

Investigating and mapping spatial patterns of arsenic contamination in groundwater using regression analysis and spline interpolation technique

Pham Quy Giang, Kosuke Toshiki, Shoichi Kunikane and Masahiro Sakata

ABSTRACT

The present study investigates the hypothesis that arsenic concentrations correlate with tubewell depth, and examines the effectiveness of spline interpolation, specifically completely regularized spline (CRS) and spline with tension (SWT) in estimating the magnitude of arsenic contamination in groundwater in Thanh Tri, a densely populated district located in the southern part of Hanoi City, Vietnam. Groundwater sampling conducted in 72 tubewells drilled into shallow aquifers yielded an average arsenic concentration of 82 µg/L with a maximum of 395 µg/L, far higher than the World Health Organization (WHO) guideline value of 10 µg/L. The average concentration in the lower Pleistocene aquifer was 86 µg/L, slightly higher compared with the average of 78 µg/L in the upper Holocene aquifer. Interestingly, regression analysis revealed that in the Holocene aquifer, depth of wells influenced arsenic concentrations significantly. Such an influence, however, was insignificant in the lower Pleistocene aquifer. Both the CRS and SWT spatial interpolation models resulted in plausible predictions for the arsenic concentration data. The problem of arsenic contamination in the study area should be considered seriously, as 99% of the area was estimated to be affected by arsenic levels exceeding the WHO guideline value.

Key words | arsenic, groundwater contamination, regression analysis, spline interpolation

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INTRODUCTION

Groundwater in the Hanoi area was first utilized for water consumption domestically and industrially in 1909 (Dan & Dzung 2002). It appeared to be the perfect answer to any water demand, especially drinking water. As Hanoi's population increased and its economic sectors expanded, groundwater extraction grew to meet rapidly increasing water needs, and recently groundwater has become the main water supply, with a consumption level of 780,000 cubic meters per day (MONRE 2009). However, along with this increasing water consumption, a growing problem of groundwater contamination by arsenic, one of the most dangerous contaminants, has been highlighted by numerous recent reports. The first report by Berg *et al.* (2001) presented arsenic contamination in private tubewell water samples with a peak concentration of 3,050 µg/L, and an average of

159 µg/L, far higher than the World Health Organization (WHO) guideline value of 10 µg/L. This made Hanoi a new focus of attention regarding arsenic contamination around the world, along with other countries and regions such as Bangladesh, India, Mongolia, Taiwan, Chile, Argentina, Mexico, and Hungary. According to Berg *et al.* (2001) and Postma *et al.* (2007), elevated arsenic concentration in the groundwater in the Hanoi area is likely to be naturally occurring. In addition, waste from industrial, agricultural and mining activities may have also contributed to this contamination (Thanh 2002). It is apparent that millions of people who use this groundwater untreated are at risk of chronic poisoning from these high concentrations of arsenic.

Detection of groundwater arsenic contamination and understanding of its distribution patterns are therefore

critically important to prevent the widespread use of unsafe water and protect local people from poisoning. Recently, investigations using a variety of methods have been conducted in seriously affected areas surrounding the city of Hanoi (Heimann *et al.* 2007; Postma *et al.* 2007; Larsen *et al.* 2008; Buschmann & Berg 2009; Jessen *et al.* 2009). However, the powerful and cost-effective technique of spatial interpolation, which can be used to reliably estimate the values of unsampled areas using data from areas with known values, has seldom been employed. In this study, we employed spatial interpolation and analytical statistical techniques to examine spatial patterns of arsenic contamination in groundwater. The dependence of arsenic in shallow alluvial aquifers on well-depth was determined using regression analysis. In addition, arsenic distribution was examined and mapped using spline interpolations, specifically completely regularized spline and spline with tension prediction methods based on 72 data points. These prediction maps provide a decision support tool to identify the areas where countermeasures are needed in the light of risks to local residents.

STUDY SITE AND METHODS

Study site

Thanh Tri is a suburban district located on the southeastern side of Hanoi, on the western bank of the Red River (Figure 1). It covers an area of 98.22 km² and has a total population of 241,000 (General Statistics Office of Vietnam 2009). The terrain of Thanh Tri is mostly low-lying, gradually and slightly descending towards the southeast following the flow direction of the Red River. The area is underlain by unconsolidated sediments dating from the Holocene to the Pleistocene age (Dan & Dzung 2002).

The upper Holocene aquifer consists of clayey and sandy layers with a thickness up to 10 m in its upper section, and is made up of various sands and gravel in its lower section. The average thickness of this aquifer is 13.3 m in the study area. The transmissivity of this aquifer ranges from 20 to 800 m²/day and the water level is 3–4 m below the surface (Thanh 2002). However, due to groundwater pumping, the water level is sometimes lower. Groundwater losses

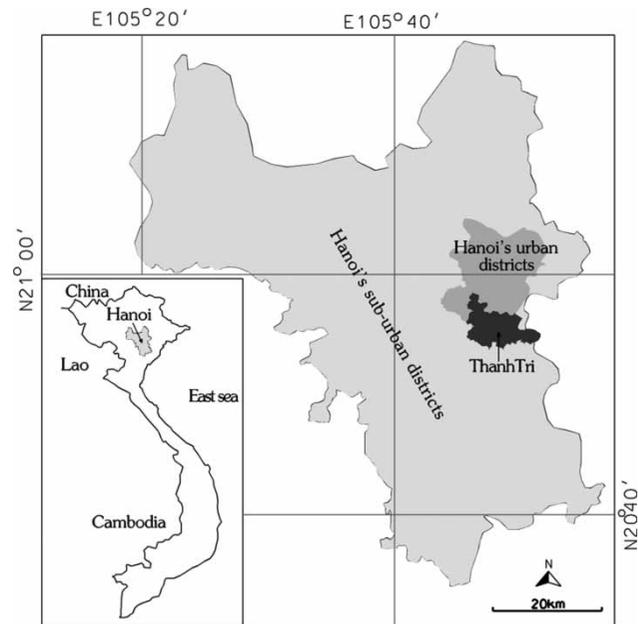


Figure 1 | Geographical location of Thanh Tri district.

usually occur in the dry season through percolation into the lower aquifers, discharge into rivers, and evaporation into the air (Dan & Dzung 2002; Thanh 2002). The groundwater in this aquifer is quite fresh. The total dissolved solids (TDS), mainly calcium carbonate, is below 0.5 g/L (maximum 0.445 g/L). In most areas, the iron concentration ranges from 0.4 to 10,000 µg/L, and manganese ranges from 200 to 2,000 µg/L. However, ammonia is very high in Thanh Tri, with a concentration up to 100,000 µg/L (Dan & Dzung 2002).

The lower Pleistocene aquifer is situated deeper in the stratigraphic sequence. It is widely distributed under the Hanoi area with abundant water volume. The depth to the top of this aquifer varies between 10 and 35 m in Thanh Tri district. However, the water level may change seasonally. The seasonal fluctuation of the groundwater head in the Pleistocene aquifer is pronounced compared with a slight change of water level in the Holocene aquifer. The Pleistocene aquifer comprises two sub-layers: the upper sub-layer is made up of medium to coarse sand mixed with gravel; the lower sub-layer consists of cobbles and pebbles mixed with sand. The thickness of the Pleistocene aquifer is from 10 to 35 m. Its transmissivity ranges from 200 to 1,600 m² per day. This aquifer is the main source of the groundwater supply for Hanoi City (Dan & Dzung 2002).

Groundwater sampling and analysis

Groundwater sampling was conducted in 72 family-based tubewells spreading over the whole study area following the systematic random sampling method (one sample in each 1 km × 1 km square block). The location of each sampling well was determined by using a hand-held global positioning system (GPS) device. Information on the depth of each tubewell was provided by the head of the household. This information is usually recorded in the drilling contract or drilling bill because the construction price is determined by the length of PVC pipe used for the tubewell installation. Generally, crystal clear water samples were collected in 500 mL plastic bottles and acidified to pH < 2 by sulfuric acid (H₂SO₄) to stimulate transformation of insoluble compounds (e.g. As₂O₃) to soluble compounds, such as H₃AsO₃ and H₃AsO₄. Finally, APHA.3500-As. Silver diethyldithiocarbamate technique (ISO 2590:1973) was employed to determine total arsenic concentration.

Application of interpolation techniques

With the main aim being to reliably estimate and visually represent the spatial distribution of arsenic, the spline interpolation technique was employed. The platform for this method was geographic information system (GIS) software package ArcGIS 9.2 and its extension, namely Geostatistical Analyst. Spline interpolation estimates the values of unsampled points using a mathematical function that minimizes overall surface curvature, resulting in a smooth surface that passes exactly through the input points.

The formula for the surface interpolation of the spline function is as follows:

$$S(x, y) = T(x, y) + \sum_{j=1}^N \lambda_j R(r_j) \quad (1)$$

where $J = 1, 2, 3, \dots, N$. N is the number of data points. λ_i are coefficients found by the solution of a system of linear equations. r_j is the distance from the point (x, y) to the j th point.

$T(x, y)$ and $R(r)$ are determined differently, depending on the selected option as shown below.

For the *regularized* option:

$$T(x, y) = a_1 + a_2x + a_3y \quad (2)$$

$$R(r) = \frac{1}{2\pi} \left\{ \frac{r^2}{4} \left[\ln\left(\frac{r}{2\tau}\right) + c - 1 \right] + \tau^2 \left[K_0\left(\frac{r}{\tau}\right) + c + \ln\left(\frac{r}{2\pi}\right) \right] \right\} \quad (3)$$

For the *tension* option:

$$T(x, y) = a_1 \quad (4)$$

$$R(r) = -\frac{1}{2\pi\varphi^2} + \left[\ln\left(\frac{r\varphi}{2}\right) + c + K_0(r\varphi) \right] \quad (5)$$

where τ^2 and φ^2 are the parameters entered at the command line. r is the distance between the point and the sample. K_0 is the modified Bessel function. c is a constant equal to 0.577215. a_i are coefficients found by the solution of a system of linear equations.

The regularized method creates a smooth, gradually changing surface with values that can lie outside the sample data range. Meanwhile, the tension method controls the stiffness of the surface according to the character of the modeled phenomenon. It creates a less smooth surface with values more closely constrained by the sample data range (ESRI 2003).

Method of model evaluation

To evaluate the prediction of each interpolation technique, the method of cross-validation (CV) was used. In this method, the interpolated results are analyzed by comparing predicted value (P_i) and measured value (M_i) for every sampling point location (n points) in order to assess systematic error, which is calculated as the mean prediction error (MPE).

$$\text{MPE} = \frac{1}{n} \sum_{i=1}^n (M_i - P_i) \quad (6)$$

The accuracy of the prediction is indicated by root mean square error (RMSE), defined as follows:

$$\text{RMSE} = \sqrt{\frac{1}{n} \sum_{i=1}^n (M_i - P_i)^2} \quad (7)$$

The closer to zero the MPE and the smaller the RMSE value, the more accurate the prediction.

RESULTS AND DISCUSSION

Magnitude of arsenic contamination

The results of the water laboratory analysis are summarized in Table 1, and illustrated in the map in Figure 2. Arsenic samples were divided into seven levels based on concentration. The first level was set under 10 µg/L, the guideline value recommended by the WHO (2001) as well as the current Vietnamese drinking water standards. The second level was set below 50 µg/L, the standard in some arsenic-contaminated countries such as India, Bangladesh, and Cambodia (Yamamura 2001).

Overall, arsenic concentration ranged from 5 to 395 µg/L, with an average of 82 µg/L, far higher than the WHO guideline value of 10 µg/L; 59 samples, accounting for 82% of the total number of samples, contained arsenic levels over 10 µg/L. Meanwhile, the proportion of samples exceeding the threshold of 50 µg/L was 51%. Compared with the results of Berg *et al.* (2001), which reported the peak and average values of arsenic concentration

in Thanh Tri district to be 3,050 and 432 µg/L, respectively, the level of arsenic contamination found in our study was considerably lower, but still far higher than the standards. Thus, our findings support those of Berg *et al.* (2001) in affirming the seriousness of the arsenic problem in the study area, and indicate that the majority of local households are most likely using unsafe water.

Spatial and geohydrological patterns of arsenic contamination

Arsenic contamination had a distinct spatial distribution within the district (Figure 2). The south-western portion, including the three communes of Ta Thanh Oai, Vinh Quynh, and Dai Ang, where three contaminated rivers, namely the Nhue, Hoa Binh, and To Lich flow through (Kikuchi *et al.* 2009), could be considered the hot-spot as all of the five highest concentrations (>300 µg/L) came from this area. Also, only one of 16 samples taken from the area contained a level of arsenic below the WHO guideline value of 10 µg/L. In addition, the commune of Van Phuc located beside the Red River was found to be highly contaminated, with the highest arsenic level exceeding 200 µg/L. In contrast, the 'cold-spot' appeared in the northernmost part of the district (Tan Trieu commune), where most samples contained arsenic levels below the guideline value. The other samples with the lowest concentration category of below 10 µg/L were found in Thanh Liet and Tam Hiep in the north and in Dong My, Duyen Ha, and Tu Hiep in the east of the district.

Family-based tubewells in Thanh Tri district are usually drilled into the two shallow aquifers described above, namely the Holocene and Pleistocene aquifers. Arsenic is present in both aquifers with averages far above 10 µg/L (Table 1). The upper Holocene aquifer yielded a level of arsenic from 5 to 317 µg/L, with an average of 78 µg/L; 86% of wells in this aquifer were found with arsenic levels over 10 µg/L. Meanwhile, the concentration in the lower aquifer (Pleistocene) varied between 6 and 395 µg/L, with a higher average of 86 µg/L. However, the ratio of wells in this aquifer containing more than 10 µg/L of arsenic was 77%.

Table 1 | Arsenic concentration

Level of As (µg/L)	All wells		Holocene		Pleistocene	
	No. of wells	%	No. of wells	%	No. of wells	%
<10	13	18	5	14	8	23
10–50	22	30	10	27	12	34
50–100	20	28	14	38	6	17
100–150	5	7	2	5	3	9
150–200	4	6	3	8	1	3
200–300	3	4	2	5	1	3
>300	5	7	1	3	4	11
Total	72	100	37	100	35	100
Min As (µg/L)	5		5		6	
Max As (µg/L)	395		317		395	
Average As (µg/L)	82		78		86	

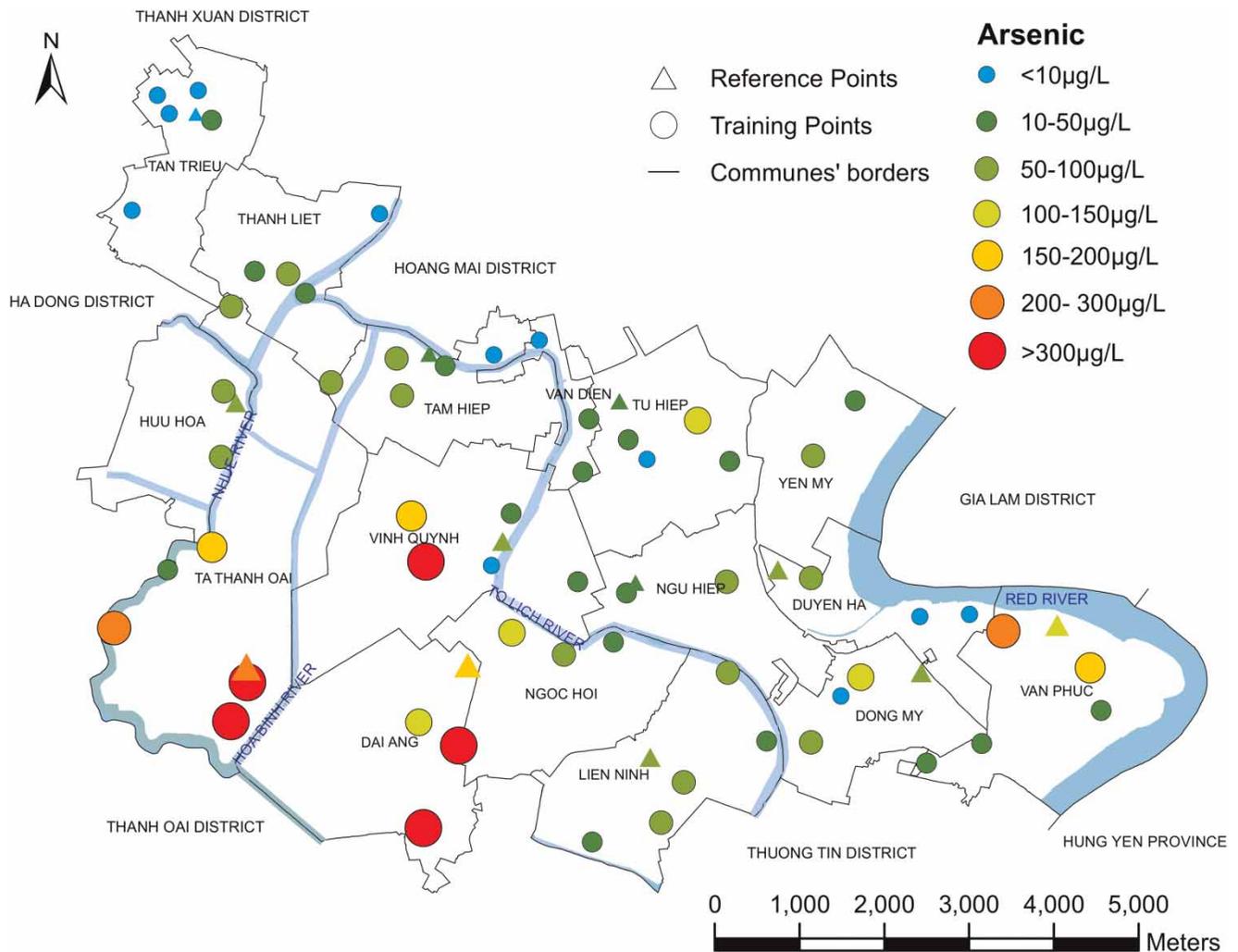


Figure 2 | Arsenic concentrations in investigated wells.

Relation between arsenic concentration and well depth

Based on a study of arsenic-contaminated groundwater in Bangladesh, Polizzotto *et al.* (2006) suggested that arsenic may be released via oxidation-reduction cycling in surface soils or sediments, and thus could subsequently be transported to well-depth through a sandy aquifer. In addition, when groundwater abstraction from a lower aquifer (Pleistocene) causes vertical drawdown of water from an upper aquifer (Holocene), arsenic mobilization becomes influenced by the process. Consequently, arsenic concentrations gradually increase in the lower aquifer. On the other hand, in a study in Red River Delta, Vietnam, Berg

et al. (2001) found that the concentration of arsenic decreased along with an increase in well depth. In relation to such findings, an important question for the present study is whether there is a relationship between arsenic level and well depth in Thanh Tri district. In order to answer this question, arsenic concentration data related to well depth was analysed statistically in SPSS software version 17.0. The curve estimation of regression function was used, with arsenic concentration as the dependent variable and well depth as the independent variable. The confidence level was set at 95% (a p value of 0.05). The results of this analysis are presented in Table 2 and Figure 3.

In the Holocene aquifer, the influence of depth on arsenic concentration was significant with a p value of 0.048 and an R^2 value of 0.173, meaning that 17.3% of the variance in arsenic concentration can be predicted from depth of wells. Thus, arsenic concentrations tended to be lower in deeper wells. In fact, seven of eight samples containing a level of arsenic higher than 100 $\mu\text{g/L}$ were found at depths of less than 25 m. This finding implies that, when considering the Holocene aquifer alone, local households that use groundwater from shallower wells may be at higher risk than those who use groundwater from deeper wells. This, however, is not applied to the Pleistocene aquifer because the influence of depth on arsenic concentration in this aquifer was insignificant ($R^2 = 0.083$, $p = 0.196$).

Spline interpolation implementation

As previously introduced, two spline interpolators, completely regularized spline (CRS) and spline with tension (SWT), were employed for mapping arsenic concentration. These are fast interpolators, but optimally calculating their parameter function is a time-consuming process. Parameters

for smoothing interpolated surfaces were automatically determined using ‘optimized parameter’ in the parameter setting interface of these methods. Thus, the optimal parameter for CRS was 0.03529, and for SWT was 0.05083. Prediction for each data point was conducted based on 15 neighbours (at least 10 neighbours).

Before generating arsenic concentration surfaces, cross-validations were carried out in order to assess prediction accuracy. Mean errors of CRS and SWT were -1.317 and -1.67 $\mu\text{g/L}$, respectively. Negative mean errors reflect the underestimation of the two methods for arsenic concentration data. This can also be seen from the error graphs (Figures 4(a) and 5(a)) where most of the predicted values over 70 $\mu\text{g/L}$ have errors below zero. The RMSE of CRS was 77 $\mu\text{g/L}$, while SWT had a slightly smaller error of 76 $\mu\text{g/L}$.

The predicted graphs showing summaries of the measured values versus the predicted values are presented in Figures 4(b) and 5(b). The thin lines are 1:1 lines, and the bold lines are best fitted lines. In good spline models, it is expected that the best fitted line as well as the scatter points are close to the 1:1 line. The predicted graphs for

Table 2 | Model summary for testing dependence of arsenic concentration on well depth

Equation		Model summary					Parameter estimates	
		R square	F	df1	df2	Sig.	Constant	b1
Linear	Holocene	0.173	1.200	1	35	0.048	161.630	-3.374
	Pleistocene	0.083	0.009	1	33	0.196	108.234	-0.521

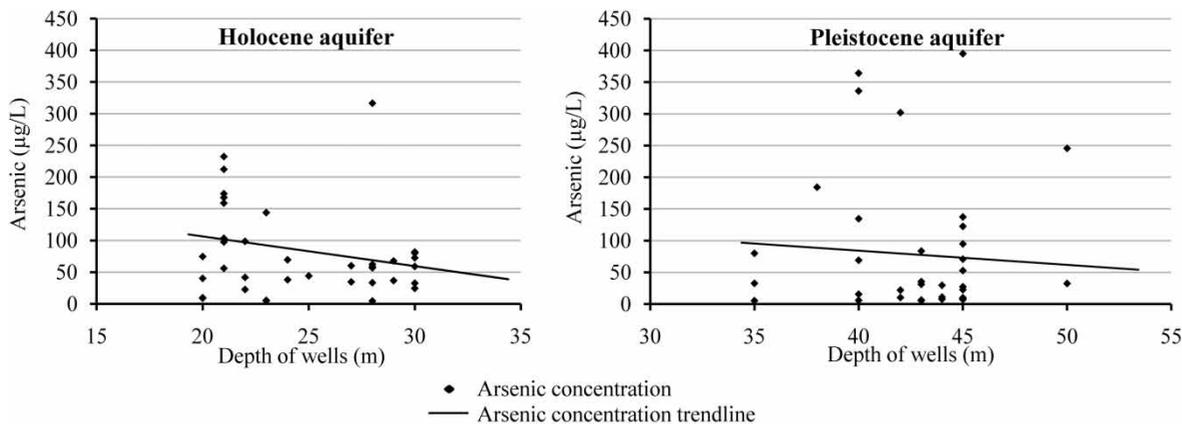


Figure 3 | Trendlines of arsenic concentration in specific aquifers with increase in well depth.

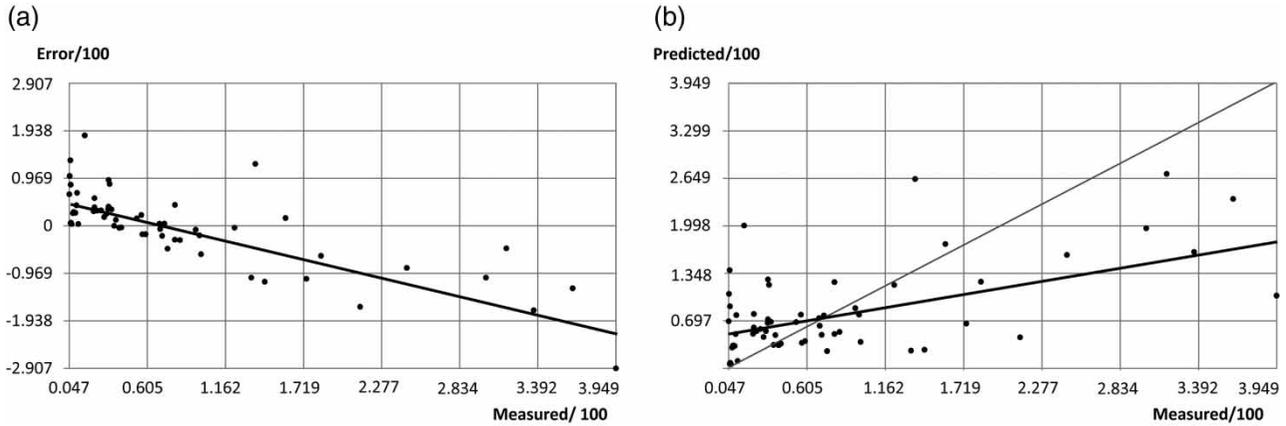


Figure 4 | Cross validation of CRS: (a) error graph; (b) predicted graph.

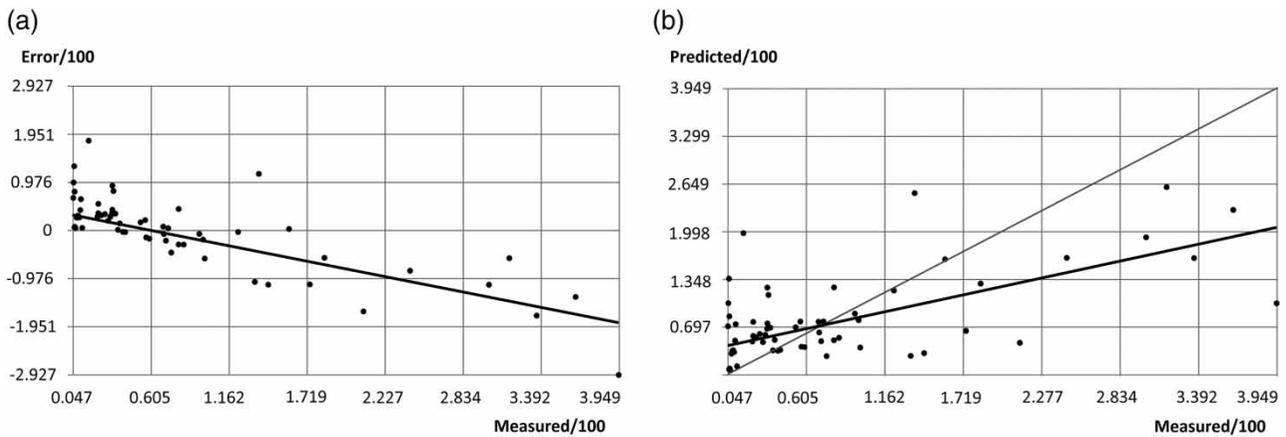


Figure 5 | Cross validation of SWT: (a) error graph; (b) predicted graph.

CRS and SWT are similar. The fitted line gets further from the 1:1 line as arsenic values increase, indicating that the higher the arsenic values are, the lower the autocorrelation they perform. In addition, we can conclude that these two methods made better predictions for lower values than for higher ones, as the arsenic values of less than about 100 $\mu\text{g/L}$ are close to the 1:1 line, while the higher values are located further apart.

It is also important to acknowledge that some points have considerable errors. This is due to the fact that spatial interpolation, and spline in particular, is more accurate when the sample density is greater (Isaaks & Srivastava 1989; Englund *et al.* 1992; Stahl *et al.* 2006) and sampling points are distributed evenly (Collins & Bolstad 1996). In

the present work, to ensure an even sample distribution, we determined sample locations based on the systematic random sampling method (one sample was selected randomly in each 1 km \times 1 km square block). However, tubewells were only available in residential areas, and not in paddy fields or water bodies, which occupied large parts of the study area. Therefore, some square blocks had no tubewell for sampling. This insufficiency made samples sparse and unevenly distributed, which resulted in less accurate predictions.

The interpolated surfaces produced by CRS and SWT are very similar, except in some small areas (Figure 6 and Table 3). According to such interpolations, more than 99% of the district is contaminated by arsenic levels above the

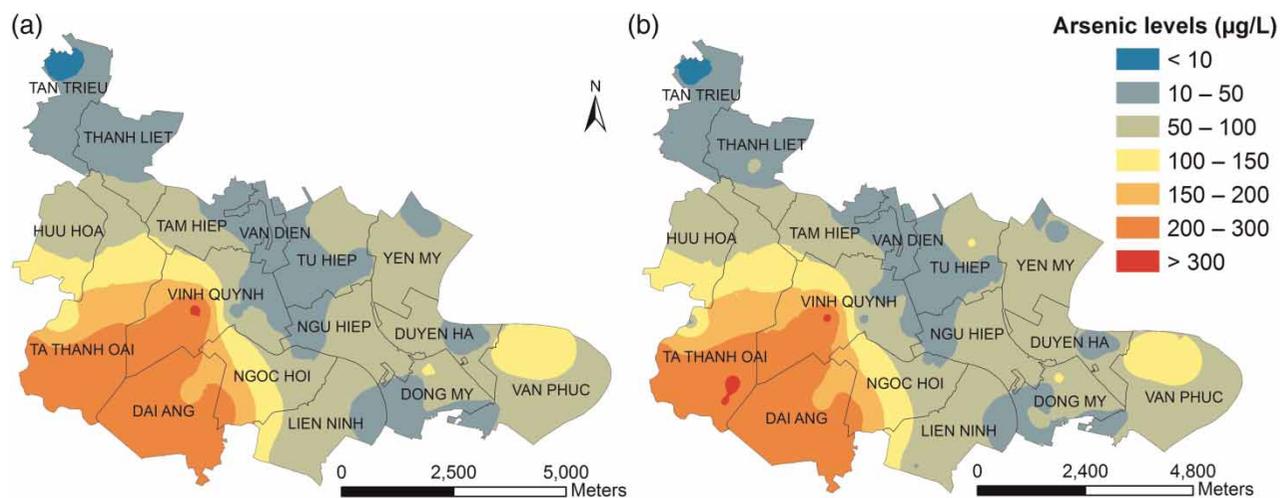


Figure 6 | Surface of arsenic concentration produced by CRS (a) and SWT (b).

Table 3 | Delineated area under different arsenic levels according to predictions of CRS and SWT

Arsenic levels ($\mu\text{g/L}$)	CRS		SWT	
	Area (ha)	% of area	Area (ha)	% of area
<10	45.99	0.72	34.11	0.54
10–50	1,596.93	25.12	1,464.80	23.04
50–100	2,539.74	39.95	2,708.04	42.59
100–150	733.30	11.53	720.89	11.34
150–200	397.51	6.25	417.71	6.57
200–300	1,040.22	16.36	995.19	15.65
>300	4.35	0.07	17.30	0.27
Total	6,358.04	100	6,358.04	100

WHO threshold. The small safe area which has arsenic concentrations below $10 \mu\text{g/L}$ is located in the northernmost part of the district (a part of the commune of Tan Trieu). In general, high levels of arsenic concentrate in the south-west, decreasing gradually towards the other parts of the study area.

Comparison of models

To compare the performance of CRS and SWT, the symmetric mean absolute percentage error (SMAPE) method was employed using 12 extra reference points, locations and arsenic levels of which were displayed in Figure 2.

This process assesses each method by comparing their SMAPE values calculated as follows:

$$\text{SMAPE} = \frac{1}{N} \sum_{i=1}^N E_i \quad (8)$$

where

$$E_i = \frac{|M_i - P_i|}{M_i + P_i} * 100(\%) \quad (9)$$

where P_i is the predicted value, and M_i is the measured value of arsenic concentration at reference point i . The detail of the comparison is presented in Table 4.

Both CRS and SWT yielded low values of SMAPE, 7% and 6%, respectively. This result is similar to that of cross-validation, where the RMSE of SWT was slightly smaller than that of CRS. However, the predictions from both methods presented here are reliable as their errors were relatively insignificant.

CONCLUSION

Arsenic contamination of water resources has become an important environmental issue and human health concern in the city of Hanoi. This study utilized statistical and interpolation techniques to analyze the geographical and

Table 4 | Prediction errors at reference points

Reference point	CRS			SWT	
	M_i ($\mu\text{g/L}$)	P_i ($\mu\text{g/L}$)	E_i (%)	P_i ($\mu\text{g/L}$)	E_i (%)
P ₁	36	46	12	45	11
P ₂	68	66	1	64	3
P ₃	60	62	2	63	2
P ₄	168	177	3	174	2
P ₅	62	70	6	71	7
P ₆	58	53	5	60	2
P ₇	44	48	4	49	5
P ₈	82	75	4	75	4
P ₉	233	300	13	288	11
P ₁₀	8	10	11	11	16
P ₁₁	10	12	9	10	0
P ₁₂	103	121	8	113	5
SMAPE		7%		6%	

geological patterns of arsenic in the two shallow Holocene and Pleistocene groundwater aquifers which are important sources of residential water supply in the city. More than 80% of the tubewells investigated were found to be contaminated by arsenic levels exceeding the WHO guideline value of 10 $\mu\text{g/L}$. In addition, contaminated areas with arsenic level over 10 $\mu\text{g/L}$ occupied 99% of the studied district.

The interpolation techniques and the statistical model of estimation curve used in this study were found to be efficient approaches to help explore and remedy arsenic contamination in Hanoi as they provide plausible projection and visual representation of arsenic distribution patterns. These techniques provide a first step towards helping environmental professionals prioritize their limited budgets for improving field investigation strategies, implementing groundwater cleanup programs, and designing long-term effective countermeasure systems.

ACKNOWLEDGEMENTS

The authors would like to thank Philip Hawke, lecturer of the Graduate School of Integrated Pharmaceutical and Nutritional Sciences at the University of Shizuoka for his advice on the English in the manuscript.

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First received 2 August 2012; accepted in revised form 21 June 2013