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Study of Arsenic Contamination of Ground Water of Patori Block of Samastipur District

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ABSTRACT

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In this present paper, we studied about the arsenic contamination in ground water of Patori block of Samastipur district. The main aim and objective of the recent work is to identify areas within Samastipur district, where ground water is significantly contaminated by Arsenic.

Keywords : Arsenic, Contamination, Water Pollution.

I. INTRODUCTION

Arsenic is a metalloid element (Atomic number 33) with one of naturally occurring isotope of atomic mass 75 & having four oxidation states (-3, 0, +3, +5). In the Aqueous Environment, +3, +5 oxidation states are most prevalent as the oxyanions Arsenite (H_3AsO_3 or $H_2AsO_3^-$ at $pH^{\sim}$ 9-11) & arsenate (H_2AsO_4- or $H_2AsO_4^{2-}$ at $pH^{\sim}$ 4-10). Arsenic is widely distributed in nature and mainly occurs in the form of inorganic or organic compound. Different forms of Arsenic have different toxicities with arsine (AsH_3) gas being the most toxic form of the inorganic oxyanions, arsenite are the most toxic form and arsenate, the less toxic form of Arsenic. The organic (Methylated Arsenic) forms are considered the least toxic form. Exposure to inorganic compounds of Arsenic may occur in a number of ways through certain industrial effluents, pesticides, chemical alloys, and combustion of fossil fuels, occupational hazards in mining and dissolution in drinking water.

The most commonly found Arsenic compounds in drinking water sources are Arsenate (+5) or Arsenite (+3). However, ground water is very prone to chemical and other types of contamination from natural sources or by anthropogenic activities.

Arsenic contamination of ground water (a major source of drinking water) is a worldwide problem due to its most damaging effects on human health which vary from various skin ailments to serious & dangerous diseases viz skin lesions and cancer continues exposure to arsenic can cause damage to the human cardio-vascular, gastrointestinal, dermatitis, hepatic pulmonary, neurological, reproductive and respiratory system [1-2].

Keeping in view the above cited dangerous effects of Arsentic contamination in ground water; it has become essential to undertake close monitoring of some of the affected Blocks of Samastipur District in

Bihar. Besides eco-friendly & economical treatment technologies have to be explored for its removal & mitigation from groundwater so as to provide safe drinking water to human beings. Arsenic occurs in ground water as a result of mineral dissolution, industrial discharge, or the application of fungicides, insecticides etc. It is a naturally occurring high concentration of Arsenic in deeper levels of groundwater which has become a high-profile problem in recent years due to the use of deep tube wells for water supply in the Ganges Delta, parts of Thailand, Taiwan, Argentina, Chile and China have also been affected by Arsenic contamination in ground water.State wise names of the districts (Partly/severely affected) form where ground water contamination by Arsenic (Above 0.05mg/l) has been reported is hereby presented (Source: Central Ground Water Board). In Bihar state, the several district like Begusarai, Bhagalpur, Bhajpur, Buxar, Darbhanga, Katihar, Khagaria, Kishanganj, Lakhisarai, Munger, Patna, Purnia, Supaul, Samstipur, Saran, Vaishai are affected by Arsenic.

In India approx. 40 million people are within the risk zone of Arsenic with in contamination [3]. Out of 610 samples collected in Patna Distt. 457 samples were reanalysed by AAS-HG Method at Sriram Institute of Industrial Research New Delhi. [4] of the 457 sample tested at SIIR, 390 samples were confirmed to level as here >10 ppb; as indicated in Table:-

Table-1

Distt.	No of water sample with more than 10 ppb Arsenic	Highest reading in ppb
Patna	392	724
Bhojpur	656	1861
Vaishali	270	360
Bhagalpur	1402	608

As reported from various sources several blocks of Samastipur distt. viz. Mohanpur & Shahpur Patori blocks lying in the vicinity of river Ganga are significantly affected by Arsenic contamination of ground water resulting in severe harmful effects on human health & causing clinical manifestations viz: hyperkeratosis, bronchitis, gangrene in limbs, skin, lesions & even skin cancer in the long run [5-6].

Consequently, in the present programme three blocks namely Shahpur Patori was subjected to arsenic contamination study and its removals were undertaken. Sixteen villages in each block were chosen, water samples collected and analysed for arsenic concentration alongwith with some other parameters. Before presenting the outcome of the present investigation, it would be appropriate here to present an account of earlier work done in the field.

II. MATERIALS AND METHOD

Bimetallic adsorbent

[6] investigated a Fe–Ce bimetal oxide adsorbent for arsenic removal using X-ray powder diffraction (XRD), transmission electron micrograph (TEM), Fourier transform infrared spectra (FTIR), and X-ray photoelectron spectroscopy (XPS). The bimetal oxide adsorbent exhibited a significantly higher As (V) capacity than the individual Ce and Fe oxides (CeO_2 and Fe_3O_4) prepared by the same procedure. Various mechanisms were proposed based on the results obtained from XRD, FTIR, TEM, and XPS.

Metal-chelated ligands

Fryxell *et al.* [38] reported the synthesis and use of metal-chelated ligands immobilized on mesoporous silica as novel anion binding materials. Nearly complete removal of arsenate and chromate from solutions containing more than 100 mg/L was achieved in the presence of competing anions under a variety of conditions. Anion loading was more than 120 mg (anion)/g of adsorbent. A binding mechanism based on computer modeling was also proposed. First Cu(II) ions bonded to ethylenediamine ligands to form surface octahedral complexes on the mesoporous silica. This gave rise to positively charged hosts with three-fold symmetry that match the geometry of tetrahedral anions. The anion binding involved initial electrosteric coordination, followed by displacement of one ligand and direct binding with the Cu(II) center.

Highly ordered mesoporous silica, SBA-15 impregnated with iron, aluminum, and zinc oxides were used for arsenic removal [7]. A 10 wt.% aluminum-impregnated sample (designated to $Al_{10}SBA$-15) had 1.9–2.7 times greater arsenate adsorption capacities over a wide range of initial arsenate concentrations and a 15 times greater initial sorption rate at pH 7.2 than activated alumina. Surface complexation modeling of arsenate adsorption edges, at different pH values, indicated that the monodentate surface-bound complex (AsO_4^{2-}) was dominant in $Al_{10}SBA$-15. Conversely, bidentate surface complexes of $HAsO_4$ and AsO_4– were dominant on activated alumina at pH 7.2. $Al_{10}SBA$-15 had a single fast-rate initial adsorption step at pH 7.2, while activated alumina had both fast and slow arsenate adsorption steps.

Fe(III)-Octolig-21 composite was prepared from dried Octolig-21 and used for arsenic remediation [8]. Octolig-21 is an immobilized ligand containing (polyethyleneamino) groups bound to a silane that is covalently bound to silica gel. A stream of ingoing water containing 50 ppb As over a 1 kg composite might last for months before column would lose effectiveness at the flow rate of 5 L/h.

Yoshitake *et al.*[41] utilized diamino group-functionalized MCM-41 and MCM-48 for arsenate remediation. Fe^{3+}, Co^{2+}, Ni^{2+}, and Cu^{2+} were captured by these diamino moieties. Elemental analyses were consistent with 2:1 and 1:1 coordinations of the ligand to metal cations. The mono-, di-, and triamino-functionalized silica chlorides were denoted by H/N-, H/NN-, and H/NNN-mesoporous silicas, respectively, where mesoporous silica is MCM-41 or MCM-48. The average forms of Fe and Cu on NN-mesoporous silicas were $Fe(en)_2$ and $Cu(en)_2$, respectively, while Co^{2+} was mostly bound to one en ligand. Ni^{2+} was adsorbed on unfunctionalized mesoporous silicas, resulting in low N/Ni^{2+} ratios. Fe^{3+} and Co^{2+} were superior to the other cations after complexation on silica, achieving complete removal of As from the solutions as evaluated by *K*d. Fe^{3+} and Co^{2+} achieved high adsorption capacities, and selectivities in the solution in the presence of SO_4^{2-} and Cl^-. The highest adsorption capacity of arsenate, 2.5 mmol/(g of adsorbent), was achieved on Fe/NN-MCM-48, in which each Fe^{3+} bound to an average of 2.7 arsenate anions. The adsorption capacities of M/NN-MCM-48 (M = Fe, Co, Ni, Cu) were much larger than those of M/NN-MCM-41, though the As/M stoichiometries are almost the same. The adsorption of nitrate, chromium (VI), arsenic(V) and selenium (VI) by a secondary and tertiary amine-modified coconut coir (MCC-AE) has been reported [9-11]. Batch adsorption-ion-exchange experiments were conducted using 200 mg of MCC-AE, initially containing chloride as the resident anion, and 50mL of different anion-containing water solutions with varying concentrations. At low pH, SeO_4^{2-} remained as the only divalent anion, while monovalent species $H_2AsO_4^-$ and $HCrO_4^-$ predominated in their respective exchanging ion solutions. MCC-AE exhibited a preference for $Cr_2O_7^{2-}$ and SeO_4^{2-} compared to the resident Cl^- ion. The maximum As(V) adsorption was 0.086 mmol/g versus 0.327, 0.459, and 0.222 mmol/g, for Cr(VI), NO_3^- and Se(VI) anions, respectively. Comparative adsorption experiments were also conducted using commercial Amberlite IRA-900 quaternary amine chloride anion-exchange resin (exchange capacity of 4.2 mequiv./g). The maximum adsorption of ions in IRA-900 was about 3 times higher for NO_3^-, 9 times higher for Se(VI), 10 times higher for As(V) and 9 times higher for Cr(VI), than those exhibited by MCC-AE. Differences in the ion-exchange behavior of MCC-AE and IRA-900 were probably due to their different amine functionalities.

Cation-exchange resins

Ce(IV) resins- PHA and IDA (Amberlite IRC-718) chelating polymers were loaded with cerium (IV) chloride at pH 5.65 and 6 [11]. Ce(IV)-Amberlite IRC-718 removed 99% of As(V) from a 374.5 ppm solution at pH 3.25, while Ce(IV)-PHA removed only 81% at pH 2.75.

Cu(II) resins- Cu(II)-loaded sorbents were prepared from two commercially available resins, Amberlite IRC-718 and pyridyl/tertiary ammonium groups (Dowex 2N), by Ramana and Sengupta [12]. Copper loadings were 0.85 and 1.6 mmol/g, respectively. Cu(II)-Dowex 2N removed As(V) in presence of 250 mg SO_4^{2-}/L at pH 8.5 where Amerlite IRA-900 was ineffective.

Fe(III) resins- The adsorption of As(III) and As(V) on an iron(III)-loaded chelating resin containing lysine-N^{α},N^{α}-diacetic acid functional groups (Fe-LDA) was investigated [13]. Arsenic(V) was strongly adsorbed at 2–4, while arsenic(III) was moderately adsorbed between pH 8 and 10. Maximum Langmuir sorption capacities of 0.74 mmol/g for As(V) at pH 3.5 and 0.84 mmol/g for As(III) at pH 9.0 were obtained. Both As(III) and As(V) were almost quantitatively recovered from the resin with 0.1 mol/L sodium hydroxide. During regeneration, less than 0.1% of the ferric ions are leached into the alkaline solutions. [14] examined an iron-loaded iminodiacetate chelating resin and a silica/iron(III) oxide composite material for As(V) remediation. The composite exhibited a higher As(V) adsorption capacity than the iminodiacetate resin. Exposure of the composite to a magnetic field caused the adsorption of As(V) to change in a complex way. The sorption capacity decreased in acidic conditions when the magnetic field was applied. However, at pH 7.0 the magnetic field intensified As(V) adsorption. Anionic metal remediation using alginic acid pretreated with Ca^{2+} and Fe(III) was investigated by [15]. Spherical gel beads (2mm in diameter) were formed by dispensing this biopolymer solution drop wise into 0.1M $CaCl_2$. The polycarboxylate Ca^{2+} beads were then washed and equilibrated with 0.1M$FeCl_3$ to achieve partial substitution of Fe(III) for Ca^{2+}. The resulting Ca-Fe containing beads were effective for As(V) removal Optimum arsenic removal and stability by these beads was achieved at pH 4. As(V) removal efficiency increased with increasing Fe content. At an initial As(V) con centration of 400μg/L, up to 94% removal was achieved at pH 4 after 120 h. [16] studied As(III) and As(V) removal on polymeric/inorganic hybrid particles composed of spherical macroporous cation-exchange polymer beads, containing nanoscale hydrated Fe oxide agglomerates that were uniformly and irreversibly dispersed. The new hybrid ion-exchange sorbent combined excellent mechanical and hydraulic properties of spherical polymeric beads with selective As(III) and As(V) sorption properties at neutral pH without any pre- or post-treatment. Efficient *in situ* regenerationwas accomplished with caustic soda and a subsequent short carbon dioxide-sparged water rinse.

The new sorbent possesses excellent attrition resistance properties and retained its arsenic removal capacity over several cycles. [15] modified polystyrene and polyHIPE (PHP) by iron hydroxide coatings. Modified media, capable of removing arsenic from the aqueous stream, led to a residual As concentration below 10μg/L. PolyHIPE (PHP) was the more effective arsenic sorbent. Zouboulis and Katsoyiannis [16] also tested biopolymers (alginate) as sorbent supports, for the removal of arsenic. Alginate, a biopolymer extracted mainly from brown seaweed, is a linear polysaccharide of (1→4)-linked μ-L-guluronate (G) and μ-D-mannuronate (M) residues arranged in a non-regular, block-wise pattern along the linear chain. A bed of calcium alginate beads was treated (doped/coated) with hydrous ferric oxides. Three modified alginates viz., calcium alginate beads coated with iron oxides and calcium alginate beads doped and coated with iron oxides were tested. The most efficient was Ca-Fe-doped alginate. An iron (III)-loaded iminodiacetate resin (LEWATIT TP 207) (168 mg Fe/g resin) was employed and a maximum of 60 mg As/g resin was adsorbed at pH 1.7 [17].

III. EXPERIMENTAL

To understand the severity of contamination and consequent health effects in West Bengal, we studied Samastipur district and undertook a detailed surgery in one of the block namely Patori. It is shown in Map:-(1).

Map:-1 Blocks of the Samastipur district

Table-2

Experimental data of water samples collected froms Patori Block, Samastipur District.

Name of Village	Arsenic concentration(in mg/L)
Araia	0.01
Bahadurpur patori	0.02
Bishunpur paharpur	0.06
Chandpur	0.04
Darwa	0.061
Fatehpur	0.03
Hasanpur	0.072
Lodipur	0.05
Mirzapur	0.65
Saidpur	0.008
Sarhad Madho	0.022
Mukundpur	0.032
Chak Saho	0.041
Chak Raman	0.05
Ismailpur	0.061
Shahpur Undi	0.049

All the samples were collecting in poly propylene bottles. Before collecting the samples, bottles were through cleaned by 8M HNO_3, followed by repeated washing with deionized water. Adding 1:1 HNO_3 for analysis of arsenic and other heavy elements preserved 1 L of each of the collected water samples. The different physical and chemical parameters were pH, electrical conductivity (EC), turbidity, total dissolved solid (TDS), total hardness (TH), calcium (Ca^{2+}), magnesium (Mg^{2+}), chloride (Cl^-), fluoride(F^-), iron (Fe), sulphate(SO_4^{2-}), nitrate(NO_3^-) and arsenic (As). The reagents used in the study were all analytical grade reagents and demonized water was used throughout for the reagent preparation. The pH of the water samples was determined in the field at the time of sample collection by using portable pH meter (Merck, India). Fluoride contents were determined by SPANDS method using spectrophotometer (Analytik Jena, Specord-40, Germany), Sullphatae was determined by spectrophotometer method (Analytik-Jena, Specord-40, Germany). Total hardness (TH) of the samples was determined by titrimetric method.

IV. RESULTS AND DISCUSSION

The experimental results relating arsenic concentration of water samples from one block namely Patori have been represented in table-(1) and Figure-(1) in the experimental section.

Arsenic - Water contaminated by arsenic is a burning problem affecting the whole world. Arsenic at high concentrations in water can cause skin lesion, cancers, vascular diseases and hypertension and diabetes mellitus [18]. It is observed that drinking water with more than 300µg/L arsenic for several years may cause arsenic skin lesions [19]. Ingestion of inorganic as in an established cause of skin bladder and lung cancer [20]. The results reveal that out of one block of Samastipur district of Bihar, Patori block shows maximum (31.28%) groundwater contamination by arsenic. In this study an average of 28.93% water samples have arsenic concentration above the permissible limit of 0.05mg/L. the maximum value recorded was 0.072 mg/L in Patori block. The variation of arsenic content indifferent sampling points from different blocks of Samastipur district is shown in figure-(1).

pH- The pH of 90% of the water samples analyzed were within the desirable limit of 6.5 – 8.5 given by WHO standard and most of the samples were slightly alkaline in nature. In this study pH of water samples from, Patori block varied from 7.1 to 7.9. The maximum 8.1 in Patori block.

Electrical conductivity and total dissolved solid- The EC, which is highly correlated, with TDS is ranged from 0.55 to 0.87 mmho/cm. Indian standards for drinking water propose no standards for EC, however, the standard for TDS are proposed which are 500 to 2000mg/L. it is seen that TDS of water samples from Patori varies from 311 to 597. The maximum value recorded is 616mg/L form Patori block.

Turbidity- It is caused by clay organic matter, phytoplankton and other microscopic organisms and makes the water unfit for domestic purposes, food and beverage industries. it is seen that TDS of water samples from Patori varies from 0.5 to 1.5. The minimum value recorded is 0.5 for Potri block. Turbidity of all samples found to be below the permissible limit 10 NTU.

Total hardness- It is the measure of the capacity of water to precipitate soap. Though hardness is not harmful to health, it has been suspected to be plying some role in heart disease. In this study, total hardness of all samples found to be below the permissible limit 600mg/L for drinking water. It is seen that total hardness of water samples from Patori block varies from 250 to 330 mg/L. The minimum and maximum values recorded were 214 and 330mg/L from Patori block.

Calcium- In order of abundance it is the fifth element which is commonly present in all water bodies where it usually comes from the leaching of rocks. Calcium is very essential for nervous systems and for formation of bones. In this study calcium concentration of water samples from Patori block varied from 27.1 to 91.7mg/L. The concentration of calcium in portable water ranges from 75 to 200mg/L.

Magnesium- Magnesium tolerances by human body are lower than that of calcium. High concentration of magnesium in drinking water gives unpleasant taste to the water. The concentration of magnesium in potable water ranges from 30-100mg/L. In this study magnesium concentration of water samples from Patori block varied from 14.9 to 48.6 mg/L.

Chloride- Chloride in excess imparts a salty taste to water and people who are not accustomed to high chloride can be subjected to laxative effects (Ravi Prakash and Krishna Rao, 1989)[122]. In ground water it may be contributed from minerals, like apatite, mica and hornblende and also from the liquid inclusion in the igneous rocks (Das and Malik, 1988)[123]. In this study chloride concentration of water samples from Patori block varied from 4.2 to 17.5 mg/L.

Fluoride - High concentration of fluoride in drinking water can cause an adverse effect on human beings. Continuous consumption of water having high fluoride content can cause diseases, like fluorosis, dental carries and bone diseases (Mariappan *et.al.*, 2005)[124]. The concentration ranges observed in the is study were 0.11 to 0.8 mg/L for block. The variation of fluoride content with different sampling points.

Iron- Iron is considered to be the most essential element to all organisms. It is present in hemoglobin and myoglobin systems. Presence of iron in water can cause staining laundry and porcelain. It gives stringent taste to water when water contains iron concentration above the permissible limit 1mg/L of drinking water. In this study iron concentration of water samples from Patori block varied from 0.18 to 0.65 mg/L. The maximum concentration observed was 1.11mg/L, in Patori block. The variation of iron content with different sampling points is presented.

Sulphate- It is the common ion in water. Sulphate can produce bitter taste at high concentration. Sodium and magnesium sulphate exert a cathartic action in human beings. It is also associated with respiratory diseases. The permissible limit and desirable limit of sulphate in drinking is 200 to 400mg/L. respectively. In this study minimum recorded value of sulphate were 0.34mg/L and maximum 22.3mg/L in Patori block. It shows that sulphate content of the three blocks of Samastipur district is all within the permissible limit of drinking water.

Nitrate- High nitrate in drinking water may cause methamoglobehemia among infants in which the skin becomes blue due to poor oxygenation of blood hemoglobin. Carcinogenous disease, like nitro-semines result due to higher nitrate in drinking water. It is seen that nitrate of water sample from Patori block varies from 1.6 to 3.0 mg/L. All samples are found to be below desirable limit 45.00mg/L.

V. CONCLUSIONS

Groundwater samples collected from these blocks shows that pH of 90% water samples analyzed were within the desirable limit of 6.5-8.5 given by WHO standard and most of the samples were slightly alkaline in nature. Similarly, TDS, turbidity, TH, Ca^{2+} and Mg^{2+} concentration in the water samples are all within the permissible limit of drinking water SO_4^{2-}, Cl^-, F^-, Fe and NO_3^- content in the groundwater samples from these blocks are also found within the permissible limit for drinking water. From the study it shows that groundwater of the region is contaminated by arsenic. The results reveal that out of one block, Patori block shows maximum (31.25%) groundwater contamination by arsenic. Though the groundwater of the two villagess has been contaminated by arsenic, no symptoms of arsenic poisoning are observed till date. This is most probably due to the presence of As^{5+} in groundwater in high amount as compared to As^{3+}. But in near future, the problem of Arsenicosis may arise and, therefore, speciation of arsenic should be taken into consideration.

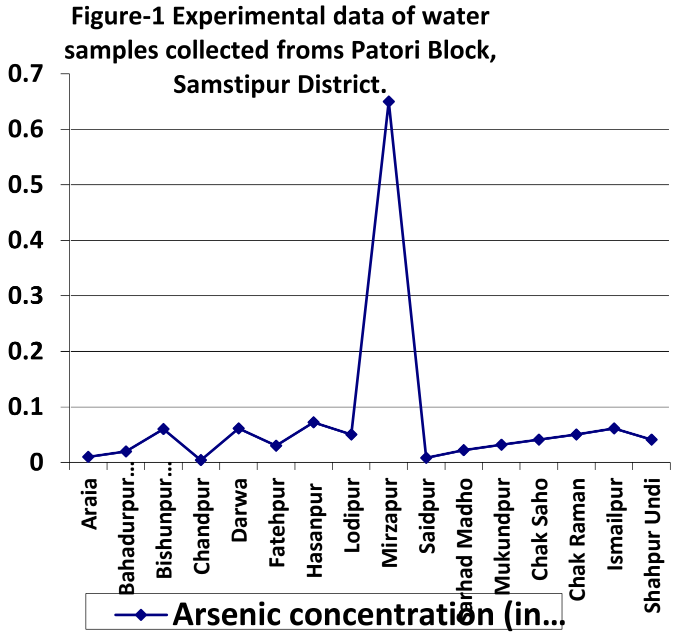
Figure-1 Experimental data of water samples collected froms Patori Block, Samstipur District.

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