

Validity of the Latest Research Findings on Causes of Groundwater Arsenic Contamination in Bangladesh

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Abstract: *The groundwater arsenic contamination problem in Bangladesh is the worst in the world. Two hypotheses are prevailing as the cause (mobilization) of groundwater arsenic contamination in Bangladesh. One is called “pyrite oxidation” and the other is “oxy-hydroxide reduction.” According to the first hypothesis, groundwater arsenic contamination is human-made, which has a relationship with excessive groundwater withdrawal. On the other hand, according to the second one, the contamination is natural. The British Geological Survey (BGS) conducted an investigation to identify the cause of groundwater arsenic contamination in Bangladesh. Based on their findings, they rejected the pyrite oxidation hypothesis and accepted the oxy-hydroxide reduction hypothesis. However, several experts are opposing their research findings. Therefore, a case study is undertaken to examine the validity of the BGS findings. Although we obtained results which are similar to the BGS, the errors in the estimates of percent of wells contaminated, the key parameter of these analyses show that the present as well as BGS results are not dependable to make any conclusion about the cause of contamination. Moreover, the validity test shows that space-dependent relationships among different hydro-geological parameters for the heterogeneous aquifer system are not valid to represent time-dependent phenomena. Therefore, the conclusion made by the BGS is not valid. Still, there is a possibility that the pyrite oxidation hypothesis could be accepted. For a final conclusion we must observe these relationships in a time domain by generating reliable time-dependent field data.*

Keywords: Bangladesh, groundwater, arsenic contamination, causes, pyrite oxidation, oxy-hydroxide reduction.

Introduction

Groundwater arsenic contamination in Bangladesh is now a well-known fact. In terms of extent and severity of the problem, Bangladesh is the most vulnerable country in the world. Out of the 64 districts, 61 have arsenic concentration above the maximum permissible limit (0.05 mg/l), and, therefore, at least 21 million people are now exposed to the arsenic poisoning. It is believed that the source of arsenic in Bangladesh groundwater is geological and the cause of contamination is over-exploitation of groundwater, which facilitates oxidization of pyrites or arsenopyrites and ultimately releases arsenic into groundwater. The hypothesis called “pyrite oxidation” came from West Bengal, a state of India, near to the west border of Bangladesh that has similar sub-surface geology and environmental factors, and is also a serious problem of groundwater contamination with arsenic. In West Bengal, it is believed by several authors (Acharyya, 1997; Sinha Roy, 1997) that the source of arsenic in groundwater is geological and the cause of contamination is “pyrite oxidation.”

To save the lives of million of people of Bangladesh, finding the source and cause(s) of contamination is an

emergent need. Accordingly, the Government of Bangladesh through the Department of Public Health Engineering (DPHE) began a project entitled “Groundwater Studies for Arsenic Contamination of Bangladesh” with the British Geological Survey (BGS) in 1997 for rapid investigation of the problem to discover the source and cause of contamination based on a field survey as well as existing field data. Recently, the BGS has published its findings and they concluded that the source of arsenic in Bangladesh groundwater is geological, but denied the “pyrite oxidation” hypothesis. Rather, the BGS accepted the “oxy-hydroxide reduction” hypothesis. The “oxy-hydroxide reduction” hypothesis to explain the mobilization of arsenic from the source to the groundwater for Bangladesh basin was first proposed by Nickson et al. (1998). The BGS reports mentioned that groundwater arsenic contamination in Bangladesh is a natural process that has no relationship with the excessive groundwater withdrawal.

Several experts are opposing the possibility of arsenic mobilization by the “oxy-hydroxide reduction” process. According to the reduction hypothesis, organic carbon takes oxygen from iron hydroxide, and arsenic is released

into groundwater. Adel (2000) offered the following question to the authors (Nickson et al., 1999): "Why was arsenic contamination detected in the 1990s if organic carbon has been taking oxygen forever?" Adel also mentioned that, to support the reduction hypothesis, Nickson et al. (1999) showed six percent organic carbon content in aquifer sediments, which goes beyond the boundary of scientific ethics. Khalequzzaman. (1999) raised a question on pH-Eh values which were used by the BGS researchers to show the relationship between arsenic contamination and reducing conditions in support of the reduction hypothesis because there was no information about pH-Eh while samples were collected. Bridge and Husain (1999) opposed the reduction hypothesis as there was no reported poisoning prior to the 1980s although a significant number of people were using groundwater as drinking source since the 1960s. Bridge and Husain (1999) mentioned that no prior reported poisoning supports the hypothesis that the poisoning of the groundwater is recent. Increased irrigation due to the reduction of surface water flow, caused by a multitude of dams built in India on streams crossing the border into Bangladesh, resulted in repeated lowering and rising of the groundwater table. Also, the drilling of thousands of tubewells to supply domestic and irrigation water resulted in repeated wetting and drying of arsenic bearing sulfides in organic rich sediments.

Thus, there are confusions about the reduction hypothesis, while the BGS reports show some evidence to reject the pyrite oxidation hypothesis. Therefore, the validity of their findings needs to be checked very carefully and these two opposing theories must be reconciled. Keeping this in mind, a case study has been undertaken to determine the validity of the research findings of the BGS.

Mobilization of Arsenic to Groundwater

Pyrite Oxidation

Arsenic is assumed to be present in certain sulphide minerals (pyrites) that are deposited within the aquifer sediments. Due to the lowering of water table below deposits, arseno-pyrite oxidized in the vadose zone releases arsenic as arsenic adsorbed on iron hydroxide. During the subsequent recharge period, iron hydroxide releases arsenic into the groundwater. According to this hypothesis, the origin of arsenic-rich groundwater is human-made.

The theory suggests that, for a particular location (more specifically, for a particular well), factors associated with pyrite oxidation in time domain are: (1) presence of pyrite in the sediments; (2) positive relationship between the cumulative arsenic concentration and the cumulative decline of water table; and/or (3) positive relationship between the yearly change of arsenic concentration and the yearly decline of groundwater table. In the above relationships, parameters other than water level, which have a direct positive relationship with water level, such as water abstraction, or direct negative relationship with water level,

such as actual recharge, may be used. Equivalent well intensity that may have indirect influence on water level may also be used. Due to the data limitation, the age of groundwater development may be used as an indication of the cumulative decline of water level or cumulative water abstraction.

If the above relationships are sought for the spatial domain and other than water level parameters are considered, care must be taken about the internal relationships between those parameters and water level as, for example, if water abstraction shows a false negative significant relationship with water level, then it would likely have a false relationship between water abstraction and arsenic contamination.

Oxy-hydroxide Reduction

Arsenic is assumed to be present in alluvial sediments with high concentrations in sand grains as a coating of iron hydroxide. The sediments were deposited in valleys eroded in the delta when the stream base level was lowered with the lowered sea stand during the last glacial advance. The organic matter deposited with the sediments reduces the arsenic-bearing iron hydroxide and releases arsenic into groundwater. According to this hypothesis, the origin of arsenic rich groundwater is a natural process, which implies that the arsenic in groundwater has been present for thousands of years without being flushed from the delta.

The theory suggests that, for a particular location, factors associated with oxy-hydroxide reduction are: (1) the presence of arsenic containing coatings of iron hydroxide and a high level of organic matter in the sediments; (2) spatial positive correlation between the arsenic concentration and the strongly reducing conditions; and/or (3) spatial positive correlation between the arsenic concentration and the depth of contamination.

Research Findings of the BGS

In Rejection of Pyrite Oxidation Hypothesis

The BGS investigated five correlations: (1) the correlation between gross water abstraction and percent of wells contaminated; (2) the correlation between maximum water level below ground surface and percent of wells contaminated; (3) the correlation between aquifer transmissivity and percent of wells contaminated; (4) the correlation between sulphate concentration and arsenic concentration; and (5) the correlation between potential recharge and percent of wells contaminated, to see the validity of the pyrite oxidation hypothesis. They found no significant but negative trends of relationships for correlations 1 to 4 and no significant but positive trend of relationship for correlation 5 (BGS, 1999). Therefore, they rejected pyrite oxidation hypothesis.

However, due to the lack of time-dependent data, the BGS considered spatial correlations. In fact, the pyrite

oxidation hypothesis is a time-dependent phenomenon. Thus, the validity of rejecting this hypothesis based on the space-dependent relationships needs to be investigated. Moreover, the appropriateness of the selection of parameters also needs to be studied because, for example, the high amount of water abstraction at a certain location will surely cause a high level of water level depletion, but for the same amount of water abstraction, a different level of water level depletion could be observed in space due to the heterogeneity of the aquifer materials. Bridge and Husain (2000) mentioned that the correlation between sulphate concentration and arsenic concentration does not indicate the rejection or acceptance of pyrite oxidation. The sulphate concentration in water depends on many factors, such as pyrite grain size, pyrite abundance, reaction rate, migration time, etc. In West Bengal, there is an evidence of pyrite oxidation with low concentration of sulphate (BGS, 1999). Moreover, the key parameter, percent of wells contaminated, which was used for the abovementioned analyses, contains errors since it was estimated mainly from the field-kits results, and field-kits cannot detect groundwater arsenic if the concentration is less than 0.20 mg/L (BGS, 1999). In addition to these, the estimation procedure of water abstraction is not clear and the study used 1994 water level data to show the relationship with the 1998 arsenic contamination data. The BGS used the potential recharge, estimated by Master Plan Organization (MPO), to show the relationship with percent of wells contaminated. Regarding the potential recharge estimated by MPO, Mott MacDonald International (MMI) in their Deep Tubewell Project II report (MMI, 1992) mentioned that the MPO recharge estimates are too low on the Old and Middle Megna floodplains and too high on parts of the Old Brahmaputra floodplain. Thus, it is necessary to recheck the relationships obtained and to see the validity of using percent of wells contaminated which were estimated mainly from the field-kits results.

In Support of Oxy-hydroxide Reduction Hypothesis

The BGS investigated three correlations: (1) the correlation between extractable iron concentration and arsenic concentration; (2) the correlation between arsenic concentration and the reducing conditions (arsenic and pH-Eh relationship); and (3) the correlation between arsenic concentration and depth of contamination. For both of the first and second correlations, they found significant positive relationships, and therefore accepted oxy-hydroxide reduction hypothesis (BGS, 1999). However, about the appropriateness of the consideration of extractable iron concentration, Bridge and Husain (2000) pointed out that the correlation between extractable iron and arsenic does not necessarily imply the adsorption (oxy-hydroxide reduction) hypothesis and the availability of pH-Eh data are questionable. Thus, the first two relationships became confusing. On the other hand, Bridge and Husain (2000) also

found a significant negative relationship for the third correlation, which contradicts acceptance of oxy-hydroxide reduction hypothesis.

Studies on Age of Wells and Arsenic Contamination

The BGS investigated two relationships; one between the age of wells and concentration of arsenic contamination and another between the age of wells and percent of wells contaminated. For both of the relationships they found significant positive relationships (BGS, 1999) that indirectly support pyrite oxidation hypothesis because aged wells would have higher cumulative decline of water level and cumulative water abstraction value. But the BGS did not accept this relationship as a cause of the pyrite oxidation since they did not get spatial significant positive correlations between maximum depth to water level and percent of wells contaminated. Both of the parameters employed, however, may have errors because 1998 water level data may not be the same as 1994 water level data and percent of wells contaminated may be erroneous due to the field-kits measurement.

Case Studies

Selection of the Study Area

The Deep Tubewell Project II area of Bangladesh (Figure 1) is selected for this study. It comprises 60 thanas (sub-districts) of 11 different districts in the northeast area of Bangladesh. The rationale behind this selection is this is the only area of Bangladesh where geological, hydrological, and environmental precise data (relative to the other areas) are available. Mott MacDonald International (UK) worked for this project from 1983 to 1991, which also worked for the BGS to produce reports on groundwater arsenic contamination of Bangladesh. The MMI's arsenic study initially covered 41 districts of Bangladesh excluding about two-thirds of the Deep Tubewell Project II area and made its conclusion based on the results for these 41 districts, though it finally produced arsenic data for all 64 districts in its main report (BGS, 1999). Therefore, results for the previously unselected area could be compared with those for the selected one.

Selection of the Parameters

Hydro-geological and water resources parameters, such as water level fluctuation, total (gross) water abstraction, net water abstraction and potential recharge, are selected to observe the relationships with the arsenic contamination. As the indices of arsenic contamination, percent of wells contaminated and mean arsenic contamination estimated by the BGS from the field and laboratory test data are considered. Unfortunately, both of these may have errors due to the contamination measurement by the field-kits and due to the inadequate number of samples. But there is no alternative, because although accurate arsenic



Figure 1. Study area and selected thanas (sub-districts).

concentrations for individual random samples (wells) measured by the BGS laboratory in the UK are available, their respective hydro-geological parameter values are not available. The arsenic contamination measured by the field-kits during 1994 to 1997 was compiled in 1998 and additional measurements by the BGS laboratory under the regional survey were made in 1998. But data of maximum depth to groundwater level were not available. Therefore, rather than maximum water level, estimated water level fluctuation is considered to see the relationship with arsenic contamination. On the other hand, water level data are available for 1990, which were correlated with respective water abstractions to see the nature of its well-known relationship (i.e., for a particular location, positive significance in time domain) in the spatial coordinates for heterogeneous specific yield values of aquifer system. The validity of the relationships that use percent of wells contaminated is examined by the nature of the expected and true errors if only field test data or a combination of field and laboratory test data are used to estimate percent of wells contaminated.

Sources of Data

The following data sources are used:

- Arsenic contamination data for 1994 to 1997 (field test data compiled in 1998) and 1998 (regional survey data) and other arsenic-related information from the British Geological Survey reports – main report and supplemental Volumes 1-3 (BGS, 1999).
- Data on performance evaluation of field-kits from the study report of School of Environmental Studies and Dhaka Community Hospital (SOES-DCH, 2000).
- For each thana (sub-district), information on numbers of deep tubewells (DTWs), shallow tubewells (STWs), manual suction lift wells (MSLWs), and their respective command area data (including command area data of low lift pumps also) for 1997 from the National Minor Irrigation Development Project report (NMIDP, 1998).
- For each thana, information on numbers of DTWs, STWs, MSLWs, and their respective command area data (including command area data of low lift pumps also), potable water, and maximum depth to water level (below ground surface) data for 1990 from the study report of Deep Tubewell Project II (MMI, 1991).
- For each thana, information on total area, effective rainfall, crop water requirement, percolation rate, recharge amount, specific yield, and storage coefficient from the study report of Deep Tubewell Project II (MMI, 1991).

Estimation Procedure

Total Water Abstraction

Total (gross) water abstraction could be estimated in two different ways: (1) number of different tubewells multiplied by their respective actual capacities and total operation times, or (2) total command areas of DTWs, STWs and MSLWs multiplied by the irrigation water requirement.

For Bangladesh, information on actual well capacity and total operation time are not available at all. Therefore, the procedure (b) is followed. Even for each thana, data on actual irrigation water requirement are absent. For the study area, irrigation water requirement is therefore estimated using the data of crop water requirement, effective rainfall and percolation rate in the Deep Tubewell Project II report (MMI, 1991). As in more than 85 percent of the area where high yielding Boro rice is cultivated, the report considered Boro rice crop for the estimation of crop water requirement, which requires an additional 150 mm water for puddling. Potable water use is also considered while estimating total water abstraction. Results are presented in depth unit (mm). Two values are obtained in this way one for 1997 and another for 1990. Total water abstraction for 1990 is estimated to see the relationship with the

maximum depth to water level below ground surface for 1990 in spatial coordinate, but that for the year 1997 cannot be estimated because of the lack of water level data.

Net Water Abstraction

Net water abstraction is estimated based on the complete water balance study of the upper shallow aquifer. Horizontal through-flow is assumed to be negligible due to the low head gradient in the study area. At first, gross water abstraction from the shallow aquifer is estimated, assuming that all of the STWs and MSLWs water and 90 percent of the DTWs water are withdrawn from the shallow aquifer. This assumption for the DTWs is based on the fact that water released from the deep aquifer is 10 percent at maximum when considering available piezometric data of the deep aquifer and their respective storage coefficient values. From the total gross water, irrigation return flow is subtracted, and potable water is added (for 1997 after making corrections in terms of yearly population growth rate, 2.1 percent (BBS, 1998), to obtain the net abstraction. Finally results are presented in depth unit (mm).

Total and Net Water Level Fluctuations

Total and net water level fluctuations are estimated dividing the total water abstractions and the net water abstractions by the specific yield values (in decimal), respectively. Both results are presented in depth unit (m).

Equivalent Well Intensity

Well intensity should have effect on water level decline. It is assumed that all the DTWs, STWs and MSLWs have effect on the upper aquifer. To estimate equivalent tubewell intensity, the following relationship is used:

Equivalent well intensity = (No. of STWs + No. of DTWs × (average command area of a DTW/average command area of a STW) + No. of MSLWs × (average command area of a MSLW/average command area of a STW))/total area of the thana

The command area of one type of tubewell in a thana is estimated by dividing total command areas of that type of tubewell by its total well number. Then the average command area of that type of tubewell for overall study area is obtained by averaging all of these estimated command areas.

The Student *t*-Test

In each correlation, the significance of the regression is evaluated by the *t*-test. In the *t*-test, at first a null hypothesis H_0 is selected such that there is no significant relationship between two parameters, and then *t* statistic is estimated using the following formula (Gupta and Kapoor, 1984):

$$t = \frac{r\sqrt{n-2}}{\sqrt{1-r^2}} \quad (1)$$

where *r* equals observed correlation coefficient, and *n* equals number of observations.

After getting the *t* value, the tabulated $t_{\alpha/2}$ value is collected for *n*-2 degrees of freedom from any standard statistics book like Walpole and Myers (1985) for a α -level of significance. The value is usually set at 0.05. If $|t| > t_{\alpha/2}$ then H_0 is rejected to recognize a significant relationship between two parameters.

Results and Discussions

Spatial Relationships

Relationships between Hydro-geological parameters and Arsenic Contamination

Investigations of some relationships between hydro-geological parameters and arsenic contamination are carried out for the study area to make a comparison with the latest BGS research findings. The selected parameters are: estimated gross and net water level fluctuations, estimated gross and net water abstraction, potential recharge, and estimated equivalent well intensity. Actual water level data for the years of interest and aquifer transmissivity values are, however, not available for comparison. The percent of wells contaminated, obtained from the field and laboratory test data (BGS, 1999), is taken as an index of arsenic contamination. And the mean arsenic concentration from the laboratory test data (BGS, 1999), is taken as another index though it is not available for all of the selected 60 thanas. The results of regression analyses for these two indices are shown in Figures 2 and 3, respectively. The significance of the regressions is evaluated by the student *t*-test.

For the study area, one can see from Figure 2 that when the percent of wells contaminated is considered as contamination index, the regressions for all of the abovementioned hydro-geological parameters become negative. Except for potential recharge and total water abstraction, *t*-tests confirm significant, negative relationships between arsenic and other parameters. When mean arsenic concentration is considered as contamination index, all other parameters except for potential recharge show negative relationships with the arsenic index (Figure 3). However, *t*-tests show no significant relationships. For both of the indices, only a small proportion of the variation could be explained by the above relationships since r^2 values are between 0.37 and 0.14 for three cases and less than 0.1 for the remaining nine cases. These results agree with the research findings of the BGS, except for the fact that in this study percent of wells contaminated gives a negative but insignificant relationship with the potential recharge. However, none of the above relationships support pyrite oxidation hypothesis, because for pyrite oxidation we should expect a significant negative relationship between arsenic and potential recharge and significant positive relationships between arsenic and the remainder of the parameters.

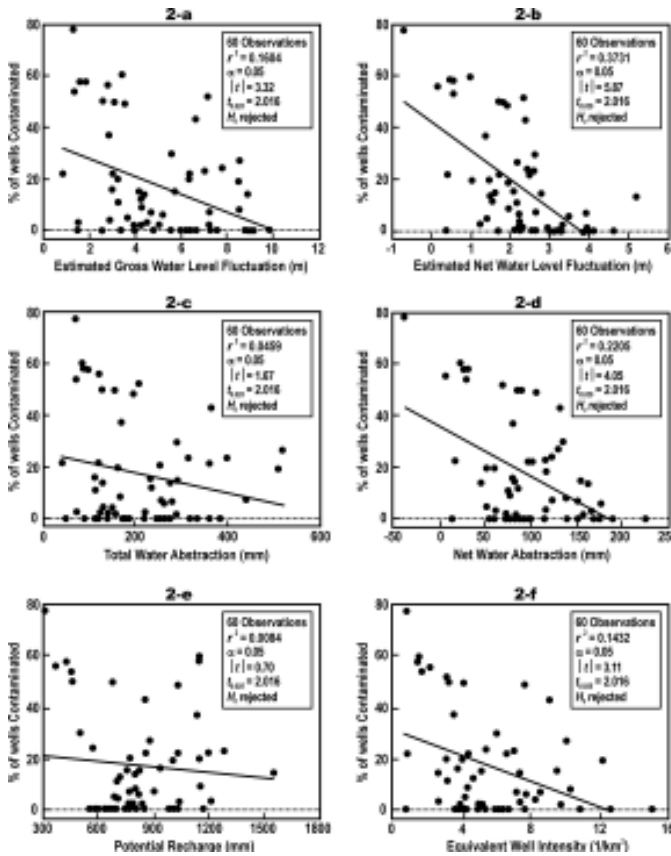


Figure 2. Correlations between hydro-geological parameters and percent of wells contaminated.

The above relationships do not show any evidence of accepting pyrite oxidation hypothesis. Before rejecting it, however, we should thoroughly investigate the appropriateness of the selection of parameters and the validity of the space-dependent relationships for the time-dependent phenomenon. Moreover, we know that neither mean arsenic concentration nor percent of wells contaminated is appropriate an index of arsenic contamination because of inadequate samples and the possibility of error.

Time-Dependent Relationship

Relationship between Arsenic Concentration and Age of Wells

The BGS investigated variation of arsenic with time. Due to the unavailability of time series data, the BGS used age of wells to infer change of arsenic over time. In its study, the BGS found positive significant relationship between age of wells and absolute arsenic concentration by the *F*-test, which is a well-known significance of regression test. But when percent of wells contaminated was used instead of absolute arsenic concentration, the *F*-test did not show any significance of this relationship. Therefore, we reinvestigated the same relationship and evaluated significance of the regression by the *t*-test. In our observation, the *t*-test confirms that there is, in fact, a sig-

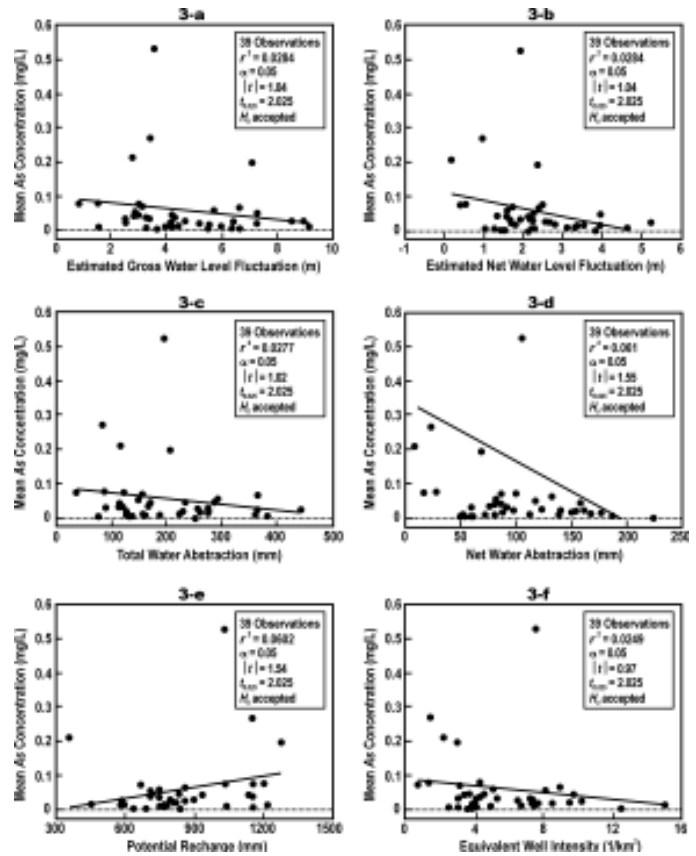


Figure 3. Correlations between hydro-geological parameters and mean arsenic contaminated.

nificant relationship between age of wells and arsenic concentration (Figure 4). This relationship gives an important indication of mobilization of arsenic by pyrite oxidation. For more confirmation, however, further investigation is needed into different relationships among hydro-geological parameters in time domain.

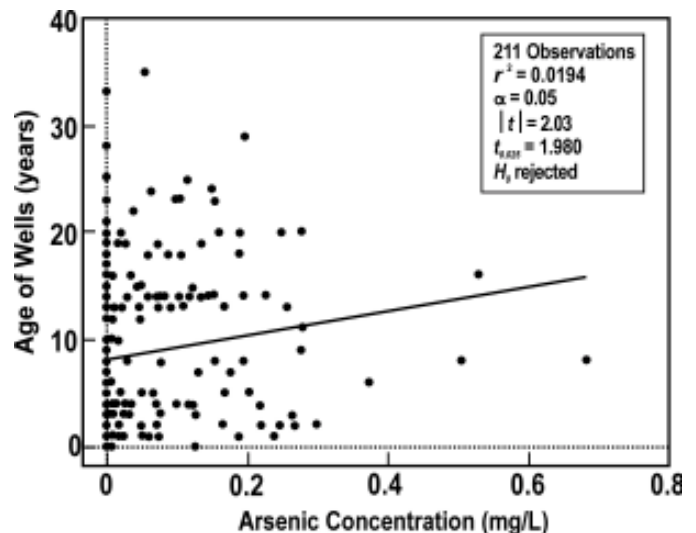


Figure 4. Correlation between arsenic concentration and age of wells.

Errors in Percent of Wells Contaminated

In the BGS analyses, the key parameter, percent of wells contaminated, was estimated using the field test data of different organizations, which were mainly collected through the field-kits by unskilled persons. On the basis of studies completed by different organizations on the performance evaluation of field-kits, the BGS reports that field-kit cannot reliably detect groundwater arsenic if its concentration is less than 0.2 mg/L (BGS, 1999). In the BGS main report, it is mentioned that the semi-quantitative kits almost never significantly over-estimate the true arsenic concentration and are sometimes quite accurate, but also frequently under-estimate the concentration. But the reports did not say how much error we could expect in percent of wells contaminated due to the under-estimation by the field-kits. On the basis of BGS comment that field-kits only under-estimate percent of wells contaminated, an attempt is made to see the possible expected errors in percent of wells contaminated that would be estimated from the field-kits results. Random samples of the regional survey (BGS, 1999) that have arsenic concentration in the range of 0.05 to 0.20 mg/L would be erroneous if they would be measured by the field-kits. A percentage of such samples in the total samples is considered as percent of expected errors. Similarly the expected percent of wells contaminated could be estimated from the regional survey data with arsenic concentration of higher than 0.2 mg/L if samples would be measured by the field-kits. It is found that for all 250 thanas the expected percent of wells contaminated is 14.12 percent with standard deviation 21.55, whereas the expected percent of errors is 19.55 percent with standard deviation 20.69. This means that if we present expected percent of errors in terms of percent of wells contaminated, it would be 138.45 percent. The mean percentages of expected errors for different ranges of percent of wells contaminated are presented in Figure 5.

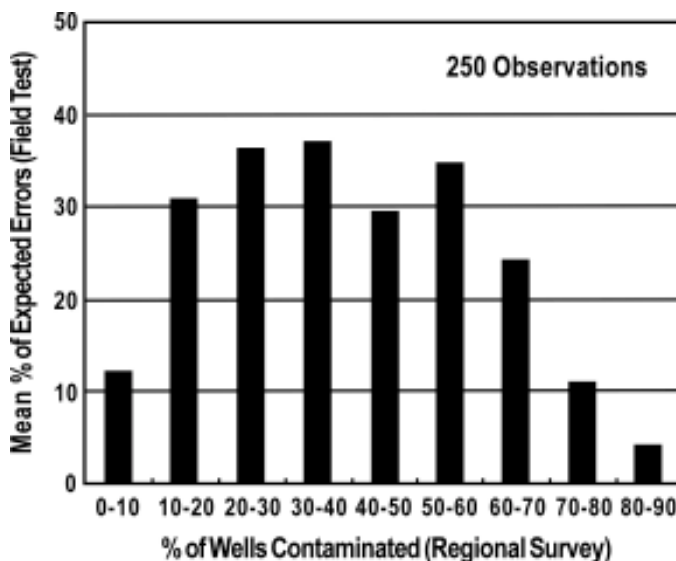


Figure 5. Expected errors in percent of wells contaminated.

Comparisons of field-kits tests with the laboratory tests (regional survey) were made for different threshold arsenic concentrations by BGS. Results showed quite poor correlations with maximum r^2 of 0.21 and 0.52 for >10 and >50 data points per thana, respectively (BGS, 1999). But BGS reported that they obtained the best fits (r^2 of 0.48) for the combined indices of arsenic concentration (a location showing arsenic concentration >0.05 mg/L by either field test or existing laboratory test) with the regional survey due to the large sample size. But BGS did not present any figure in their reports to support this finding. Therefore, an attempt is made to determine the goodness of the fit with the regional survey if combined field and laboratory tests results are used to estimate percent of wells contaminated. The relationship obtained is presented in Figure 6. In Figure 6, r^2 value is found to be 0.53 showing moderate fitness of the data. But if we carefully observe this relationship, we could see that when percent of wells contaminated from regional survey data exceeds 30 percent, there is a quite poor correlation. It could be possible to show by making a separate relationship using data percent of wells contaminated exceeding 30 percent that r^2 value would be 0.20.

From the above analyses (Figures 5 and 6), we can gauge about errors in percent of wells contaminated. However, neither Figure 5 nor Figure 6 represent true situation because Figure 5 is drawn using the expected errors and Figure 6 represents a comparison where the sampled wells and the sampling strategies are not the same. The true situation could be understood by the detailed analysis of School of Environmental Studies (SOES) and Dhaka Community Hospital (DCH) (SOES-DCH, 2000). A performance evaluation of field-kits was done jointly by SOES and DCH using water samples collected from 2,866 tubewells which were colored red (contaminated-arsenic concentration >0.05 mg/L) or green (safe-arsenic con-

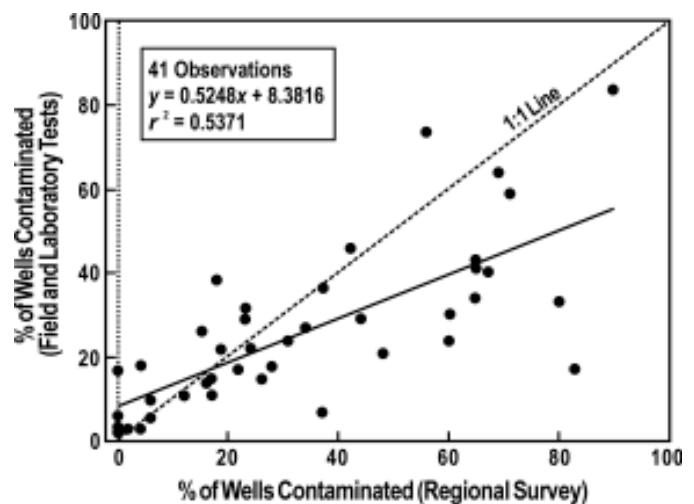


Figure 6. Comparison between percent of wells contaminated estimated by the regional survey data and field and laboratory tests data.

centration <0.05 mg/L) after testing with field-kits. For comparison, SOES-DCH used FI-HG-ASS system laboratory analysis that can determine arsenic up to 0.002 mg/L with 95 percent confidence level. Using SOES-DCH data. A figure is presented to show the percent of wells violated in field-kits measurement. Figure 7 shows that contaminated and safe wells, identified by field-kits, are 60.12 percent and 39.88 percent, respectively. When contaminated (red) wells are rechecked by FI-HG-ASS system laboratory analysis, 30.50 percent (50.73 percent of the total red wells) is considered as safe (green), and the remaining 29.62 percent (49.27 percent of the total red wells) are considered truly contaminated. Thus, if we represent violation (over-estimation) in the field-kits in terms of truly contaminated wells identified from the red wells, it would be 102.97 percent. On the other hand, when safe (green) wells are rechecked, 2.93 percent (7.35 percent of the total green wells) is regarded as contaminated (red) and therefore the percentage of truly safe wells is reduced to 36.95 percent (92.65 percent of the total green wells). Thus, the rate of violation (under-estimation) in the field-kits for the green wells is 7.93 percent. Therefore, by combined analysis, true percent of contaminated wells for all of the 2,866 samples could be obtained by adding percent of wells violated in the green wells and the actual percent of wells contaminated found in the red wells. Similarly, true percent of safe wells for all samples could be estimated. The truly contaminated and safe wells for all samples are found to be 32.55 percent and 67.45 percent, respectively. Therefore, combined analysis shows that there are 84.70 percent (in terms of actually contaminated wells) over-estimation and 40.87 percent (in terms of actually safe wells) under-estimation in the field-kits results. The above findings show that overall field-kit do not underestimate but over-estimate actual status of groundwater arsenic contamination. The reason may be the semi-quantitative nature of field-kits, as presented in Figure 8 (drawn from SOES-DCH data), where the highest percentage of violation (74.1 percent) could be observed in low concentration range (0.03 to <0.05 mg/L) of arsenic. The mentality of unskilled field-kit workers is also sometimes responsible for such over-estimation. SOES-DCH received information from a field coordinator that when field-kit workers get several safe tubewells in the same area after testing by field-kits, they are not happy to have the entire tubewells safe. Consequently, they decide to color at least a few tubewells red (contaminated) (SOES-DCH, 2000). Whatever the reason, field-kits significantly mis-identify actual status of tubewells. Therefore, the percent of wells contaminated, estimated from field-kit tests or combination of field-kit and laboratory test results, is not dependable.

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Validity of the Space-Dependent Relationships

To determine the validity of the space-dependent relationships among different hydro-geological parameters, other than water level and arsenic contamination, internal relationships between total water abstraction and maximum depth to water level below ground surface (i.e., depth of the lowest water table in the year from the ground surface, assuming elevations of ground surface at all locations are the same since the study area is almost plane) and between net water abstraction and maximum depth to water level below ground surface are investigated and are illustrated in Figures 9 and 10, respectively. From Figures 9 and 10, it is apparent that there is a significant negative relationship between total water abstraction and maximum depth to water level and there is no relationship between net water abstraction and maximum depth to water level. Both of these are opposing the fact that the same is true for a particular location if considered over time. This is

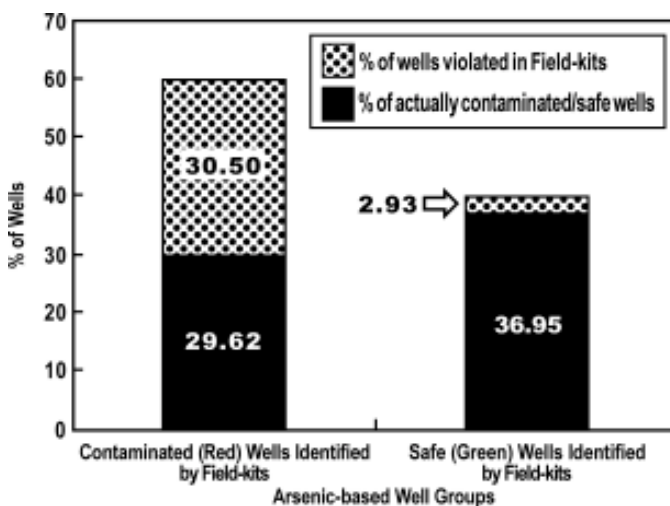


Figure 7. Comparison between actual percent of wells contaminated and percent of wells violated by field-kit measurement.

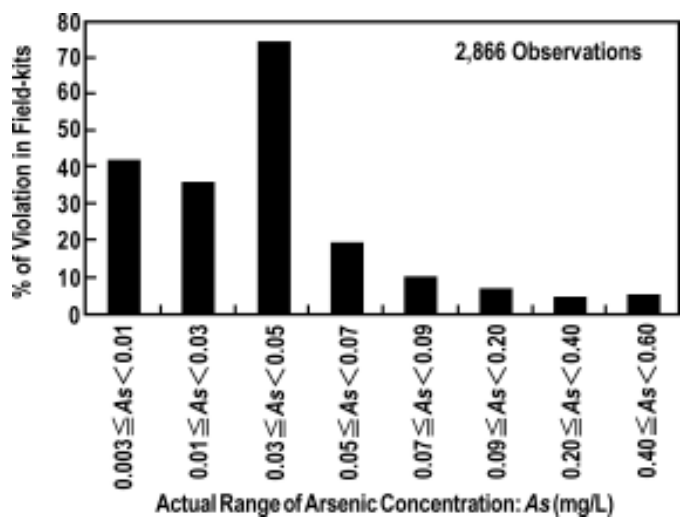


Figure 8. Percentage of wells violated by field-kit measurement for different range of actual arsenic concentration.

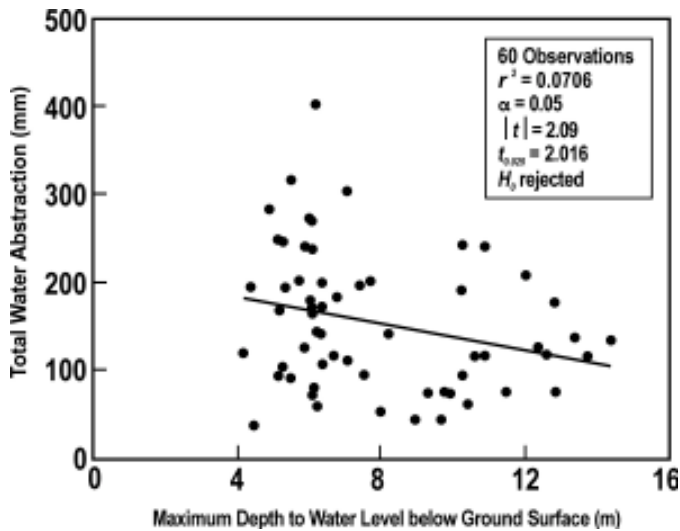


Figure 9. Spatial correlation between maximum depth to water level and total water abstraction.

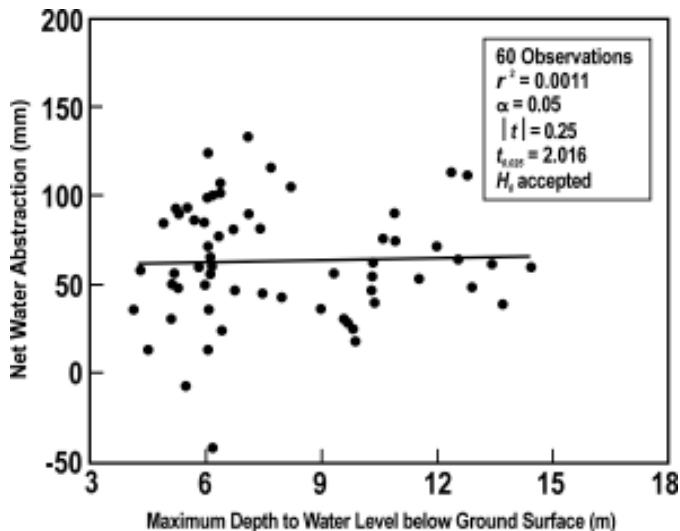


Figure 10. Spatial correlation between maximum depth to water level and net water abstraction.

mainly due to the location factor (heterogeneous specific yield values). When a specific yield value for a particular location is constant over time, we may consider water level as a function of water abstraction only and expect positive significant relationship between them. If the same is considered over space, water level is not a function of water abstraction only since location factor (specific yield value) also affects water level. Thus, if after ignoring location factor we consider the relation of water level and water abstraction, there is a possibility of having a false relationship. The true relationship could be visualized by making a three-dimensional representation of this relationship (x : is water level, y is water abstraction, and z is location factor) obtained from the time series data for all of the locations. We may obtain a similar false relation for water

level and arsenic contamination also, because arsenic contamination for a particular location would only be the function of water level if their relationship is considered in time. But in space, arsenic contamination would be the function of water level and location factor (arsenic in subsurface geology). The above findings indicate that for a heterogeneous aquifer system space-dependent relationships are not representative of the time-dependent relationships. Therefore, employing these to identify the cause of arsenic contamination in groundwater will not be valid. Hence, to determine the actual cause of groundwater arsenic contamination, reliable time series data must be generated. As it is difficult to represent arsenic contamination of a thana accurately by either percent of wells contaminated or mean arsenic concentration, and as maximum depth to water level and other hydro-geological data of a thana are averaged one, it would be wise to select some random samples (wells) in collecting data of arsenic contamination and water level as well as all other necessary information in time domain. The final conclusion could be made by the relationships obtained only from these data.

Conclusions

An investigation was undertaken for a study area of Bangladesh to identify the cause of groundwater arsenic contamination. Correlations between different hydro-geological parameters and arsenic contamination were investigated and the results were compared with the BGS findings. The validity of the relationships was also evaluated. From this study, the following conclusions could be made:

- Spatial relationships between hydro-geological parameters and arsenic pose no evidence of pyrite oxidation as a cause of groundwater arsenic contamination in Bangladesh. This result agrees with the BGS research findings. However, in the validity test, these space-dependent relationships become invalid. Moreover, error analysis shows that the key parameter of these relationships, percent of wells contaminated, contains much error.
- Positive significant correlation between absolute concentration of groundwater arsenic and age of wells suggests validity of the pyrite oxidation hypothesis.
- Studies on neither pyrite oxidation nor oxy-hydroxide reduction hypotheses by the BGS using the existing data are enough to draw any final conclusion.
- Final conclusion could be made only by the time-dependent relationships using reliable field data.

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