



Tracing of heavy metal sources and mobility in a soil depth profile via isotopic variation of Pb and Sr



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ABSTRACT

This study aimed to investigate soil profiles from four localities in Quanzhou, southeast China, namely an industrial zone (P51 profile), an agricultural zone (paddy field; P105 profile), a commercial residential area (P115 profile), and a traffic zone (P16 profile). Fe, Mn, Zn, Pb, Ni, Cr, and Cu concentrations were determined using inductively coupled plasma-mass spectrometry. Community Bureau of Reference sequential extraction was used for heavy metals speciation. The pollution sources in the soil profiles were examined using a Pb and Sr isotope tracing technique. The results showed that Pb and Mn were present throughout the P105 soil profile, while Zn and Cu in the surface soils exceeded background values. Pb pollution occurred deep in the soil profile, while Cu and Zn were enriched in surface soils, but not in the lower soils. In the P16 profile, Mn and Ni occurred throughout the entire soil profile, Pb occurred in the lower 100 cm of the profile, and Zn occurred in the topsoil. These values exceeded background levels, which was possibly due to contamination by traffic. In the P115 profile, only Mn in surface soils exceeded background levels, and there was only slight overall contamination by other metals. Heavy metals speciation analyses demonstrated that Mn, Cr, and Pb largely occurred in the form of non-residual fractions. Pb isotope tracing showed that sludge and bedrock parent materials were the main sources of Pb in the industrial and agricultural samples. Combining Sr and Pb isotope tracing confirmed the Pb isotope analysis.

1. Introduction

Anthropogenic contamination in soils represents a significant risk to human health when mobilised into drinking water and the food chain through plants (Domy, 2001). Tracing sources of heavy metals pollution may improve the ability to effectively measure heavy metals pollution in soil. Therefore, scholars have focused on researching heavy metals pollution in soil.

At present, there are three main discriminant methods for examining pollution sources, namely the statistical method (Micó et al., 2006; Zhao et al., 2007a; Martínez et al., 2008; Franco-Uría et al., 2009), geochemical mapping method (Nanos and Rodríguez-Martín, 2012; Khalil et al., 2014; Guagliardi et al., 2015), and isotope tracer method (Kayhanian, 2012; Walraven et al., 2015). The first two methods are mainly to analyse and evaluate the total amount of heavy metals and their chemical forms in soil and plants, but they cannot effectively identify the soil-plant heavy metal multi-source system. In

addition, these two methods require a large sampling area, and the workload is very large; thus, there are clear limitations in tracing pollution sources. Hence, it is impossible to distinguish effectively when the multi-source system is mixed. In contrast, isotopic technology has an advantage in tracing multi-source pollution.

However, the isotopic composition of different elements is also widely used as a clear indicator for different sources of origin, including in soil (Faure, 1986). Pb is a typical heavy metal that constitutes a severe threat to human health and the environment, and can be detected by electrochemical sensors (Zhu et al., 2014; Zeng et al., 2016). More importantly, due to the isotopic signature of Pb, Pb isotopes often serve as tracers of environmental pollution (Bove et al., 2011; Grezzi et al., 2011; Klaminder et al., 2011; Morton-Bermea et al., 2011; Zhang and Luo, 2011; Kayhanian, 2012). For example, the isotopic composition of Pb in soils reflects the mixing of all possible Pb sources (Komárek et al., 2007). Similarly, analysis of Sr isotope ratios is a powerful tool in provenance studies and has been successfully applied

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to inorganic (Graham et al., 2000) and organic samples (Barbaste et al., 2002). Methods for analysing Sr isotope sources have a wide range of applications (Prohaska et al., 2005; Bullen and Bailey, 2005).

Due to the fairly regular vertical distribution of heavy metals in soil profiles, heavy metals pollution is mainly caused by impacts on surface soil, with fewer impacts on the subsoil. As time passes, heavy metal pollutants from farming activities, under the influence of eluviation, consistently move deeper, thereby contaminating the subsoil. Therefore, only studying the total heavy metals in the surface soil is insufficient to clarify the pollution characteristics of the soil. This part of the soil profile can help assess heavy metals migration, but it is not sufficient to assess how heavy metal pollutants from both natural and human sources reach lower depths. It is difficult to resolve how both the geological background and human factors influence their interaction, which makes it very difficult to determine the complete source of soil contamination (Chu and Ma, 2012). It is in this context that the present study of both surface soils and subsoils in four soil profiles from southeast China was conducted. The variation in heavy metals concentrations and Pb and Sr isotope compositions at different depths were used to distinguish different sources of Pb and Sr.

Quanzhou is located in the southeast of Fujian Province on the west coast of the Taiwan Strait. The region has experienced rapid economic development as a result of China's reformation and more open society, with industrialisation and urbanisation having occurred relatively quickly. The region's climatic conditions (subtropical climate), geological background (granitic region), and industrial structure are representative of many cities in southeast China. With the rapid development of industrialisation and urbanisation, soil pollution in Quanzhou has become increasingly serious in recent years (Ye et al., 2014), thereby posing increasing difficulties for the Quanzhou government and people.

In this study, the concentration and speciation of heavy metals (Cr, Cu, Zn, Ni, Pb, Fe, and Mn) in soil profiles from different functional areas (an industrial zone, agricultural zone, commercial residential area, and traffic zone) of Quanzhou, southeast China, were examined. Sr and Pb isotope compositions and the main sources of heavy metal pollutants in the profiles were also analysed in order to provide data for the analysis of sources of metals pollution of urban soils.

2. Materials and methods

2.1. Sample collection and preparation

Vertical profiles through the soil horizons were collected by digging pits down to the bedrock. Samples were collected using a cylindrical layered profile sampler, which collected soil with a thickness of 20–30 cm per layer. They were collected at four locations, namely an industrial zone (P51), an agricultural zone (paddy field; P105), a commercial residential area (P115), and a traffic zone (P16). A total of 25 samples were carefully stored in clean plastic vessels before processing and analysis and were labelled with the sampling location, date, number, and name of the person conducting the sampling.

In the laboratory, the soil samples were air-dried in a controlled clean environment, and litter, roots, animal residues, and other debris were removed. The soil was spread into a thin layer and then repeatedly turned to accelerate drying. The samples were then ground with an agate pestle and mortar and sieved with a 200-mesh nylon sieve. The material that passed through the sieve (fine fraction) was stored in sealed polyethylene bags for future use, while the retained sieve materials (coarse fraction) were placed back into the original bags.

2.2. Sample analysis

2.2.1. Physical and chemical parameters testing

The pH measurements were conducted with a soil and water ratio of 5:1, stirring for 1 min, and settling for 30 min. Then, the pH of the

supernatant was measured using a pH meter with a glass electrode (Thermo Scientific Orion 828). The average value was used as the result of the measurement.

The total organic carbon (TOC) was measured using a TOC analyser (Multi N/C-2100, Analytik Jena, Germany), and CaCO₃ was used to draw the TOC standard curve. Before the sample was measured, 1 mol/L HCl solution was pre-acidified to achieve the removal of inorganic carbon. Two to three soil samples with a mass of about 10–50 mg were placed in the sample boat and sent to the high temperature combustion furnace using a special iron hook. The mean value was finally obtained when the relative deviation was < 10%.

2.2.2. Total heavy metals testing

Total heavy metals in the soil were extracted using the method described by Adamo et al. (2005). Sample solutions and reagent blanks were analysed for Cr, Mn, Ni, Cu, Zn, Pb, and Fe using inductively coupled plasma-mass spectrometry (ICP-MS) (ELAN9000, Perkin-Elmer, USA) at the Chinese Academy of Sciences. Background corrections and matrix interference were monitored throughout the analyses. All experiments involving the soil samples were conducted in duplicate. To enhance the detection of heavy metals in the soils during the total extraction method, standard material ESS-3 (GSBZ50013-88) was used for parallel sample analysis. The analytical results of the selected metals of interest showed good agreement between the reference and analytical values of the reference materials. In addition, the recovery rates for the selected metals from the standard reference materials were approximately 91–103%, except for Cr (81.92%).

2.2.3. Community Bureau of Reference sequential extraction

A modified Community Bureau of Reference (BCR) three-step sequential extraction procedure (Ure et al., 1993) was used to obtain the species of heavy metals (Cr, Ni, Cu, Zn, Mn, Pb, and Fe). Residual fraction digestion was completed using a microwave-assisted acid digestion procedure. After every successive extraction, the extraction solution was centrifuged at 3000 rpm for 10 min and then filtered. The fractions extracted were as follows: acid-soluble fraction (F1), reducible fraction (F2), oxidisable fraction (F3), and residual fraction (F4). Heavy metal concentrations were determined using ICP-MS. Blanks were included in each step of the analysis. The results were expressed as mg/kg dry soil for the total analysis and as species percentages. All pieces of apparatus were pre-cleaned with deionized water in ultrasonic waves for 30 min.

2.2.4. Measurement of Pb isotopes

The Pb isotope ratio samples were prepared in the Analytical Laboratory of Beijing Research Institute of Uranium Geology. Pb separation and purification were conducted using the analytical procedure DZ/T 0184.12-1997. The Pb isotope composition was measured using a VG354 thermal ionization mass spectrometer. Throughout the process, Pb certified reference material with an order of magnitude of 10⁻⁹ (NBS981) from the National Bureau of Standards was used to check the accuracy of isotopic relationships, calibrate instruments, and conduct isotopic correction. Pb isotopes were measured using the method described by Hu et al. (1999). The ratio of ²⁰⁸Pb/²⁰⁶Pb of NBS981 was 2.1681 ± 0.0008, the ratio of ²⁰⁷Pb/²⁰⁶Pb was 0.91462 ± 0.00033, and the ratio of ²⁰⁷Pb/²⁰⁴Pb was 15.491 ± 0.0097.

2.2.5. Measurement of Sr isotopes

Samples were decomposed in Teflon bombs using HClO₄ + HF and then separated using a cation exchange technique. The Sr isotope analysis of the samples was conducted with a VG354 thermal ionization mass spectrometer at the Analytical Laboratory of Beijing Research Institute of Uranium Geology. Numerous analyses yielded a mean value of 0.71022 ± 0.00004 (2σ) for the NBS 987 Sr standard with a range of 1 × 10⁻⁹ to 2 × 10⁻⁹ g for the blank value of Sr (Hu et al., 2005).

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