INTERNATIONAL JOURNAL OF PLANT, ANIMAL AND ENVIRONMENTAL SCIENCES
IJPAES
VOLUME-2, ISSUE-1, JAN-MAR-2012 ISSN 2231-4490
Accepted: Dec-2011 *Review Article*

PUBLIC HEALTH, ACTS AND METHODS OF REMEDIATION OF URANIUM FROM GROUND WATER

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ABSTRACT: Uranium is natural radionuclides present at mBq/lto Bq/l concentrations in many aquifers worldwidedue to leaching and transport mechanism from natural deposits. Human activities such as mill tailings, nuclear industry, coalcombustion and the use of phosphate fertilizers can also alter the natural composition of groundwater. Uranium through water transferred to the human food chain and its ingestion and inhalation causes many health problems. Manybioindicators are used to predict the path and durationof uranium remain in the body, which permits the risks calculation to specific tissues, organs as well as the whole body. So, a combination of both biological and non-biological methods seems to be the most promising way to efficiently remediate the uranium from groundwater.Hydrogeologists increasinglyrely on bioremediation of contaminated groundwaterbased on immobilization of the metal by bioreduction or biosorption. This review deals with the trafficking, prevention and possible remediation of uranium from groundwater.
Keywords: Uranium leaching, Bioindicators, Trafficking models, Biosorption, Remediation

INTRODUCTION

Due to the population explosion and climatic changes like drought in the world, many countries have intensively increased underground water sources exploitation for supplying water to populations as well as for developing their agriculture and industry. So, it is necessary to study the pollution of these water reservoirs to avoid any excess of exploitation[1]. Few years back in India, especially in Punjab and Himachal state using standard scientific techniques, viz., fission track and laser fluorimetry reported maximum uranium concentration ~100 µg/L in the handpump water samples from Giana village (Solan, H.P.). Recently even higher values of uranium concentrations approaching 1000 ppb have been reported in certain regions like KaramgarhSattran (Bhatinda, Punjab). Uranium is the heaviest naturally occurring elements having average concentration of 0.0003% (3mg/kg) in the earth crust, present in all soil, rocks such as volcanic rocks, granites, dark shales, sedimentary rocks that contain phosphate, metamorphic rocksand seawater (3.3 ppb of uranium by weight (3.3 µg/kg) [2]. The uranium concentration in groundwater depends on lithology and other geological conditions of the region and also contamination from human activities such as the use of phosphate fertilizers, mining, and combustion from coal or other fuels [3].Commonuranium-bearing mineralsareAutunite, Carnotite, Coffinite, Tyuyamunite, Uraninite and Uranophane. The natural weathering of rocks such as granite dissolves the natural uranium, which goes into groundwater byleaching and precipitation called illuviation process [2]. Nuclides of uranium emit alpha rays of high ionization power and therefore it may be hazardous if inhaled or ingested[4]. According to the Environmental Protection Agency, drinking uranium-rich water over many years may cause kidney damage and increase the chances of getting cancer because of non-biodegradable naturesof uranium[5].The provisional guideline value for tolerable daily intake of uranium for drinking water is 30 µg/l[6].

URANIUM LEACHING

Plant root respiration and microbial oxidation of organic matter in soils produce CO_2, resulting in formation of carbonic acid with water. It reacts with the calcium carbonates (calcareous soil) to form bicarbonate which is an efficient leaching agent for uranium from soils and sediments.Leaching also depends upon intrinsic conditions of an aquifer such as alkalinity and oxygenated water, flow-system, hydraulic conductivity, porosity, pumping rate, and other geochemical conditions[7]. Alkalinity increased in shallow ground water due to agricultural development which accumulate salts left after evaporation of irrigation water.Movement of the contaminated water is hindered by the presence of fine-grained material, but is not completely inhibited by this method[8].Other chemicals like humic acids formed due to agricultural product decay are also known for leaching metals.Uranium also enters an aquifer when it gets dissolved in oxygenated waters, such as rainwater. The oxygenated water percolates downward and form uranium soluble compounds that can interact with other chemical elements and compounds which determine its movementand toxicity in the environment[9].Status of groundwater uranium of different countries was given below (Table 1).

Table 1.Status of groundwater uranium in different countries.

Country	Name of the city/state	Year	Concentration of uranium reported
USA [10]	Kleberg County, Texas	2011	160 and 771 ppb
USA [11]	Grand Junction, Colorado	2010	0.11 mg/l(MCL= 0.044 mg/l)
South Africa [12]	AngloGold's South Uranium plant	2005	15,000 ppb
Japan [13]	Malvesi conversion plant	2008	110-200µg/l
Australia [14]	Queensland	2011	0.71 Bq/l
India [15]	KaramgarhSatran, Dhillawan, Giana and Guddavillages of Bathinda, Punjab	2011	644 µg/l, 463.6 µg/l, 292.65 µg/l and 165.85 µg/l respectively
India [16]	Jaduguda, Jharkhand	2011	3.5-208 mBq/l
India [4]	Sirsa and Bhiwani, Haryana	2011	19.14µg/l
India [17]	Peddagattu and Seripalli, Andhra Pradesh	2010	0.2-118.4 ppb
China [18]	Hengyang, Hunan Province	2008	20mg/l
China [19]	Seoul Gyeonggi and Daejeon, South Korea	2004	322 µg/l and 402.30 µg/l respectively
Pakistan [20]	Muzaffarabad and Reshian	2004	0.03-6.67 µg/l
Bangladesh [21]	Bualda, Fulbaria, Jamjami, and Komlapur	2009	< 0.2-10 µg/l

URANIUM TRANSFER IN HUMAN FOOD CHAIN

The transfer of uranium to the food chain of humans depends upon geological origin of the soils and the groundwater basin as well as the living area of the flora and the drinking water reservoir. On average, granite weathering soils produce the significantly uranium-richest vegetable forage and foods mostly stored in young plants[22]. Absorption of radionulides in the gastrointestinal tract of all higher animals depends upon composition of food and the nutritional status of the animal. Sugar, starch and fat rich foodstuffs are uranium-poor (fruits, seeds, flour) whereas leafy vegetables, tea and herbs are found to be rich in uranium content. The transfer factors (TF) of U from soil to different parts (root, stem, leaf, seed and fruit)have proved that roots have markedly higher concentration than those for leaf, stem, fruit and seed plants [23]. People ingest about 1–2 µg of natural uranium everyday through food and take in about 1.5 µg for every liter of water they drink. Uranium is also dispersed by ocean currents in the sea which contaminate fish entering the food chain. Bacteria, phytoplankton and single celled organisms adsorbed or absorbed radionuclides and subsequently ingested by zooplankton which later passes to fishes. Detritus also serve as an important reservoir for radionuclides in terrestrial and aquatic ecosystems which can recycle radionuclides into soluble forms for uptake by primary producers and other biota [24].

HEALTH HAZARDS OF INGESTED URANIUM

All uranium isotopes (U-234, U-235, and U-238) are radioactive and undergo radioactive decay by emission of an alpha particlethat has little penetrating ability, accompanied by weak gamma radiation until a stable product is formed. The release of radiation during the decay process raises health concerns which are chronic rather than acute [25]. The extent of damage depends on the solubility of the compound and the route of exposure. On the basis of the toxicity of different uranium compounds in animals, it was concluded that more watersoluble compounds (uranyl nitrate, uranium hexafluoride, uranyl fluoride, uranium tetrachloride) were the most potent systemic toxicants. The poorly water-soluble compounds (uranium tetrafluoride, sodium diuranate, ammonium diuranate) were of moderate-to-low systemic toxicity, and the insoluble compounds (uranium trioxide, uranium dioxide, uranium peroxide, triuraniumoctaoxide) had a much lower potential to cause systemic toxicity but could cause pulmonary toxicity when exposure was by inhalation [26].

Kidney damage: Uranium can enter the body by inhalation or ingestion, or under it may enter through cuts in the skin. After inhalation in lungs, it gradually dissolved into the bloodstream and approximately 10% of it will initially concentrate in the kidneys[27].Very high uranium intakes (about 50 to 150 mg) can cause acute kidney failure and death. At lower intake levels (around 25 to 40 mg), damage can be detected by the presence of protein and dead cells in the urine, but there are no other symptoms. There is possibility that at lower intake levels, the kidney repairs itself over a period of several weeks after the uranium exposure has stopped.Study onexperimental animal's shows that the proximal convoluted tubules (PCT) were mostly damaged and predominant in the distal two-third [28].

Carcinogenicity : Radiation may cause cancers at high doses rates [greater than 50,000 mrem (milirem)] include leukemia, breast, bladder, colon, liver, lung, esophagus, ovarian, multiple myeloma, and stomach cancers but currently there are no data for the occurrence of cancer following exposure to low doses and dose ratesbelow about 10,000 mrem [100 mSv (milliievert)][29].

Diabetes : The global pandemic of diabetes which is increasing almost exponentially each year began with the atomic bombs dropped on Hiroshima and Nagasaki in 1945[30]. Diabetes is a syndrome of impaired insulin secretion or decreased sensitivity of the tissues to insulin. Type 2diabetes is fundamentally due to magnesium deficiencies but is also being triggered by UO_2.Type 3 Diabetes is caused by mutation in the hepatocyte nuclear factor-1-alpha gene (142410), which maps to chromosome 12q34[31].Researchers have even investigated in vivo the effects of a chronic exposure to depleted uranium on vitamin D(3) metabolismas it has been shown to impair insulin synthesis in humans and involved in the pathogenesis of both forms of diabetes[32].

Brain and neurological effects: Nerves can act as a unique channel, carrying inhaled uranium from the nose directly to the brain, found in a study with rats.This study provides information that the inhaled isotope accumulated at 2 to 3 times higher levels than the injected isotope in the olfactory (smell) paths from the nose to the brain and in the frontal cortex and hypothalamus of the brain.The scientists then chemically damaged the olfactory nerves of the nose and found 3 times less uranium in the olfactory system than the rats with intact olfactory nerves. These finding suggests that inhaled uranium can travel directly from the nose along the olfactory nerves to the front of the brainwhichcan cause unstable gait, weakness, tremors, equilibrium disturbances, rigidity in limbs, anorexia, irritability, decreased IQ, memory changes, hyperactivity, respiratory effects and hypothermia[33].

Bioindicator of ingested uranium in humans: Hair is an excellent matrix in human biomonitoring for uranium. It has numerous advantages such as easy collection, transportation, storage and procedure for radiochemistry is considerably easier and less time consuming than other methods. The only major disadvantage is the difficulty in distinguishing between internal and external exposure[34]. Some other study shows kidney[35] and teeth used as bioindicator whichwere determined by means of a high-resolution inductively coupled plasma mass spectrometer (ICP-MS) [36].

RADIONUCLIDES CONTROL ACTS[37] (Table 2).

Table 2. Regulatory control acts and their functions.

Control acts	Functions
Safe Drinking Water Act (1996)	EPA direct standards for maximum contaminant limit in drinking water that may adversely affect human health.
Low-Level Radioactive Waste Policy Act (1985)	Encourage state to provide disposal facilities for commercial low-level waste generated within its borders.
Uranium Mill Tailings Radiation Control Act (1978)	EPA set standards for the cleanup and disposal of contaminants at closed uranium and thorium mill tailings sites.
Underground Injection Control Program(1974)	Designed to prevent ground water contamination from injection wells.
Federal Water Pollution Control Act (1972)	Prohibits the discharge of radioactive wastes into navigable waters without a permit.

PREVENTION FROM URANIUM MOVEMENT TO GROUNDWATER

The water found in and adjacent to uranium orebody in groundwater is naturally contaminated and unsuitable for drinking.Leaching of uranium from mining can be prevented by immobilization[38]by using two inexpensive reactive materials namely Zerovalent ironnanoparticles (nano-Fe(0)) and magnetite nanoparticles (nano-Fe_3O_4)[39]and Phosphate-induced metal stabilization (PIMS) using apatite[40].When production of uranium terminates from underground, the well must befilled with cement,covered with 2 meters of clay and topsoil to reduce radiation levels near orebody region. Another technique involves surface treatment of recycled groundwater before it is reinjected into the aquifers and water quality must be restored to regulatory standards levels using groundwater sweeping technique [41].Surface soil must be decontaminated and the ground surface must also be re-vegetated.

METHODS OF REMEDIATION

I. **Physical process**include use ofactivated alumina (pH 5.0-6.0), anion exchange method (pH 6-8), lime softening (pH >10.6)[41].

II. **Electrokinetic method**is based on electromigration and electroosmosis.Electromigration is the electrochemical process in which the contaminants are ionic or surface charged andElectroosmosisdirect the flow of water.Itsefficiency depends on particle size, ion mobility, contaminant concentration,type of species, their solubility in the specific soil, electrical charge, total ionic concentration, location and availability of organic matter in the soil[42].

III. **Chemical process**is based on precipitation ofU(IV) which depends on the type and concentration of the organic ligand present in solution. Briefly, ≥95% of the U(IV) precipitated by acetate and malonate, 80–89% by aluminium coagulation at pH 6,< 20% of the U(IV) by oxalate and citrate and little (≈10%) by tiron solutions.Polyphosphates are the most efficient inorganic chelators, others areDiphenylphosphinic acid, carbonate, phosphate, EDTA, EDDA, citric acid, Alizarin S, Nitrilotriacetic acid, Iminodiacetic acid etc.[42].

IV. **Bioremediation**

a) Biosorption is a biotechnological innovationin which passive uptake of toxicants by dead/inactive biological materials or by materials derived from biological sources is done. It is advantageous over bioaccumulation processes as toxicant accumulation may interrupt metabolism resulting in death of the organism[43], solvedisposal problem of fermenting industry, absence of toxicity limitation, no need of growth media and nutrient and metal get easily adsorbed. Pretreatment method of dead cells can also increase the metal uptake capacity of the organism (Table 3). This method has some limitations such as metal desorption is required when metal absorptive sites are blocked, dead cells limit the biological process improvement by genetic engineering and there is no potential for biologically altering the metal valency state of the organism.

Table 3. List of dead microorganism used as biosorptive agent.

Organisms- method	Conditions
Saccharomyces cerevisiae- dried and finely ground(pore size 100µm) [44]	Contains abundant chitin–chitosan units and histidine as matrix for interaction of U(VI) ion,highest uptake was observed at 1g biomass in 100mg/L at pH 5 and 100 µm particle size.
Rhodotorula Glutinis- heating at 60℃ [45]	Sorption capacity of uranium increased by 31% in methanol-treated biomass and 11% in formaldehyde-treated biomass at an initial concentrationof 140 mg/L.
Aspergillus fumigates- sodium alginate immobilized [46]	U (VI) ions could beeffectively adsorbed by the active immobilized *A. fumigatus* beads. The maximum percentage removal of U (87.8%) was observed when the initial U (VI) concentration was 60 mg/L.
Penicilliumcitrinum- dried and finely ground [47]	Exhibited the highest uranium sorption capacity at an initial pH of 6.0 at 5 h, capacity increased from 103.1 to 127.3 mg/g with an increase in temperature from 298-318K at an initial uranium concentration of 50 µg/mL.
Arthrobacter sp. G975 [48]	Can uptake up to 90% of the U(VI) at 150.2 ± 71.4 mg/g and decreased with increasing bicarbonate concentrations.
Cystoseiraindica- treated with HCl and $CaCl_2$ [49]	0.1 M $CaCl_2$solution at pH4, increased the uranium uptake capacity more than 30% (371.39 mg/g).

b) Microbial reductionusemicrobial community such asFe(III)-reducing bacterial (FeRB), nitrate-reducing bacterial (NRB), and sulfate-reducing bacterial (SRB), fungi and algae as electron donorfor both active U(VI) reduction and maintenance of the stability of reduced U [50]. But have some limitations as it is limited to biodegradable compound and the products of biodegradation may be more persistent or toxic thanthe parent compound.Microorganisms require mediaand are highly specific so that their growth and activity must be stimulated.It often takes longer time than other treatment options, such as excavation and removal of soil or incineration.

c)Phytoremediationis an eco-friendly approach comprised of two components, one by the root colonizing microbes and the other by plants themselves, which accumulates the toxic compounds to further non-toxic metabolites (Table 4).Primary limitation of phytoremediation is the contact of root zone to the contaminated media, takesseveral years while excavation and disposal or incineration takes weeksto months.Other limitations are high concentrations of contaminants may inhibit plant growth, leaching cannot be fully prevented and adding organic chelators lead to soil pollution. Hyperaccumulator plants after germination, increases metal content in biomass and ifinsects feed on them, may develop resistance to metals[51]. Pollen dispersal ofhyperaccumulators from the site of remediation develops into weeds as most hyperaccumulators evolved under extreme conditions.

These limitations can be improved through site specific soil condition and plant research.Plants havingmetalmust be incinerated or composted. After incineration, the ash must be disposed of in a hazardous waste landfill.Gas emitted during incineration or smelting process can lead to the formation of metal oxide which is both toxic and carcinogenic. So, through smelting or electro-winning processes, metals can be extracted from the bio-ore which is a rich and potentially valuable ore than extracted metal [52, 53].

Table 4.Phytoremediation of uranium.

Plants	Condition
Arabidopsis halleri[54]	Can accumulate~35 mg/kg U in roots and ~17 mg/kg U in shoots and soil-to-plant transfer factor (TF) were 1.2 for roots and 0.6 for shoots.
Helianthus annuus L. and*Phaseolus vulgaris* L. var. vulgaris [55]	For *H. annuus,* more than 97% of uranium removed by rhizofiltration within 24 h having initial conc. 543 µg/L and for *P. vulgaris,* 70% of the uranium was removed having initial concentrations in groundwater were 80 and 116 µg/L at pH 3–5.
SenecioglaucusL. [56]	Grown in the soil collected from the coastal sand dunes contain 3.69 ppm uranium concentrations and the soil-to-plant TF value was 0.07.
*Phragmitesaustralis*and *Cyperusiria*[57]	*P. australis* accumulate the highest concentration (820µg) of U in the shoot and had soil-to-plant transfer factor (TF) is 16.6 and *C. iria* accumulate 36.4 µg/g of U in the shoot and had a TF of 5.48.
*P. sylvestris, Piceaabies*and *Acer pseudoplatanus*[58]	Trees grown on a uranium mill tailings waste pileshowedhighest activity concentrations infoliage, followed by shoots and wood and the activity concentrations in trees were from 0.01-5.4 Bq $kg^{-1}$for^{238}U, all activityconcentrations were calculated on dry weight basis.
Wheat straw [59]	NaOH treated straw at 303 K and pH 3.0, show adsorption up to 1.20 mg/g at approximately 120 min.
Hydrillaverticillata (L.f.) Royle [60]	Maximum accumulation being 78 mg g^{-1} DW upon exposure of 100 mg L^{-1} U after 24h, due to presence of guaiacol peroxidase, ascorbate peroxidase, catalase, proline, total phenolics and also the constituents of thiol metabolism (viz., cysteine and glutathione)

CONCLUSIONS

Uranium is an emitter of ionizing radiation. It's continuously elevatingconcentrationin the groundwater is causing carcinogenic, mutagenic and teratogenichealth hazards. So, there is need of efficient remediation of U(VI)-contaminated sites which require the use of several physical and chemical methods in parallel or sequential.In this process, bioremediation method is used frequently as it is safe and cheap but it is crucial to prevent the reduced uranium from being reoxidized and therefore re-dissolved in groundwater. It has other limitations also which can be improved through site specific soil condition and research works.Surface soil should decontaminate to defined limits and disposed of at a licensed site after uranium production termination from mills. Also there is heightened and urgent need for further work on predictionmodels for uranium trafficking in groundwater and in human.Above all, there should be strict regulatory controls over use and disposal of uranium contaminated materials.

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