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ABSTRACT

This paper shows the situation and mechanism of arsenic contamination of groundwater at the worst contaminated areas in UP (Uttar Pradesh) state, India, which is obtained from the integrated arsenic mitigation project by University of Miyazaki under the Japan International Cooperation Agency (JICA) Partnership Program (JPP). The project has been executed from 2008 until now. The integrated mitigation, such as the raising awareness of villager, installing of alternative water supply units and healthcare of arsenocosis patients, have been executed at the 2 villages. The symptom of the arsenocosis patients was not so severe, which will be, therefore, improved by drinking arsenic-safe water supplied through arsenic removal units, installed by this project.
We have obtained following results for the situation and mechanism of arsenic contamination of groundwater, objected in connection with the installation of arsenic removal units:(1) Groundwater is almost contaminated with arsenic in deep tubewell (depth: about 30m), but scarcely in shallow tubewell (depth: about 10m). (2) Arsenic contaminated groundwater is under the reduced condition with the oxidized condition for no-arsenic contaminated groundwater. (3) Arsenic concentration shows almost linear correlation with concentrations of Fe^{2+} and NH_4^+-N. (4) Ground is composed of sand with high arsenic content at around 25m depth. (5) Arsenic exists mainly in the phase of reducible fraction or weak acid soluble fraction but no oxidizable fraction in the ground.

INTRODUCTION

Arsenic contamination of groundwater, in Asia, is seen in the basins of the great rivers, originating in the Himalayan Mountains and the Tibetan Plateau, such as the Ganges River, the Indus River, the Mekong River, the Haw River, and the Yellow River [1], where people depend on the drinking water for groundwater. A thermally altered metamorphic zone in the Higher Himalaya, containing various types of minerals, is considered as the source of arsenic [2].
Arsenic pollution of groundwater in Ganges River basin, West Bengal, India, and Bangladesh is known for long. The detection of the arsenic pollution is in 1982 and 1993, respectively. The investigations and countermeasures have been performed [3~6].
On the other hand, in Mekong River basin, Vietnam, Cambodia and Laos, arsenic pollution was first confirmed around 2000 and countermeasures just began under help such as UNICEF and GIST (Gwangju Institute of Science and Technology) [7,8]
The authors have elucidated the mechanism for arsenic contamination and developed the safe water devices in Bangladesh since 1997 together with the NGO “Asia Arsenic Network” (AAN) [9]. AAN has implemented the Arsenic Mitigation Project with Japan International Cooperation Agency (JICA) in Bangladesh from 1999 until now. The University of Miyazaki has conducted activities for arsenic mitigation in Uttar Pradesh State, India, under a JICA

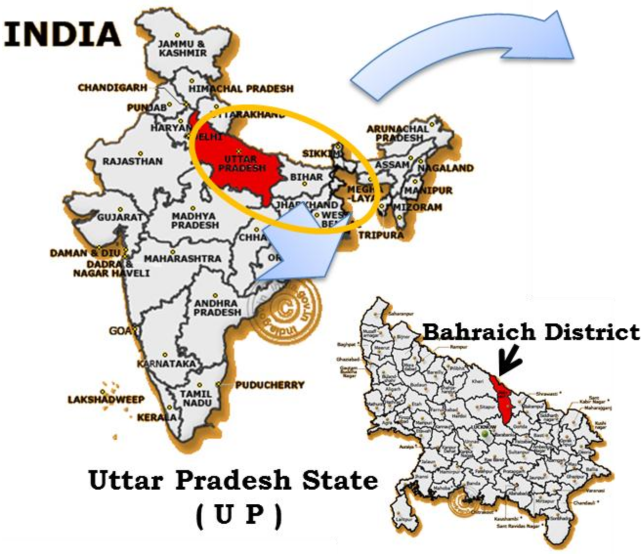

Fig. 1 . Location of Utter Pradesh State

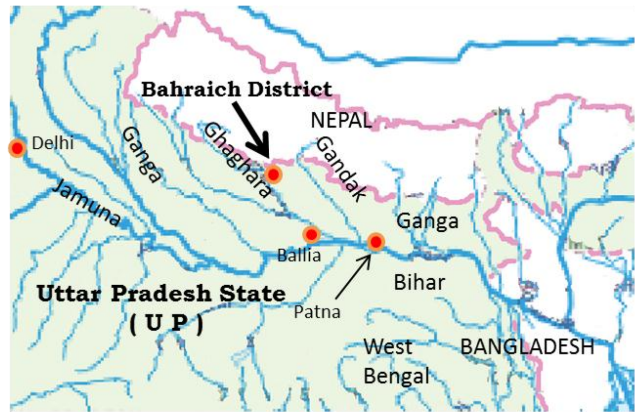

Fig. 2. Location of Project Area, Bahraich District

technical cooperation project from 2008 until now, in collaboration with AAN.

Uttar Pradesh State (abbreviated as UP State hereafter) is located at north of India bordering on Nepal, Geographical area of which is about 4,700km^2, where two big rivers are running from the northwest to the southeast. The former is the Ghaghara River flowing down from the arsenic affected Terrai plane, and the latter is the Ganges River as shown in Fig.1 & 2. The arsenic contamination in UP State was first recognized in 2003 at Ballia District, where both of the Ghaghara and the Ganges are joining.

Arsenic-contaminated tubewell water is detected in the 20 Districts out of 70 Districts in UP State by UP government under the assistance of UNICEF. The government survey was, however, performed only for the government tubewells (GTWs), and private tubewells (PTWs), numerous compared with GTW, were not checked at all. In regard to arsenocosis patients, the number of patient is unknown yet, because the medical examination has not been executed until now.

Our project area is in Bahraich District (See Fig.2), severely arsenic affected one in the above-mentioned 20 Districts. The project is an integrated arsenic mitigation with 3 activities: (1) Raising awareness of villagers for poison of arsenic through a Signboard Installation, Street play program (play drama in the place in which people gather), Flip chart program (tell a story illustrated with picture cards) Small group meeting program etc., (2) Identification of arsenocosis patients after training local medical doctors for diagnosis of chronic arsenic poisoning, (3) Installation of arsenic removal unit after checking all tubewells used in the villages.

Though the groundwater of the Ganges medium basin is contaminated with arsenic, few reports [10~12] are obtained. We will, therefore, introduce the situation and mechanism of arsenic contamination of groundwater, obtained from the 1st phase and 2nd phase of JICA arsenic mitigation project (1st :2008~2010, 2nd 2011~2013) as a report.

This paper mainly shows the data obtained in the activity (3) mentioned above.

SITUATIONS OF ARSENIC CONTAMINATION IN UP STATE

Fig.3 shows the ratio of arsenic polluted GTW (As>50ppb) in the 20 Districts obtained from the above mentioned government survey. There are 3 severe contaminated Districts: Kehri, Ballia, and Bahraich.

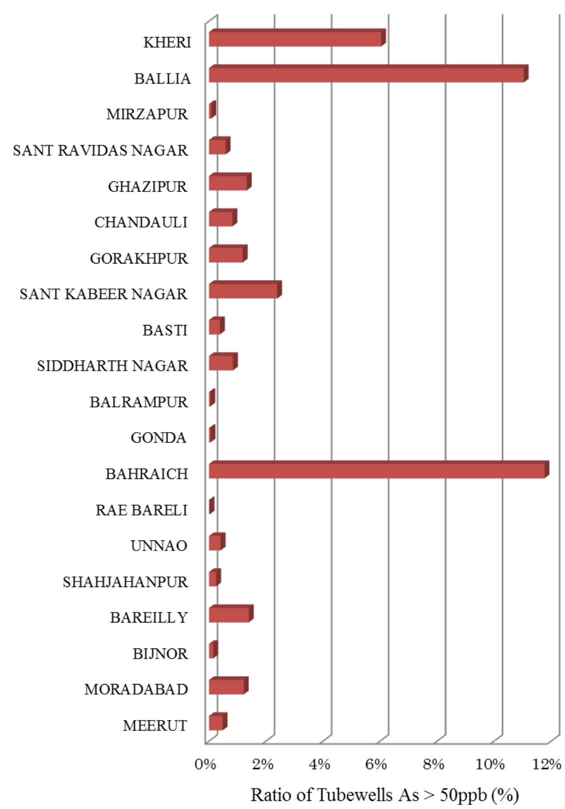

Fig. 3. Ratio of As>50ppb TWs in Districts of UP State

after that in Ballia, and 500 of deep wells were installed in Ballia and 250 wells in Kehri.

The ratio of arsenic contaminated GTWs is highest in Bahraich District, where we have been performed the JICA project. The project area belongs to Tejwapur Block in Bahraich District, which is composed of 14 Blocks. The arsenic contamination is seen in the 10 out of 14 Blocks. Fig.4 shows the ratio of GTW of As>50ppb in the 10 Blocks. It is clear that the arsenic contamination is highest in the Tejwapur Block.

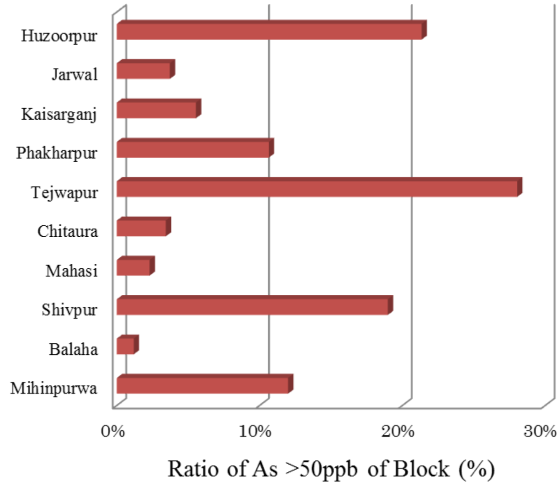

Fig. 4. Ratio of As affected TW in Blocks of Bahraich District

Tejwapur Block has 80 villages. In 9 out of 80 villages, GTW of As>50ppb is detected. Fig.5 shows the situations of arsenic contamination in the 9 villages. The ratio of As>100ppb is highest in Newada village, followed by Chetra village. The JICA project area is in the both villages with 27 habitations in Newada village and 7 habitations in Chetra village.

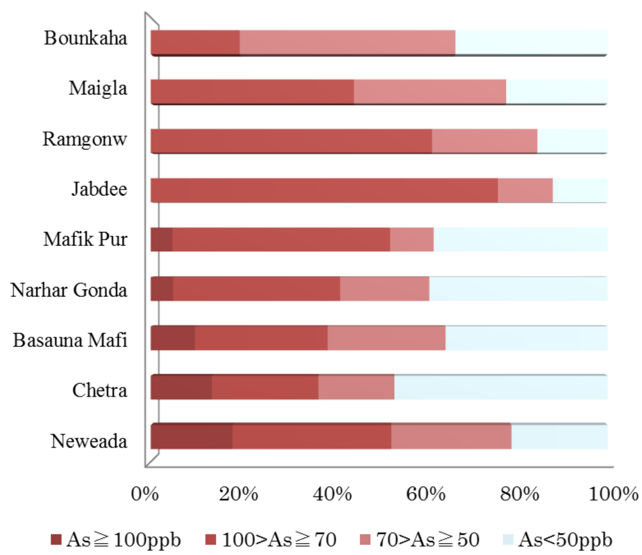

Fig. 5. Ratio of TW of As >50ppb in the 9 villages of Tejwapur Block

ARSENIC CONTAMINATION OF GROUNDWATER IN THE PROJECT AREA

Arsenic Contamination of Groundwater

We had measured the arsenic concentration of all tubewells in the project area, which is composed of 34 habitations of 2 villages. The number of tubewells in the 34 habitations is 118 of GTW and 720 of PTW. The arsenic concentration measured is shown in Fig.6 and Fig.7.

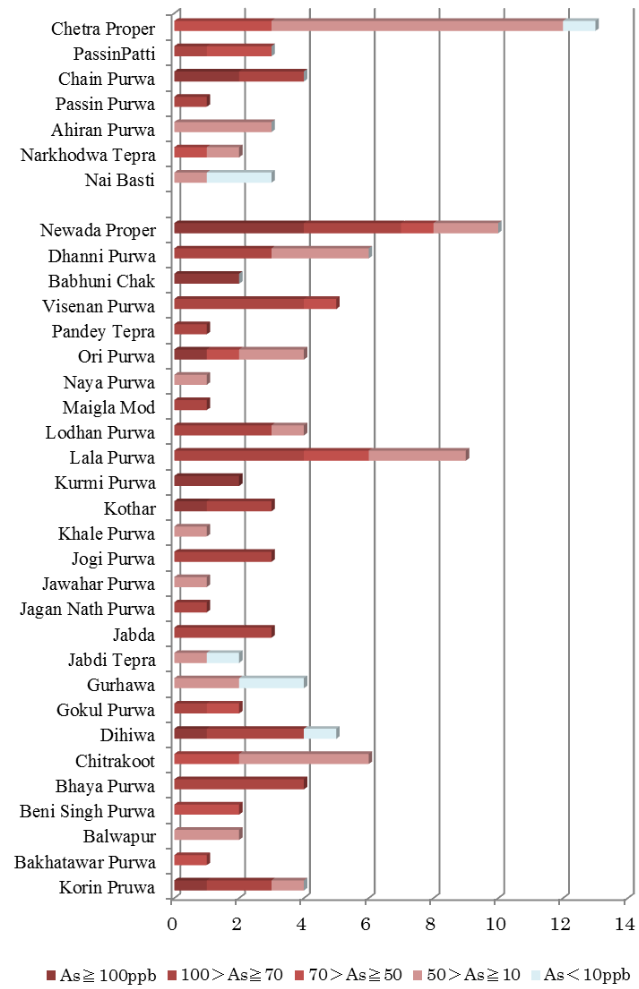

Fig. 6. As Contaminations of Government TWs in 34 Habitation (total Number :118)

GTWs (Depth: about 30m) are almost contaminated with arsenic, in which 62% of TW shows As≧50 ppb and 94% for As≧10 ppb on the average in the 34 habitations. The highest contamination is seen in Newada Proper and the lowest in Nai Basti.

On the other hand, the arsenic contamination in PTW (Depth: about 10m) is overall low. PTW of As≧50 ppb is 9% and 38% for As≧10 ppb on the average in the 34 habitations. The high arsenic contamination is, however, seen in Passin Patti. d Babhuni Chak and Lala Purwa, which should be remarkable.

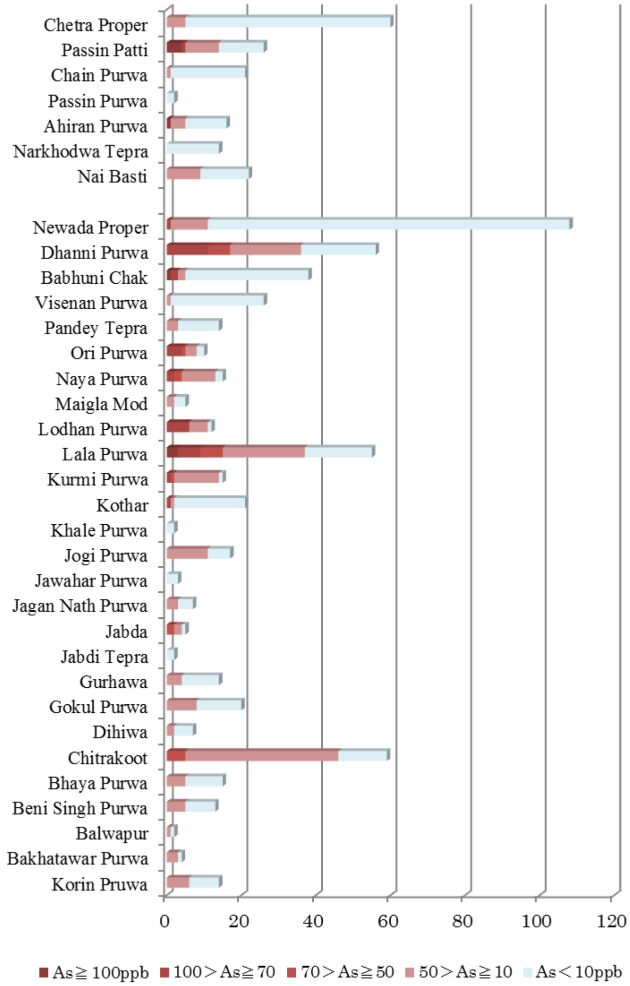

Fig. 7. As Contaminations of Private TWs in 34 Habitation (total Number :720)

Mechanism of Arsenic release

We had examined water quality for 11 GTWs an 12 PTWs. A part of the results is shown in Fig. 8(a) and (b). The symbols in the figures are explained in Table 1. From these figures, it is understood that most of arsenic-safe water in PTWs is in oxidized conditions because of positive values of ORP and little of Fe^{2+}. And, PTW is dirtier than GTW with much dissolved ions from high EC.

Fig.9 shows the relation of total arsenic concentration and total iron concentration in the GTW (□) & PTW(○). The concentration between As and Fe shows a liner relation with some scattered data in the both tubewells.

The arsenic valence in the arsenic contaminated GTWs & PTWs was all trivalent, As (Ⅲ), which shows the reduced condition in groundwater.

From these data, we consider that arsenic, which had been absorbed with iron in underground, was released into groundwater under the reduced conditions.

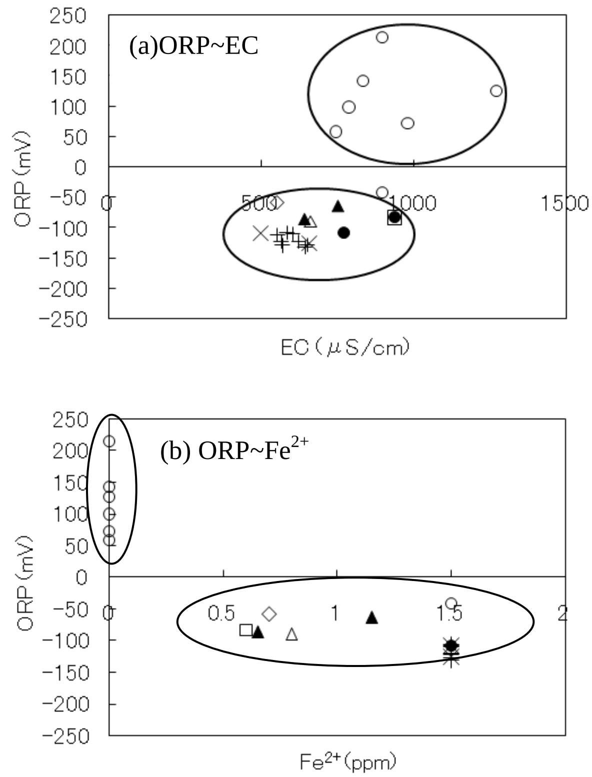

Fig. 8. Relation between PTW without As and GTW with As

Table 1. Arsenic Concentration in Fig. 8. (mg/L)

GTW	×	0.1≦As
	+	0.05≦As＜0.1
	◇	0.01≦As＜0.05
PTW	▲	0.1≦As
	●	0.05≦As＜0.1
	□	0.01≦As＜0.05
	△	0＜As＜0.01
	○	As＝0

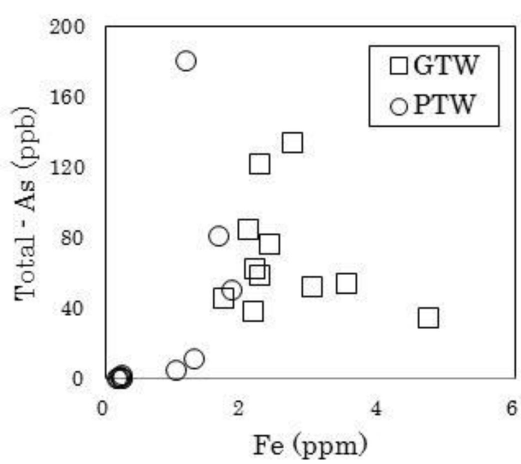

Fig. 9. Relation between concentration of As and Iron

Relation between arsenic and nitrogen

We found roughly a linear correlation between concentrations of arsenic and ammonia in the above data. So, we collected more samples to check the correlation.
Fig.10 shows the NH_4^+-N exits in both shallow tubewell (PTW) and deep tubewell (GTW). It may be considered the source of nitrogen is from cow dung on the garden or fertilizer in the cultivated field. The concentration of NH_4^+-N shows fairly a linear correlation with that of arsenic from Fig.11. It may be considered that influences of microorganism activities on arsenic release under the reduced conditions, which will be a research theme in future.

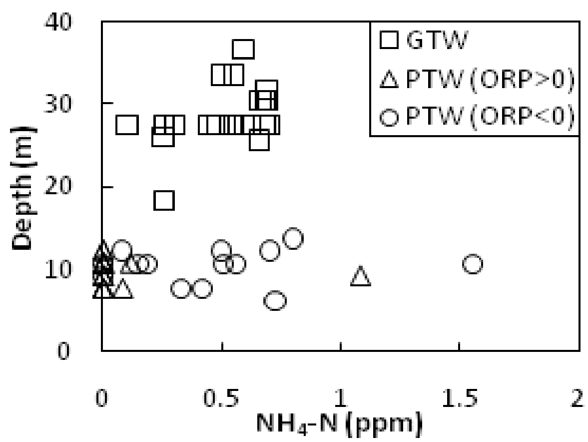

Fig. 10. NH_4-N detected both PTW and GTW

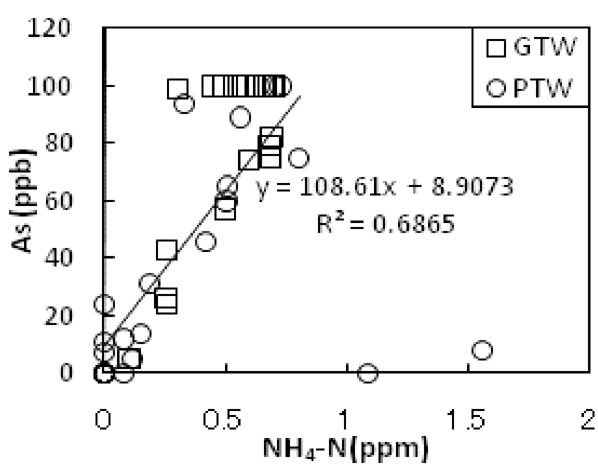

Fig. 11. Correlation of NH_4-N with As

ARSENIC CONTENT IN UNDERGROUNG

We had test borings, showing the geological profiles with the alternation of fine and medium sand until 80m depth without any silt and clay layers. The boring survey and soil analysis was carried out on 6 sites in habitations in order to grasp the soil characteristic and distribution of arsenic.

Specifically, the soil samples collected every 1 m by hand boring were checked about the condition and texture, and arsenic content and mineral composition were measured by EDX and XRD. And distribution of arsenic and some elements was estimated by the sequential extraction.
In the boring survey, specific classification of soil texture was shown that most topsoil (1 ~ 3 m) was a mixed layer of clay and sand, and 10 ~ 30 m of soil was fine sand that thin mixed layers of gravel and sand and a mixed layer of clay and sand sometimes appeared.

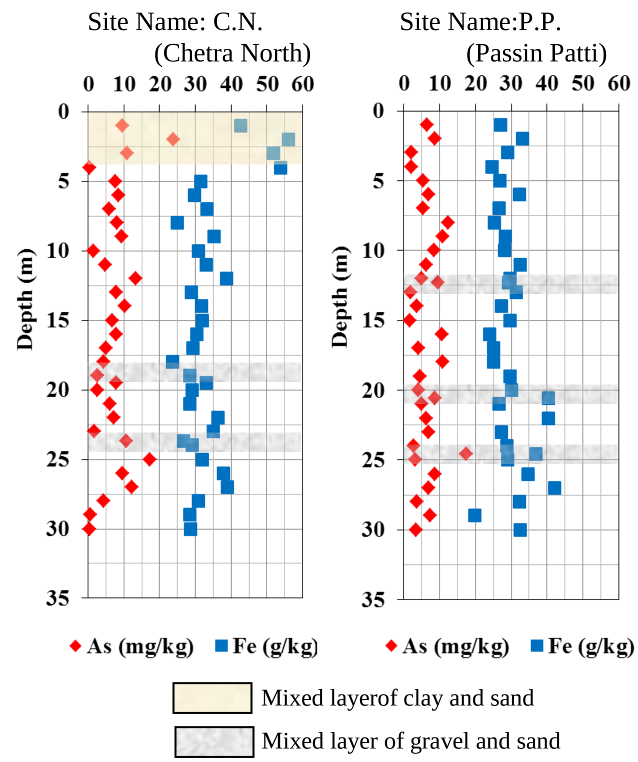

Fig. 12. Contents of Arsenic and Iron in underground

Fig.12 shows the distributions of arsenic and iron contents along to soil depth until 30m, in which the boring was performed near to the arsenic contaminated GTW (depth: about 30m).
From Fig.12 it is seen that arsenic concentration is harmony with that of iron and arsenic concentration was high in gravel layers at the depth of more than 20m that was the range of intake depth of GTW.
Fig.13 shown the mineral component of the boring samples.12 samples that collected from topsoil, sand layer near intake depth of PTW, and mixed layers of gravel and sand near intake depth of GTW, were analyzed in XRD. As results of each X-ray diffraction pattern, mineralogical composition in soil was mainly quartz and mica, including calcite, chlorite, albite and doromite. And, comparatively high peak of calcite was detected in mixed layers of gravel and sand. On the other hand, minerals of iron which has high affinity to arsenic wasn't detected .

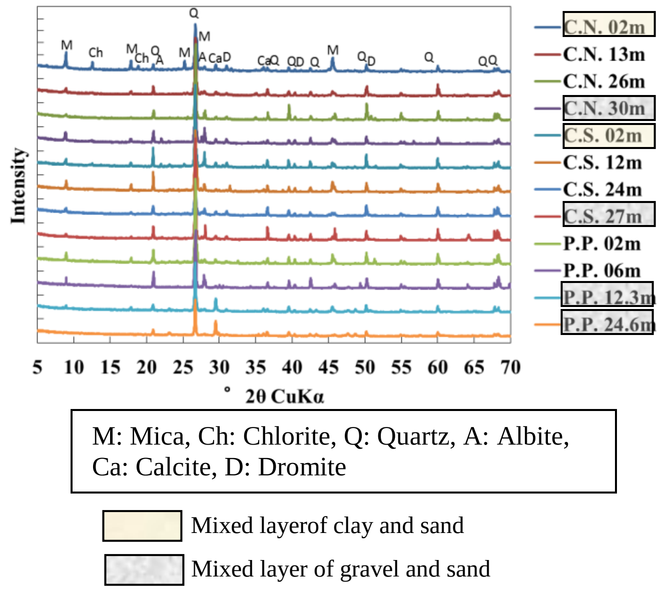

Fig. 13. XRD pattern of samples for sequential extraction

The modified BCR sequential extraction procedure [13] was applied to get the chemical combining form of arsenic in soil of the boring sample at the 25m depth. We used 4 steps of sequential extraction to estimate 1) Water soluble fraction, 2) Exchangeable & weak acid soluble fraction, 3) Reducible fraction, and 4) Oxidizable fraction.

Table 2. Procedure and condition of the sequential extraction method

Sequence	Fraction	Solvent / reaction time	Target
Step 1	Water soluble	Deionized water [H_2O], 6 hrs	Soluble salt (present in soil solution), easily exchangeable ion
Step 2	Acid-soluble	0.11 mol/L acetic acid [CH_2COOH], 16 hrs	Bound to carbonates, exchangeable ion
Step 3	Reduction soluble	0.5 mol/L Hydroxyl ammonium Chloride [ClH_4NO], 16 hrs	Bound to Fe - Mn oxides
Step 4	Oxidative soluble	8.8 mol/L Hydrogen peroxide (85°C) [H_2O_2], 1 hr 1.0 mol/L Acetic ammonium [CH_2COONH_4], 16 hrs	Specially bound to organic matter, sulfides

From Fig.14 it may be said that 1) Arsenic has the highest extractability in the step 3, meaning arsenic mainly exits in the oxidized form with iron, aluminum and manganese, 2) There is no arsenic with sulfide as pyrite because no arsenic extraction in the step 4, and 3) It may be estimated that arsenic exits with carbonate from step 2 (Extraction of arsenic with calcium in step 4), because of much calcareous soil (Kankar) in UP State [14]. Therefore, it was considered that arsenic in soil didn't bind to organic matter and sulfide and was included in mainly carbonates and iron minerals by adsorption, ion exchange and other mechanism. And it was guessed with the cause of arsenic concentration in PTW being lower than GTW that arsenic in topsoil was relatively distributed in a high ratio in STEP 3 including iron minerals.

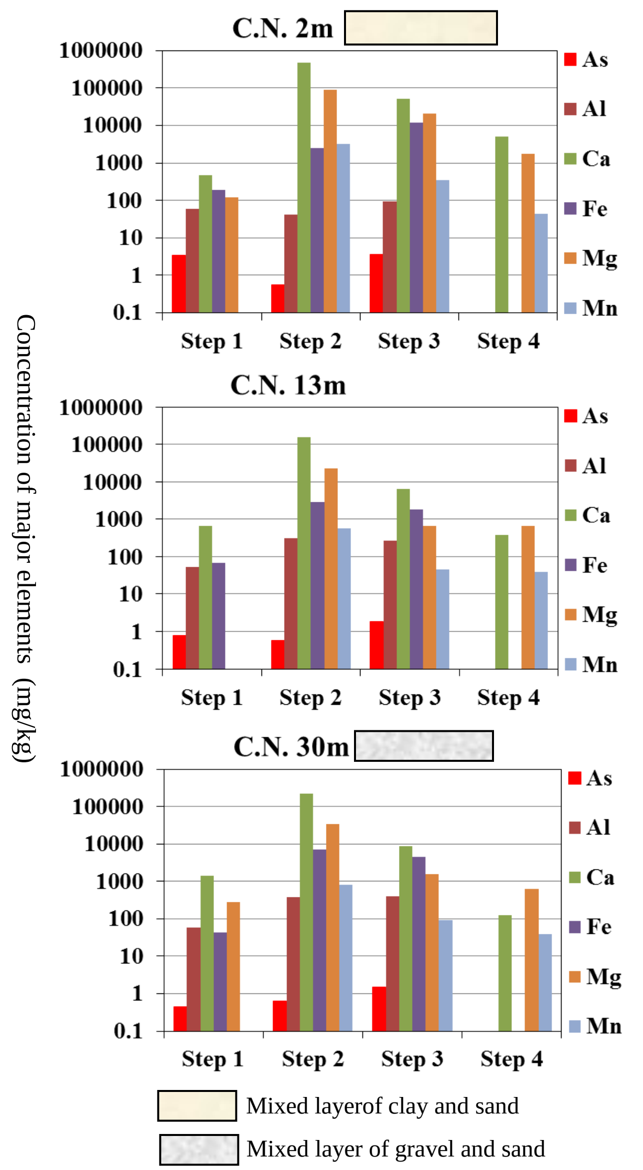

Fig. 14. Distribution of arsenic and major component in each fraction

ARSENIC RELEASE MECHANISM

From the results, it may be said that arsenic, mainly fixed in Fe- and/or Mn-oxides in the underground, was released to groundwater under the reduced condition through microorganism activities with the existence of nitrogen acting on the metabolism of microorganism. It is similar mechanism in Bangladesh [15], although there is different in soil profile, abundant clay in Bangladesh and no clay in our project area, UP State.

ALTERNATIVE WATER SUPPLY SYSTEM AND ARSENICOSIS PATIENTS

In the project we installed 10 units of alternative water supply: 3 filters for dugwells to treat the Fecal coliform bacteria and 7 arsenic removal plants, GSF, for government tubewells to remove arsenic.
GSF (Gravel Sand Filter) is a community-based arsenic removal unit, removing arsenic by co-precipitating of arsenic with iron in the gravel tank after aeration of groundwater. GSF has been developed by us [16], and more than 50 GSFs are now operated at the arsenic affected villages in Bangladesh.
1750 out of 7,000 villagers had drunk arsenic-affected tubewell water (As>50ppb) in the project area. We had the medical examinations for detecting the arsenocosis patients together with training local doctors, from which 64 patients were identified. As their symptoms of chronic arsenic poisoning were very mild, they will be improved through drinking the arsenic safe water.

Fig. 15. Photo of Gravel Sand Filter (GSF) in the village

CONCLUSION

In this paper, the following results are obtained.

a) The government tubewells (depth: 30m) are almost contaminated with arsenic and the private tubewsells (depth: 10m) are overall not affected with arsenic.
b) The arsenic contaminated tubewells are under reduced condition and the non-arsenic tubewells are under the oxidized condition, meaning that arsenic is leached out into groundwater under reduced condition.
c) Arsenic concentration has roughly linear correlation with those of iron and ammonia in the groundwater.
d) Ground is composed of sand until 80m with high arsenic content at around 25m depth, where similar release mechanism as Bangladesh might be considered, although there is different in soil profile: abundant clay layer in Bangladesh and no clay in the project area, UP State.
f) 1750 out of 7,000 villagers had drunk the arsenic contaminated water, and 64 of arsenocosis patients have been identified.

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