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An exploration of an integrated stochastic-fuzzy pollution assessment for heavy metals in urban topsoil based on metal enrichment and bioaccessibility

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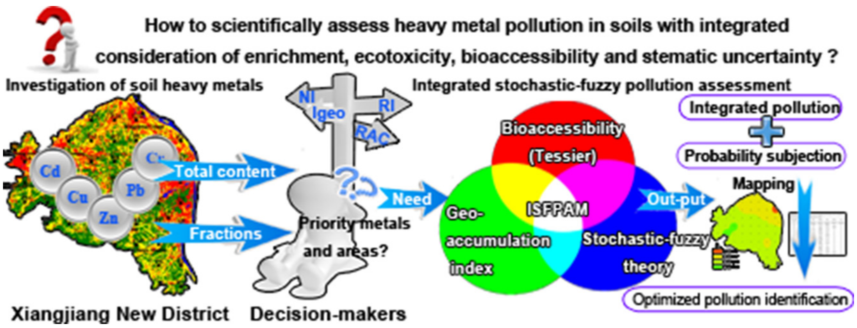
HIGHLIGHTS

- Soil metal enrichment, ecotoxicity and bioaccessibility need integrated evaluation.
- Integrated stochastic-fuzzy pollution assessment method (ISFPAM) was established.
- ISFPAM integrated I_{geo}, stochastic-fuzzy theory and double weight system.
- Cd, Cu and Pb was identified to be priority pollutants in topsoil from XND.
- ISFPAM show better resolving ability compared with *SF*, *NI*, I_{geo}, *RI* and *RAC*.

GRAPHICAL ABSTRACT



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ABSTRACT

An integrated stochastic-fuzzy pollution assessment method (ISFPAM) for soil heavy metal was established based on geo-accumulation index (I_{geo}), stochastic-fuzzy theory and double weight system under synthetical consideration of metal ecotoxicity and bioaccessibility. The pollution characteristics of the topsoil heavy metals (Cu, Zn, Cd, Pb and Cr) in Xiangjiang New District were evaluated by the widely-used Single factor index (*SF*), Nemerow index (*NI*), I_{geo}, Potential ecological index (*PERI*), Risk assessment code (*RAC*) and the ISFPAM. The results of *SF*, *NI*, I_{geo}, *RI* and *RAC* of the studied metals revealed the following orders: Cd > Zn > Cr > Cu > Pb, Cd > Zn > Pb > Cr > Cu, Cd > Cr > Cu > Zn > Pb, Cd > Cu > Pb > Cr > Zn, and Cd > Pb > Cr > Zn > Cu, respectively. The different pollution assessment methods outputted the differentiated conclusions to some extent except the judgment for Cd. Results based on ISFPAM indicated that metal pollution degrees decreased in the order of Cd (5.91, Grade 6) > Cu (2.81, Grade 3) > Pb (2.66, Grade 3) > Cr (1.58, Grade 2) > Zn (0.69, Grade 1). By detailed comparison analysis, the double weight system and stochastic-fuzzy theory made ISFPAM better resolving ability to find out priority heavy metals and areas with relatively higher enrichment, ecotoxicity and bioaccessibility under efficient parameter uncertainty control. Cd, Cu and Pb were regarded as the priority control metals, especially Cd. Simultaneously, the reliabilities of heavy metal pollution corresponding to adjacent pollution grades were quite close in some sites, which recommend recheck for avoid misleading the decision-makers.

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1. Introduction

Rapid urbanization is a worldwide phenomenon which always goes with intensive industry, economic activities and environmental problems, especially in developing countries (Li et al., 2016; Li and Jia, 2018). Urban soils play a core role in urban environments and, the pedosphere acts like pivot for material and energy interchanges among the atmosphere, hydrosphere, biosphere and lithosphere (Hu et al., 2018; Li et al., 2018a). Furthermore, urban soils serve as one of most key reservoir or sink for toxic metals and other pollutants which could be resulted from both natural and anthropogenic activities (Li et al., 2015a; Wang et al., 2018). Excessive inputs of heavy metals into urban soils have attracted worldwide extensive attention owing to their toxicity, persistence and bioaccumulation (Dong et al., 2018; Islam et al., 2017). Therefore, it is of importance to study pollution characteristics of heavy metals in urban soils and evaluate their pollution degrees to protect the urban environment and human health.

In recent years, various works have been done to explore the heavy metal distributions and sources in urban soils as well as their bioaccessibility or bioavailability based on metal chemical fractions (Gu and Gao, 2018; Li et al., 2015b; Li et al., 2018a; Luo et al., 2015; Tepanosyan et al., 2017), correlativity with soil physicochemical properties (Li et al., 2015a; Liu et al., 2016), pollution and eco-risk levels (Cheng et al., 2014; Luo et al., 2012b; Wei and Yang, 2010), primary sociometric causative factors (Liu et al., 2016; Wang et al., 2018) and corresponding remedy technologies (Kim et al., 2016). Environmental pollution assessment of heavy metals in unban soil is an important reference for identifying the pollution degree and formulating pollution prevention strategies. Nowadays, the Enrichment factors (*EF*), Single factor index (*SF*), the Nemerow index (*NI*), the Geo-accumulation index (I_{geo}) were frequently used as indexes of heavy metal enrichment degree (Han et al., 2018; Li and Ji, 2017; Lin et al., 2018). Moreover, the potential ecological index (*PERI*) and risk assessment code (*RAC*) were widely utilized to identify the ecological risk and ecological bioaccessibility of heavy metals, respectively (Chen et al., 2018; Hakanson, 1980; Li and Ji, 2017; Zhu et al., 2012). Unfortunately, these widely-used methods mostly evaluate from a certain aspect, without scientifically considering soil metal enrichment, bioaccessibility and ecotoxicity comprehensively. Similar weaknesses of the previously developed pollution indices were discussed by some experts (Karbassi et al., 2008; Roudposhti et al., 2016). The *PERI* assess ecological risk caused by heavy metals from total content and ecotoxicity based on abundance of the elements in the earth's crust, ignoring effects on bioaccessibility caused by varying metals' chemical fractions and binding states (Huang et al., 2016; Zhu et al., 2012). By contrast, the *RAC* emphasizes bioaccessibility of heavy metals based on metals' chemical fractions, but there is some discrepancy among the bioaccessibility, enrichment and ecotoxicity (Matong et al., 2016; Li et al., 2018b). Moreover, the above deterministic methods probably lead to biased assessment conclusion or even an unreliable one due to the randomness and fuzziness in environment system caused by natural change and human activities (Hu et al., 2016a; Li et al., 2012a). New, some improved models have been developed to include uncertainty control methods (such as fuzzy method, stochastic method, grey theory, etc.), but these computational models tend to be too laborious or impractical for regulatory purposes (Hu et al., 2016b; Li et al., 2012a; Li et al., 2018b). Obviously, when there are different theoretical foundations, the evaluation results and conclusions differ to some extent, which may confuse decision makers or even mislead the final policy (Li et al., 2018c; Matong et al., 2016). Therefore, it is of significance to explore a high-efficiency and feasible pollution assessment method synthetically considering the complexity of soil metal toxicity and bioaccessibility, together with spatial differences in metal background concentrations (Karbassi et al., 2008; Roudposhti et al., 2016).

The objectives of this study were: (i) to establish an integrated stochastic-fuzzy pollution assessment method (ISFPAM) for soil heavy metal based on geo-accumulation index under synthetical consideration of heavy metal ecotoxicity and bioaccessibility; (ii) to investigate the Cr, Cu, Pb, Zn and Cd concentrations and their chemical fractions in topsoil throughout the Xiangjiang New District, Middle China; (iii) to evaluate the spatial pollution state of topsoil heavy metals based on ISFPAM, and identify the priority pollutants and hotspot areas. (iv) to make comparative analysis on evaluation results between ISFPAM and other widely-used pollution evaluation methods to verify the scientificity and feasibility of ISFPAM.

2. Materials and methods

2.1. Study area

The Xiangjiang New District (XND), which was called Xiandao District before 2015, is a municipal district of Changsha city with a total area of approximately 1200 km^2 and the permanent resident population of over 1.34 million. The district belongs to subtropical monsoon climate with the annual average temperature 16.8–17.2 °C and the average annual precipitation 1358–1370 mm (Li et al. 2015). The Changsha city is an important center of economy, culture, transportation, education and manufactory located in central south China. From 2010 to 2015, the urbanization rate in Changsha city increased from 67.69% to 74.4% and that in XND increased from 80.08% to 84.58%. Moreover, the land in XND was traditionally associated with agriculture, forest, industrial and residential uses, and based on the land use mapping determined from a remote sensing image of XND from United States Geological Survey (USGS), the proportion of urban construction land in XND increased obviously from 1990 to 2014, especially in 2000–2014 (Fig. S1). From 2011 to 2016, the Gross Domestic Product (GDP) of XND rapidly increased from 80.93 billion to 180.10 billion, respectively. Moreover, GDPs of the primary industry, the secondary industry and the tertiary industry in year 2015 were 4.65 billion, 108.41 billion and 47.19 billion, respectively. In 2007, XND was approved by the Chinese State Council as a national pilot district of constructing the resource conservation and environment friendly society. Therefore, XND was selected as the study area (Fig. 1) which has been experiencing rapid urbanization and economic development with an obvious decline of environmental quality in recent years (Chen et al., 2011; Li et al., 2016; Wang et al., 2010).

2.2. Samples collection and chemical analysis

A total 156 samples (3 parallel samples for each sampling site) of topsoil (0–20 cm) from 52 sampling sites were collected at intervals of 5 km throughout XND in October 2013. Each sample was a composite of 5 random subsamples from nearby 5 m^2 area. The sampling sites, with artificially adding sites in areas under high high-density population distribution, contained 25 farmland sampling sites (F1-F25), 15 construction land sampling sites (U1-U15), and 12 woodland sampling sites (W1-W12) (Fig. 1). During soil sampling, the planned regular sampling was not possible to be exactly followed because of topographic problems and mountainous terrain of this study area, but care was taken to preserve a uniform distribution of sampling sites as possible. A hand-held Global Positioning System (GPS) was applied for navigation with geographic coordinates (latitude and longitude). The map of the final sampling locations was shown in Fig. 1. For each sampling site, >500 g samples were collected using a stainless steel auger with scale and then placed in a polyethylene bag for transportation to the laboratory.

The soil samples were naturally dried to a quality of basic stability in the indoor ventilated and shade place of the laboratory. Then, the samples were crushed and grinded successively until soil sieved through a 2 mm nylon sieve to remove stones, dead organisms and coarse debris. Afterwards, 50 g preliminary processed soil was grinded with agate mortar and further sieved through a 0.15 mm nylon sieve. The second sieving soil samples were stored in brown polyethylene bags and