H-6 Studies for Plan to Maintain the Global Environmental Protection to the Widely Arsenic-Affected Groundwater

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1. Introduction

Bangladesh and West Bengal, India are two areas of the most harmful damages against the arsenic in the world. For instance, Karim[1)] estimated that about 35million people in Bangladesh are affected by arsenic contamination. Although many local, national and international organizations have been studying about arsenic contamination and doing a variety of activities to overcome it, tens of millions of people are still suffering from arsenic. Therefore, arsenic removal technologies for the contaminated groundwater are of great concern to mitigate the adverse effect of arsenic in human beings. People in those areas use the arsenic-contaminated water for drinking, cooking and other

house-hold purpose and continue to get the serious influence caused by the arsenic.

Environmental cycling of arsenic in groundwater, especially after its usage, will be studied in arsenic-polluted area at West Bengal, India, in order to establish scientific basis for the mitigation / remediation of the pollution.

Frmers in those areas mainly use the arsenic-contaminated underground water for irrigating on the drying season from October to March. So There are the big problem caused by the arsenic contamination of irrigated land and of agricultural products.

The arsenic removing pilot plant using local arsenic contaminated groundwater was installed and its arsenic removal efficiency had been investigated.

It is urgently important that, supply a safe drinking water for the people who suffer from arsenic pollution.

As for arsenic, removing by physical processing is common, and there is little biological application. As an aid of arsenic removal in Bangladesh where is deficient in energy and technology, application of genetic engineered microbe was considered and the arsenic perception biosensor was also produced. Furthermore, as hygiene education is important for spreading the simple equipment for arsenic removal, hygienic situations and senses, customs, sociality, etc. were investigated at the start.

Along with increasing uses of ground water through wells, serious intoxication associated with arsenic contamination in the water has been observed in various areas in Asia including Bangladesh, West Bengal of India, China, Inner Mongolia etc. Among those who have been drinking the contaminated water, people who manifested some typical symptoms of arsenic poisoning have been increasing so that appropriate counteractions should be considered and implemented urgently.

Recently, moreover, some experimental evidence showing a difference in methylation process in the arsenic metabolism between animals and humans was indicated, suggesting, although indirectly, the epidemiological findings associated with carcinogenic property of arsenic among humans.

2. Research Objective

1) Evaluation against Harmful Effects of arsenic to Human Health

In order to estimate the total exposure of arsenic on the arsenic -contaminated area of both Mushidabad district, West Bengal, India and Samata village, Bangladesh, we collected 249 samples of urines and hairs and drinking waters from 66 families. We determined the arsenic species from urines, 17 elements including arsenic in hairs and inorganic arsenic in drinking waters. We reported the harmful influence of arsenic to the habitants in West bengal, India and Bnagladesh.

2) Studies for plan to maintain the global environmental protection to the widely arsenic- affected groundwater

Arsenic is easily reduced and methylated by biological activity to volatile forms. In addition, we noticed that usage of cow dung for cooking fuels in arsenic polluted area may cause in house arsenic pollution due to volatilization of arsenic contaminated in cow dung. This year, we sampled SPM in houses and analyzed water-soluble arsenic concentrations in them and their species as well as arsenic concentrations and species in straws and cow dung.

3) Studies for Harmful Effects of arsenic to agricultural land and agricultural products

In orde to catch the movement of arsenic from the arsenic-contaminated underground water to the irrigated land or the agricultural products, we visited an arsenic-contaminated area that locates to Mushidabad, West Bengal, India and collected the soils and the agricultural products. After digesting all the samples, 17 elements including arsenic were determined by ICP/MS.

4) Contributing Study Topic: Study on the development of large-scale arsenic removal plant in drinking water supply system.

Currently, small-scale arsenic removal facilities, which are for a household or a small sized community, have been interested in and installed by many NGOs. They are useful as a kind of tentative countermeasures. However, from a long-term perspective, it is crucial to investigate the applicability of low cost and large-scale arsenic removal methods. This is the main objective of the study. To be concrete, a pilot plant treating the local arsenic contaminated groundwater was installed and the removal efficiency of arsenic and other substances had been investigated.

5) Research on the Environmental Plan for the Earth Environment Preservation to the Over

(1) Performance evaluation of arsenic removal by drinking water supply systems in Bangladesh

The research objective is to conduct a field investigation on the evaluation of performance of existing arsenic removal plants in a rural area in Bangladesh in collaboration with NGO and BUET (Bangladesh University of Engineering and Technology) and to develop an affordable and appropriate arsenic removal system for drinking water supply.

(2) Development of the Arsenic Removal Technique in Small Scale Drinking Water Apparatus such as Well

The arsenic removal method using genetic engineered *Escherichia coli* (*E. coli*) and development of measurement technology (biosensor) were performed. Moreover, the social background for hygiene education was grasped.

The arsenic removal by cell surface engineering was tried using the specific bonding to inducible substance of ArsR repressor protein. The arsenic luminescence sensor using the gene regulation function of ArsR repressor protein was developed. The sensor could be analyzed As(Ⅲ) and As(Ⅴ), individually from the characteristic of luminescence, and the membrane transportation mechanism of arsenic to cell was considered.

On the other hand, the actual situations and the problems in Bangladesh for hygiene education were reexamined from the previous research and the hearing.

6) Workshop to dissolve the problems related to arsenic pollution of drinking water in other regions

As part of the main study, this part was responsible to organize workshops with inviting researchers from each of the polluted areas in order to exchange information and discuss the possible political and technological counteractions. The major points of our concern was how much confident are the monitored data on contamination and possible health effects and what type of counteractions are possible. Finally, it is important to summarize appropriate recommendations to policy-makers as well as researchers.

3. Results and Discussion

1) Evaluation against Harmful Effects of arsenic to Human Health

(1) In the surveys of 2000 or 2001 refered to West Bengal, India, we could know that 9 out of 12 families or one out of 19 families used the dangerous drinking water over the safe water of guidline of 50 ppb in India and arsenic level in drinking waters. In the suvey of 2002 refered to Bangladesh, the drinking waters obtained from 31 families were safe. But the arsenic in 8 out of 13 tubewell's water for the household's purpose were over 50 ppb.

(2) The ratio of (MMA+DMA)/total arsenic in urines has been already reported that the normal values ranged from 70 to 90 %. We found out 2 persons in the suvey of 2000, 11 persons in 2001 and 2 persons in 2003 who had the abnormal metabolic efficacy against arsenic.

(3)When estimating the urinary arsenic compounds such as As(III), DMA, MMA and As(V) having the normal metabolit against arsenic, the relationship between As(III) and DMA, As(III) and DMA and MMA were in good agreement($P<0.05$). Higher were the arsenic level in drinking water, higer became the urinary total arsenic.

(4) Comparing the urinary arsenic compounds from male with those from female within the same couples, the relationship of urinary As(III), DMA and MMA were in good agreement($P<0.05$).

(5)Comparing the urinary total arsenic or DMA with the age having the normal metabolit against arsenic, both data gave the good relationship.

(6)Relationship between the arsenic concetrations in hairs and those in drinking waters were in good agreement($P<0.05$).

(7) When estimating the difference of the arsenic concentrations in one spot urines against in the 24-hour urine, there were no difference.

2) Studies for plan to maintain the global environmental protection to the widely arsenic- affected groundwater

Around 15 ng As/m3 of water-soluble arsenic, mainly inorganic (arsenate), was found in SPM in houses. Fare amount of arsenic was present in straw and fresh cow dung, both of which show similar chemical patterns. Methylation seemed to proceed during drying process of cow dung under sun, but methylated species, except trimethylarsine oxide, disappeared during burning. The results show that arsenic pollution through respiration of in house air might have significant health effects and that more detailed research is necessary for proper assessment and management to protect human health from arsenic pollution through irrigation.

3) Studies for Harmful Effects of arsenic to agricultural land and agricultural products

The suveyed spots were Domkal, Jalangi, Hariharpar and Raninagar-II blocks in Mushidabad district, west Bengal, India.

(1) Comparing the arsenic concentration in the surface soil with that in the irregated water from the irrigated lands in Domkal and Jalangi blocks, both data were in good agreement ($P<0.05$).

(2) The arsenic in the irrigated land affected rising the arsenic level in the agricultural products. The arsenic concentrations of the peel of potatos and onion were higher than those in their meats.

(3) The arsenic concentrations in the irrigated water in Hariharpar and Raninagar-II blocks ranged from 0.019 to 0.12 mg/l. The mean of total arsenic per year scattered from the arsenic-contaminated groundwater was1.79 kg/year and the total arsenic per ha was 5.02 kg/ha.

(4) The average of arsenic concentrations in surface soil, soil around the root of agricultural plants and soil ranged from 0 to 30 cm depth were 14.2, 13.7 and 14.8 mg/kg, respectively. The increase of the depth was lowering the arsenic concentrations in soil.

(5) The average of arsenic concentrations in the agricultural lands for peper, ginger and arum were 15.3(ranged from 12.1 to 17.5 mg/kg), 17.5(ranged from 12.2 to 21.0 mg/kg) and 15.3 (ranged from 11.4 to 19.8 mg/kg)mg/kg, respectively.

(6) Relationship between the arsenic concetrations in soil around the agricultural plants or in soils of each depth and the arsenic concentrations in irrigated water were in good agreement ($P<0.05$).

(7) The average of arsenic concentrations from the roots, steams and leaves cultivated in arsenic-contaminated land were 996(ranged from below 0.04 to 4850 ng/g), 297(ranged from below 0.04 to 2900 ng/g) and 246 ng/g(ranged from below 0.04 to 1600 ng/g), respectively. Comparing the arsenic concentration in irrigated water with those in roots, steams and leaves, both data were in good agreement($P<0.05$).

4) Contributing Study Topic: Study on the development of large-scale arsenic removal plant in drinking water supply system.

The large-scale drinking water treatment is focused on in this study. The arsenic removal pilot plant was installed in Nawabganj, which is located on the west of Bangladesh. The diagram of the pilot plant is shown in Fig.1. The coprecipitation and filtration method is adopted in the pilot plant. This is because the running cost is relatively low (ca. \$0.05/m^3) and the iron ion, which is usually coexisting with arsenic in groundwater, can be utilized for coprecipitation of iron and arsenic. Nanofiltration (NF) membrane separation system is also equipped considering the advanced treatment of arsenic. During the study, arsenic removal efficiency under a variety of operational conditions had been investigated treating local groundwater which is arsenic contaminated.

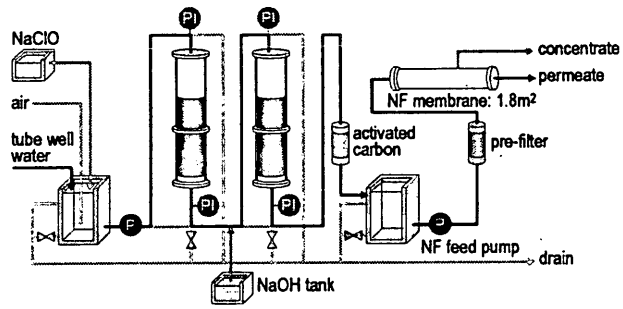

Fig.1 Diagram of the pilot plant

In addition to the pilot plant experiment, the periodic surveys of 10 shallow tubewells (hereafter Tubewells) had been conducted to investigate the arsenic contamination situation in Nawabganj area and the characteristics of coexistent ions and substances. Tubewell surveys had been conducted six times from November 2000 to April 2002.

As the result of tubewell surveys, it was found that 7 Tubewells out of 10 exceeded the Bangladesh arsenic standard, which is 0.05mg/L, in Nawabganj area. In addition to the arsenic, higher concentrations of iron and manganese than WHO guideline values for drinking water guideline were detected in the Tubewells. Other than Tubewells, uranium over the WHO guideline value was detected in some dugwells.

As for the seasonal fluctuation of arsenic concentration, arsenic concentration tends to be relatively low in the dry season and it becomes high in the beginning of the rainy season. During the rainy season, arsenic concentration decreases again toward the beginning of the dry season.

Regarding the relationship between iron and arsenic in groundwater, many studies have indicated that there is strong correlation between them. However, in the case of our survey, they are not always correlated. There were some tubewells that contain high concentration of arsenic but low iron.

As the result of the arsenic removal plant operation, good arsenic removal could be seen during whole operation period, that is, arsenic concentrations in treated water have been under the Bangladesh water quality standard (0.05mg/L).

5) Research on the Environmental Plan for the Earth Environment Preservation to the Over

(1) Performance evaluation of arsenic removal by drinking water supply systems in Bangladesh

We conducted a filed survey in August 2001, March 2002, and October, 2002 in Narayaganj and Manikgonj district in Bangladesh. Two types of most promising technology, so far, of arsenic removal plants, i.e., AIRP (Arsenic and Iron Removal Plant) and IARP (Iron cum Arsenic Removal Plant.), were investigated. AlRP consists of a raw water tank which is connected to a contaminated tube well up-flow sand filtration unit and a treated water tank. IARP adds a down-flow sand filtration unit after a similar up-flow sand-filtration instead installing a treated water tank. Comparing both types of the plant, it is generally concluded that up-flow sand-filtration is not sufficient in satisfying the treated water below Bangladesh drinking water standard for As, i.e., 0.05mg/L, while an additional down-now sand-filtration improves satisfactorily treated water quality in terms of As. However, it needs further treatment, if we want to get safer water of As content below WHO guidehne (0.01mg/L). We need a strategic approach of such a stepwise improvement of arsenic removal plant. In this sense, it is recommended, first, to have a set of treatment consisting of iron-arsenic co-precipitation followed by an up-flow and down-flow sand-filtration in sequence. Then it seems rational to an adsorption device as a further improvement, if necessary. It was observed that treated water was deteriorated a little bit at the tap probably due to As-release from accumulated sludge particles in the receiver part, of the treated water. Therefore, it is strongly recommended to add a valve for washing the accumulated sludge periodically. The quality of treated water is satisfactory in most cases, however, an additional process (such as adsorption resins) may be effective to secure 0.05 mg/l. It is recommended to install an additional process to remove As in order to satisfy WHO guideline of 0.01 mg/l.

(2) Development of the Arsenic Removal Technique in Small Scale Drinking Water Apparatus such as Well

① In the arsenic removal by cell surface engineering, ArsR repressor protein was specifically combined with arsenic.

As a result of experiments, when the amount of removal of pTV+*lamB-arsR* and pTV+*lamB* was measured, *lamB-arsR* has removed more arsenic. In the arsenic removal capability of pTV+*lamB-arsR*, the arsenic concentration with solution of 0.5 mg/L decreased to the maximum amount 1.54 μg.

② pUCD615 + *arsO.P.* plasmid was developed. In this procedure, both *arsR* gene which collected from *E. coli* and *O/P* of *ars* operon were introduced to the up-stream of *lux* operon which express the luciferase luminescence.

As a result, As(Ⅲ) concentration was determined in the region of 1 μg/L - 1 mg/L, and this detection sensitivity was the same as the quantity limit of the silver-diethyldithiocarbamate method.

③ Arsenic absorption ability of vegetable fibers of *Cocos nucifera* L and *Phapis humilis* were excelled.

Arsenic absorption of *Noottopteris antique* was distinguished in plant ferns.

④ Hygiene situations and sociality for hygiene education in Bangladesh were followings: Lack of education and awareness among the majority of people as they do not know or understand importance of hygienic practices, Lack of coordination and cooperation among governmental, non-governmental and donor organizations in planning, policy making and implementing water supply and sanitation (WSS) activities, Lack in the supply and demand process of the WSS facilities/components, Inadequate funds and poor economic status, Lack of appropriate technologies, Lack of understanding of the existing technologies, Lack of monitoring and evaluating the system, Lack of appropriate training and education programs, Lack of support for small NGOs from big NGOs, governmental and other concerned organizations, Lack of importance given to women's involvement at all levels: policy making and implementing (governmental and NGOs) as well as community and household levels.

6) Workshop to dissolve the problems related to arsenic pollution of drinking water in other regions

Two workshops and one symposium were organized in collaboration with United Nations University and WHO.

In Shanxi province where there is a hot spot area polluted with arsenic, for example, arsenic measurements have already been performed in general and people whose drinking water was contaminated with arsenic have been recommended to use safe water from neighbor households. However, it was evident that poisoned people showing some typical symptoms such as keratosis, melanosis as well as skin cancer. Our concern was a possible increase of various cancers as suggested by some epidemiological studies from now on even among the people who have not been exposed to not-too-high level of arsenic in drinking water. Moreover, there was case of arsenic intoxication due to exposure to indoor air pollution wish arsenic through low-quality coal burning for heating and cooking in Guizou.

As for the pollution in Bangladesh or other countries including Thailand, the information has been summarized by UNICEF for the last several years, showing a large number of people have been affected.

4. Conclusion

1) Evaluation against Harmful Effects of arsenic to Human Health

The arsenic standard of drinking water in both India and bangladesh is below 50 ppb. Ten out of surveyed 52 families used the arsenic-contaminated water over the arsenic standard. The urinary total arsenic from these families were high. The relationship between three kinds of arsenic metabolites such as As(III), DMA and MMA were in good agreement($P<0.05$).Fourteen out of surveyed 249 people got the damage of arsnic metabolism. Comparing the urinary arsnic coumponds

from the males with those from the females using the same couples, the urinary arsenic patters were the same ones. There were no difference of arsenic metabolism between the male and female. The rise of the ages gave the decrease of the urinary total arsenic.The relationship between the arsenic in hairs and the arsenic concentration in drinking water were so in good agreement that the change of the arsenic in hairs should reflect the change of arsenic concentration in drinking water.

3) Studies for Harmful Effects of arsenic to agricultural land and agricultural products

Conparing the arsenic concentrations in irrigated waters with those in surface soil, soil around the agricultural land and soil collected from each depth, there were so the good relationships that the results suggested that the arsenic level in agricultural land should depend on the arsenic concentrations in irrigated waters. When estimating the arsenic concentrations in each depth of 5 cm from the surface to 30-cm depth, the increase of depth led the decrease of arsenic concentrations in soils. The reslut suggested that the arsenic contamination of agricultural land should be due to the arsenic contamination of irrigated waters and that the arsenic contamination should progress from the surface to deep part.The arsenic concentrations of the meat of potatos and onions were so lower than those of their peels that there might be the reserve of arsenic in peels. The arsenic level of agricultural products cultivated in high arsecic contaminated land were high.

4) Contributing Study Topic: Study on the development of large-scale arsenic removal plant in drinking water supply system.

Fig.2 shows the arsenic concentrations after each process under adequate operational condition. The arsenic concentration of the raw water was reduced to 44 □g/L after Sand Filter column 1 (SF1). Therefore, it is possible to remove arsenic and iron simultaneously in the sand filtration process, if the iron contained in the raw water is converted to iron hydroxide flocs by aeration in the oxidizing tank. The arsenic removing performance of sand filtration is much affected by the concentration ratio to the coexisting iron. Therefore, higher arsenic removal performance may be obtained by adding the iron coagulants.

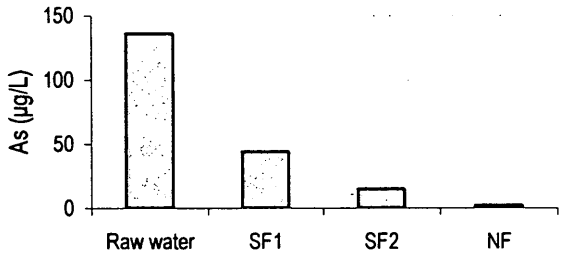

Fig.2 Arsenic removal after each

As for the advanced treatment using NF equipment, arsenic can be removed stably less than 0.5μg/L. Although NF equipment has the demerit of high cost (ca. $3/m^3$), there may be some demand of perfectly arsenic-free water in such as hospitals and schools.

5) Research on the Environmental Plan for the Earth Environment Preservation to the Over

The following knowledge was acquired in considerations about arsenic removal by genetic engineered *E. coli* and hygiene concepts, etc.

Example of application of the heavy metal removal by the repressor protein in cell surface engineering was not found out, and not only arsenic removal but the application to other heavy metal removal was suggested.

Luminescence biosensor that arsenic was taken in the *E. coli* cell instantaneously could be constructed. Furthermore, it will be possible to raise the sensitivity adding mutant in the *O/P* part.

On the other hand, although the requirements of hygiene ideas and education are recognized, school attendance rate is improving recently. At the present, support activities are activating, however, a community leader's (landowner and society for men) controlling power is strong, and if the present conditions are not broken, advance of women and social development cannot be achieved.

6) Workshop to dissolve the problems related to arsenic pollution of drinking water in other regions

As common to all of the cases reported in the workshops, it was needless to say, one of the most important action is to stop exposure to arsenic in the water and another is to remove arsenic from the water if alternative safe water is not available. To this end, monitoring and education of the public must be the primary. Moreover, epidemiological study on the polluted populations as well as health administrations for protecting them from further exposures. In relation, capacity building in terms of education of the public as well as trainings of the professionals is also emphasized. Researches on the possible chronic effects like cancers in various organs should be stressed in order to improve the knowledge help for considering safer standards.

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