Predicting Geogenic Arsenic Contamination in Shallow Groundwater of South Louisiana, United States

Ningfang Yang,*† Lenny H. E. Winkel,‡§ and Karen H. Johannesson†

†Department of Earth and Environmental Sciences, Tulane University, 101 Blessey Hall, New Orleans, Louisiana 70118, United States
‡Eawag, Swiss Federal Institute of Aquatic Science and Technology, Überlandstrasse 133, P.O. Box 611, 8600 Dübendorf, Switzerland
§Institute of Biogeochemistry and Pollution Dynamics, ETH Zurich, Universitätstrasse 16, 8092 Zurich, Switzerland

Supporting Information

ABSTRACT: Groundwater contaminated with arsenic (As) threatens the health of more than 140 million people worldwide. Previous studies indicate that geology and sedimentary depositional environments are important factors controlling groundwater As contamination. The Mississippi River delta has broadly similar geology and sedimentary depositional environments to the large deltas in South and Southeast Asia, which are severely affected by geogenic As contamination and therefore may also be vulnerable to groundwater As contamination. In this study, logistic regression is used to develop a probability model based on surface hydrology, soil properties, geology, and sedimentary depositional environments. The model is calibrated using 3286 aggregated and binary-coded groundwater As concentration measurements from Bangladesh and verified using 78 As measurements from south Louisiana. The model’s predictions are in good agreement with the known spatial distribution of groundwater As contamination of Bangladesh, and the predictions also indicate high risk of As contamination in shallow groundwater from Holocene sediments of south Louisiana. Furthermore, the model correctly predicted 79% of the existing shallow groundwater As measurements in the study region, indicating good performance of the model in predicting groundwater As contamination in shallow aquifers of south Louisiana.

INTRODUCTION

Arsenic (As) is a highly toxic and carcinogenic metalloid and is among the few elements that can be mobilized at common groundwater pH values (i.e., 6.5 ≤ pH ≤ 8.5) under both oxidizing and reducing conditions.1 Long-term consumption of As-contaminated groundwater can cause serious health effects, such as increased risk of cancers (e.g., skin, lung, bladder, and kidney),2 infant mortality,3 and reduced intellectual and motor function in children.4,5 At present, more than 140 million people worldwide are drinking As-contaminated groundwater (i.e., As ≥ 10 μg/L),6 and the most severely affected region is the Ganges–Brahmaputra–Meghna (GBM) delta in Bangladesh and India, which has, therefore, been the focus of substantial investigation.7–10 Geogenic processes are the main cause for the locally elevated As concentrations in the groundwater from the GBM delta.6–10 Aquifers with elevated groundwater As concentrations commonly consist of Holocene riverine and deltaic sediments that are under reducing conditions, poorly drained, and associated with high levels of organic matter content.6,7,10,11 Therefore, it is reasonable to expect that other areas that share similar combinations of these geologic, geochemical, and hydrological characteristics may also be at risk of groundwater As contamination.

The Mississippi River (MR) delta in south Louisiana, U.S.A., represents a large river and delta complex that shares broadly similar characteristics to the GBM delta. For example, shallow aquifers in the MR delta are composed of relatively young (e.g., Holocene), organic-rich (e.g., peat), alluvial/deltaic sediments,12 all of which are considered important factors in
controlling the spatial distribution of groundwater As contamination.\textsuperscript{1,13,14} Groundwater As contamination has generally not been reported in the MR delta region in south Louisiana, which likely reflects the fact that the chief drinking water sources for local major population centers (i.e., New Orleans and Baton Rouge) are surface water from the Mississippi River and deep groundwater, respectively. Nonetheless, a recent study reported high As concentrations in shallow groundwater from alluvial aquifers of the Mississippi River valley in southeast Arkansas.\textsuperscript{15} Our preliminary data (Supporting Information (SI) Table S1) show that shallow groundwater As concentrations in south Louisiana range from less than \(10 \mu g/L\) to as high as \(100 \mu g/L\), which demonstrates the possibility that shallow groundwater in south Louisiana is at risk of As contamination. It is important to note that historic application of arsenical pesticides to control the bow weevil in cotton-growing regions of the Gulf south (e.g., Arkansas, Texas, north and east Louisiana) complicates the interpretation of the source of the high As groundwater in southeast Arkansas and possibly south Louisiana. Most of the region of south Louisiana in this contribution is dominated by sugar cane, rice, and crayfish (\textit{Procambarus clarkii}) cultivation and consequently does not belong to the historic cotton belt. Therefore, in this contribution we are primarily interested in identifying potential As-contaminated shallow groundwater caused by natural sources and processes under reducing shallow aquifers with flat topography and low hydraulic gradients in the MR delta in south Louisiana. Specifically, logistic regression is applied to develop a risk model using surface hydrology, soil properties, geology, sedimentary depositional environments, and groundwater As concentration data from Bangladesh. The model is then applied to south Louisiana where negligible As concentration data are available to predict the probability of shallow groundwater As contamination from natural sources and processes in south Louisiana. This model can guide scientists and government agencies in targeting the high risk areas, initiating early protection measures, and hence, preventing chronic As poisoning in local populations.

\section*{METHODS}

\textbf{Groundwater Arsenic Data.} Because As concentration data for shallow groundwater from south Louisiana are sparse, we employed data from Bangladesh to build and calibrate the model. Groundwater As concentration data from Bangladesh were obtained from the British Geological Survey (BGS) and the Department of Public Health Engineering (DPHE),\textsuperscript{16} which consisted of 3448 As measurements from shallow aquifers (<100 m). Arsenic concentration data of south Louisiana consisted of 78 As measurements from shallow aquifers (<50 m), of which 55 data points were compiled from the United States Geological Survey (USGS)\textsuperscript{17} and 23 samples were measured as part of the current study. The details on groundwater sampling, preservation, As analysis, and the analytical results are described in the Supporting Information (Method). The As data from south Louisiana were used to validate the model.

\textbf{Independent Variables.} On the basis of previous investigations,\textsuperscript{13,14,18–21} a total of 15 independent variables, which were considered potentially closely related to groundwater As contamination, were initially compiled for the logistic regression analysis (SI Table S2). Due to differences in resolution and data format, the 15 variables (SI Table S3) were uniformly converted to a raster format at 1 km resolution using ArcGIS (Version 10) and then classified into the following four categories.

\textbf{Geology.} Detailed geologic maps of Bangladesh and south Louisiana were obtained in digital format from the USGS and the Louisiana Geological Survey, respectively (SI Table S2). On the basis of geology and sedimentary depositional environments, the data provided on the geologic maps were reclassified into five categories, including pre-Holocene deposits, deltaic deposits, organic-rich deposits, alluvial deposits, and tidal deposits (Figure 1).

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure1.png}
\caption{Classified geologic maps of Bangladesh (a) and south Louisiana (b).}
\end{figure}
Shallow groundwater As contamination. 13, 14, 18 and investigation, including predicting the probability of As concentrations in shallow groundwater. 19 and were previously shown to be strong predictors of elevated (i.e., 1 km × 1 km), and distance to rivers is the shortest distance from the center of each raster cell to rivers. Both variables can influence hydrological processes within aquifers and were previously shown to be strong predictors of elevated As concentrations in shallow groundwater. 19

Development of the Arsenic Prediction Model. Groundwater As concentrations are point measurements at different well depths, whereas the other independent variables are either vector or raster variables with coarser spatial resolution. Because the 15 independent variables (SI Table S3) were all converted to a raster format with a pixel size of 1 km, the point measurements of As concentrations were also aggregated using the geometric mean to a resolution of one point per pixel with a pixel size of 1 km. Data aggregation reduced the data set of Bangladesh to 3286 individual values, which were binary-coded for later analysis using the World Health Organization drinking water standard for As (i.e., 10 μg/L) as the threshold. The aggregated data were binary-coded because our goal is to develop a model predicting the probability of groundwater As contamination, such that there are only two possibilities: (1) contaminated (As ≥ 10 μg/L), binary-coded as 1); or (2) uncontaminated (As < 10 μg/L, binary-coded as 0). These binary-coded variables are the dependent variables in the study.

Logistic regression is a probabilistic statistical method, which can be used to predict binary outcomes from various independent variables (i.e., numerical or categorical). 22 Logistic regression has been widely used in environmental assessment and investigation, including predicting the probability of groundwater As contamination. 15, 14, 18–21 In this contribution, logistic regression is applied to develop a risk model based on the relative importance of geology and sedimentary depositional environments, topography, hydrology, and soil properties in exerting controls on groundwater As concentration in Bangladesh. The model is subsequently applied to the MR delta in south Louisiana, where little information exists regarding the possibility and geographic extent of As contamination of shallow groundwater by natural sources. The statistical significance of each independent variable in predicting the probability of As contamination in shallow groundwater is tested using stepwise logistic regression. The stepwise regression begins with adding the most significant independent variable to the model. After testing the abilities of the remaining independent variables in explaining the remaining model error, the next most significant independent variable is added. This procedure is repeated until no other independent variables left can significantly explain the remaining model error. Ultimately, only the statistically significant independent variables are included in the final model. The dependent variable is the natural logarithm of the odds, p/(1 – p), where p is a numerical value representing the probability of the occurring event. The log transformation converts the dependent variable constrained between 0 and 1 into a continuous variable that is linearly related to the combination of the independent variables, such that

\[ Y = \ln(\text{odds}) = \ln\left(\frac{p}{1-p}\right) = C + \sum_{i=1}^{n} \lambda_i X_i \] (2)

Here, Y is the log transform of the dependent variables, p/(1 – p), which is related to the odds of occurrence of an event, C is the intercept of the regression, and $\lambda_i$ are the weighting coefficients of each independent variable, X. 22

Once the model is developed, its predictive performance under different probability cutoff values is evaluated using the receiver operating characteristic (ROC) analysis. The ROC curve describes the predicative performance of the logistic regression model as the probability cutoff value varies. The optimal probability cutoff value is a value at which the model obtains best predicative performance, which is used to classify the probability map into a risk map showing areas that are predicted to be at risk of groundwater As contamination and regions that are not likely to be at risk. It is critical to note that a good fit of the regression model does not necessarily mean a good predictive performance of the model. Consequently, the best way to validate the predictive ability of the model is to use a separate data set that is not employed in developing the model. 23 In this study, groundwater As data from Bangladesh (n = 3286) were used to build the model, whereas data obtained from south Louisiana (n = 78) were used for validation of the model. Because the current amount of groundwater As concentration data for south Louisiana is sparse, full validation of the predicative ability of the model for this region awaits additional planned groundwater sampling and analysis.

## RESULTS

Arsenic Prediction Model. Statistical significance of each independent variable was tested at a 95% confidence level using the stepwise logistic regression. The results (SI Table S3) showed that only 7 of the 15 compiled independent variables were significant in predicting As-contaminated shallow groundwater in Bangladesh (Table 1) and therefore were retained in the final model to evaluate the probability of shallow groundwater As contamination in south Louisiana. The
concentrations greater than or equal to 10 μg/L (true positive), classically the area under the curve (AUC) is a measure of ability to correctly classify samples in the group with As concentrations ≥10 μg/L. Specificity is the correctly classified samples in the group with As concentrations <10 μg/L. The optimal probability cutoff value is obtained at the point where sensitivity and specificity intersects.

As prediction model was evaluated using ROC analysis. The performance of the model at different probability cutoff. Sensitivity is the correctly classified samples in the group with As concentrations ≥10 μg/L. Specificity is the correctly classified samples in the group with As concentrations <10 μg/L. The optimal probability cutoff value is obtained at the point where sensitivity and specificity intersects.

Figure 2. Performance of the model at different probability cutoff. Sensitivity is the correctly classified samples in the group with As concentrations ≥10 μg/L. Specificity is the correctly classified samples in the group with As concentrations <10 μg/L. The optimal probability cutoff value is obtained at the point where sensitivity and specificity intersects.

stepwise logistic regression indicates that pre-Holocene deposits (λ = −3.42) and distance to rivers (λ = −1.86) are inversely correlated to shallow groundwater As contamination (As ≥ 10 μg/L), of which, pre-Holocene deposits play the most important role in the model. In contrast, Holocene sedimentary depositional environments contribute positively to the model, of which, deltaic deposits (λ = 1.62) play the most important role, followed by organic-rich deposits (λ = 0.82), and then recent alluvial deposits (λ = 0.51) in terms of relative importance for predicting shallow groundwater As contamination in Bangladesh. Of the soil variables, medium-textured soil (λ = 1.16) and clay content (λ = 0.11) of the top 1 m soil layer are positively correlated to shallow groundwater As contamination, meaning that both of these parameters are important indicators of elevated As concentrations in shallow groundwater in Bangladesh.

**Probability Map and Risk Map.** The performance of the As prediction model was evaluated using ROC analysis. Specifically, the area under the curve (AUC) is a measure of the model’s performance, with values typically varying between 0.5 and 1. Despite the fact that our model only considers surface parameters (i.e., 2D data sets and not 3D data sets that include the vertical dimension), the AUC value of 0.76 (SI Figure S1) indicates that the model does a good job at predicting groundwater As contamination for shallow aquifers (<100 m) in Bangladesh. Sensitivity measures the model’s ability to correctly classify groundwater samples with As concentrations greater than or equal to 10 μg/L (true positive), whereas specificity measures the model’s ability to correctly classify groundwater samples with As concentrations less than 10 μg/L (true negative). Classification results of the As prediction model under different probability cutoff values are presented in Figure 2, which shows that sensitivity is inversely related to specificity. To obtain a model with better predicative performance, a value of 0.5, where the model has both higher sensitivity and specificity, is selected as the optimal probability cutoff value to classify the As probability map into the binary risk map.

The map showing the predicted probability of groundwater As contamination in Bangladesh is presented in Figure 3. Figure 3a shows that areas at high-risk (probability ≥0.5) of As contamination are predicted to occur along the three major rivers (Ganges, Brahmaputra, and Meghna; 0.5–0.7) with alluvial deposits, in the northeast Sylhet basin (0.6–0.8) with organic-rich deposits, and in the Ganges delta (>0.7) with deltaic and organic-rich deposits (Figure 1a). Figure 3b presents the As risk map of Bangladesh, which shows areas at high-risk (probabilities ≥0.5) and low-risk (probabilities <0.5) of groundwater As contamination. The model predicted high- and low-risk areas are in good agreement with the known spatial distribution of groundwater As contamination. Of the 3286 aggregated and binary-coded As concentration measurements, 68% are correctly predicted by the logistic regression model. More specifically, 73% of the groundwater samples with As concentrations greater than or equal to 10 μg/L are correctly predicted (sensitivity), whereas 65% of the groundwater samples with As concentrations less than 10 μg/L are correctly predicted (specificity). Consequently, below we apply the same model and approach to south Louisiana to evaluate the probability that elevated As concentrations in shallow groundwater may also characterize the region including the MR delta. Again, the existing As concentration data for shallow groundwater in south Louisiana are rare, making the logistic regression an appropriate approach to evaluate the possibility of naturally sourced As contamination of local groundwater. Moreover, it is important to stress that the model does not allow us to predict shallow groundwater in south Louisiana that may be at risk of anthropogenic-sourced As contamination.

**Application of the Arsenic Prediction Model to South Louisiana.** Figure 4 shows the results of the model predictions applied to south Louisiana. The probability map (Figure 4a) shows that regions of south Louisiana where shallow groundwater (i.e., < 50 m) exhibit a high probability (i.e., probability ≥0.5) of having elevated As concentrations. These regions include areas underlain by alluvial deposits along the Mississippi and Atchafalaya rivers (0.6–0.8), deltaic deposits in the lower MR delta (>0.8), and organic-rich marshes along the coast in extreme south Louisiana (>0.8) (Figure 1b). The

| Table 1. Significant Independent Variables Used in the Final Logistic Regression Modela |
|---------------------------------|-----------------|-----------------|--------------|
| **category**                    | **variables**   | **λ**           | **Ward-value** | **p-value**  |
| hydrology                       | distance to rivers | −1.86           | 124.56        | <0.001       |
| geology — sedimentary depositional environments | pre-Holocene deposits | −3.42           | 55.49         | <0.001       |
|                                 | deltaic deposits | 1.62            | 50.83         | <0.001       |
|                                 | organic-rich deposits | 0.82           | 12.41         | <0.001       |
|                                 | alluvial deposits | 0.51            | 6.25          | 0.0125       |
| soil                            | medium-textured soils | 1.16           | 45.50         | <0.001       |
|                                 | clay content     | 0.11            | 41.26         | <0.001       |
|                                 | intercept        | −4.11           | 44.63         | <0.001       |

Weighting coefficients (λ) of the independent variables were used in the model to calculate probabilities of shallow groundwater As contamination. Large λ values represent greater relative importance, small values represent lower relative importance, positive values represent positive correlation, and negative values represent negative correlation. The model predicted high-risk areas in the Ganges delta (>0.8) with deltaic and organic-rich deposits (Figure 1a). Figure 3b presents the As risk map of Bangladesh, which shows areas at high-risk (probabilities ≥0.5) and low-risk (probabilities <0.5) of groundwater As contamination. The model predicted high- and low-risk areas are in good agreement with the known spatial distribution of groundwater As contamination. Of the 3286 aggregated and binary-coded As concentration measurements, 68% are correctly predicted by the logistic regression model. More specifically, 73% of the groundwater samples with As concentrations greater than or equal to 10 μg/L are correctly predicted (sensitivity), whereas 65% of the groundwater samples with As concentrations less than 10 μg/L are correctly predicted (specificity). Consequently, below we apply the same model and approach to south Louisiana to evaluate the probability that elevated As concentrations in shallow groundwater may also characterize the region including the MR delta. Again, the existing As concentration data for shallow groundwater in south Louisiana are rare, making the logistic regression an appropriate approach to evaluate the possibility of naturally sourced As contamination of local groundwater. Moreover, it is important to stress that the model does not allow us to predict shallow groundwater in south Louisiana that may be at risk of anthropogenic-sourced As contamination.

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risk map (Figure 4b) indicates that an area of 23,881 km² in south Louisiana may be at risk of shallow groundwater As contamination (probability ≥0.5) from natural sources and processes. To validate the performance of the model, 78 binary-coded As concentration measurements for shallow groundwater (<50 m) were compared to the risk map predicted values (Figure 4b). The comparison results showed that the model correctly predicted 79% of the total groundwater samples. Specifically, 88% of the shallow groundwater samples that have As concentrations greater than or equal to 10 μg/L are correctly predicted by the model, whereas 77% with As concentrations less than 10 μg/L (specificity) are correctly predicted. Consequently, the performance of the model in south Louisiana is comparable to its performance in Bangladesh. Nevertheless, additional sampling of shallow groundwater in south Louisiana is required to test the accuracy of the model for extensive regions of the study area for which shallow groundwater As concentration data do not currently exist.

II DISCUSSION

Parameters Contributing to the Arsenic Prediction Model. Groundwater As contamination is commonly associated with shallow aquifers composed of Holocene sediments in areas with flat topography and low hydraulic gradients. Specifically, the chemically reducing Holocene sedimentary depositional environments that characterize the deltas and floodplains of large rivers are thought to influence the development of As-contaminated groundwater. Winkel et al. demonstrated the statistical significance of Holocene deltaic, alluvial, and organic-rich deposits in predicting As contamination in shallow groundwater from major river deltas of South and Southeast Asia. Our model confirmed the importance of hydrology, soil, geology, and sedimentary depositional environments in predicting groundwater As contamination in the shallow reduced aquifers of the Ganges-Brahmaputra-Meghna delta in Bangladesh, as well as the Mississippi River delta in south Louisiana, U.S.A. (Figure 3 and Figure 4). The positive weighting coefficients of deltaic deposits (λ = 1.62), organic-rich deposits (λ = 0.82), and alluvial deposits (λ = 0.51) indicate that the presence of deltaic deposits in the Holocene aquifers likely exert the most important controls on the development of elevated As concentrations in the associated groundwater, followed by organic-rich deposits, and then alluvial deposits. Deltaic deposits and alluvial deposits are formed in deltas and river floodplains, which commonly form in areas with flat topography and low hydraulic gradients. These characteristics provide favorable conditions for the development of chemically reducing environments where groundwater As contamination can occur. In contrast, pre-Holocene deposits are negatively (λ = −3.42) associated with groundwater As contamination, which indicates that aquifers consisting of older sedimentary deposits (e.g., Pleistocene) are expected to exhibit relatively low As concentrations in associated groundwater. Soil properties and texture are indicative of current as well as past drainage conditions within various depositional environments, and as such, they are closely related to the local hydrological processes of the aquifers. For example, shallow aquifers overlain by fine-grained sediments (i.e., silt and clay) in Bangladesh are known to be poorly drained, and therefore, the associated groundwater commonly exhibits relatively high levels of As. The positive weighting coefficient of the clay content in the top 1 m soil layer (λ = 0.11) implies that groundwater from shallow aquifers overlain by fine-grained deposits is at relatively high-risk of...
developing elevated As concentrations due, in part, to the low permeability of clay. Medium-textured soil (i.e., silt) in river basins, floodplains, and deltas is also an indicator of the presence, transport, and/or deposition of fresh, reactive materials (e.g., organic matter, Fe (III) oxides/oxyhydroxides), which favors reductive dissolution of Fe (III) oxides/oxyhydroxides under reducing conditions and subsequent release of previously absorbed and/or coprecipitated As into groundwater.29−32 Our model also confirmed that the presence of medium-textured soil was indicative of elevated As concentrations in shallow groundwater (λ = 1.16). Distance to rivers can influence the local hydrological processes and as such can be used as an indicator of relatively young (i.e., Holocene) sediments. Aquifers located close to rivers commonly consist of relatively young and freshly deposited sediments, which can provide a reducing environment that drives microbial respiration and leads to As enrichment in shallow groundwater.30−32 The negative weighting coefficient (λ = −1.86) of distance to rivers indicates that aquifers close to rivers are at high risk of groundwater As contamination, and this risk decreases with increasing distance to rivers.

The fact that the MR delta of south Louisiana shares broadly similar geologic and hydrologic characteristics with the rivers/deltas of South and Southeast Asia, in conjunction with the model prediction presented here, indicates that shallow groundwater from south Louisiana may also be at risk of As contamination as a result of similar natural sources and processes. Owing to the relatively low numbers of shallow groundwater samples that have been analyzed for dissolved As concentrations, the extent of naturally occurring As-contaminated groundwater in south Louisiana still remains poorly known. Nevertheless, the application of the As prediction model to south Louisiana presented here, and its relatively accurate predicted results for the existing groundwater As data, indicate that more sampling and analysis of As concentrations in local shallow groundwater are warranted. Therefore, it is not advisable to use local shallow groundwater as an alternative drinking water source in south Louisiana if no further assessment of groundwater As contamination is carried out.

**Limitations of the Arsenic Prediction Model.** The potentially important parameters controlling the distribution of natural groundwater As contamination inherently contain a 3D component (i.e., depth), which defines the subsurface boundary of aquifers formed under different geological periods (e.g., Pleistocene or Holocene) or under different sedimentary depositional environments (e.g., alluvial, deltaic, or tidal). However, due to a lack of readily available, accurate 3D data (i.e., depth) for the extensive region of south Louisiana examined herein, our model is based only on 2D surface data. Consequently, the model assumes that geology and sedimentary depositional environments expressed at the surface are the same as those in the shallow subsurface (<50 m). Therefore, misclassifications will occur when geology and sedimentary depositional environments expressed at the surface differ from those of the shallow subsurface. Improvements on the model predictions are expected for cases where geology, hydrology, and/or soil properties as a function of depth can be included in the regression model.14 Despite these and other limitations, the model generally does a good job at predicting groundwater As contamination in shallow aquifers of south Louisiana where scarce data are currently available.

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**ASSOCIATED CONTENT**

Supporting Information

Groundwater sample collection and analysis, arsenic concentration in shallow groundwater of south Louisiana, sources of GIS data used in this study, and the 15 initially considered independent variables in building the As prediction model are presented in the Supporting Information. Also presented is the receiver operation characteristic curve of the regression model.14 Despite these and other limitations, the model generally does a good job at predicting groundwater As contamination in shallow aquifers of south Louisiana where scarce data are currently available.

**AUTHOR INFORMATION**

Corresponding Author

*E-mail: nyang@tulane.edu. Fax: (+1) 504 865-5199. Tel.: (+1) 504 862-3193.

Notes

The authors declare no competing financial interest.

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**Figure 4.** Predicted probability of shallow groundwater As contamination in south Louisiana. (a) Continuous probability map. (b) Binary risk map showing high- and low-risk areas overlain by 78 shallow groundwater As concentration data.
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