, Dalhousie University (MSc. Thesis), 1985.
- [11] J. B. Hursh and N. L. Spoor, "Data on man. In: Handbook of Experimental Pharmacology. Uranium, Plutonium Transplutonic Elements". H. C. Hodge, J. N. Standard, and J. B. Hursh (eds), Springer- Verlag, New york, 197-240, 1973.
- [12] M. L. Zamora, B. L. Tracy, J. M. Zielinski, D. P. Meyerhof and M. A. Moss, "Chronic ingestion of uranium in drinking water: a study of kidney bioeffects in humans", *Toxicology Sciences*, 43, 68-77, 1998.
- [13] P. Kurttio, A. Auvinen, L. Salonen, H. Saha, J. Pekkanen, S. B. Makelainen, I. M. Penttila and H. Komulainen, "Renal effects of uranium in drinking water", *Environmental Health Perspectives*, 110, 337-342, 2002.
- [14] P. Kurttio, H. Komulainen, A. Leino, L. Salonen, A. Auvinen and H. Saha, "Bone as a possible target of chemical toxicity of natural uranium in drinking water" *Environmental Health Perspectives*. 113, 68-72, 2005.
- [15] P. Kurttio, A. Harmoinen, H. Saha, L. Salonen, Z. Karpas, H. Komulainen and A. Auvinen, "Kidney toxicity of ingested uranium from drinking water" *American Journal of Kidney Disease*. 47(6), 972-982, 2006.
- [16] P. Kurttio, L. Salonen, T. Ilus, J. Pekkanen, E. Pukkala and A. Auvinen, "Well water radioactivity and risk of cancers of the urinary organs" *Environmental Research*, 102, 333-338, 2006.
- [17] WHO, World Health Organization. "Uranium in drinking water, Background document for development of WHO guidelines for drinking-water quality". WHO/SDE/WSH/03.04/118, 2004.
- [18] Health Canada, "Guidelines for Canadian drinking water quality, summary table", prepared by federal-provincial territorial committee on drinking water of the federal-provincial territorial committee on health and environment, 2010.
- [19] Environmental Protection Agency of the United States of America (US EPA), "2011 edition of the drinking water standards and health advisories", EPA 820R-11-002, office of water, US EPA, Washington, DC, 2011.
- [20] AERB, DAE., Atomic Energy Regulatory Board, Department of Atomic Energy, Mumbai, India. Drinking water specifications, 2004.
- [21] J. K. Osmont and J. B. Cowart, Ground water In: Iranovich M and Harman RS (eds), Uranium – series disequilibrium: Applications to Earth, Marine and Environmental Sciences, pp 290 - 333. Clarendon Press, Oxford, UK, 1992.
- [22] M. Asikainen and H. Kahlos, "Anomalously high concentrations of uranium, radium and radon in water from drilled wells in the Helsinki region", *Geochimica et Cosmochimica Acta*, 43, 1681-1686, 1979.
- [23] A. Auvinen, P. Kurttio, J. Pekkanen, E. Pukkala, T. Ilus and L. Salonen, "Uranium and other natural radionuclides in drinking water and risk of leukemia: a case-cohort study in Finland", *Cancer Causes and Control*, 13, 825-829, 2002.

- [24] A. Auvinen, L. Salonen, J. Pekkanen, E. Pukkala, T. Ilus, and P. Kurttio, "Radon and other natural radionuclides in drinking water and risk of stomach cancer: a case-cohort study in Finland", *International Journal of Cancer*, 114, 109-113, 2005.
- [25] A. P. Gilman, D. C. Villeneuve, V. E. Secours, A. P. Yagminas, B. L. Tracy, J. M. Quinn, V. E. Valli, R. J. Wiles and M. A. Moss, "Uranyl nitrate: 91-day exposure and recovery studies in the male New Zealand white rabbit", *Toxicol. Sci.* 41, 138-151, 1998.
- [26] G. Jia and G. Torri, "Estimation of radiation doses to members of the public in Italy from intakes of some important naturally occurring radionuclides in drinking water". *Applied Radiation and Isotopes*. 65, 849-857, 2007.
- [27] Z. Karpas, O. Paz-Tal, A. Lorber, L. Salonen, H. Komulainen, A. Auvinen, H. Saha and P. Kurttio, "Urine, hair and nails as indicators for ingestion of uranium in drinking water", *Health Physics*, 88(3), 229-242, 2005.
- [28] R. Kleinschmidt, J. Black and R. Akber, "Mapping radioactivity in groundwater to identify elevated exposure in remote and rural communities", *Journal of Environmental Radioactivity*, 102, 235-243, 2011.
- [29] J. Kronfeld, D. I. Godfrey-Smith, D. Johannessen and M. Zentilli, "Uranium series isotopes in the Avon Valley, Nova Scotia", *Journal of Environmental Radioactivity*, 73, 335-352, 2004.
- [30] X. Ortega, I. Valles and I. Serrano, "Natural radioactivity in drinking water in Catalonia (Spain)", *Environment International*, 22(1), S347- S354, 1996.
- [31] R. C. Ramola, S. Singh and H. S. Virk, "Uranium and radon estimation in some water samples from Himalayas", *Nuclear Tracks and Radiation Measurements*, 15(1-4), 791-793, 1988.
- [32] S. K. Sahoo, S. Mohapatra, A. Chakrabarty, C. G. Sumesh, V. N. Jha, R. M. Tripathi and V. D. Puranik, "Distribution of uranium in drinking water and associated age-dependent radiation dose in India", *Radiation Protection Dosimetry*, 136(2), 108-113, 2009.
- [33] M. L. Zamora, J. M. Zielinski, G. B. Moodie, R. A. F. Falcomer, W. C. Hunt and K. Capello, "Uranium in drinking water: renal effects of long-term ingestion by an aboriginal community", *Archives of Environmental and Occupational Health*, 64(4), 228-241, 2009.
- [34] J. Singh, L. Singh and S. Singh, "High U-contents observed in some drinking waters of Punjab, India", *Journal of Environmental Radioactivity*. 26, 217-222, 1995.
- [35] M. Kumar, A. Kumar, S. Singh, R. K. Mahajan and T. P. S. Walia, "Uranium content measurement in drinking water samples using track etch technique", *Radiation Measurements*, 36, 479 – 481, 2003.
- [36] A. Rani and S. Singh, "Analysis of uranium in drinking water samples using laser induced fluorimetry", *Health Physics*. 91(2), 101-107, 2006.
- [37] E. Blaurock-busch, A. Friedle, M. Godfrey and C. E. E. Schulte-Uebbing, "Metal exposure in the physically and mentally challenged children of Punjab, India", *Maedica- J. Clinical Medicine*. 5(2), 105-113, 2010.
- [38] E. Blaurock-busch, A. Friedle, M. Godfrey, C. E. E. Schulte-Uebbing and C. Smit, "Metal exposure in the children of Punjab, India", *Clinical Medicine Insights: Therapeutics*, 2, 655-661, 2010.
- [39] A. Kumar, N. Usha, P. D. Sawant, R. M. Tripathi, S. S. Raj, M. Mishra, S. Rout, P. Supreeta, J. Singh, S. Kumar, H. S. Kushwaha. "Risk Assessment for Natural Uranium in Subsurface Water of Punjab State, India", *Human and Ecological Risk Assessment: An International Journal*, 179(2), 381-393, 2011.
- [40] M. Alrakabi, G. Singh, A. Bhalla, S. Kumar, S. Kumar, A. Srivastava, B. Rai, N. Singh, J. S. Shahi and D. Mehta, "Study of uranium contamination of ground water in Punjab state in India using X-ray fluorescence technique" *J. Radioanal. Nucl. Chem.*, 294, 221-227, 2012.