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Source discrimination of atmospheric metal deposition by multi-metal isotopes in the Three Gorges Reservoir region, China☆

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Concentrations of heavy metals, as well as isotopic compositions of mercury (Hg) and lead (Pb), in mosses (*Bryum argenteum*) from the Three Gorges Reservoir (TGR) region were investigated to decipher the sources of atmospheric metals in this region. Higher contents of metals (0.90 ± 0.65 mg/kg of Cd, 24.6 ± 27.4 mg/kg of Cu, and 36.1 ± 51.1 mg/kg of Pb) in the mosses from TGR were found compared with those from pollution-free regions. Principal component analysis (PCA) grouped the moss metals into four main components which were associated with both anthropogenic and natural sources. The ratios of Pb isotopes of the mosses (1.153–1.173 for $^{206}Pb/^{207}Pb$ and 2.094–2.129 for $^{208}Pb/^{206}Pb$) fell between those of the traffic emissions and coals. Similarly, the compositions of $\delta^{202}Hg$ (−4.29~-2.33‰) and $\Delta^{199}Hg$ (within ±0.2‰) were comparable to those of the coals and coal combustion emissions from China and India. These joined results of Pb and Hg isotope data give solid evidences that the coal combustion and traffic emissions are the main causes of metal accumulation in the TGR region.
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1. Introduction

The Three Gorges Reservoir (TGR) is the world's largest reservoir with a length of 600 km and an area of 1080 km^2 (Wu et al., 2003) (Fig. 1). In addition to serious natural calamities (e.g., floods and droughts) (Zhu et al., 2017), the TGR region has suffered significant anthropogenic impacts (Han et al., 2015), among which, the contaminations of heavy metals arouse most concern due to their high toxicity (Ye et al., 2011; Han et al., 2015; Bing et al., 2016; Gao et al., 2016; Wei et al., 2016; Wang et al., 2017). It is well known industrial emission, shipping activity, and sewage discharge were predominate sources of metal contamination in the TGR (Ye et al., 2011; Wang et al., 2015; Zhao et al., 2015). Traffic exhaust and coal combustion are important emitting sources of atmospheric deposition of heavy metal in the TGR (Han et al., 2015). However, few studies have focused on the atmospheric deposition of heavy metals in the TGR. Thus, there exists a serious knowledge gap in understanding the sources of heavy metal in the TGR.

With the rapid development of mass-spectrometric technique, non-traditional metal isotopes have been widely employed to trace the sources of heavy metals in various environmental settings, such as air, soil, water, and plants (Zhu et al., 2001; Chen et al., 2005; Weiss et al., 2008; Liu et al., 2011; Wu et al., 2011; Yin et al., 2013a; b). Mercury (Hg) is the only metal to possess both mass-dependent (MDF) and mass-independent fractionation (MIF) (Bergquist and Blum, 2009; Yin et al., 2014), which allows the sources, migration, and transformation of Hg to be effectively traced based on its isotopic compositions (Estrade et al., 2010; Liu et al., 2011; Yin et al., 2013b). However, we still know little about the isotopic composition of atmospheric Hg. For example, samples from different atmospheric environments showed significant variations in Hg isotopes ($\delta^{202}Hg$ varied within 4‰, $\Delta^{199}Hg$ varied within 5‰) (Carignan et al., 2009; Yu et al., 2016). Most of the atmospheric samples in the world have negative $\Delta^{199}Hg$ values, but unusual fractionation of both odd and even Hg isotopes (positive $\Delta^{199}Hg$ and significant MIF of $\Delta^{200}Hg$) in precipitation, snow, and lichen has also been reported recently (Chen et al., 2012). By contrast, the variation of Pb isotopes in different sources was independent of the physical and chemical conditions during both the transportation and transformation processes (Bollhöfer and Rosman, 2001; Cheng and Hu, 2010; Sun and Zhu, 2010). Therefore, Pb isotopes have been successfully applied to trace the sources of Pb pollution in sediments/soils, aerosols, dusts, coals, ores, and fuels (Patterson, 1955;

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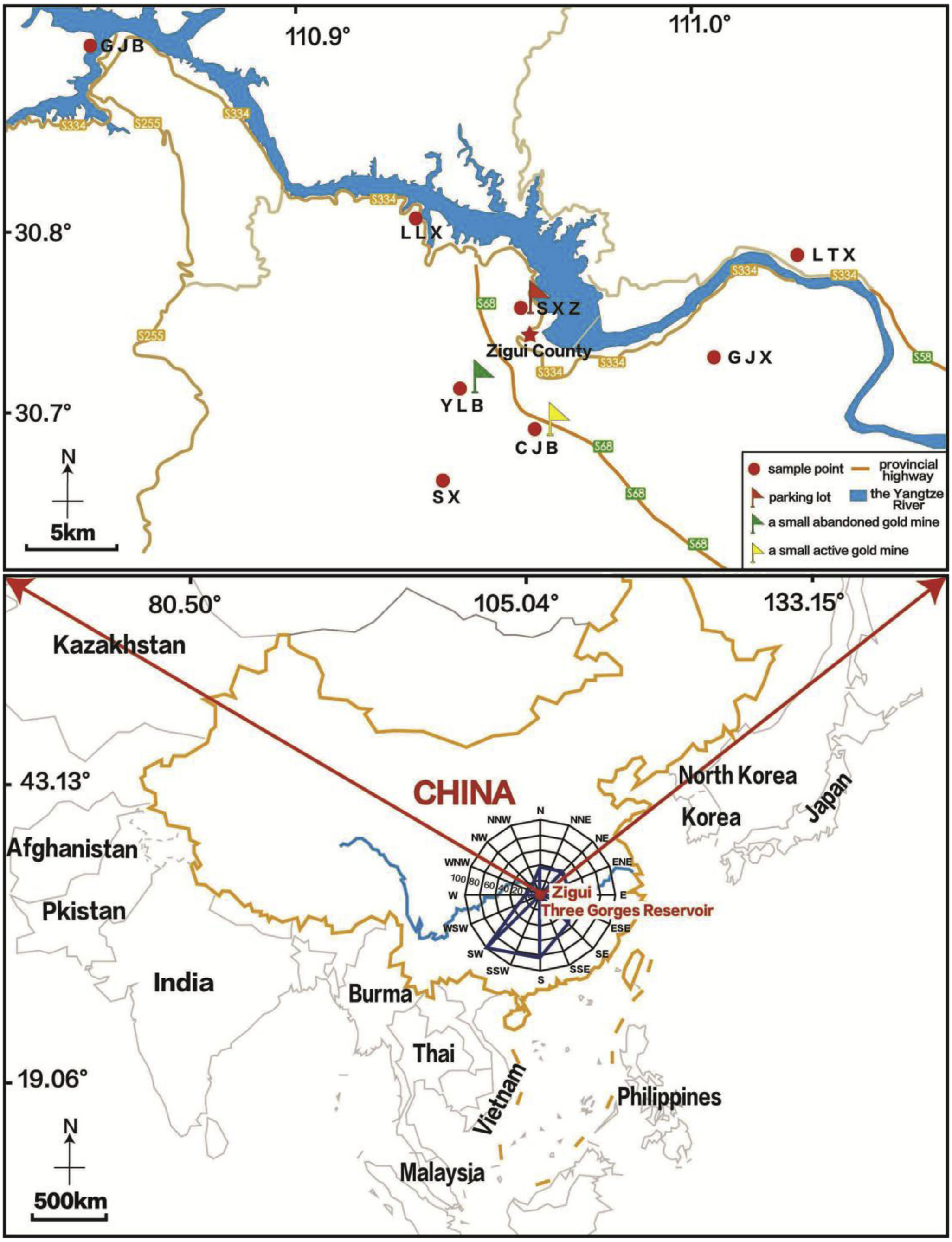

Fig. 1. Map of sampling sites in Zigui, the Three Gorges Reservoir region (TGR).

Chow and Patterson, 1962; Chow and Johnstone, 1965; Shirahata et al., 1980; Bollhöfer and Rosman, 2001; Cheng and Hu, 2010; Zhu et al., 2013; Bory et al., 2014; Shotyk et al., 1998, 2015, 2016; Bi et al., 2017; Xu et al., 2017). But in some cases, the Pb isotopic compositions of different pollution sources (e.g., coal combustion and Zn/Pb ores) are too similar to be clearly discriminated (Bi et al., 2017; Xu et al., 2017). So, the combined use of Hg and Pb isotopes may be more effective and accurate in pollution source tracing than using a single metal isotope alone.

Unlike higher plants, moss plants obtain nutrients mostly from surface adsorption of atmospheric flux and have a limited uptake of minerals from soil due to the absence of root system or cuticle layer (Rühling, 1994). Thus, moss analysis can provide a simpler and cheaper way to study heavy metal fallout compared with conventional precipitation analysis (e.g. practices, aerosols, rainfall, and snow). More importantly, a normally higher level of heavy metals in mosses than in air, rain, and snow makes them insensitive to contamination during the sampling process and the analysis process (ICP Vegetation, 2005). To date, moss plants have been widely used to monitor air pollution (Qiu et al., 2005; Fernández et al., 2015; Kempter et al., 2017), and the isotopes of Pb and Hg in mosses have been considered as effective indicators for tracing pollution sources (Bergquist and Blum, 2009; Bing et al., 2014; Yin et al., 2014; Shotyk et al., 2015). Different species of moss plants have different heavy metal adsorption abilities depending on their mat density (Sucharová and Suchara, 1998). However, in the TGR region, only one dominant moss species (*Bryum argenteum*) has been found, which has been successfully used to monitor

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