

## Contributed Articles

# A Forensic Look at Groundwater Arsenic Contamination in Bangladesh

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10 A comprehensive forensic analysis of AS contamination in Bangladesh was undertaken using geographic information system (GIS), univariate, and bivariate statistics. The assessment provided the background needed for fine-tuning emerging/evolving methods of spatial mapping based on nonlinear deterministic dynamics and multivariate factorial analyses. Data were derived from a nation-wide AS survey completed in 1999 by the British Geological Survey (BGS) in collaboration with the Department of Public Health Engineering (DPHE) of Bangladesh. It was found that the standard deviation of arsenic concentration is highest along the river flood plains of the major rivers. The distribution of arsenic concentration was found to be symmetric in regions of very high mean concentration, while the regions of low and moderate contamination reveal a highly skewed distribution, suggesting the need for data transformation at these points prior to spatial interpolation. The mean value of Ca and Mg concentrations showed a strong spatial similarity in the southwest region of Bangladesh. This spatial similarity was also reflected in terms of skewness of the data. It was inferred that the mapping of AS concentrations at nonsampled locations on the basis of multivariate factorial or regression models may not require the dual presence of both Ca and Mg parameters; hence, one of them is redundant. Both the skewness and kurtosis of the distribution of Si concentration appeared very similar and highly heterogeneous in space suggesting that Si data requires careful transformation prior to modeling only in the smaller scale but not for country-wide/regional purposes. The age of wells was found to be nonlinearly correlated in the southeast region. The correlation of AS with Fe appeared strongest in the southwest region indicating that the oxidation–reduction hypothesis may be more appropriate as a release mechanism in the southwest region. In another regard, the sulphate concentration was found to be more correlated to AS in the northwest Pleistocene region of Bangladesh, suggesting that the pyrite oxidation hypothesis may be appropriate. Overall, our forensic analysis using GIS revealed useful findings for data transformation and selection of parameters towards spatial mapping based on multivariate factorial and nonlinear deterministic models in the resource-poor settings of developing countries.

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25 Keywords: arsenic, Bangladesh, groundwater, forensics, GIS, univariate and bi-variate statistics, spatial mapping

### Introduction

30 The large-scale AS contamination in the groundwater aquifers of Bangladesh needs no renewed introduction. Since its discovery in 1993, numerous studies have steadily improved our understanding of the contamination scenario in the following areas: 1) the spatial extent, mapping, and management (Biswas et al., 1998; Burgess et al., 2000; Mukherjee and Bhattacharya, 2002; Hossain and Sivakumar, 2006; Hossain et al., 2006; in press ); 2) 35 probable mechanisms of contamination (Harvey et al., 2002; van Geen et al., 2003a, 2003b; McArthur et al., 2001, 2004; Nickson et al., 1998); 3) effect on public health (Yu et al., 2003), and 4) 40 Q3 the socioeconomic effect (Nahar et al., in press ). It is now esti-

40 mated that at least 50% of the country’s population is affected by this large-scale contamination with the most vulnerable victims being the villagers (Ahmed, 2003).

45 When it comes to spatial mapping for cost-effective management, and probable mechanisms of contamination, a significant amount of uncertainty or lack of consensus among scientific researchers continues to exist in current literature. For example, it has been reported that the widespread application of geostatistical techniques such as kriging for the spatial mapping of AS contamination results in an interpolated ‘field’ subject to greater uncertainty at nonsampled locations due to measurement and sampling errors of in situ tests (Hossain et al., 2006; in press). In a 50 recent assessment of ordinary kriging, Hossain et al. (in press) reported that the probability of successful prediction of safe wells is 72% (for WHO safe limit—10 ppb) and 78% (for Bangladesh safe limit—50 ppb). Their findings from this study exemplified that, while mainstream geostatistical approaches (e.g., ordinary 55 kriging) may not provide the most accurate prediction of mean

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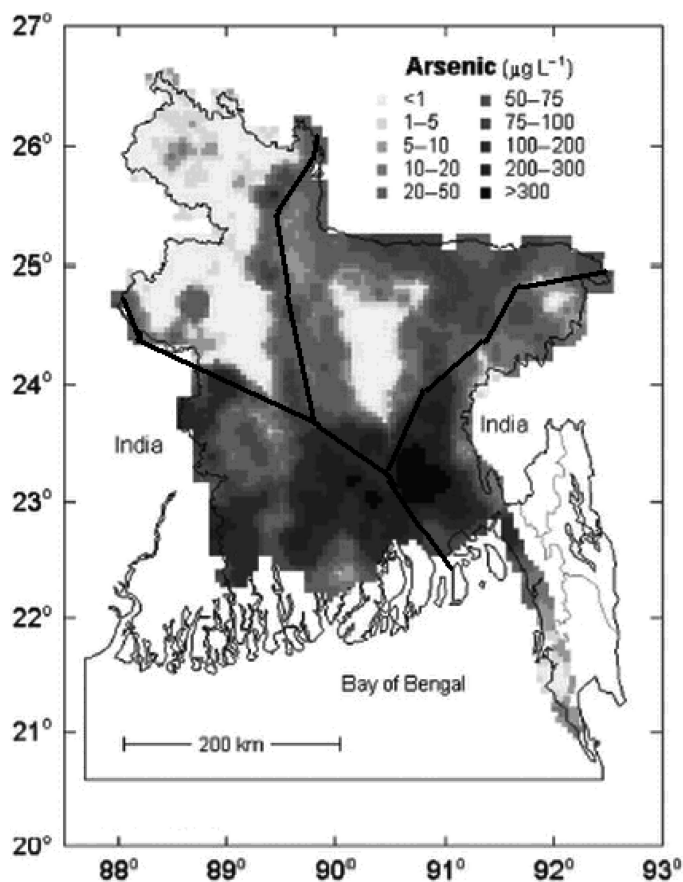
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AS concentration at non-sampled locations, they can identify an approximate strategy for management of AS contaminated shallow groundwater if applied carefully.

60 Similarly, the actual causes of high AS concentration in the groundwater of Bangladesh have not yet been clearly pinpointed. Among the few hypotheses initially proposed to explain the possible mechanism of AS release, the scientific community appears to have converged on two major hypotheses: 1) *pyrite oxidation hypothesis* (Kinniburgh and Smedley, 2001)—oxidation of AS mineral ‘arsenopyrite’ (FeAsS) or AS-rich ‘pyrite’ resulting in release of AS in groundwater, and 2) *oxy-hydroxide reduction hypothesis* (Nickson et al., 1998)—reduction of AS-rich Fe-oxihydroxides leaching the AS that remains at adsorbed state on  
Q4 its surface. In a recent e-mail survey conducted among experts to identify the prevailing views on the possible mechanism of AS release, 58% of the respondents supported the oxyhydroxide reduction hypothesis, 33% supported the pyrite oxidation hypothesis, and 75% of the respondents believed that overextraction of  
70 groundwater had some relationship with the contamination of AS in groundwater in Bangladesh (Akmam, 2002).  
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Given the prevailing uncertainty that currently exists on the probable causes of contamination and the limitations of mainstream geostatistical methods for spatial mapping and management, the recent collective works of Hossain et al. (2006; in press), Hossain and Sivakumar (2006) and Hill et al. (in press) have argued for the adoption of more enhanced and value-added techniques to derive cost-effective strategies for management of the contaminated groundwater resources. For example, Hossain and Sivakumar (2006) have argued that the geostatistical treatment of AS contamination in space as a regionalized (stochastic) random variable constitutes an incomplete analysis of its spatial variability. Because Bangladesh is essentially a riverine (dendritic) country with numerous ‘small’ floodplains (see Figure 1 for the network of major rivers), and further because the geology shows presence of pockets of Holocene-like and Pleistocene-like deposits scattered throughout the country, there is adequate reason to anticipate chaotic (nonlinear deterministic dynamic) behavior in the spatial pattern of AS contamination. Their study  
85 indicated correlation dimension (CD) values (Grassberger and Procaccia, 1983) ranging anywhere from 8 to 11 depending on the region, suggesting that the AS contamination in space, from the chaotic point of view, is a medium- to high-dimensional problem. In a follow-up study, Hill et al. (in press) demonstrated, using logistic regression, possible physical connections between the nonlinear deterministic CD value and the mathematical modeling of risk of AS contamination in groundwater. They reported that the uncertainty associated with prediction of wells as safe and unsafe by logistic regression model declines systematically  
90 as the total number of influencing variables increases from 7 to 11, suggesting that CD can act as a rapid proxy to improve spatial mapping of contamination under a resource-limited scenario.  
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In this study we have therefore attempted to undertake comprehensive forensic analysis of AS contamination in Bangladesh using Geographic Information System (GIS), univariate and bivariate statistics. Although our assessment is entirely data-based  
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**Figure 1.** Study region showing the mean AS concentrations based on the British Geological Survey–Department of Public Health Engineering (BGS-DPHE, 2001) data. The solid dark line represents the approximate longitudinal axis of majors rivers and flood plains in Bangladesh.

and mostly qualitative, we believe that such type of analyses—which has not been reported yet in literature to the best of our knowledge—can develop the knowledge base necessary for cost-effective techniques on spatial mapping. For example, the  
115 use of correlation dimension analyses in deterministic modeling requires improved understanding of geochemical parameters that co-exist with AS contamination. Hence, prior knowledge of the spatial variation of correlation coefficient would be beneficial. Similarly, the use of enhanced (i.e., value-added and  
120 more complex) geostatistical techniques such as factorial kriging (Goovaerts, 1997) requires knowledge of cross-correlation among geochemical parameters responsible for AS contamination (Lin et al., 2006).

It is important to highlight at this stage that, prior to the ad-  
125 vent of easy-to-use GIS tools, such an analysis over a broad region (a country) would have been almost difficult, if not impossible, to carry out to gain the ‘big picture’ snap shot on the preliminary forensic status of contamination. To the best of our knowledge, such a GIS-based assessment that aims to search  
130 for ‘connections’ among contamination variables from a data-based yet ‘forensic’ perspective is absent in literature as far the AS contamination of groundwater in Bangladesh is concerned.

Q5 While the study presents only a baseline (i.e., “a ball-park”) on  
 135 the forensic nature without a clear revelation on the physics of  
 the AS contamination, the findings from such an assessment  
 along with the limitations that may be revealed, can only justify  
 further the need for more detailed physically based studies in-  
 volving GIS, forensic tools and contamination theory to advance  
 140 spatial mapping tools for developing countries.

The objectives of our study are therefore as follows:

1. To compare the univariate statistical behavior of pertinent  
 geochemical parameters with AS concentrations and thereby  
 establish qualitative connections on the type of data transfor-  
 145 mations needed prior to spatial mapping.
2. To qualitatively analyze the bivariate statistical behavior of  
 pertinent geochemical parameters with AS and thereby es-  
 tablish connections on the choice of variables for multivari-  
 ate modeling (such as logistic regression or factorial kriging  
 150 etc.).

We expect our findings to provide the background needed for  
 fine-tuning the emerging/evolving methods of spatial mapping  
 based on nonlinear deterministic dynamics, multivariate factor-  
 ial analyses. This article is organized as follows. In the following  
 155 section, we describe the data used for the study, followed by a  
 description of the tools and methods. In the results and discus-  
 sion section, we present our forensic findings on univariate and  
 bivariate analyses and discuss their implications. Finally, in the  
 conclusion we present the major findings of our study and the  
 160 suggested extensions of our work.

### Study Region, Data, and Methods

Our study region was Bangladesh excluding the dense forests  
 in the southwest and southeast (Figure 1). Geologically, the  
 region is made up of mainly old oxidic Pleistocene and rela-  
 165 tively young anoxic Holocene deposits. Our dataset on AS con-  
 centration of groundwater was derived from the BGS-DPHE  
 (2001) study comprising 3,534 wells (Figure 1). The dataset  
 was available at: <http://www.bgs.ac.uk/arsenic/Bangladesh.html>  
 Q6 (accessed). In the overall scheme of our investigation, the BGS-  
 170 DPHE (2001) survey currently represents the most quality-  
 controlled database of AS measurements available for any kind  
 of country-wide forensic and GIS analyses. AS measurements  
 of BGS-DPHE 2001 survey were based on the atomic absorp-  
 tion spectrophotometric method, which is currently considered  
 175 a reliable method used for AS testing (Rahman et al., 2002;  
 Hossain et al., 2006; Goovaerts et al., 2006). In another regard,  
 even though the Bangladesh Arsenic Mitigation Water Supply  
 Q7 Project (available at: [www.bamwsp.org](http://www.bamwsp.org) (accessed)) is host to an  
 extensive archive on the status of approximately two to three mil-  
 180 lion wells, this larger dataset is not amenable to forensic analyses  
 due to the following two reasons: 1) use of semiquantitative field  
 kits with very large measurement errors (Hossain et al., 2006);  
 and 2) lack of geospatial information on individual wells that

were sampled (Project Director of National Arsenic Mitigation  
 Council – NAMIC, Bangladesh, personal communication,). 185

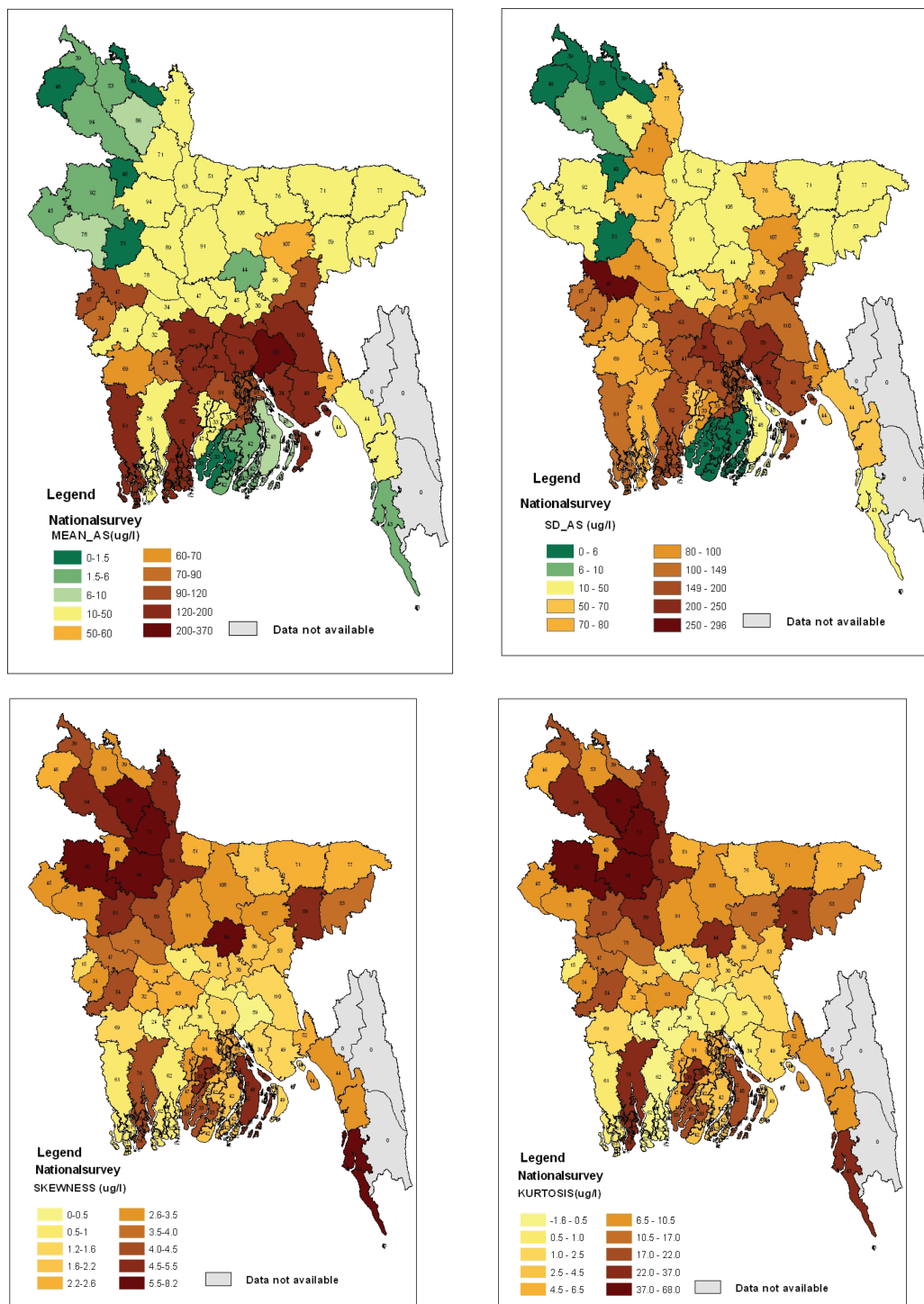
We assumed the in situ (field measured) data on AS concen-  
 Q8 tration and pertinent geochemical and hydrologic variables to be  
 reasonably stationary at the time scales of intermediate manage-  
 ment efforts (i.e., 5–6 years) for which the improved schemes on  
 spatial mapping are intended in developing countries. This is an 190  
 acceptable assumption until a more structural solution is devised  
 for the AS problem in Bangladesh. While the incorporation of  
 a dynamic contaminant transport model would undoubtedly be  
 relevant for our investigation (Davis et al., 2004), there are sev-  
 eral limitations to spatial mapping of the dynamic AS field that 195  
 the current state-of-the-art on AS data measurement protocols  
 and scientific know-how can not address. These are: 1) mecha-  
 nism of AS release in the groundwater for the shallow aquifers  
 of Bangladesh is not well understood and actively debated  
 by the greater scientific community (see for example, Harvey 200  
 et al., 2002; van Geen et al., 2003; McArthur et al., 2004); Q9  
 2) groundwater flushing rates are insignificant in Bangladesh  
 given its flat terrain (in the timescales of millennia; refer to  
 the report by BGS-DPHE, 2001); 3) there are no country-wide  
 well-monitoring programs that record the temporal fluctuations 205  
 of AS concentration in groundwater. In fact, to the best of our  
 knowledge, a second measurement of AS concentration at a  
 cluster of wells on a regional basis has not been performed due to  
 the prohibitive costs involved. We further justify our assumption  
 of stationarity based on Goovaerts et al. (2005) who reported an 210  
 absence of a trend or seasonality for naturally occurring AS con-  
 centration in southeast Michigan over a period of 10 years. Davis  
 et al. (2004) has also argued against fitting standard contaminant  
 process models without a scientific basis in the absence of mech-  
 anistic understanding of field scale variability of AS could be 215  
 dangerous.

Table 1 shows the potential list of ‘culprit’ parameters that Q10  
 were analyzed in this study. These variables were co-sampled by  
 BGS-DPHE (2001) along with AS concentration in well water.  
 The minimum (i.e., basically detection limits) and maximum 220  
 values of these variables (Table 1) indicate the range of variabil-  
 ity across Bangladesh. The parameters chosen were (in mg/L):  
 1) Fe; 2) Mg; 3) Ca; 4) SO<sub>4</sub>; 5) Si; 6) Na; and 7) Zn. Additional  
 parameters of ‘age’ (in years) and depth (in meters) of wells  
 were also selected (not shown in Table 1). Our choice of vari- 225  
 ables is primarily dictated by literature reports on the causes of

**Table 1.** The geochemical parameters for forensic analysis. (Note: ‘Depth’ and ‘age’ are not shown herein as they are not geochemical parameters)

Variable	Mean	Minimum (~ Detection Limit)	Maximum
Ca (mg/L)	51.590	0.100	366.000
Fe (mg/L)	3.353	0.005	61.000
Mg (mg/L)	20.750	0.040	305.000
Na (mg/L)	88.936	0.700	2700.000
Si (mg/L)	20.519	0.030	45.200
SO <sub>4</sub> (mg/L)	5.917	0.200	753.000
As (ppb)	55.205	0.500	1660.000

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Figure 2. Univariate statistical behavior of AS concentration in terms of four central moments: 1) mean (top left), 2) standard deviation (top right), 3) skewness (bottom left), and 4) kurtosis (lower right). SD, standard deviation.

AS mobility (e.g., Welch et al., 2000; Harvey et al., 2002; van  
Q11 Geen et al., 2003a; McArthur et al., 2004; Zheng et al., 2004),  
the availability of accurate data and the suitability for spatial  
230 mapping (see Lin et al., 2006).

For GIS analysis we used the software ArcGISTM (Ormsby  
et al., 2004) that has built-in capability to produce univariate

and bivariate statistics of geo-located data and graphical pres-  
entation. For univariate statistics, we analyzed the data for: 1) Q12  
mean; 2) standard deviation; 3) skewness, and 4) kurtosis (i.e., 235  
excess kurtosis). For bi-variate statistics we basically derived  
correlation measures for 1) Pearson's measure, and 2) Spear-  
man's rank correlation. We were particularly interested in rank



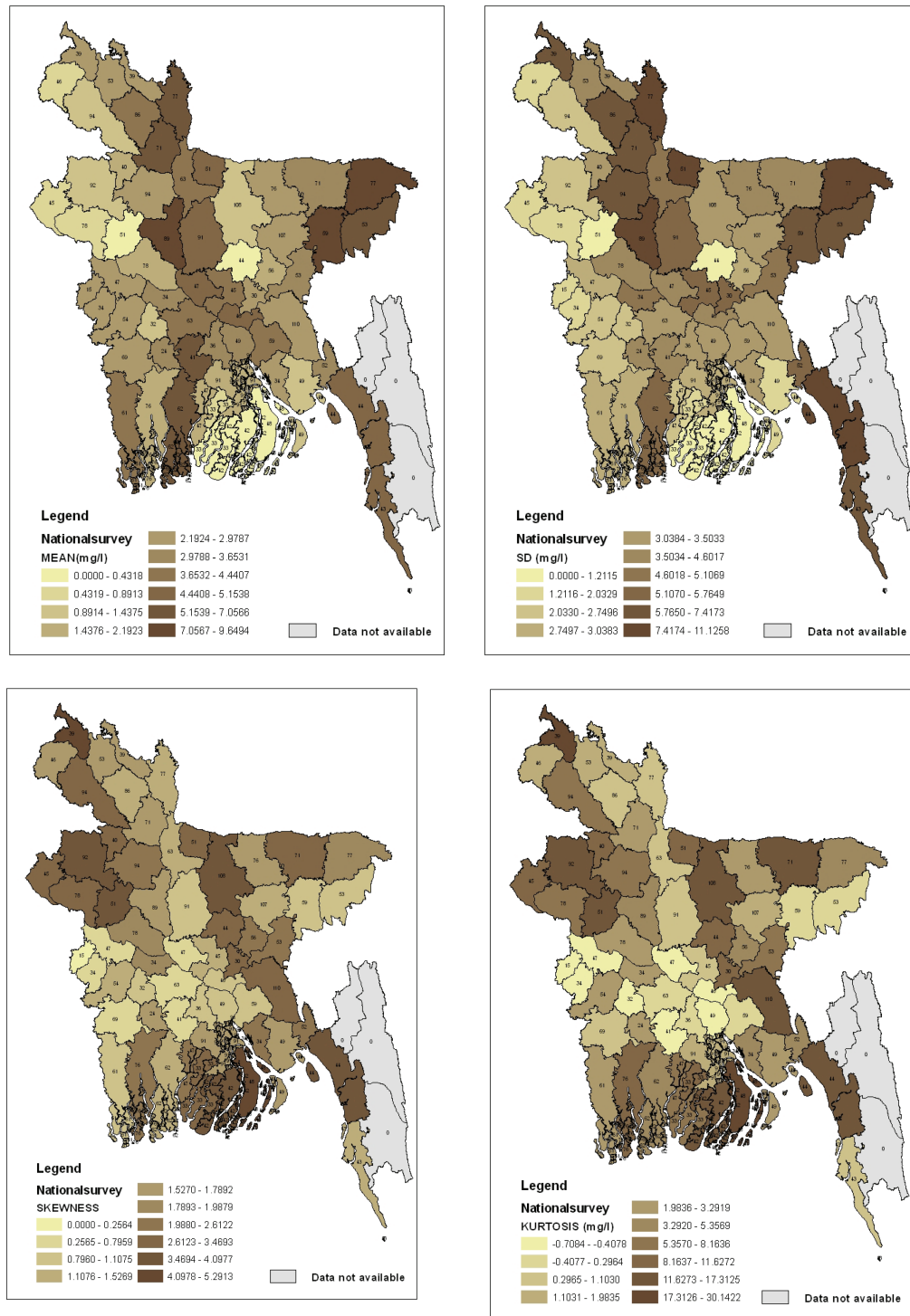


Figure 3. Univariate statistical behavior of Fe concentration in terms of four central moments: 1) mean (top left), 2) standard deviation (top right), 3) skewness (bottom left), and 4) kurtosis (bottom right).

240 correlation because of its ability to identify nonlinear covariation that is not usually detectable by the Pearson's measure. Unfortunately, ArcGISTM does not automatically yield the level of significance of the statistical analyses; hence they could not be reported in the herein study. However, such levels of confidence

are important and should be reported in future work. Because the statistical measures we quantified are commonly used and 245 available in any standard textbook on statistics, their theoretical definition is avoided herein. The statistical analyses were conducted for each district using all the individual well data within

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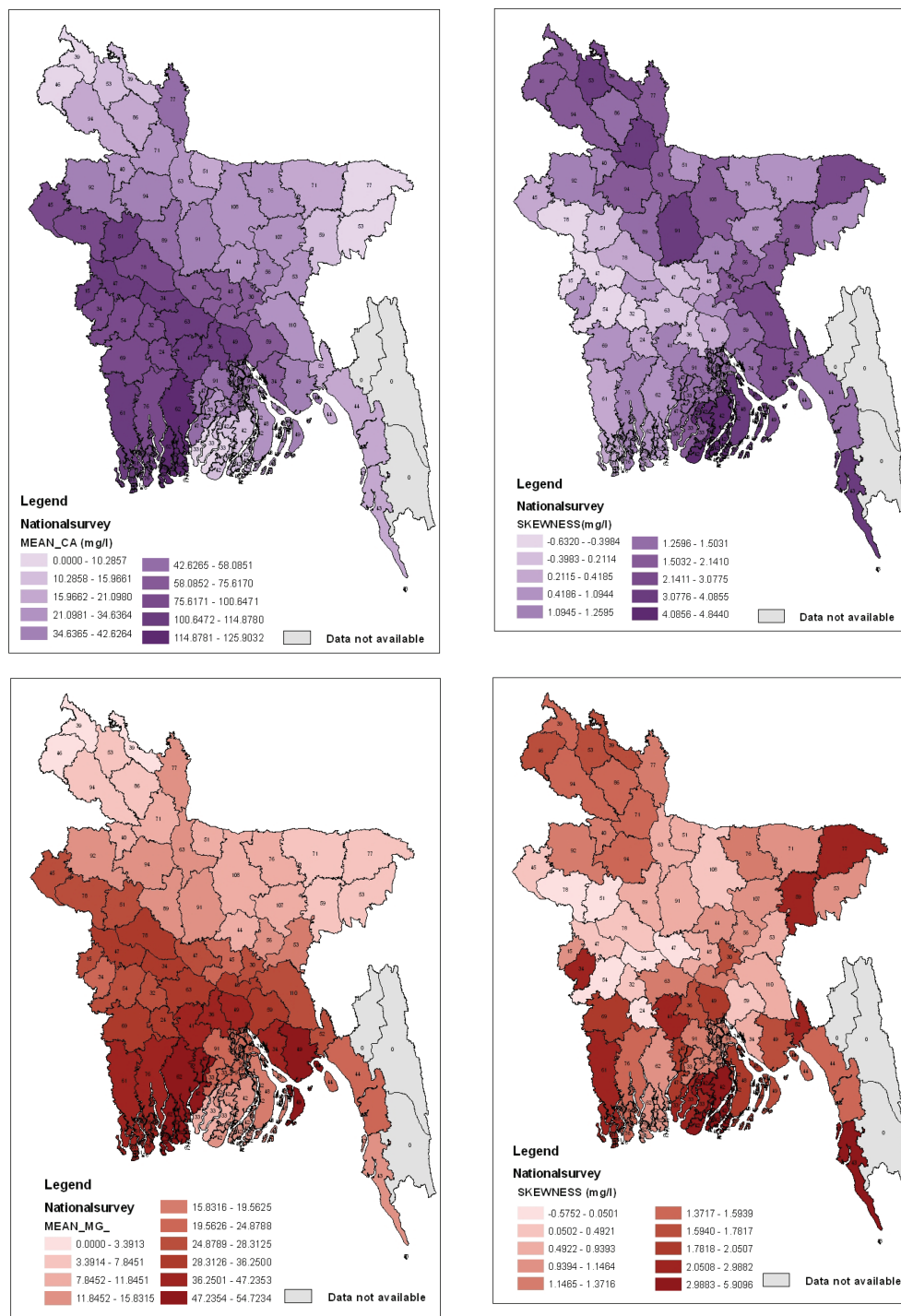


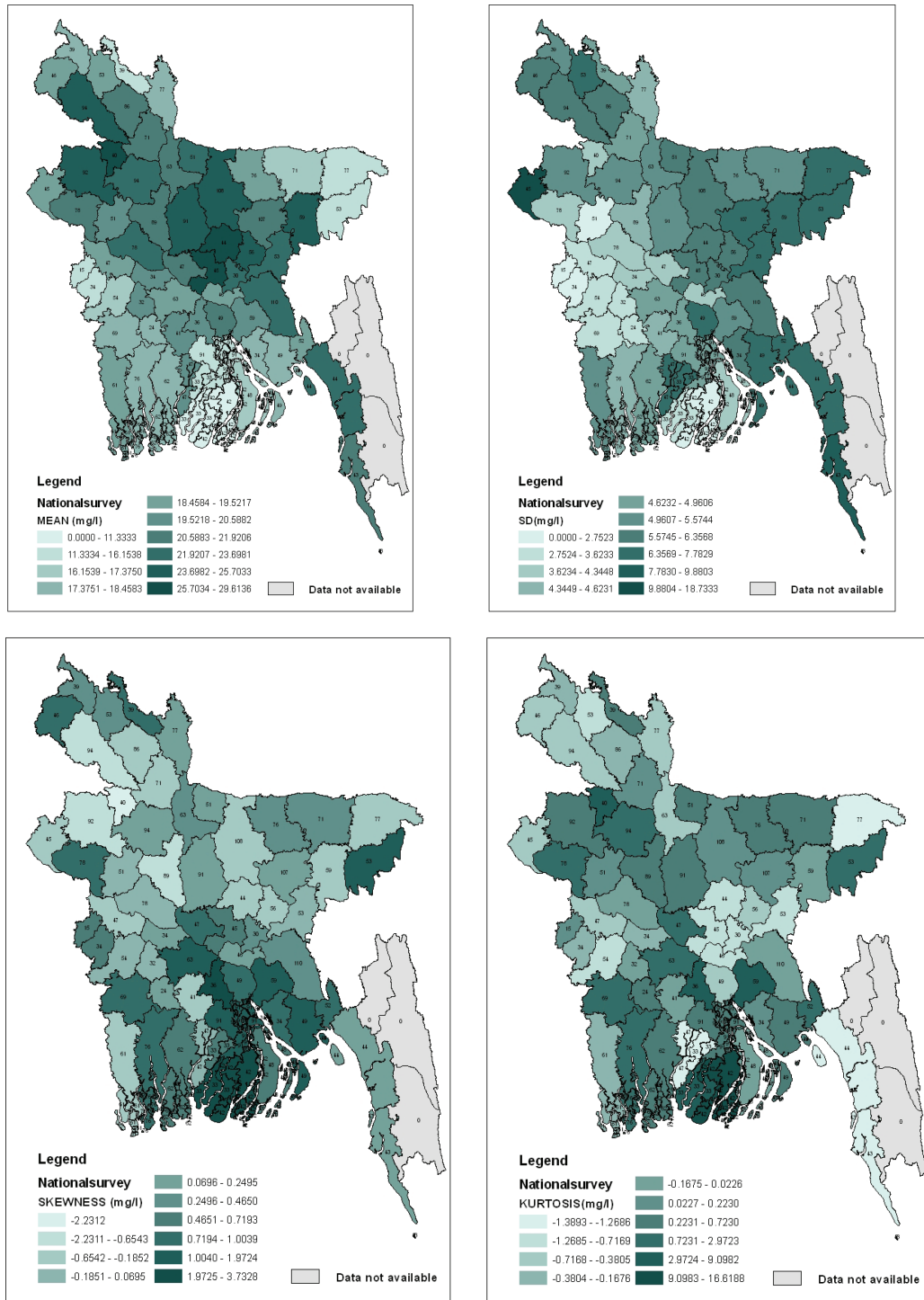
Figure 4. Univariate statistical behavior of Ca (top panels) and Mg (bottom panels) in terms of: 1) mean (left panels), and 2) skewness (right panels).

that administrative unit. For example, the computation of the mean or the standard deviation yielded one singular value reported for each district based on an average of approximately 40–100 data points. The sensitivity of these statistical analyses to sample size, while no doubt important, could not be evaluated due to the inherent paucity of data.

**Results and Discussion**

*Univariate Analyses*

As part of univariate analyses, we first show the statistical behavior of AS concentration in space in terms of the four moments: mean, standard deviation, skewness, and kurtosis (Figure 2).



**Figure 5.** Univariate statistical behavior of Si in terms of: 1) mean (top left), 2) standard deviation (top right), 3) skewness (bottom left), and 4) kurtosis (bottom right).

260 The mean AS concentration (Figure 2, top left panel) indicates  
 the already-known phenomenon that AS contamination is geo-  
 logically dependent, with the highest and lowest concentrations  
 abounding in the Holocene southcentral and Pleistocene North-  
 west regions, respectively. However, a look at the standard  
 265 deviation reveals that slightly more refined picture where the vari-

ability is highest along the river flood plains of the Brahmaputra  
 and Ganges rivers (Figure 2, top right panel). Although the skew-  
 ness and kurtosis maps appear qualitatively similar, we observe  
 that the distribution of AS concentration is symmetric usually  
 in regions of very high mean concentration (Figure 2, compare  
 270 bottom left panel with top right panel), while the regions of low

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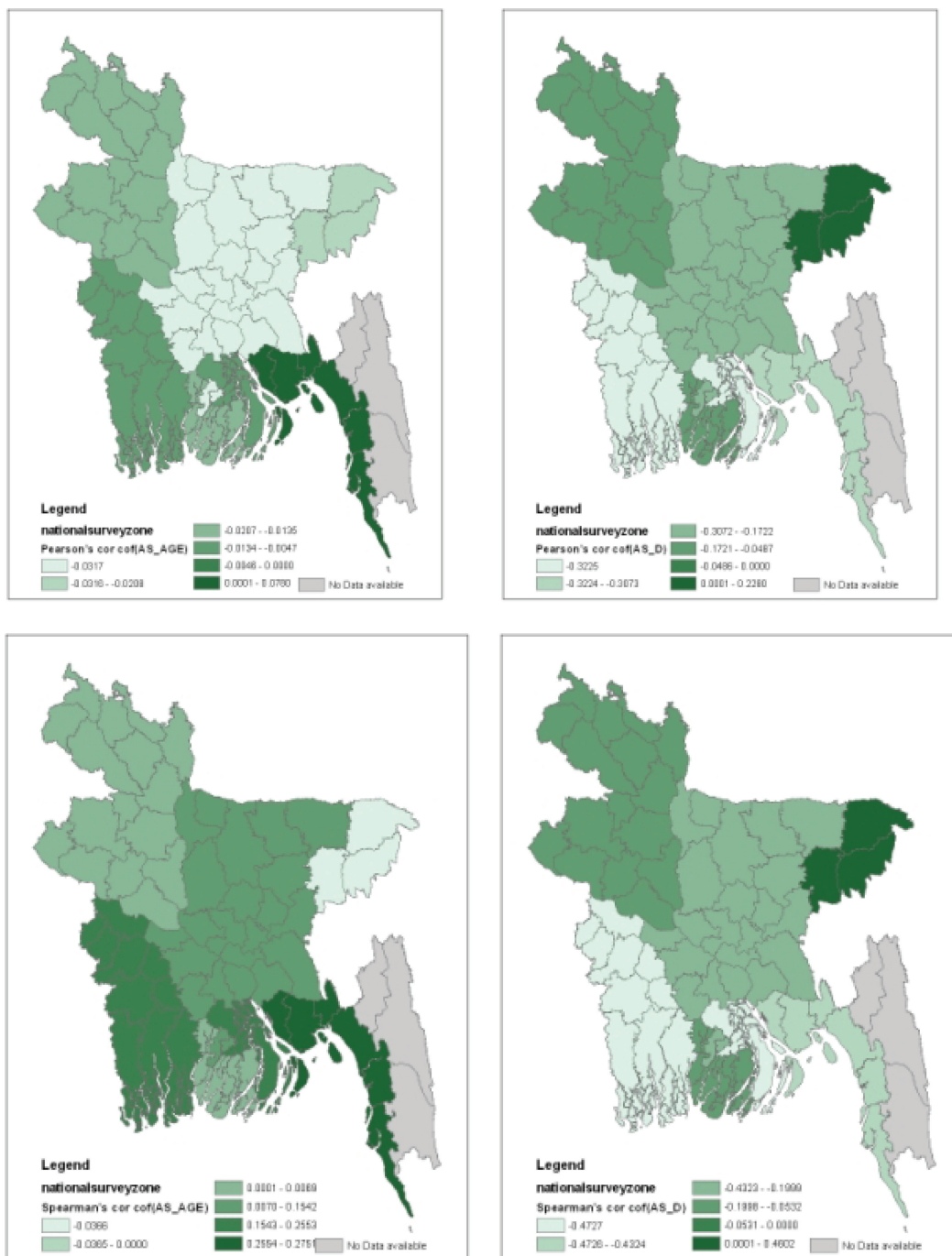


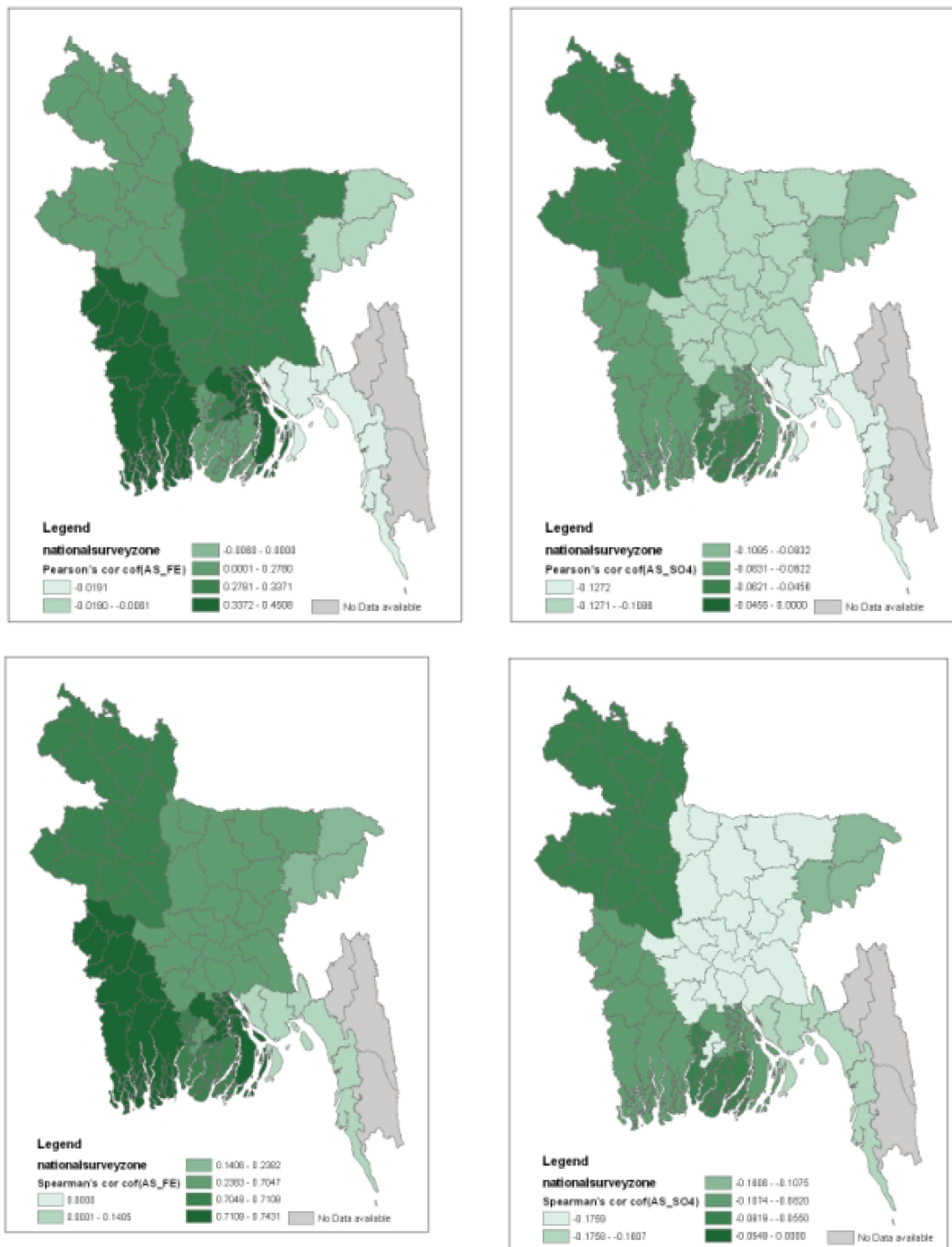
Figure 6. Bivariate statistical behavior of arsenic with age (left panels) and depth (right panels) of wells in terms of: 1) Pearson's correlation coefficient (top panels), and 2) Spearman's rank correlation (bottom panels).

and moderate contamination usually reveal a highly skewed distribution. This indicates the need for data transformation in those regions prior to the use of spatial mapping techniques such as kriging (Hossain et al., in press). In another regard, the AS distribution tends to flatten in regions of high contamination. This may be an indication of the presence of a uniform distribution.

Figure 3 shows the univariate statistical behavior of Fe in groundwater. This parameter is physically important in further-

ing our understanding of AS release mechanism hypothesis based on oxidation–reduction. In general, the mean Fe concentration appears to be high in regions with high AS concentration (Figure 3, top left panel) although there is no overwhelming similarity observed. However, the spatial map of skewness and kurtosis appears very similar to that for AS (Figures 2 and 3, compare bottom panels). This indicates that Fe concentration data may require similar kind of transformation as





**Figure 7.** Bivariate statistical behavior of AS with Fe (left panels) and sulphate (right panels) of wells in terms of: 1) Pearson's correlation coefficient (top panels), and 2) Spearman's rank correlation (bottom panels).

AS prior to application of geostatistical or nonlinear mapping techniques.

290 The mean value of Ca and Mg concentrations shows a strong spatial similarity (Figure 4, left panels) in the southwest region of Bangladesh. This spatial similarity is also reflected in terms of skewness of the data (Figure 4, right panels), although the kurtosis values were not found to be as similar (not shown herein). We

infer that the mapping of AS concentrations at nonsampled loca- 295  
 tions on the basis of multivariate factorial or regression models may not require the dual presence of both Ca and Mg; hence, one of them is redundant. We failed to observe any tangible behavior in the statistical behavior of Na and Zn in space and hence they are not reported herein. Finally, Figure 5 shows the univariate 300  
 statistical behavior of the parameter Si, which is as important

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as Fe in the analyses of hypotheses of AS release. Although the mean Si concentration appears to exhibit little similarity with AS concentration, an interesting pattern emerges on the univariate statistical behavior. Both the skewness and kurtosis of the distribution appear very similar yet highly heterogeneous in space. This may be an indication that Si data will require careful transformation prior to any nonlinear or multivariate modeling only in the smaller scale but not for country-wide/regional assessment (Figure 5).

*Bivariate Analyses*

Based on preliminary univariate analyses (reported in the preceding section), we performed covariance analyses among the following pairs of parameters: 1) AS with age of wells; 2) AS with depth of wells; 3) AS with Fe; and finally 4) AS with sulphate. Furthermore, we present our bivariate findings in terms of correlation statistics of Pearson's (for linear correlation) and Spearman's rank (for nonlinear correlation), and finally in terms of administrative regions of Bangladesh. These larger administrative regions are called *divisions*.

In Figure 6, the correlation of AS with depth (right panels) and age of wells (left panels) are shown. It appears that only the northeast and southeast regions are noticeably correlated. In particular, the age of wells is only nonlinearly correlated in the southeast region while the rest of the country shows very weak correlation (Figure 6, left panels). The correlation of AS with Fe appears strongest in the southwest region (Figure 7, left panels), and this correlation appears more nonlinear (Figure 7, compare top left with bottom left panels). This possibly indicates that the oxidation–reduction hypothesis may be more appropriate as a release mechanism in the southwest region. In another regard, the sulphate concentration appears more correlated to AS in the northwest Pleistocene region of Bangladesh, suggesting that the pyrite–oxidation hypothesis may be more appropriate there (Figure 7, right panels). Overall, the Spearman's rank correlation coefficient appears to be a better alternative for screening dependent variables such as Fe and sulphate for multivariate modeling and factorial kriging.

**Conclusion**

A comprehensive forensic analysis of AS contamination in Bangladesh was undertaken using GIS, univariate and bivariate statistics. Our assessment provided the background needed for fine-tuning emerging/evolving methods of spatial mapping based on nonlinear deterministic dynamics and multivariate factorial analyses. The data were derived from the nation-wide AS survey completed in 1999 by the BGS in collaboration with the DPHE of Bangladesh. The study found that the standard deviation of AS concentration is highest along the river flood plains of the major rivers. The distribution of AS concentration was found to be symmetric in regions of very high mean concentration while the regions of low and moderate contamination reveal a highly skewed distribution. The mean value of calcium and magnesium concentrations showed a strong spatial

similarity in the southwest region of Bangladesh. This spatial similarity was also reflected in terms of skewness of the data suggesting that the mapping of AS concentrations at nonsampled locations on the basis of multivariate factorial or regression models in Bangladesh may not require the dual presence of both Ca and Mg parameters and hence, one of them is redundant. Both the skewness and kurtosis of the distribution of Si concentration suggested that Si data would require careful transformation prior to any nonlinear or multivariate modeling only in the smaller scale but not for country-wide/regional assessment. The age of wells was found to be only nonlinearly correlated in the southeast region. The correlation of AS with Fe also appeared strongest in the southwest region indicating that the oxidation–reduction hypothesis may be more appropriate as a release mechanism in the southwest region. In another regard, the sulphate concentration was found to be more correlated to AS in the northwest Pleistocene region of Bangladesh, suggesting that the pyrite-oxidation hypothesis may be more appropriate. Overall, our forensic analysis has revealed a useful knowledge base for data transformation and selection of geochemical parameters towards spatial mapping based on multivariate factorial and nonlinear deterministic models.

Our study is not without limitations. Most of the inference is qualitative, and more definitive quantification would require the use of detailed mathematical methods (such as hypothesis testing). Furthermore, this study presents mostly from a data-based perspective without formulating physically based mechanistic hypothesis. Nevertheless, we believe the forensic findings from our study can be a useful first step towards developing more emerging spatial mapping techniques in light of uncertainty that exists on probable release mechanisms and mainstream geostatistical methods for mapping. Very recent work of Lin et al. (2006), Goovaerts et al. (2005), and Hill et al. (in press) reveal the importance of prior knowledge on data transformation, distribution analysis, principal components, and dominant variables for more accurate spatial mapping techniques based on factorial or nonlinear deterministic modeling.

Another limiting aspect of the study is that we have attempted to draw regional-level conclusions without looking closely in to the effect of spatial scale. For example, an open question remains as follows: What role do the spatial resolution and sampling of contamination data play in the statistical inferences on the forensics? The paucity of quality-controlled data (approximately 50 samples per district; see section discussing study region, data, and methods) makes it impossible to investigate if statistical analyses at scales smaller than the district level would significantly alter our forensic findings. Natural extensions of this work would therefore be: 1) significance testing of the univariate and bivariate statistical measures; 2) PCA; 3) establishment of connections between geochemical parameters based on mechanistic and physically based hypothesis; and 4) investigating the role of spatial scale (with respect to geologic variability) of AS contamination data on the univariate and bivariate statistical analyses. Some work has already begun along the suggested line and we hope to report them in the near future.

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