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Spatial autocorrelation analysis of monitoring data of heavy metals in rice in China

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ABSTRACT

The aim of this study was to analyze the spatial distribution of heavy metal monitoring data in paddy rice to provide a scientific basis for food safety risk assessment and suggestions for possible risk management. In this study, the spatial distribution data of heavy metals (cadmium, lead, total arsenic, total chromium, and total mercury) in rice collected in the main production provinces of China were analyzed by multidimensional visualization and aggregation analysis. The spatial correlation of elemental contamination was also identified. Monitoring data of cadmium, lead, and arsenic content in different varieties of rice was compared with their mean confidence intervals. Results showed that cadmium content in rice was higher than the limit value in some areas of Hunan, Sichuan, Guangxi and Anhui Provinces in China. With respect to other heavy metals, a small area of Sichuan Province experienced lead levels in rice higher than the limit value. Also, the arsenic level in rice was higher than the limit value in Jiangxi Province, a northern area of Liaoning Province and most parts of Guangzhou and its surrounding areas. In contrast, chromium was only detected at excessive levels in southern Sichuan Province. In addition, a small part of the eastern Sichuan Province was found to have excessive levels of arsenic. Moran's I index of cadmium, arsenic, chromium, lead, and mercury in rice was 0.50, 0.55, 0.21, 0.09, and 0.05, respectively, which revealed a spatial autocorrelation. Overall, there was moderate aggregation of cadmium and arsenic in the monitoring areas, while lead, chromium and mercury showed low aggregation. Geographically for the provinces, the high aggregation of cadmium in rice was evident in Hunan and Jiangxi Provinces and Guangdong border areas. The arsenic in Jiangxi Province and border areas of Jiangxi and Guangdong Provinces also showed high level of aggregation. Meanwhile, the parameter testing of the samples showed that the concentration of cadmium and lead were significantly higher in late Indica rice compared to early Indica rice, while the arsenic and chromium showed the opposite effect. In view of the high levels of certain heavy metals in rice in some provinces, more refined dietary intake assessments of rice as consumed are necessary to determine if populations are exposed to levels that exceed the health-based guidance levels.

1. Introduction

Rice is the main food of more than half of the world's population, especially in developing countries in Asia, for which rice provides more than 70% of the daily caloric intake from food (Qian et al., 2010). Therefore, the quality and safety of rice are closely related to health and the quality of life. Pollution and natural contamination of rice with heavy metals or other potential toxins are important factors that may threaten health and safety, i.e., whether or not the rice can be consumed without risk. Therefore, strict monitoring and control of potentially toxic elements in rice are very important.

Over the past three decades, rice cultivation in China has been expanding, reaching 303,117 million hectares by 2013 (Dong et al., 2015; Zhong, 2015). However, assessing heavy metal contamination of rice is complicated, because the distribution patterns of contamination of the elements in rice are different depending on both the kinds of rice and the region in which it is grown. The heavy metal content in the soil and irrigation water, pollution from surrounding industry and agriculture practices can significantly affect the content of heavy metals in rice (Duan, Gao, Jiang, & Wu, 2005;

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Zhao et al., 2009, 2010). The absorption characteristics of elements in rice also vary greatly due to different elements and different varieties of rice. Under certain conditions when there is a mixture of pollutants, the absorption of heavy metals in rice will increase (Wang & Liang, 2000). Some of these challenges have led to difficulties in the formulation of rice pollution management policies, which, in turn, makes it difficult for the relevant regulatory authorities to develop a localized management and monitoring policies.

Heavy metals enter the body in a variety of ways, with the primary routes being inhalation (breathing), dermal contact, and diet intake. Relative to respiratory and dermal contact, dietary intake is considered the major source of exposure due to large amount of food consumed, dietary types and content of heavy metals in food, and other factors (Crasnuck & Scholz, 2005). Heavy metal pollution in the environment has a strong correlation with the presence of these harmful elements in rice and has become the focus of ecological and health concerns. The intent of this research work was to use monitoring data of heavy metals in rice from the main production areas of China to analyze the contents of cadmium, lead, arsenic, chromium, and mercury in paddy rice both by location (i.e. space) and by species. Furthermore, we try to provide clues for the sources and trends of rice contamination and provide a scientific basis for risk assessment and the formulation of heavy metal management and monitoring policies.

2. Material and methods

2.1. Data source

The data contained in this paper were collected from a 2015 monitoring study in China that covered 18 cities in 19 provinces. The main monitoring data included cadmium, lead, arsenic, chromium and mercury of rice and its products. The monitoring results included a total of 2151 samples and a number of rice varieties, including early Indica Rice, late Indica Rice and Japonica Rice.

2.2. Method

ArcGIS (geographic information system software; Esri, Redlands, California, USA) was used to produce a thematic map showing areas where potentially harmful contamination occurred (Mitchel, 2005). To effectively monitor the concentration of heavy metals in rice and reduce possible adverse health impacts, it is necessary to understand the geographical distribution of the compounds of interest. This study uses the following methods:

(1) Global spatial autocorrelation analysis mainly uses Moran's I (Griffith, 1987) coefficients to reflect the degree of spatial clustering of attribute variables in the whole study area. The application software *OpenGeoDa* (open source; geodacenter.github.io) is used for cluster analysis. The global spatial autocorrelation analysis examines whether a given region is a cluster region. Moran's I coefficient is used to reflect the degree of clustering.

Moran's I is expressed as (Harry & Prucha, 2001):

$$I = \frac{n\sum\limits_{i=1}^{n}\sum\limits_{j=1}^{n} w_{ij}(x_i - \bar{x})(x_j - \bar{x})}{\left(\sum\limits_{i=1}^{n}\sum\limits_{j=1}^{n} w_{ij}\right)\sum\limits_{i=1}^{n}(x_i - \bar{x})^2} \tag{1}$$

where n represents the number of regions of the study object space; x_i represents the attribute value in the i-th region, x_j represents the attribute value in the j-th region, $\bar{x}$ represents the average value of the attribute value of the studied region; x_{ij} represents the spatial weight matrix, and i is not equal to j.

The *Z*-score of Moran's I is:

$$Z = \frac{I - E(I)}{\sqrt{Var(I)}} \tag{2}$$

where E(I) represents the expectations of Moran's I, Var(I) represents the variance of Moran's I. When $|Z| > 1.96$, $p < 0.05$, we reject the null hypothesis, and spatial autocorrelation (Getis & Ord, 1992) is present. Moran's I coefficient is [-1, 1]. When the value is greater than 0, it indicates that there is a spatial positive correlation between the study area. Values indicate a stronger spatial autocorrelation with numbers closer to 1. When values less than 0, values approaching -1 indicates that the spatial negative autocorrelation is stronger. Random distribution exists when the value is closer to zero.

(2) Localized Moran's I coefficient (also called LISA-local indicator of spatial autocorrelation) and local Getis coefficient (G_i) were used to reflect the specific accumulation area and spatial aggregation of harmful elements in rice.

The localized Moran's I coefficient provides the determination of the correlation of each spatial unit. For the i-th region, LISA of Moran's I is defined as follows (Zhang, Luo, Xu, & Ledwith, 2008):

$$I_i = \frac{x_i - \bar{x}}{S^2}\sum_{j=1}^{n} w_{ij}(x_j - \bar{x}) \tag{3}$$

where i≠j, and $s^2 = \frac{1}{n}\sum_{i=1}^{n}(x_i - \bar{x})^2, \bar{x} = \frac{1}{n}\sum_{i=1}^{n}x_i$.

The LISA statistic of Moran's I was tested using the *Z*-score test:

$$Z = \frac{I_i - E(I_i)}{\sqrt{Var(I_i)}} \tag{4}$$

The LISA coefficient is used to determine the presence or absence of spatial clustering in the heavy metal elements. A LISA coefficient >0 indicates the existence of a spatial positive correlation between the local space unit and the adjacent spatial unit, which is expressed as “high-high” or “low-low”; a LISA coefficient <0 indicates negative correlation between the performance of the “low-high” or “high-low” aggregation.

The Getis coefficient is used to detect the “hot” or “cold” areas of the spatial distribution of harmful elements. The local Getis coefficient is defined as:

$$G_i^* = \frac{\sum\limits_{j=1}^{n} w_{ij}x_j - w_i^*\bar{x}}{s\sqrt{\left(ns_{1i} - w_i^{*2}\right)\Big/(n-1)}} \tag{5}$$

where $w_i^* = \sum_{j=1}^{n} w_{ij}, s_{1i} = \sum_{j=1}^{n} w_{ij}^2$.

When $G_i^* > 0$, the aggregation area is a hot spot area with high value (a hot zone with respect to one of the classes of harmful elements). If $G_i^* < 0$, the aggregation area is a low-value area; i.e., a so-called “cold region” with respect to harmful element pollution. Values close to or equal to zero indicate no accumulation in a given area.

(3) The distribution of detection values for different heavy metal elements in rice was estimated. Heavy metal content of many