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Mapping of spatial multi-scale sources of arsenic variation in groundwater on ChiaNan floodplain of Taiwan

Yun-Bin Lin [a], Yu-Pin Lin [b,*], Chen-Wuing Liu [c], Yih-Chi Tan [d]

[a] Department of Recreation and Health-Care Management, ChiaNan University of Pharmacy and Science, Rende Shiang, Tainan County 71710, Taiwan, ROC
[b] Department of Bioenvironmental Systems Engineering, National Taiwan University, 1 Sec. 4 Roosevelt Road, Taipei 10673, Taiwan, ROC
[c] Department of Bioenvironmental Systems Engineering, National Taiwan University, Taipei 10673, Taiwan, ROC
[d] Department of Bioenvironmental Systems Engineering, and Hydrotech Research Institute, National Taiwan University, Taipei 10673, Taiwan, ROC

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Abstract

This study applied multivariate factorial kriging to derive the characteristics of the spatial variations of groundwater arsenic distributions at different scales on the ChiaNan floodplain, Taiwan. Seven variables (dissolved oxygen, oxidation-reduction potential, alkalinity, sulfate, iron cations, manganese cations and total organic carbon) and Arsenic were adopted to analyze the mechanisms of arsenic enrichments in groundwater. The hydrogeological environment had spatial and quantitative influences on arsenic enrichments at different scales. The regional scale was set to 32 km referring to the extension distance of flow paths to reflect the effects of flushing in the aquifer, while the local scale was set to 16 km referring to the farthest distance of seawater intrusion to determine the influence of seawater intrusion. The results of factorial kriging suggested that arsenic releases resulted partially from pyrite oxidation during the flushing at the regional scale and partially due to the siderite dissolution at the local scale. Overall, the alkalinity dominated arsenic distribution in groundwater at both the regional and local scales. The multivariate factorial kriging results also demonstrated that seawater intrusion slightly affected the increase of arsenic in groundwater, accounting for only 17.3% of total variation. However, the interaction of seawater intrusion and arsenic distribution in space indicated that seawater intrusion restrained the distribution of arsenic from the areas where seawater was located. High dissolved oxygen was found at where over-pumping induced drawdown cones occurred and also limited the spatial variation of arsenic. Our findings indicate that multivariate factorial kriging can be a useful mapping tool to improve understanding of the mechanism of arsenic release in groundwater at different scales. And the results conducted from the application of multivariate factorial kriging in southwestern Taiwan reveal the important influences of the hydrogeological processes, either artificial or natural, on the arsenic variations in groundwater.

Keywords: Multivariate factorial kriging; Spatial sources of arsenic variation; Arsenic contamination of groundwater; Flushing; Seawater intrusion

1. Introduction

Black foot disease was generally found on the ChiaNan floodplain during the early 1960s. This disease

* Corresponding author. Tel.: +886 2 2368 6980; fax: +886 2 2368 6980.
E-mail address: yplin@ntu.edu.tw (Y.-P. Lin).

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was found to be strongly correlated with the direct ingestion of high arsenic (As) groundwater from deep wells mostly screened 100–280 m below ground. Large amounts of data whose subjects were assigned exposures according to exposure categories instead of having individual measurements of As exposures was collected in southwestern Taiwan and the data was generally recognized as the ecological data (Tseng et al., 1968; Chen et al., 1994; Guo and Valberg, 1997). The statistical analysis of ecological data initiated the quantitative risk assessment of cancer mortality of skin cancer and some internal cancers, including lung, bladder and liver due to chronic ingestion of As in groundwater. The assessment was adopted to define the maximum contaminant level of drinking water (National Research Council 1999, 2001). The World Health Organization (World Health Organization, 1993) and the U.S. EPA (National Research Council, 2001) reduced As standards for drinking water from 50 to 10 ppb after consulting studies regarding the carcinogenicity of As in drinking water conducted in southwestern Taiwan.

At present, the groundwater is not the primary sources for direct ingestion or domestic use in Taiwan. However, the study concerning the statistical correlations between high As and seawater samples in littoral zones in Taiwan which was done by Liu et al. (2003) via conventional factor analysis motivates further investigation of spatial correlations between distributions of As and seawater intrusion via geostatistics. Aquaculture is the primary agricultural sector in the littoral zones in southwestern Taiwan, including the ChiaNan floodplain. Groundwater provides a stable and low priced source of water. Large quantities of groundwater have been extracted from the aquifer to meet fishpond needs. The serious seawater intrusion problem was induced by these extractions. The seawater intrusion seemed to restrain the distribution of As from the areas where seawater was located. Pumping from high As area raises a serious issue of edible safety of aquaculture products in the littoral zones of southwestern Taiwan.

The variation of As in groundwater is complex, Smedley and Kinniburgh (2002) established the classification table which lists the risk factors being tested with the highest priority in various hydrogeological environments. The variations of As in groundwater are related to past and present hydrogeological conditions, which may include the flushing process in aquifers, and seawater intrusion induced by over pumping. These artificial or natural hydrogeological processes perform at different scales. Hossain and Sivakumar (2006) used the chaotic analysis to determine the dimensions of the spatial variability of As in groundwater. The value of dimensions represents the number of variables needed in modeling the spatial variability of As in groundwater and was found to be highly correlated with the sampling interval. The analytical results indicated that different mechanisms involve in at different scales. The assessment of the oncoming risk of cancer incidence due to As exposures relies on the understanding of the release mechanism at different scales which generates the fluctuations of As concentrations over time in groundwater. Mapping of multivariate spatial variation of groundwater quality can effectively improve our understanding of the As release mechanism at different scales and clarify the spatial relationships between the disease and groundwater As in Taiwan. Moreover, the results of multivariate spatial analysis can also be applied to support the management of water containment and ensure the edible safety of aquaculture products in the littoral zone of southwestern Taiwan.

Geostatistical methods, such as kriging, provide a linear un-biased estimate at the unsampled grid nodes. The estimates are weighted sums of the adjacent sampled variables. The weights are determined by the spatial structures of experimental variograms and are selected by minimizing the estimation variance (Cressie, 1990; Wackernagel, 1995). Spatial structures of experimental variograms are modeled using the linear combination of various allowable basic models with different ranges. The modeling procedure is also termed 'regionalization' in the univariate case and 'co-regionalization' in the multivariate case. Moreover, the modeled matrix of the experimental variograms should be positive semi-definite in the multivariate case (Goovaerts et al., 1993; Goovaerts, 1997). Goulard and Voltz (1992) proposed the linear model of co-regionalization (LMC) to minimize the weighted sum of the squared differences between the experimental variogram and the variogram model, and to maintain the positive semi-definition of the modeled matrix. The LMC resulting matrices at different scales or ranges were then applied to principal component analysis (PCA) to examine the relationships between variables at each scale. Finally, the spatial variations of the principal components at different scales, regionalized factors, are mapped by factorial kriging. The dominant causation generating the relationships between variables is then deduced from the PCA results and maps of regionalized factors. Therefore, the spatial variations of multi-variables associated with various processes at different scales can be distinguished via Multivariate factorial kriging (MFK) which is a multivariate geostatistical method for dealing with multivariate problems with