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Methodology of spatial risk assessment for arsenic species associated with sampling and analysis results optimization

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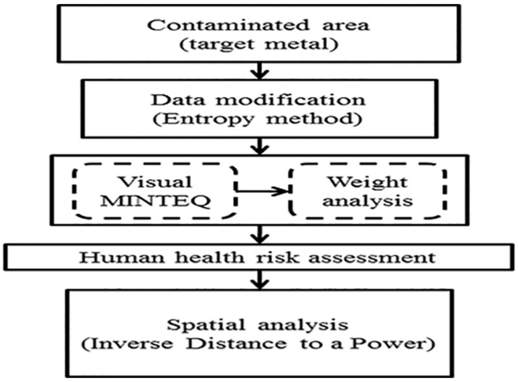
HIGHLIGHTS

- A method was proposed to optimize sampling and analysis results for spatial health risk of arsenic species in groundwater.
- An application of the proposed method for estimating As species was studied.
- The health risk distribution of As species showed severe health risk to human.
- $HAsO_4^{-2}$, H_3AsO_3, $H_2AsO_4^-$, $H_2AsO_3^-$ and $AsS(OH)HS^-$ caused main risk to human.

GRAPHICAL ABSTRACT



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ABSTRACT

The conventional risk assessment methods may be defective for more effective health risk assessment due to ignoring heavy metal species and the accuracy and integrity of sampling and analysis results. Using the accurate and integral data to quantify the human health effects of metal species can provide great support for more effective health risk assessment. This study presents a new methodology to optimize sampling and analysis results for implementing the spatial human health risk of heavy metal species in contaminated sites. The method integrated Entropy method and Inverse Distance to a Power (IDW) for obtaining the effective risk, and mapping the visual risk distribution of metal species. The results of its application with ingesting arsenic via oral route on adults showed that carcinogenic and non-carcinogenic risks of As were influenced by its species. The risk of $HAsO_4^{-2}$, H_3AsO_3, $H_2AsO_4^-$, $H_2AsO_3^-$ and $AsS(OH)HS^-$ exceeded threshold that was significantly harmful to human health in study area. This method broadened the scope of human health risk assessment and provided a basis for government policy-making and site remediation.

1. Introduction

Excess heavy metals in aquatic environments have become a global trouble (Serkan, 2012). Once aquifers become polluted, contamination is persistent and difficult to remedy due to their large storage, long residence times and physical inaccessibility further affecting human health

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(Foster and Chilton, 2003; Li et al., 2014). With elevating levels of arsenic in the environment, arsenic has attracted increasing concern due to its negative impacts to human and ecological health (Yang et al., 2011). In recent years, arsenic has been reported that over 137 million people in >70 countries were affected by arsenism for groundwater in worldwide (Yang et al., 2012). Studies indicated cancer of skin, lung and urinary bladder are important cancers associated with chronic arsenic toxicity (Guha Mazumder, 2008). Therefore, it is of great importance to effectively assess the potential human health risk for heavy metals in groundwater.

Numerous studies had conducted to quantify the health risk of heavy metals (As, Pb, Cr, Mn, etc.) in groundwater (Damodharan and Reddy, 2013; Abdelhafez and Li, 2014; Zhu et al., 2015; Liu et al., 2016). In general, health risk was calculated by the total concentration of heavy metals (Krishna et al., 2014; Wongsasuluk et al., 2014; Abdelhafez and Li, 2015), as well as the concentration of heavy metal species detected through extraction experiment in recent years (Ren et al., 2015; Tepanosyan et al., 2017). The risk of concrete heavy metal species (i.e. $H_2AsO_3^-$) to human only considered by Zhang et al. (2017a, 2017b) using a methodology-metal species weighted human health risk assessment (MSRA) to quantify and distinguish the contribution of metal species risk on human in site-specific groundwater with chemical equilibrium model. Moreover, to determine data uncertainty and preserve spatial analysis of heavy metals, many methods combined with human health risk assessment to reveal the uncertainty and the health risk distribution of heavy metals in some studies. Geographic Information System (GIS) was used to assess the data uncertainty with Fuzzy Set Theory, Gray Theory Method and Neural network method, as well as the risk distribution (Kontic et al., 2002; Bień et al., 2005; Poggio and Vrščaj, 2009). Likewise, a spatial decision support system software called DESYPE with the Monte Carlo analysis was developed to characterize the uncertainty and variability in risk estimates by repeatedly sampling the probability distributions of the risk equation inputs and using these inputs to calculate a range of risk values (Carlon et al., 2008; Critto and Agostini, 2009; Sourdis et al., 2013).

However, some insufficiencies were found from the published literature on the human health risk assessment for heavy metals. Firstly, the inaccuracy of sampling and analysis results caused by artificial errors and experimental errors may be neglected, as well as the integrity of sampling and analysis results (Ferson and Ginzburg, 1996). The results without the consideration of the relationship between each sampling points lead to the risk overestimated (Li et al., 2011). Secondly, the spatial analysis of metal species was no-considered (Sun et al., 2010; Xie et al., 2011), but it is particularly important to be recognized for policy-making and site remediation. Owing to the concentration of heavy metals species are specific and non-homogenous spatial distribution with different toxicity, mobility and bioavailability (Giubilato et al., 2014). For example, As (III) is reported to be 25–60 times more toxic than As (V), and several hundred times more toxic than organic arsenides (Garelick et al., 2005; Muñoz and Palmero, 2005; Basiri et al., 2011). Thirdly, existed methods for optimizing data uncertainty are complicated and inconvenient. With the results of no-satisfying the requirements for simply use to optimize data uncertainty. Subsequently, such effects may result in the inappropriate risk management and decision-making.

To address the existed problems, it is necessary to develop a method with the ability of taking into account not only the inaccuracy and integrity of sampling and analysis results, but also the spatial risk of metal species. Therefore Entropy Method and Inverse Distance to a Power (IDW) were incorporated to efficiently and visually quantify the health risk distribution of heavy metal species on human in this paper.

The objective of this paper was to: (1) optimize sampling and analysis results from contaminated site to obtain the accurate and integrity analysis results of heavy metal (As); (2) simulate the heavy metal species and compare the relationship between the concentration of heavy metal species and heavy metal (As); (3) evaluate the carcinogenic and non-carcinogenic spatial risks of heavy metal species (As); (4) provide theoretical support for government policy-making and site remediation.

2. Materials and methods

2.1. Overview of the methodology

A methodology of sampling and analysis results optimization and spatial human risk assessment for heavy metal species in contaminated groundwater was proposed. The framework of proposed method is shown in Fig. 1 including four parts: source; data optimization; human health risk assessment; spatial analysis. Data optimization is aimed at obtaining the accurate and integral sampling and analysis results. The optimized results were used for species simulation which was in order to obtain the concentration of each metal species through Visual MINTEQ (Gustafsson, 2012a). The results of species simulation were subsequently modified based on MSRA. In accordance with the modified concentration of heavy metal species, the carcinogenic and non - carcinogenic risks of heavy metal species were calculated by modified average daily dose. Finally, the human health risk spatial distributions of heavy metal species were obtained based on Inverse Distance to a Power.

2.2. Study area

The research area is located in Jiangxi province between 27°47′ Latitude and 114°40′ Longitude, with a size of 2.4156 km^2. It was used to placing slag for 9 years and slag accumulation was >5 million tons. There is a village in a 3 km from the slag with a 7 hundred population. Moreover, the area is near several tributaries. In this region, the major drinking water source is groundwater and it is mainly supplied by atmospheric precipitation. The composition of the slag is complex and generally contains harmful elements. These substances were oxidized, leached, dissolved, infiltrated underground causing groundwater pollution. Consequently, the local people and ecosystem can be exposed to severe environmental pollution.

2.3. Sampling and chemical analysis

The total 12 groundwater samples were collected from a series of boreholes and sampling locations were presented in Fig. 2. All groundwater samples were collected from the shallow depths ranging from 6.5–20 m. The latitude and longitude were recorded via global positioning system (GPS). Samples were collected in pre-cleaned polyethylene bottles using a submersible pump, with over 100 L being pumped and thrown away. The pHs of samples were measured in situ using pH meters (PHS-3C, LEICI). The UV/Visible Spectrophotometer (Model UV 1601Shimadzu) was used to determine the concentration of nitrates and sulphates at a wavelength of 220 nm. The concentrations of chloride (Cl^-) were determined by titration method (ISO 9297-2007), moreover the concentration of alkalinity (CO_3^{2-} and HCO_3^-) was detected using acid-base titration method. The concentrations of calcium (Ca^{2+}), K^+, Na^+ and Mg^{2+} were measured by atomic absorption spectrometry, using a Perkin Elmer 3100. The concentrations of arsenic were determined by using atomic absorption spectrophotometer (spectra AA220FS, Varian, New Jersey, USA). and other cations were determined by inductively coupled plasma-mass spectrometry (ICP-MS, Agilent 7500cx, Agilent Inc., USA).

2.4. Data optimization

The measured concentration of target heavy metal was optimized based on Entropy method, an objective way to determine weight. The entropy indicates the disorder degree of a system, with the capacity of