

## 2.2 Medical examination and support of Toroku patients by UOM

The Faculty of Medicine, University of Miyazaki (UOM), has been performing the medical examination and health check of the chronic arsenic poisoning patients in Toroku village for 35 years from 1972 just after of the official recognition of the Toroku arsenic disease. From 1974 the staff of other faculties of UOM has been supporting the movement of Toroku arsenic patients, as the member of NGO, “Toroku Support”, with the citizen more than 30 years. They have supported Toroku patient’s fights for the company in responsibility and the law system to take care of patients, as the hands & foote of patients. UOM staff has also investigated the contamination of water and soil in Toroku.

The NGO, “Toroku Support”, was developed to “Asian Arsenic Network” (AAN) in 1994 after the 20 years support for the recovery of the human rights of Toroku patients. AAN began to transfer the long experience of Toroku patients to the victims in the arsenic affected area in Asia. Many staff of UOM is the member of AAN and has supported the activities of AAN in the professional fields of Engineering, Agriculture, Medicine and Social System.

Here, the situation of arsenic contamination and mitigation measures in Ganges basin are only mentioned out of the whole arsenic contamination problems in Asia.

## **3. Arsenic Contamination in Ganges basin**

### 3.1 Arsenic contamination in Bangladesh

#### *(1) Situation of arsenic contamination*

It was at the west region along the Ganges, border to India, in 1993 that the arsenic contamination of groundwater was first detected in Bangladesh, where the 8 chronic arsenic poisoning patients were confirmed in 1994 at the first time in Bangladesh. Arsenic contamination is now seen across the nation, in 61 out of 64 districts, excluding the Chittagong mountainous region in the southeast (BGS *et al.*, 2001). It is presumed that 29,000 thousands people are drinking tube well water beyond the standard, 50µg /L and the number of chronic arsenic poisoning patients amounts to 375 thousands (Ahmed *et al.*, 2002).

The survey for arsenic contamination has been performed on a nationwide scale to grasp the whole situation of arsenic contamination under World Bank help since 2002. It was found that 30% of tube well was contaminated with arsenic and there were 15% of villages where the ratio of arsenic contaminated well was higher than 80%. 38,500 chronic arsenic poisoning patients were confirmed, too. Now, an

alternative arsenic-safe water supply system is to be installed in about 10 thousands villages, where the ratio of arsenic contaminated well was higher than 80%.

(2) Research and countermeasure by UOM

UOM and AAN had first visited Bangladesh in February 1996 and had made detail surveys in a model village, Samta village, Jessore district, in order to get the mechanism and countermeasure for arsenic contamination, from 1997 ~2000 in collaboration with NGO, RGAG (Research Group for Applied Geology, Japan), NIPSOM (National Institute of Preventative & Social Medicine, Bangladesh) and BUET (Bangladesh University of Engineering and Technology).

The arsenic contamination map, Fig.1, showing the arsenic concentration of all tubewells in Samta village, made clear that more than 90% of tube wells had arsenic concentration above 0.05mg/l (Tanabe *et al.*,2001). The arsenic contamination map became a typical method to investigate the mitigation for arsenic contamination in the arsenic affected villages in the subsequent programs by World Bank and JICA, etc.

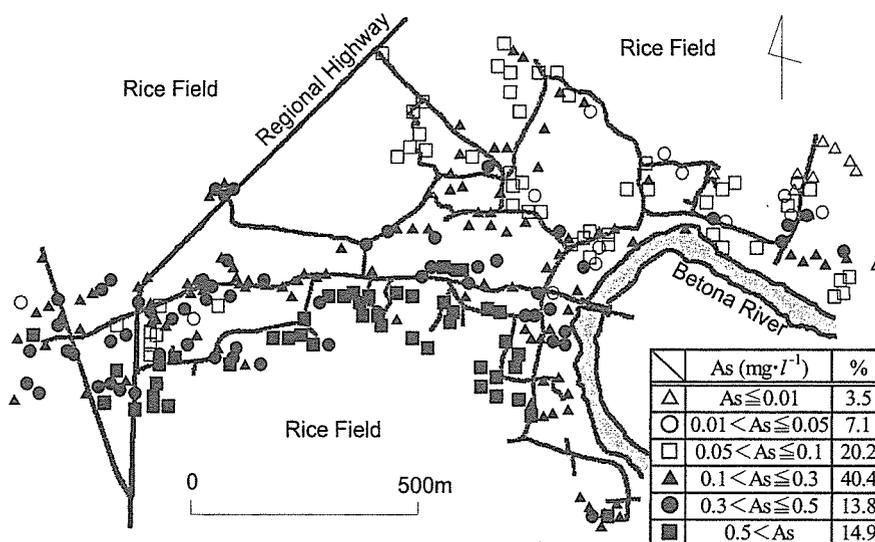


Fig. 1 Distribution of arsenic concentration of groundwater in Samta village

We had examined the water quality & water flow of groundwater in Samta village to harmonize the arsenic distribution of the arsenic map, and got the results that the arsenic in the underground was released to groundwater under the reduced condition in groundwater. The medical examination was performed by the medical

doctor group of UOM and University of Kumamoto with NIPSOM, too.

We also had developed the Pond Sand Filter (PSF) as an arsenic-safe water supply unit. PSF is comprised of two systems as shown in Fig.2 (H.Yokota *et al.*,2001). One is a Horizontal Roughing Filter (HRF) developed by the All Indian Institute of Hygiene and Public Health (AIIH&PH) (Nath *et al*, 1997), and the other is a Slow Sand Filter (SSF).

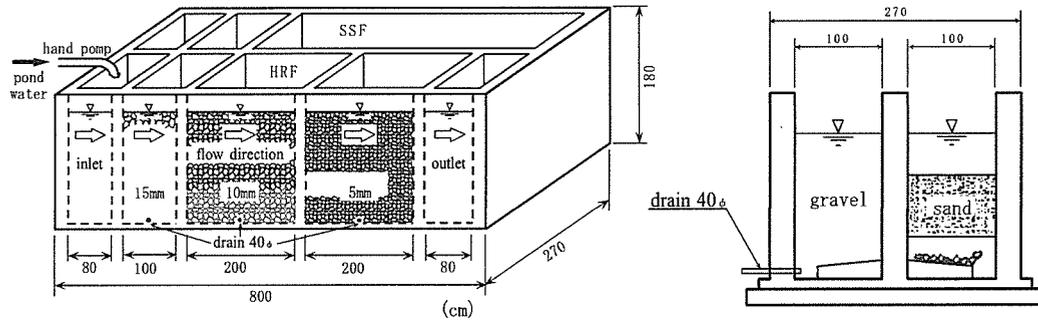


Fig. 2 Structure and size of PSF

The performance of PSF is very good, and PSF is now a representative unit for the community water supply system. About 30 units of PSF have been installed in Bangladesh until now. Some members of the young arsenic committee of Samta, who had worked with us for the installation of PSF, have grown up as professional engineers for the alternative water supply system and have been working as the staffs of AAN in Bangladesh these several years.

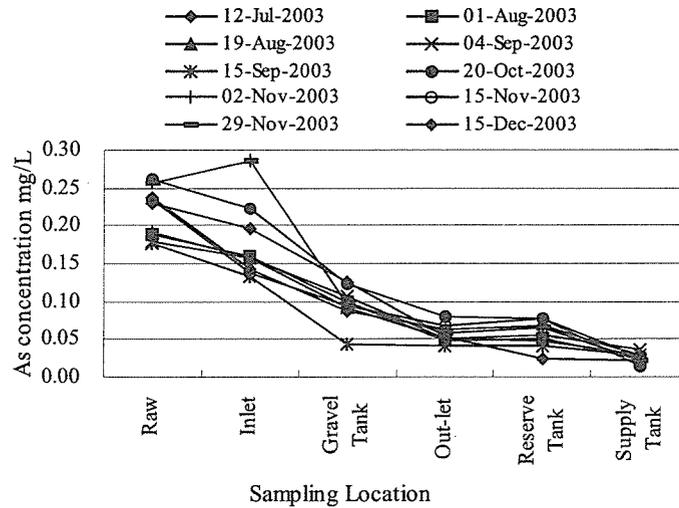


Fig.3 Arsenic removal performance for Hybrid use of PSF/ GSF

Only one fault is, however, difficult to use the pond water in dry season for the dry up of pond water. Therefore, the hybrid use of PSF, as an arsenic removal unit by putting the tube well water in PSF during dry season, named as GSF (Gravel sand filter), has been estimated from 2000 by UOM.

After element/model tests in Bangladesh and laboratory of UOM from 2000 to 2001, we installed an arsenic removal unit, GSF with a perfect same-sized unit as PSF. The performance for arsenic removal of GSF has been examined from December, 2002 in the second model village, Marua village, Chogacha upozilla, Sharsha district. The Fig.3 shows the arsenic removal performance in 2003, which shows the good utility of hybrid use of PSF/GSF (Hiroshi Yokota *et al*, 2004 ).

The good performance for the exclusive unit, GSF, is obtained under the periodical maintenance of the unit, too. And until now 4 units of GSF have supplied the arsenic safe water in Bangladesh.

The tests for arsenic sludge, discharged from GSF, have also been performed to check out characteristics of arsenic leaching from the sludge. The sludge is the material of iron co-precipitation with arsenic in the gravel spaces of the gravel tanks, which is to be discharged from the GSF during periodical maintenance.

Fig.4 shows the ratio of arsenic content, leached out from the sludge mixed with cement, to the total arsenic content in the sludge. More than 90% of arsenic content in the sludge can remain in the sludge without mixing cement. The percent value increases up to 94% and 96% in the case of mixing cement of 2% and 10% of the sludge, respectively (Miah M., Hussainuzzaman, *et al.*, 2005, 2006)

These test results will serve as the basic data for the future sludge disposal methodology development.

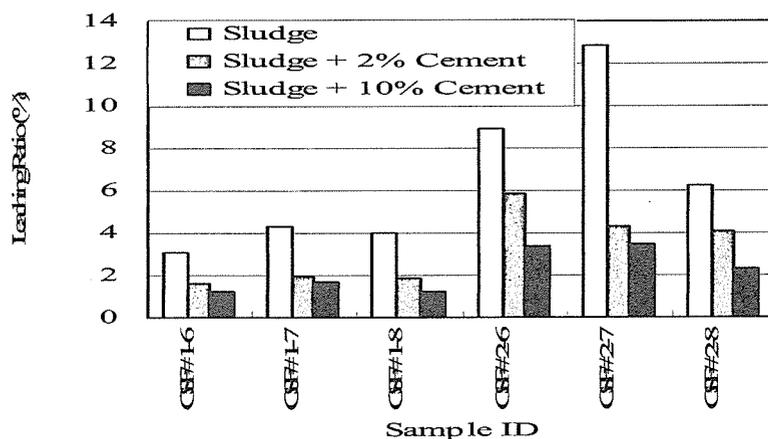


Fig.4 Arsenic leaching ratio from arsenic sludge

UOM has been visiting the GSF site these years and collected the data for getting the arsenic removal mechanism as improving & maintaining the function of GSF and the data for the treatment of arsenic sludge. These techniques have been transferred to the arsenic young committee of Marua, who had collaborated with us for the improvement and maintenance. The skilled up members of the committee are now working in JICA/AAN project and AAN Bangladesh.

### 3.2 Arsenic contamination in Nepal

#### *(1) Situation of arsenic contamination*

Nepal is divided into three zones, that is, the Himalayas mountainous region and hill country and Terrai plain in geographical features. There are 20 prefectures in the Terrai plain (75 prefectures in whole country), where 47% of the overall populations, 9.7 million people, are now living. A lot of rivers flow down from the mountainous region in the north, and join the Ganges in India .

The arsenic contamination of groundwater was investigated in three prefectures of the east part of Terrai plain (sample number: 268) under the support of WHO in 1999 and the possibility of the arsenic contamination in the Terrai plain was pointed out. The project of countermeasure for arsenic contamination started in 2002 under help of UNICEF, and the examination for 17,066 tube wells, including the preliminary examination in 2001 (4,000 tube wells: the number of all tube wells in the Terrai plain is about 200,000) have finished in July, 2002.

As a result, the well water of  $As > 50 \mu\text{g/L}$  was detected in 18 out of 20 prefectures, and it turned out that the groundwater in Terrai plain had been contaminated almost overall, though the ratio of tube well beyond  $As > 50 \mu\text{g/L}$  was 4% as the average in Terrai plain. The highest value of the ratio, 25%, was seen in Nawarlparasi prefecture (center part in the Terrai plain), where the maximum value of arsenic concentration was  $436 \mu\text{g/L}$  and 49 patients out of 1,102 people were detected. It is estimated that about 400 thousands people are now drinking the well water of  $As > 50 \mu\text{g/L}$ , though the severe arsenic contaminated area seems to be unevenly distributed (ENPHO, 2004).

#### *(2) Investigation and countermeasure by UOM*

UOM and AAN has surveyed in model villages in the Nawarlparasi prefecture from 2002 with RGAG, DWSS (Dept. of Water supply & Sewage, Nepal) and NGO, ENPHO, Nepal. Sampling test of 4 villages at Nawarlparasi in December, 2002, had showed the worst arsenic contamination in Nawarlparasi. So, we surveyed in detail the arsenic contamination of 2 out of 4 villages. The investigation data of Knowahr

(May & Decembe,2003) and Pathokauli (Decembe,2004) are shown in Fig.5 (Kimiko Tanabe *et al.*, 2004). The ratios of tube well beyond As>50µg/L in these model villages, 70% (Knowahr) and 95% (Pathokauli), were extremely high compared with 25% of the mean value in Nawarlparasi.

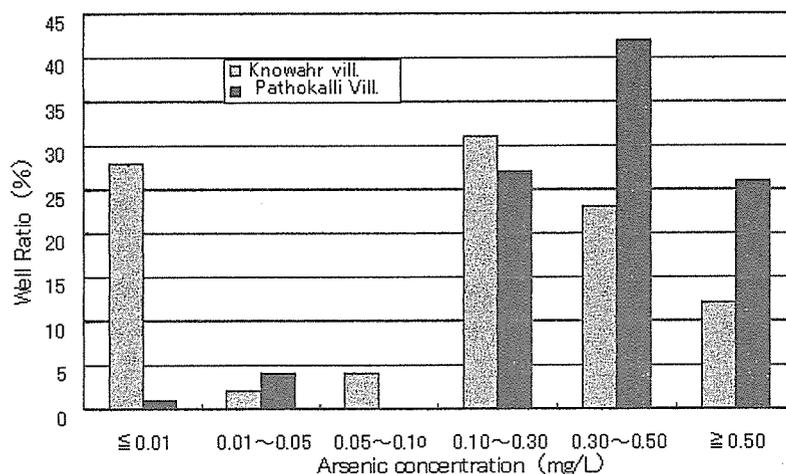


Fig.5 Arsenic concentration of wells in model villages

MIT and Texas University distributed domestic arsenic removal units under the support of World Bank at Knowhar village by using the arsenic contamination map made by us. Regarding Pathokauli village we installed 4 deep tubewells for community use as an emergency measure, which has been supplying arsenic-safe water to villagers until now.

We had medical examination in these villages. The percentages of arsenic patient in Knowahr and Pathokauli villages were 6.5 % and 18.6%, respectively. The symptom of arsenic disease showed the mild level that was major in the both villages regardless of heavy arsenic contamination of tubewell water. It is, therefore, requested to supply the arsenic-safe water quickly so that the arsenic disease is to be improved in the stage of mild symptom by drinking safe water. The political insecurity in Nepal has, however, stopped our mitigation program from 2005.

And, the agriculture survey was performed in the villages to show that in the dry season the cultivation of rice farming has not done and the irrigation water from not groundwater but river water was used for the fields of wheat and vegetables in the dry season in Nawarlparasi. So, there is no influence for arsenic contamination through the pumping groundwater up for irrigation in the dry season, which is

considered one factor of arsenic contamination in Bangladesh.

### 3.3 Arsenic contamination in Uttar Pradesh State, India

#### *(1) Situation of arsenic contamination*

The arsenic contamination in Uttar Pradesh State (UP State) along the medium basin of Ganges was first confirmed at Ballia district in 2003 through the investigation of Jadvpur University (School of Environmental Studies, 2005). The severest disaster was recognized as those in Ganges down basin of West Bengal State, India and Bangladesh, through the sample investigation of groundwater and medical examination at 5 blocks in east area of Ballia district.

CGWB (Central Ground Water Board) detected the arsenic contamination of groundwater at 4 districts in the west and 2 districts in the east of UP State, under the sampling tests of all the districts in UP State in 2003. The arsenic contamination of 4 districts in the west was along Yamuna river, a tributary of Ganges, and that of 2 districts in the east was along Ghaghara river where many rivers join from Nepal. CGWB recognized the high and wide arsenic contaminated areas in the east of Ballia district to recommend the quick installation of alternative water supply unit (CGWB,2004).

On the other hand, UNICEF surveyed in the 8 districts in the north of UP State bordering to Nepal in 2003~2004, and confirmed the arsenic contamination at 4 districts from the center to east in the north of UP State. UNICEF, in collaboration with Dept. of Water Supply, UP State (Jal Nigam), performed the sampling survey of public tubewells in Lakhimpur-kheri and Ballia districts in 2005 and showed the percentage of arsenic contamination of 35.3 % (As>0.01mg/L), 2.1 % (As>0.05mg/L) for Lakhimpur-kheri district and 26.4%(As>0.01mg/L), 4.4% (As>0.05mg/L) for Ballia district (UNICEF,2005).

Now, UNICEF/UP State finished the installation of 250 deep tubewells in Ballia district and is performing the sampling tests in 40 districts (all the districts in UP State: 70) including Gajipuru district next to Ballia district.

#### *(2) Surveys by UOM*

We, UOM and AAN had a preliminary survey of arsenic contamination in Ballia district from 7 to 10 January 2006 with our counterpart, NGO Eco-friends, India. We tested tubewell water and examined medically villagers in 4 blocks in Ballia, and decided the sites for model village of our mitigation program. The survey results in Ballia showed that 1) many villagers had little awareness about arsenic poisoning, 2) some villages were out of the arsenic-safe water supply

system planned in the UP arsenic mitigation program and 3) professional techniques were seemed to be needed in the fields of arsenic-safe water supply system and diagnosis of chronic arsenicosis patients in the State of UP (Mitsuhiro Sezaki *et al.*,2007).

From 5 to 20 September, 2006 we again surveyed UP State to get fundamental data of arsenic contamination in the sites for model villages in Ballia and Gajipur districts. The mitigation measure in Ballia district was so progressed to install 250 deep tubewells by UNICEF/UP State. And, more 250 tubewells will be soon installed in future by UNICEF/UP State. We got the arsenic contamination data for 3 villages as shown in Fig.6. We now get the unofficial decision the JICA Partnership Program, a technical cooperation program at the grass-roots level, to JICA in order to mitigate arsenic contamination in UP State. JICA will have the meeting for the official decision of the program with UP State and India. UP state has been expecting the JICA project.

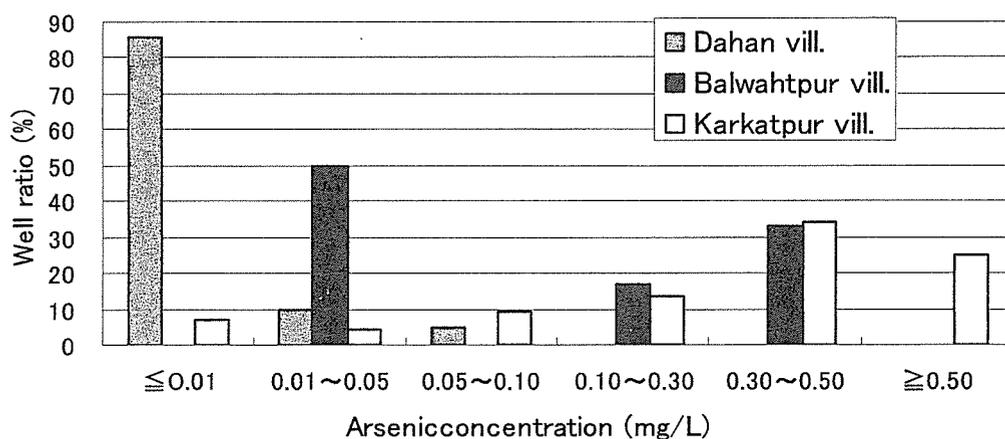


Fig.6 Arsenic concentration of wells in model villages (UPState,India)

#### 4. Collaboration between UOM and NGOs for the mitigation of arsenic contamination in Ganges basin

As mentioned above UOM started the arsenic mitigation projects in the model villages, Samta from 1997 and Marua from 2002, followed by Nepal (2002~2004) and UP State (2006~) projects as shown in Fig.7. All the projects have been performed with AAN, RGAG, and local NGOs. The main role of AAN is to coordinate the project. RGAG is the professional group for the applied geology and the co-researcher of UOM. The local NGOs are the young arsenic committees in

Samta and Marua villages, who have helped our field surveys and grown up to the cite engineers in JICA/AAN Projects. The NGOs, EMPHO in Nepal and Eco-friend in UP State are our counterparts and have worked as the coordinator and co-researcher.

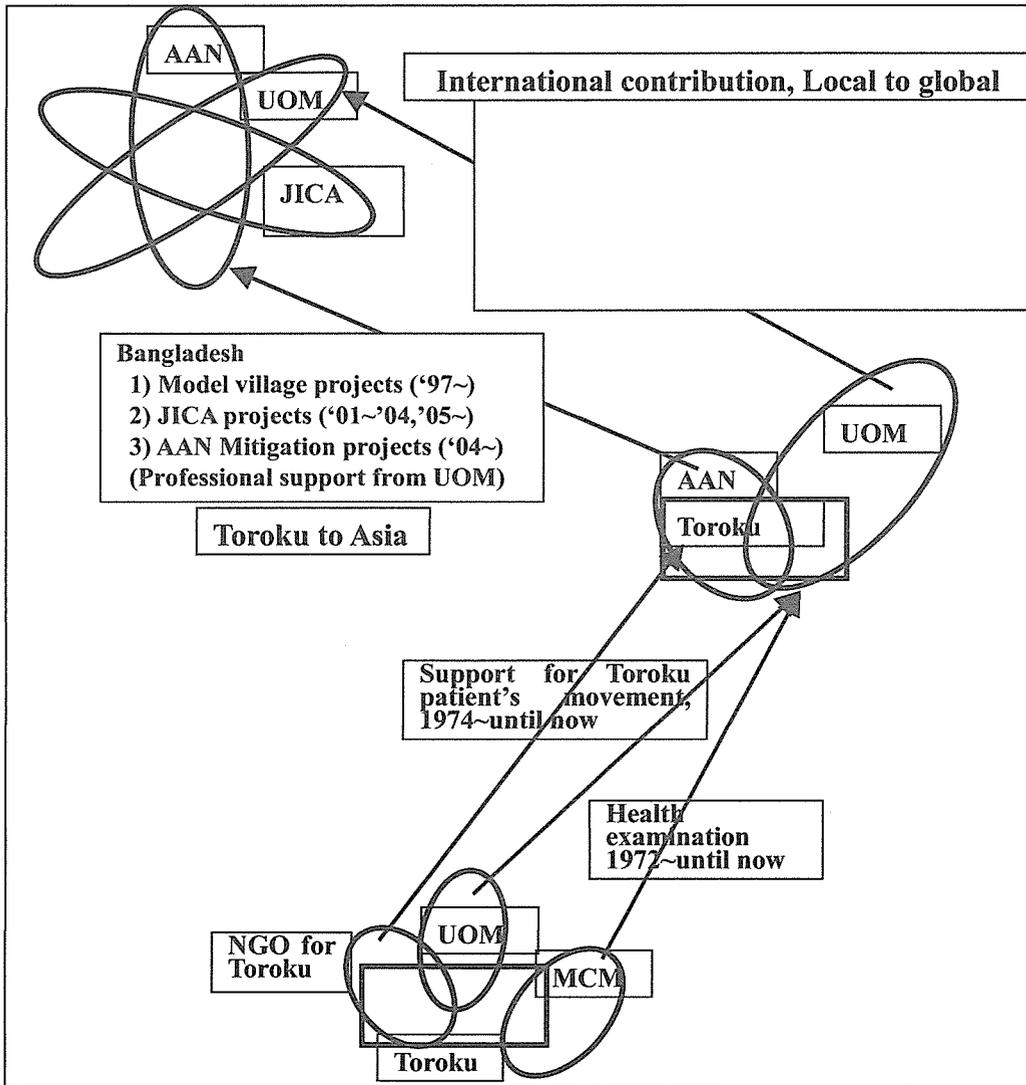


Fig.7 Collaboration between UOM and AAN from Toroku to Asia

AAN had finished successfully JICA Project from 2001 to 2004 after the model village projects and now is doing new 3 years JICA Project (PROTECO) from 2005. AAN-Bangladesh has carried another arsenic mitigation programs different from

PROTECO from 2004. The number of staff in the former (PROTECO) is about 30, and 10 in the latter. UOM has often supported those projects in the development of alternative water supply system. AAN has developed the practical technology for awareness, health care and arsenic-safe water supply unit through the JICA Project. We, UOM and AAN, RGAG have pushed forward the transfer of the practical technology combined with the theoretical knowledge of UOM and RGAG.

The Integrated approach for arsenic mitigation needs many fields of technology and science as civil engineering, agriculture, geology, chemistry, biology, medicine, sociology, etc. It is necessary not only to collaborate each other in these fields of technology and science, but also to have the professional knowledge each other across over these fields. The staff of NGO has worn the wide knowledge across over these fields to get the integrated solution in the front of arsenic mitigation.

The staff of arsenic research in UOM has been extended to the professional fields mentioned above and the arsenic mitigation in Asia is major project in the activities for international contribution of UOM.

As stated above UOM has been doing research on arsenic pollution for a long time. Beginning with Toroku arsenic disease and expanding to groundwater contamination in Asia, UOM's activities are consistent with its basic policies on education and research, and the slogan "Look at the World, Start with the Community".

One of role of university is to apply the results and potential of research to society, especially in the developing countries. In that case it needs the collaboration with NGOs home and foreign, which will lead to the formation of network & community in Asia. I think the network & community in Asia will be essential for us to proceed along the sustainable way in future.

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# ARSENIC REMOVAL FROM CONTAMINATED GROUNDWATER OF BANGLADESH WITH NATURALLY OCCURRING IRON

M. M. Hussainuzzaman, H. Yokota

Department of Civil & Environmental Engineering, University of Miyazaki, Japan

K. Tanabe

Cooperative Research Center, University of Miyazaki, Japan

**ABSTRACT:** It becomes increasingly difficult to find a sustainable arsenic free safe water source in Bangladesh. A community based arsenic removal system has been developed which can be used with most of the existing groundwater sources. The units utilize the natural iron of groundwater to remove arsenic by co-precipitation and do not need any added chemicals or power source. The paper describes those units and the directions of the future research in this aspect.

**KEYWORDS::** arsenic, naturally occurring iron, groundwater

## INTRODUCTION

Many parts of the world have arsenic problem in the groundwater; among the affected regions, Bangladesh has the most catastrophic arsenic pollution in the history of mankind. An estimated 29 million people are exposed to arsenic from tubewell water exceeding 50 µg/L (Bangladesh standard), 49 million people to more than 10µg/L (WHO guideline value) (Ahmed, 2003; BGS, 2001). It is necessary to develop low cost and sustainable safe water source for the people of this poor country. To achieve this many government and non-government organizations (NGO) are now working in Bangladesh to develop safe water options.

## BACKGROUND

University of Miyazaki has been working in Bangladesh to provide safe water to rural population since 1997. Initially, pond sand filters (PSF) were constructed to treat pond water for microorganisms and then supply it to the people. The units were working efficiently to supply safe water to the people (Yokota et. al. 2001). But during dry seasons these units could not be operated due to the lack of sufficient water in the ponds. To overcome that problem it was necessary to develop an arsenic removal unit, so that it can remove arsenic from groundwater and supply it to the people, especially during dry seasons.

There are a lot of ways to remove arsenic from groundwater (Ahmed, 2001). We along with many researchers have chosen arsenic removal by iron co-precipitation due to the availability of natural iron in the groundwater of Bangladesh (Tanabe et. al. 2001; Mamtaz and Bache, 2001; Yokota et. al. 2002). Besides,

a number of researchers (Song et. al. 2006; Wickramasinghe et. al. 2004; Han et. al. 2002; Meng et. al. 2001 and 2002; Mamtaz and Bache, 2001) have concluded that arsenic removal by iron co-precipitation is very efficient in the prevailing pH condition of Bangladesh groundwater without posing any potential risk of secondary contamination due to introduction of any new chemicals. Most of them have suggested that the naturally available iron can be used to remove arsenic from groundwater of Bangladesh.

Iron content of groundwater in that locality having those PSFs is quite high (Tanabe et. al. 2001, Yokota et. al. 2002). The University of Miyazaki has developed a system to utilize the natural iron to remove arsenic by co-precipitation using the structurally same unit of PSF. It was named as gravel sand filter (GSF). Therefore, this same unit can be used as PSF as well as an arsenic removal unit (ARU) during dry seasons.

## GSF AND ITS PRINCIPLE

Gravel sand filter (GSF) was developed with an aim to solve the low/no flow problem of the existing PSFs during dry seasons. Therefore structurally same unit has been used. Only difference was that the input was groundwater from a tubewell instead of pond water and the gravel size in the roughing filter unit was a bit smaller (5 mm diameter instead of 10 or 20 mm). So, far four GSFs with similar principles and setup have been operating in different locations in Bangladesh. Detail discussion on GSF#1 and its setup was covered in Hussainuzzaman and Yokota (2006).

In general, these units consist of inlet, gravel chambers as roughing filters, outlet, slow sand filter (SSF), and reservoir for treated water. The system gets

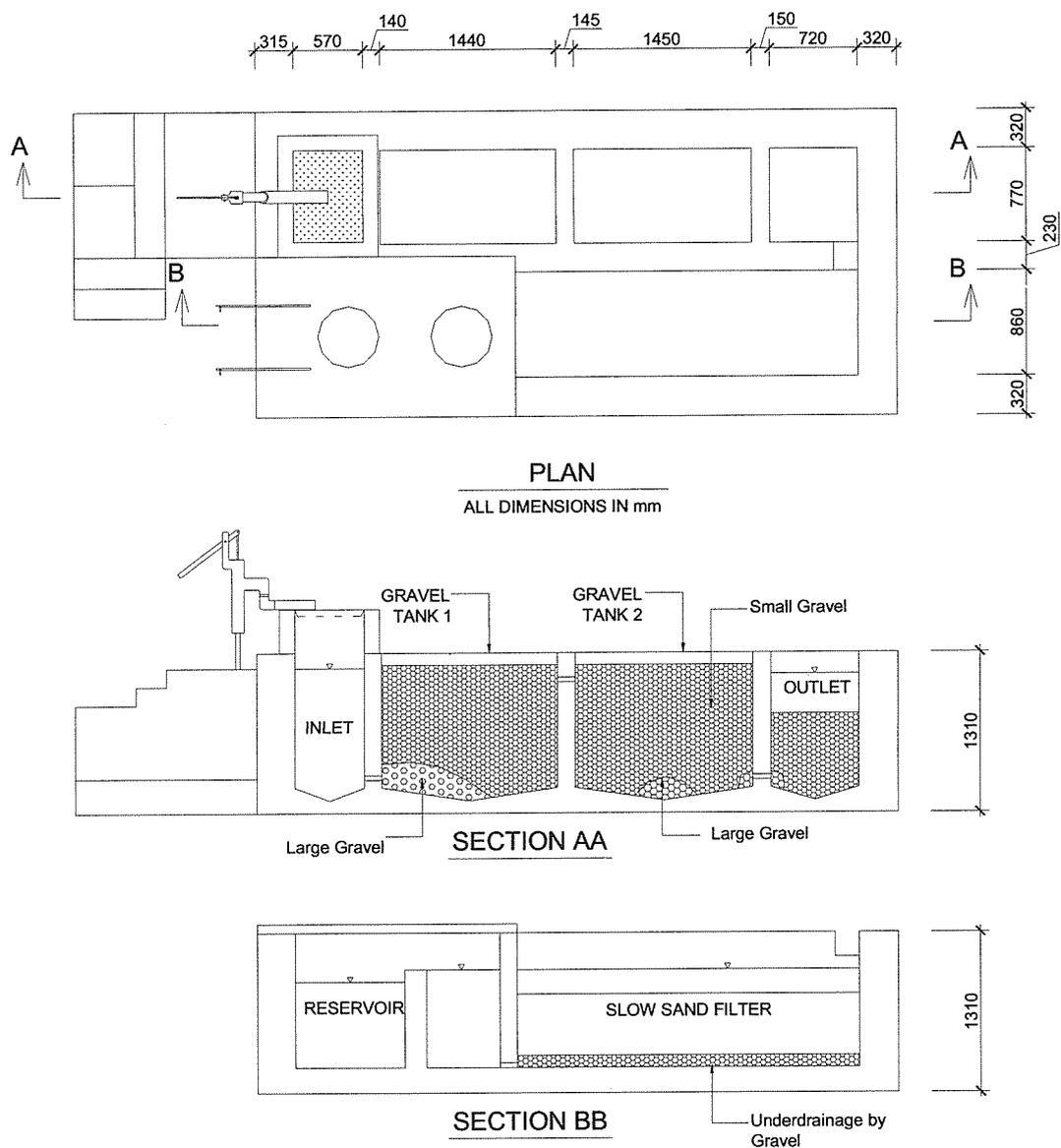


Fig.1 Sketch of the Gravel Sand Filter 1

raw tubewell water through pumping of the hand pump by the user, which gets the groundwater into the inlet. But in between the tubewell pump tip to inlet, there is some arrangements for aerations to supply the required oxygen to the water. In GSF1 and GSF2 this aeration is achieved by perforated metal trays; water comes on the tray and then showers down to the inlet. In GSF3 and GSF4 aeration is achieved through flowing the pumped water through an open channel, which is placed over the side walls of the unit, before reaching the inlet. Some aeration is also achieved through the surface of water in the inlet sections. Figure 1 shows the sketch of GSF1.

In this way of oxidation ferrous ( $\text{Fe}^{2+}$ ) turns into ferric ( $\text{Fe}^{3+}$ ) and arsenite ( $\text{As}^{3+}$ ) turns into arsenate ( $\text{As}^{5+}$ ). Solubility of the principal ferric compound i.e. ferric hydroxide [ $\text{Fe}(\text{OH})_3$ ] is very low in the prevailing

pH range and hence they form insoluble flocs which eventually settle down or being filtered out in the successive gravel chambers. But before getting removed in that way, these flocs adsorb arsenic onto its surface as arsenic species have strong affinity for iron hydroxide (Stollenwerk, K.G. 2003). Thus arsenic is removed from groundwater and relatively arsenic free water passes through the successive section of the unit to the reservoir.

Removed arsenic accumulates as arsenic rich iron sludge and they are periodically removed from the roughing filter by backwashing. The sludge removed that way goes to an underground reservoir; supernatant water goes to natural ponds from that reservoir.

## METHODS

Measurement of oxidation was measured in two ways. One was direct measurement of dissolved oxygen content of the water from different parts of the units. However this type of measurements were limited to GSF1 and GSF2. Dissolved oxygen measuring probe (Masterflex probe) was inserted to the water in the different chambers. In the gravel chambers there were sampling tubes installed to different depths and at different sections; measurement probes were inserted inside those sampling pipes to have direct measurements. These small sampling wells were purged before any kind of such measurements to ensure fresh water from the target layer enters there.

Other way of measurement was to measure the effects of oxidations i.e. the oxidized products of iron and arsenic. Ferrous ( $Fe^{2+}$ ) and ferric ( $Fe^{3+}$ ) irons were measured (HACH DR 2010) with different samples from different sections of the units. Field kit (Hironaka; <http://www.asia-arsenic.net/as3as5/ASKITNEW.HTM>) was used to measure arsenite ( $As^{3+}$ ) and arsenate ( $As^{5+}$ ). Afterwards, arsenic speciation was done during sampling using solid phase extraction cartridges (Supelco, 3-mL LC-SAX) following procedure as explained by Bednar et al. (2004). Samples were then tested with atomic absorption spectrophotometer (SHIMADZU AA 6650).

Sludge was collected from the bottom part of the inlet tank of GSF1. To collect that sludge first the wash valve, located at the bottom part of the chamber, was kept open until almost all the water drained out. Then it was shut down; at this stage the accumulated sludge in the gravel tank 1, started to come out slowly through the entrance holes at the bottom part of the partition wall between inlet and the gravel tank 1. The sludge was then being accumulated at the bottom of inlet tank. Grab samples were taken from that sludge; samples were then dried in the oven at 105°C for an hour to get rid of the aqueous part. Dried sludge was pulverized and was collected in plastic bottles. Metal extraction was conducted following Japanese standard procedure for metal extraction (JLT 046). From the extract arsenic was measured using the AAS (Shimadzu AA 6650); iron was measured by spectrophotometer (Shimadzu UV 1600) following the prescribed procedures for those machines.

## RESULTS AND DISCUSSIONS

### Effective oxidation

With no arrangements for elevated aeration the range of DO in inlet section was between 1-2 mg/L. Later with an increase of the tubewell pump height by 45 cm the DO range increased to 2.5 – 4.0 in the inlet. The measured dissolved oxygen (DO) has a general tendency to be very low (around or less than 0.5 mg/L) inside the gravels even though the concentration was quite high in the inlet sections. This may indicate that the incurred DO has been used up to oxidize the ferrous and arsenite to ferric and arsenate. Table 1 presents the

Table 1 Measured dissolved oxygen (DO) in GSF1

DO mg/L	Days in December 2004						
	5 <sub>(1st)</sub>	5 <sub>(2nd)</sub>	6 <sub>(1st)</sub>	6 <sub>(2nd)</sub>	7	11	12*
Raw	0.90	1.00	0.60	0.80	1.40	-	-
Inlet (surface)	1.40	1.40	1.40	1.20	1.00	-	2.80
Inlet (mid)	1.40	1.30	1.20	1.20	1.20	-	3.00
Inlet (bottom)	1.10	1.10	0.80	0.90	0.70	-	3.00
G-1 (80cm depth)	0.30	0.30	0.20	0.20	0.17	0.53	0.43
G-1 (40cm depth)	0.50	0.30	0.50	0.30	0.27	0.20	0.27
G-1 (surface)	0.20	0.30	0.30	1.00	0.20	-	0.10
G-2 (80cm depth)	0.20	0.10	0.10	0.10	0.10	0.10	0.17
G-2 (40cm depth)	0.30	0.30	0.30	0.10	0.20	0.07	0.20
G-2 (surface)	0.20	0.30	0.30	0.60	0.10	-	0.10
Outlet <sub>(70cm depth)</sub>	-	0.10	0.10	0.10	0.10	0.10	0.10
Outlet (surface)	0.30	0.20	0.40	2.20	0.10	0.10	0.10
SSF (surface)	2.80	1.70	2.50	3.50	1.90	1.90	1.90
SSF (70cm depth)	2.00	1.80	2.20	1.70	0.30	0.47	0.23
Supply	3.50	3.30	2.60	2.00	1.20	-	-

\*increased aeration with the pump raised by 45 cm.

The unit was operated in up-flow mode for the gravel roughing filters.

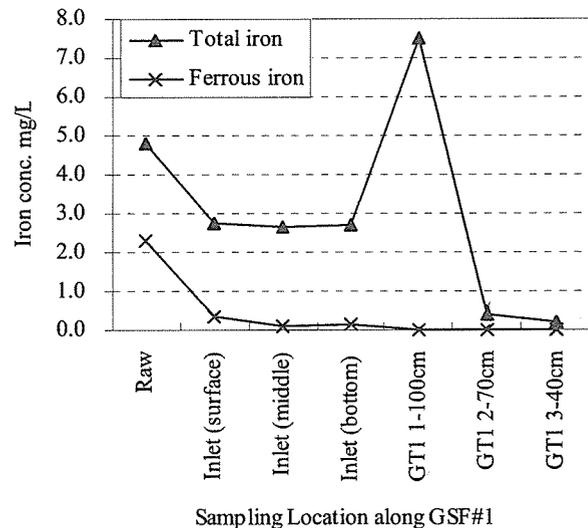


Fig.2 Oxidation of iron during 18-Jan-2004 (Diagonal flow mode)

measured DO before and after increasing the aeration by elevating the pump height. Data on 12-Dec-2004 is for measurements with raised pump.

Measured ferrous iron gives indication of prevailing oxidation. Figure 2 represents field-measured data of total and ferrous iron during 18-Jan-2004. During that time the roughing filters had diagonal flow arrangement. Represented data is the iron concentrations from raw water to the end of the gravel chamber 1. The iron concentration beyond the last point in gravel chamber 1 was too low to be considered. In the figure GT1 stands for gravel tank 1, then the cm values stands for the depth of the sampling location from the top of gravel chamber. The three locations in the gravel chambers represent the tentative diagonal flow path of water through the gravel tank 1; 1, 2 and 3 represents three

Table 2 Arsenic data showing oxidation effects as well as arsenic removal

Sampling Date	Sample ID	Total Arsenic (ppb)	Arsenic (III)* (ppb)
27-Aug-06	GSF1-RW	176	92
27-Aug-06	GSF1-Inlet(b)	154	19
27-Aug-06	GSF1-Outlet	112	67
27-Aug-06	GSF1-GT1-2	108	99
27-Aug-06	GSF1-Tap	72	0
11-Aug-06	GSF2-RW	74	-
11-Aug-06	GSF2-Tap	24	-
27-Aug-06	GSF2-RW	83	59
27-Aug-06	GSF2-Inlet	61	1
27-Aug-06	GSF2-Tap	18	0
11-Aug-06	GSF3-RW	70	-
11-Aug-06	GSF3-Tap	30	-
27-Aug-06	GSF3-RW	72	64
27-Aug-06	GSF3-Inlet(b)	75	15
27-Aug-06	GSF3-Tap	34	4
13-Aug-06	GSF4 RW	449	-
13-Aug-06	GSF4 Tap	49	-

\* Speciation sampling done by solid extraction tubes (Supelco, 3-mL LC-SAX) as described by Bednar et al. (2004).

RW = Raw Water sample; Inlet(b) = samples from the bottom of the inlet chamber; GT1-2 = samples from a location in between gravel tank 1 and gravel tank 2.

successive sections at the beginning, middle and end of that chamber. The sudden jump of total iron concentration in the GT1-1-100 cm is probably due to presence of some solid sludge particle in the sample. This unfiltered sample was collected from the sludge-clogging zone at the vicinity of water entrance holes in this chamber. Besides oxidation of ferrous irons this figure also represents the removal of iron sludge in the gravel chamber.

Besides iron, arsenic is also effectively being oxidized due to aeration. Table 2 represents arsenic speciation data during August 2006, from 4 different units of GSF located and operated with different conditions. It was not possible to collect speciation samples from GSF 4. The sample from inlet (bottom) indicates the samples from the bottom part of inlet, prior to entering the gravel sections. In each sampled cases, it is clearly visible that the trivalent arsenic has been effectively reduced, which means efficient oxidation. In case of GSF1 data shows later rise of trivalent arsenic, which should be checked again because, that unit has been reset in the previous day. Steady state condition probably has not been reached up to those later sections by the time sampling has been done.

#### Effective arsenic removal

Besides showing oxidation effect data from Table 2 also shows the arsenic removal efficiencies of those units. The treated water quality for GSF 1 and GSF4 seems high but considering the amount of arsenic

Table 3 Iron and arsenic contained in the sludge extract

Sample designations	Fe (mg/L)	As (mg/L)
Inlet Sludge 06-Jul-2005	39.195	15.46
Sludge Tank 20-Mar-2006 --1	33.652	0.93
Sludge Tank 20-Mar-2006 --2	34.390	0.91
Sludge Tank 20-Mar-2006 --3	34.331	0.92
Inlet Sludge 04-Jul-2006 --1	35.761	37.35
Inlet Sludge 04-Jul-2006 --2	35.750	47.53
Inlet Sludge 04-Jul-2006 --3	34.331	55.94

removed, compared to GSF2 and GSF3, achievement is quite good. Resulted water in GSF2 and GSF3 are very good due to lower concentrations of arsenic in the raw water. Between GSF1 and GSF4, the later have better performance due to higher iron concentrations. Besides during the sampling GSF1 was not in stable condition after the maintenance done in the previous day.

#### Co-precipitation evidence

Arsenic content in the iron sludge has been checked to ensure the removal of arsenic with iron. Table 3 represents data of analysis of sludge collected from the inlet tank and from the underground reservoir tank. It should be clarified that, though the inlet sludge was collected from inlet, but actually those comprises of sludge which was previously stored in the gravel tank 1. To collect the sludge, first the wash out valve at the bottom of the inlet tank has been opened. Sludge and water accumulated in the inlet will flow out faster than that resulted from the backflow through the gravel tank 1. When almost all the water from inlet flows out, the sludge with some water get a chance to flow into the inlet. At that stage the wash out valve of inlet is closed to collect the accumulated sludge. Therefore, this sludge in fact is the sludge from gravel tank1.

Data indicates the sludge have quite high concentration arsenic along with iron. Other parameters from the extract are yet to be tested. The accumulated arsenic may have been re-mobilized under reducing conditions in sludge reservoir resulting lower concentration in that sludge sample.

#### RESEARCH DIRECTIONS

The iron in the groundwater has been the key element to remove arsenic by this system. Mamtaz et. al. (2001) has examined the potential of removing arsenite (trivalent arsenic compound) from water by co-precipitation with naturally occurring iron. They have used FeCl<sub>3</sub> for supplying the iron to the nano pure water spiked with arsenic salt. They showed that, at a pH of 7.5, greater Fe/As ratio removes arsenite better. They also have showed that the variation of mixing energy does not affect the adsorption.

Meng and Korfiatis (2001) as well as Meng et. al. (2001 and 2002) have checked the effect of competing ions on arsenic removal by iron co-precipitation. They showed that an iron to arsenic ratio (Fe/As) of 12 was sufficient to remove almost all the arsenic (400 ppb) from simulated Bangladesh groundwater without any competing ions. But addition of competing ions representing some Bangladesh groundwater (1.9 mg-P/L and 18 mg-Si/L) has increased the required Fe/As ratio to 40. They have estimated that these competing ions have occupied about 37% of the adsorptive sites in iron.

Therefore, the available natural iron in the groundwater may not always be sufficient to remove arsenic to achieve the desired level of purification. In those cases additional iron should be supplied by some means to aid arsenic removal. Extra iron can be supplied as chemical dose (Ali et. al. 2001) of an iron compound but that option might not be very cost effective. Supplying extra iron from metallic iron would be the most cost effective way to provide such extra iron for arsenic removal.

Alternatively corroding iron from a metallic iron source might serve as a potential source of iron in such cases. Sarin et. al. (2004) have examined the iron release in drinking water from corroding supply pipes as a result of the presence of dissolved oxygen in that water. Zhang and Huang (2006) have found that in the corroding iron lepidocrocite ( $\gamma$ -FeOOH) was the preferential product in the presence of DO. When the oxide coating thickened, then inner layer transformed to magnetite ( $\text{Fe}_3\text{O}_4$ ), which retained as the only stable corrosion product once DO was depleted.

Besides these findings a number of researchers have studied arsenic removal by metallic iron (Bang et. al., 2005<sup>1</sup>; Bang et. al., 2005<sup>2</sup>; Leupin et. al., 2005; Lien and Wilkin, 2005).

Karschunke and Jekel (2002) have performed lab scale arsenic removal from drinking water by enhanced corrosion of iron. They showed that natural corrosion of fine iron wool was suitable to remove effectively high but realistic arsenic concentrations of 500 ppb from drinking water. However, the corrosion rate decreased with time and they have used two different methods to enhance corrosion. Irrespective of the mode of corrosion, a post-treatment step was used to remove the arsenic loaded rust particles from the effluent.

Leupin and Hug (2005) have used the zero valent iron or the metal iron dusts to remove arsenic by successive filtration of arsenic contaminated water through a sand filter containing that iron dust. They showed that in the first filtration step removal of arsenic was not so high, rather the present phosphate of the water has been almost completely removed. The successive steps yielded better removal, as there were no competing ions (i.e. phosphates).

Therefore, to ensure sufficient iron in the arsenic removal unit as well as to make the unit efficient to suit all kind of groundwater, in the next phase of testing we will check this possibility of using zero valent iron in the GSF.

## CONCLUSIONS

Laboratory based research reports and our actual fieldwork data have already proved that arsenic removal by natural iron of groundwater, available in Bangladesh, is a very effective and sustainable way to remove arsenic from water. But to develop a system, which can handle the fluctuations of arsenic and iron concentrations in the water, it should be ensured that sufficient iron is present in the water for arsenic removal.

## ACKNOWLEDGEMENT

Authors are deeply grateful to the support of Asia Arsenic Network (AAN) during the field works. Government of Japan provided support for this research.

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# Removal of arsenic from contaminated groundwater by Iron Oxide

K. OHE, T. OSHIMA, Y. BABA

Department of Applied Chemistry, University of Miyazaki, Japan

M. SHIMIZU, Y. MIYAKE

Department of Chemical Engineering, Kansai University, Japan

T. HORIKAWA

Department of Chemical Science and Technology, the University of Tokushima, Japan

H. YOKOTA

Department of Civil Engineering, University of Miyazaki, Japan

**ABSTRACT** The removal of arsenic from aqueous solution was carried out using various iron oxides such as  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub>(FO), magnetite(MAG), silica-magnetite(SiMAG) and magnetite containing magnesium(MgMAG). As(III) and As(V) were removed at the region of pH4-7. Arsenic was effectively removed from groundwater contaminated using FO, MAG, MgMAG and SiMAG. MAG has the highest ability against the removal of arsenic.

**KEYWORDS:** arsenic, magnetite, removal, groundwater, Bangladesh

## INTRODUCTION

In recent years, arsenic contamination in groundwater has become problem in Asian countries such as India, Bangladesh, China(Tanabe et al., 2001 and 2003, Kadono et al., 2002, Geen et al., 2002, Graziano, 2002, Hargey et al., 2006). Arsenic contamination of the groundwater in Bangladesh is confirmed at 61 in 64districts (British et al., 2001). Above 95% of drinking water depends on the groundwater in Bangladesh. To take the drinking water including arsenic causes skin cancer and damages of liver and lungs. The patient of chronic arsenic poisoning has been increasing. In the WHO guidelines, the standard value of arsenic is less than 10 mg/m<sup>3</sup> (The World Health Organization, 2004). Therefore, the developmdcent of removal for arsenic from drinking water and groundwater is very important and urgent.

In order to remove arsenic, there are various processes such as coagulation, ion exchange and adsorption (Manning et al., 1998, Ohe et al., 2005). We have focused on the adsorptive method as an effective removal method from aqueous solutions because it generates no secondary contaminated substances and it is possible to remove toxic metal ion from an aqueous diluted solution. Iron compounds such as magnetite(Ohe et al., 2005, Wakui et al. 2005), hydrous iron oxides(Manning et al., 1998) was effective the removal of arsenic. In this work, the adsorption behavior of arsenic and removal of arsenic from groundwater contaminated in Bangladesh was investigated by iron compounds.

## EXPERIMENTAL

### Material

Adsorbents were prepared by materials included chemicals of reagent grade. Magnetite(MAG) and magnetite containing magnesium(MgMAG) was prepared by precipitation method(Ohe et al.(2005)). In case of, each aqueous solution of FeCl<sub>2</sub> and FeCl<sub>3</sub> was mixed at a molar ratio of 1:2, into which an aqueous sodium hydroxide solution was added to adjust to pH11. The product

resulted was dried at 50 °C. In case of MgMAG, nitrate salt was used. An aqueous ammonium solution was added to aqueous metal ion solution. Silica containing magnetite(SiMAG) was prepared by the previous paper(Horikawa et al., 2005). Silica beads prepared by sol-gel method was added to ferrous ion solution. The suspension was stirred at 25 °C for one night. After the resulted product was dried at 110 °C, it calcined at 500 °C for 5 h under CO<sub>2</sub>.  $\alpha$ -Ferric oxide(FO) and magnesium oxide(MgO) used to compare is commercial material.

### Characterization

The resulted products were identified by Powder X-ray diffraction(XRD) measurement made on a Rigaku RINT2000/PC with Cu K $\alpha$  radiation. The pHs of their zero point of charge(=pH<sub>zpc</sub>) were determined by an acid-base titration method in 0.1mol/dm<sup>3</sup> aqueous sodium chloride solution. Specific surface area(=SSA) was calculated by Brunauer-Emmett-Teller(BET) method using the nitrogen adsorption measurement. The results are summarized in Table 1.

Table 1 pH<sub>zpc</sub> and specific surface area of adsorbents.

adsorbents	pH <sub>zpc</sub>	BET SSA <sup>2)</sup> (m <sup>2</sup> /g)
MAG <sup>1)</sup>	6.0	46.3
SiMAG	4.6	540
MgMAG	9.0	
FO		
MgO	10.6	

<sup>1)</sup> K. Ohe et al., *J.Chem.Eng.Jpn.*, 38, 671-676(2005)

<sup>2)</sup> Specific surface area calculated from BET method

### Adsorption experiment

The effective of pH was investigated using adsorbents FO, MAG, SiMAG, and MgMAG. 10mg of adsorbent

was added to 0.01 dm<sup>3</sup> arsenic solution and the sample was shaken for 24h at 30°C. The pH was adjusted using hydrochloric acid and aqueous sodium hydroxide solution. The concentrations of As(III) and As(V) before and after adsorption were measured by atomic absorption spectrophotometer on a Shimadzu AA6800.

Here, the adsorption percentage of arsenic was calculated according to the following equations, respectively:

$$A = (C_{int} - C_{eq}) / C_{int} \times 100 \quad [\%] \quad (1)$$

where  $C_{int}$  is the initial concentration of arsenic [mmol/dm<sup>3</sup>],  $C_{eq}$  is the equilibrium concentration of arsenic [mmol/dm<sup>3</sup>] and  $A$  is adsorption percentage [%]

The arsenic contaminated groundwater was collected from tube well in Marua village in Bangladesh. The groundwater was used without pretreatment. 0.05-0.3 g of the adsorbents was added into 0.04dm<sup>3</sup> of the groundwater in plastic tube. At different periods of reaction time, the filtrate was separated from suspension. The filtrate was analyzed for concentration of total arsenic and iron.

## RESULTS AND DISCUSSION

### Effect of pH on Arsenic adsorption

The effect of pH on the adsorption As(III) was shown in Fig.1. The adsorption of As(III) was affected by the solution pH. The maximum As(III) adsorption on MAG, FO, MgMAG was observed in the region of pH 4-7, while in the region of alkaline pH, the adsorption percentage of As(III) decreased. Optimum pH of SiMAG on the adsorption of As(III) was shown around pH4, whereas the adsorption decreased at above pH4. The adsorption of

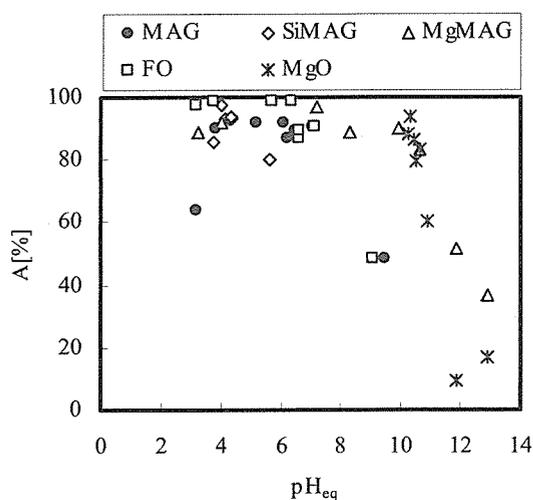


Fig.1 Effect of pH on the adsorption percentage.(Data of MAG was cited from reference(K.Ohe, 2006).

As(III) on SiMAG decreased at lower pH than that of MAG. This result is considerable to the  $pH_{zpc}$  (pH4.6) of SiMAG. In the region of alkaline pH, the adsorption behavior of As(III) on MgMAG was agree with MgO.

Main chemical species of As(III) at pH 4-7 was  $H_3AsO_3$ . Therefore the adsorption of As(III) is considered to occur between  $H_3AsO_3$  and the surface hydroxide group of adsorbent.

Fig.2 shows the influence of pH on the adsorption percentage of As(V). The adsorption behavior of As(V) was different from that of As(III) also affected by the solution pH. The adsorption percentage of As(V) was around 100% at below pH 7, except for FO. The adsorption behavior of As(V) using FO was similar to goethite(Manning et al., 1998). However in the region of  $pH > 7$ , the adsorption percentage of As(V) greatly decreased. Chemical species of As(V) at  $pH < 7$  are  $H_2AsO_4^-$  and  $HAsO_4^{2-}$ . On the other hand, the surface of hydroxide group has plus charge because  $pH < 7$  is lower than the  $pH_{zpc}$  of adsorbents. These results suggest that As(V) is mainly adsorbed by electrostatic force between As(V) anions and the surface hydroxide of adsorbent

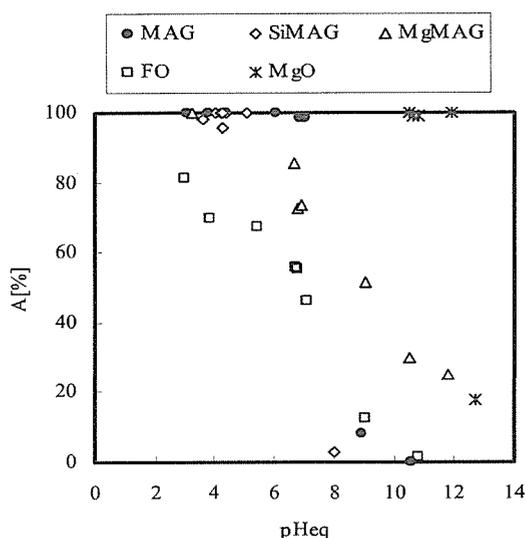


Fig.2 Effect of pH on the adsorption percentage of As(V).(Data of MAG was cited from reference(K.Ohe, 2006).

### Removal of arsenic from groundwater contaminated using iron compounds

Chemical analysis of groundwater in Marua village in Bangladesh was Table 2. The total arsenic concentration in groundwater contaminated was around 180mg/m<sup>3</sup> (As(III)=ca.90 mg/m<sup>3</sup>). The concentration was around 17-18 times higher than standard value of WHO guidelines.

Table 2 Chemical analysis of groundwater in Marua village in Bangladesh.

pH	Total Fe mg/dm <sup>3</sup>	Total As mg/dm <sup>3</sup>
7.1	4.2	0.17-0.18

Figs.3 and 4 show the removal kinetics of arsenic and the variation of pH in solution on adsorbents 100mg and 300mg, respectively. Fig.5 shows the removal kinetics of

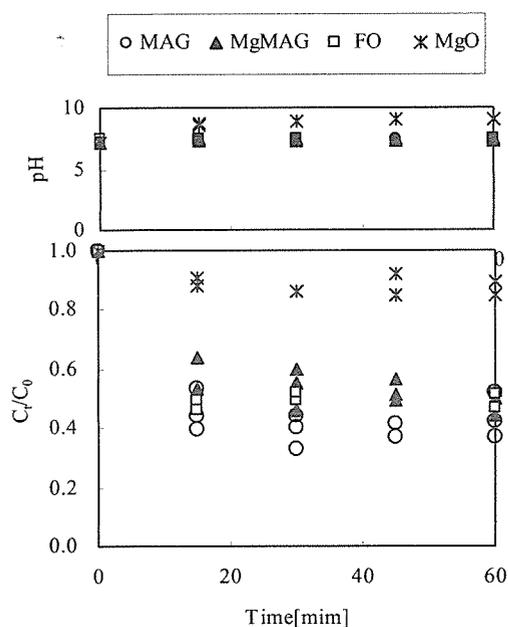


Fig.3 The effect of reaction time on the adsorption of arsenic from groundwater contaminated. Adsorbents=100mg

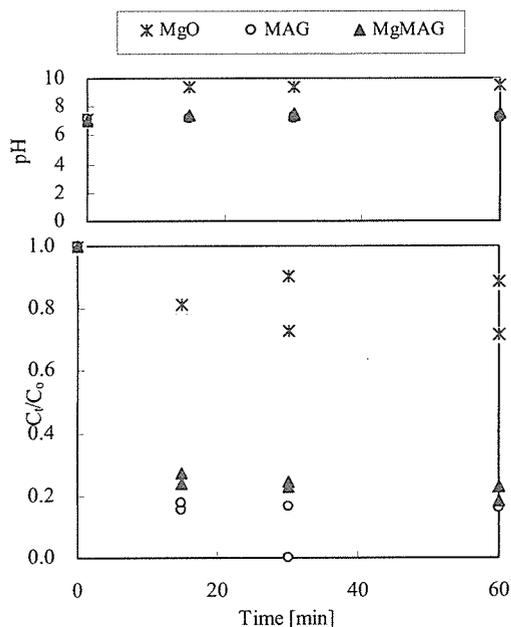


Fig.4 The effect of reaction time on the adsorption of arsenic from groundwater contaminated. Adsorbents=300mg

arsenic on SiMAG. The times required to reach the equilibrium of MAG, MgMAG and FO were fast. The residue arsenic concentration kept constant at pH7. After 60min iron in filtrate wasn't detected by atomic adsorption spectroscopy. Removal of arsenic on MgO was very low. In spite of a small amount, arsenic was removed from the groundwater using SiMAG. Therefore iron compounds were effective adsorbents. Especially MAG is stable and an excellent adsorbent for arsenic.

On the other hand, the concentration of arsenic in

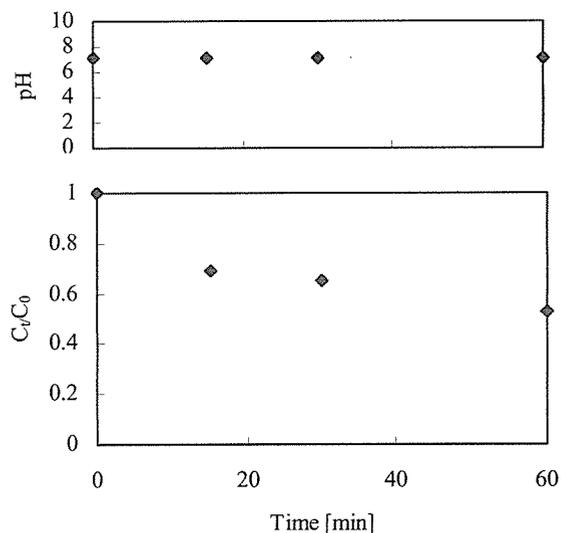


Fig.5 The effect of reaction time on the adsorption of arsenic from groundwater contaminated. SiMAG=50mg

groundwater decreased around 20-30% without adsorbent. Concentration of iron also decreased. This result suggests that coagulation/ precipitation was produced by iron ion in the groundwater because the color of the groundwater changed from clear to orange. Thus from these results, it is considered that coagulation/ precipitation and adsorption occur at the same.

From the results of Fig.1 and Fig.2, MgMAG were shown the high affinity for As(III) and As(V). But the adsorption of arsenic on MgMAG was slightly lower than that on the other adsorbents. These results are considered to be caused by the influence of co-existing ion against the adsorption of arsenic on MgMAG and the elution of Mg from MgMAG.

These results was shown that iron oxides such as magnetite are highly effective adsorbents in order to remove of arsenic from groundwater contaminated. Two important points against the removal of arsenic are shown that the pH<sub>zpc</sub> is closed to pH of groundwater and the adsorbent has high specific surface area.

#### Acknowledgements

The authors thank Mr. Miyatake for assistance of fields experiment in Bangladesh.

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