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Establishing a health risk assessment for metal speciation in soil—A case study in an industrial area in China



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ARTICLE INFO

Keywords:

Metal-speciation Human health Soil Simulation

ABSTRACT

An improved method was proposed which integrates the distribution of metal speciation simulated by chemical equilibrium model, different exposure models and average daily intake dose modified by analytic hierarchy process for human health risk assessment of metal species (MS). With the rapid development of economic and urbanization, the metals pollution had become more serious in industrial areas. Adverse effects of soil contaminants on human health in typical industrial area should be assessed to evaluate the risks of soils in these areas. The method was applied to study nickel (Ni) species health risks in soil of industrial areas. The pH possessed significant impact to determine distribution/existence and solubility of Ni species, followed by DOC. The non-carcinogenic risk (HQ) of Ni species were less than 1 in each sampling points, except Ni 2 +. In addition, the carcinogenic risk (CR) of different Ni species were less than 10^{-6} , except for FANi and Ni 2 +.

1. Introduction

Soil pollution from metals has accelerated in China in recent decades due to te high-intensity development of land resources and rapid economic development in recent decades (Lu et al., 2015; Lin et al., 2017). Metals and their compounds are frequently used as catalysts and chemical additives in industrial processes, and as such, soils can be polluted by industrial emissions (Salmani-Ghabeshi et al., 2016; Huang et al., 2017; Li et al., 2015). In addition, industrial areas are often in regions of dense human activity (Guan et al., 2017). Metal and metalloid contaminated soil is a major environmental problem, resulting in increased human exposure to these contaminants (Peña-Fernández et al., 2014). Human safety is a primary consideration in risk assessment of soil contamination. Industrial areas have been included as key governance plots in China's soil management and protection. Thus, the adverse effects of soil contaminants on human health in typical industrial areas should be assessed.

Increasing evidence shows that the hazards of metals to organisms health are determined by metal species, rather than the total concentration (TC) of metals (Gu et al., 2016; Mashal et al., 2015; Reimann et al., 2005; Reis et al., 2014; Zhang et al., 2017), because toxicity/bioavailability strongly depends on their species in natural environment (Castlehouse et al., 2010; Song and Ma, 2016). Furthermore, researches

have shown that the health risks from metals from their species, determined through speciation experimental extraction (such as RBA, Tessier and etc.) with a combination of health risk assessment models in soil (Yang et al., 2015; Dehghani et al., 2017; Li and Ji, 2017). Previous studies have shown the adverse effects of metals in soil and ignored the effects in soil solutions. However, it is widely accepted that the toxicity and bioavailability of metals in soils are conditional on their speciation in soil solution, especially on the concentration of free metal ions (Ge et al., 2000; Schneider et al., 2016) and on the solid/solution distribution of total metals (Zhang et al., 2015).

Empirical and mechanistic models (such as WHAM VI, Visual MINTEQ, PHREEQC) have been widely employed for metal species (MS) in soil systems (Rooney et al., 2007; Ge et al., 2000; Schneider et al., 2016; Zhang et al., 2015), Compared with speciation experimental extraction approaches, these models detail speciation, and are time-saving and cost-saving. On the other hand, functionally these models meet the qualification that simulating the distribution of metal speciation with variation in environmental conditions, in virtue of metals speciation formation are closely relevant to soil properties such as pH, organic matter, Mn and/or Fe content and oxidation-reduction potential (Ukowski et al., 2013; Barman et al., 2015). The empirical and mechanistic models are crucial tools in the analysis of MS in soil.

The absorption dose of metals is not equal to the actual

Abbreviations: TC, Total concentration; MS, Metal species

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concentration of pollutants (Zhuang et al., 2009; Machida et al., 2004), and traditional health risk assessments neglect the relationship between concentration and uptake of pollutants by organisms. In addition, many lines of evidence distinctly show that uptake and metal toxicity are mainly dependent on the dissolution concentration and free ion activity (Steenbergen et al., 2005; Van Gestel and Koolhaas, 2004; Zhang et al., 2013). For these reasons, it is regarded clearly and definitely that to modify the average daily dose is equivalent demanded. The analytic hierarchy process (AHP) is a multi-criteria decision making method that helps to solve a complex problem with multiple conflicting and subjective criteria (Ishizaka and Labib, 2011). This mechanism can make it possible to effectively, accurately and conveniently modify the average daily intake with influences from multiple factors.

The major objectives of this study were as follows: (1) to propose an improvement method that can quantify and distinguish the predominant MS in soil that are known to pose health risks; (2) to identify the dominant MS absorbed by organism based on an analytic hierarchy process; (3) to apply the method to an industry assessing the carcinogenic and non-carcinogenic risks of Ni speciation in soil; (4) to evaluate and compare the health risks between TC level and each species concentration; (5) to confirm the distribution of MS risks along with variation soil environmental factors at each sampling points.

2. Framework for modeling human health risks from metal species

A method for assessing health risks from MS was developed to quantify and distinguish the contribution of MS risks to human in soil (Fig. 1). Four main aspects make this approach innovative. First, a chemical equilibrium model (Visual MINTEQ) was used to provide information of MS concentration and activity. Second, an analytic hierarchy process was employed that modified the average daily dose of MS to determine the sequence of dominant species that could be intake by the organism. Third, the method compared the health risks between MS and the TC level through exposure pathways. Finally, it input different environmental conditions for simulation and then helped to understand the impact of MS risks on factors variation. Some advantages of this method can be summarized as follows: (1) It can assess the health risks of metals more accurately; (2) It offers available information to propose a hierarchical risk management strategy that provides reference for flexible and cost-efficient risk management policy-making. For example, specific treatment measures for remediation can be proposed

based on detailed MS health risk values and their characteristics. (3) It identifies the priority species ingested by organisms and provides detailed information on the study of bioavailability; (4) It can be conducive to identifying the contribution/ priority of MS risks caused by variation in soil environmental factors associated with the probability of risk in excess of the accepted level.

2.1. Modeling

Visual MINTEQ. 3.1 (Gustafsson, 2012) was used to simulate MS in soil solutions with the Stockholm Humic Model (SHM) (Gustafsson, 2001) and Hydrous Ferric Oxide (HFO) (Butler et al., 2005). The SHM has been used for speciation of many trace metals. HFO plays a significant role in MS because hydrous ferric are important ligands for metals, and have a high specific surface area and strong affinity for many elements (Schneider et al., 2016). The determined concentrations in the soil solution of cations (K, Ca, Na, Mg, Al, Fe and Ni) and anions (F⁻, Cl⁻, NO₂⁻, NO₃⁻, PO₄³⁻ and SO₄²⁻), as well as the pH, Eh and the DOC content, were used to perform the calculation. The values of MS concentration (C_i , mol/L,i = 1, 2, 3, ..., n) and activity (A_i ,i = 1, 2, 3, ..., n) (i is the number of MS) were obtained from the simulation.

2.2. Exposure assessment

Human health risks resulting from soil contaminants were evaluated based on the US EPA site-specific risk assessment method (US EPA OOSW, 2009; Us Epa, 2011, 1996). Classically, there are three central exposure pathways of metals to human body, either through oral ingestion, inhalation and dermal contact. The average daily dose of MS was modified through the analytic hierarchy process (AHP) (Cheng et al., 2002).

In this study, we use the following algorithm to select the greater average daily dose for MS through the AHP technique:

Step 1: Structure the problem as a hierarchy:

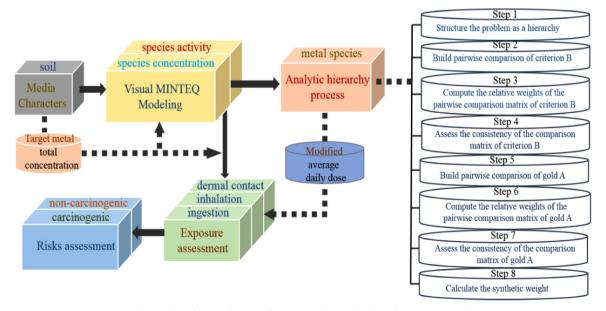


Fig. 1. Flow chart of the steps for assessing human health risk from MS in soil.

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