



Heavy metal pollution caused by small-scale metal ore mining activities: A case study from a polymetallic mine in South China

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HIGHLIGHTS

- Heavy metal pollution in a small-scale metal ore mining area was investigated.
- Pollution generally decreased with increasing distance from the mining district.
- Cu, Zn, Cd, and Pb in local farmland soils came primarily from the mining activities.
- Heavy metal pollution of local farmland soils posed high potential ecological risk.
- Significant attention needs to be paid to the dumped tailings of small-scale mines.

GRAPHICAL ABSTRACT



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ABSTRACT

Although metal ore mining activities are well known as an important source of heavy metals, soil pollution caused by small-scale mining activities has long been overlooked. This study investigated the pollution of surface soils in an area surrounding a recently abandoned small-scale polymetallic mining district in Guangdong province of south China. A total of 13 tailing samples, 145 surface soil samples, and 29 water samples were collected, and the concentrations of major heavy metals, including Cr, Mn, Co, Ni, Cu, Zn, As, Cd, Pb, and Se, were determined. The results show that the tailings contained high levels of heavy metals, with Cu, Zn, As, Cd, and Pb occurring in the ranges of $739\text{--}4.15 \times 10^3$, $1.81 \times 10^3\text{--}5.00 \times 10^3$, $118\text{--}1.26 \times 10^3$, $8.14\text{--}57.7$, and $1.23 \times 10^3\text{--}6.99 \times 10^3$ mg/kg, respectively. Heavy metals also occurred at high concentrations in the mine drainages (15.4–17.9 mg/L for Cu, 21.1–29.3 mg/L for Zn, 0.553–0.770 mg/L for Cd, and 1.17–2.57 mg/L for Pb), particularly those with pH below 3. The mean contents of Cu, Zn, As, Cd, and Pb in the surface soils of local farmlands were up to 7 times higher than the corresponding background values, and results of multivariate statistical analysis clearly indicate that Cu, Zn, Cd, and Pb were largely contributed by the mining activities. The surface soils from farmlands surrounding the mining district were moderately to seriously polluted, while the potential ecological risk of heavy metal pollution was extremely high. It was estimated that the input fluxes from the mining district to the surrounding farmlands were approximately 17.1, 59.2, 0.311, and 93.8 kg/ha/yr for Cu, Zn, Cd, and Pb, respectively, which probably occurred through transport of fine tailings by wind and runoff, and mine drainage as well. These findings indicate the significant need for proper containment of the mine tailings at small-scale metal ore mines.

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1. Introduction

Heavy metals are natural constituents of the earth's crust, and some of them (e.g., Cu and Zn) are essential for normal metabolic functions in plants, animals, and humans (McLennan, 1999; Saracoglu et al., 2009). They are rarely accumulated to levels that would pose detrimental effect to the environment and ecosystem under natural conditions (Chibuike and Obiora, 2014; Moore and Ramamoorthy, 1984). However, with fast industrialization and urbanization, soil heavy metal pollution caused by anthropogenic activities, such as industrial production, traffic, wastewater irrigation, sludge or municipal compost, pesticides and fertilizers, has occurred widely in many developing countries (Adler Miserendino et al., 2013; Ahmad and Goni, 2010; Deb et al., 2008; He et al., 2017; Hu and Cheng, 2013; Hu et al., 2013; Lin et al., 2018). In the rural regions, mining activities are well documented as one of the most significant sources of soil heavy metal pollution (Dong et al., 2011; Li et al., 2014; Li et al., 2015; Navarro et al., 2008; Rodríguez et al., 2009; Schmitt et al., 2007; Xiao et al., 2017; Zhuang et al., 2009). China holds diverse mineral resources and there are thousands of large- or medium-scale metal mines and numerous small-scale metal mines distributed in various parts of the country (Hu et al., 2011; Li et al., 2014). Small-scale mines have relatively limited exploitable reserves and low annual production rates of ores, and the mining operations may not last long. According to a directive released by the Ministry of Land Resources in 2004, open-pit mines of iron ore with production capacities <0.6 Mt/yr and lead zinc ore mines with production capacities below 0.3 Mt/yr are classified as small-scale mines in China. The mining industry has been, and continues to be an important pillar of the national economy (Shen and Gunson, 2006). Unfortunately, inappropriate exploitation and uncontrolled dumping of the mine tailings, particularly at the small-scale mines, could cause serious heavy metal pollution problems.

Mining activities could pollute the surrounding environment through a range of pathways, including physical disturbance of the landscape, spilling of mine tailings, emissions of dusts contain heavy metals into atmosphere, and generation of large quantity of acidic drainage that contains heavy metals (Cheng et al., 2009; Zhuang et al., 2009). Small-scale mining activities take place throughout the world, and they are particularly widespread in the developing countries in Africa, Asia, and South America, primarily due to poor government oversight and lack of environmental awareness (Bose-O'Reilly et al., 2010; Bose-O'Reilly et al., 2010; Bosso and Enzweiler, 2008; Charles et al., 2013; Fei et al., 2017; Hurtado et al., 2006). Small-scale mines, especially the illegal ones, can be extremely damaging to the environment and often have serious health and safety consequences for the inhabitants in surrounding communities, due to their poor practices in the mining and processing of the target minerals (Hentschel et al., 2008). In recent years, the environmental problems caused by small-scale mining activities have attracted growing attention worldwide (Adler Miserendino et al., 2013; Bosso and Enzweiler, 2008; Deb et al., 2008; Tarras-Wahlberg, 2002). It has been suggested that many of the worst heavy metal pollution problems in the world resulted from the poorly regulated small-scale operations (e.g., artisanal mining) (Harris and McCartor, 2011; Scholz, 2017; Xiao et al., 2017). Over the past decade, the pollution and ecological risk arising from large-scale mining operations have been relatively well documented in China (Zhao et al., 2012; Zhuang et al., 2013), while little attention has been paid to those associated with small-scale mining activities.

There are numerous small-scale mines scattered in the remote mountainous areas of south China, including Hunan, Guangdong, Guangxi, and Jiangxi provinces, which are rich in nonferrous metal resources (Hu et al., 2016). With limited resource reserves, combined with the mountainous terrain, these small mines are unattractive to the large mining companies. Nonetheless, small developers, and often local farmers take the opportunity of exploiting them. The mineral deposits are typically extracted in primitive ways, and the minerals

mined are usually sold to the large mining companies. In contrast to the large state-owned mines that follow standardized mining practices and observe environmental protection regulations, the small-scale mines are privately owned and often pay minimum attention to pollution control (Hilson, 2000; Hu et al., 2016). The wastes and wastewaters produced from the extraction, beneficiation, and processing of minerals are dumped and discharged out of convenience. Unfortunately, such small-scale mining activities, sometimes even illegal, have caught little attention because they are located in the sparsely populated remote regions and are not aware by the general public, even though the pollution caused by them can pose serious threats to the local environment and the health of local residents. Once the operations become uneconomical, the owners often abandon the mines without taking any conservation or protective measures. As a result, the abandoned mining sites could continue to serve as a source of pollution, even after active mining has long ceased.

This study was conducted to (1) evaluate the soil and water pollution caused by small-scale mining activities in China using a recently abandoned mine located in the remote mountainous region of Guangdong province as a case study, and (2) estimate the input fluxes of heavy metals from the abandoned mine to the farmlands in the surrounding areas, and thus demonstrate the significant need for proper containment of the mining tailings. The Yaoposhan polymetallic mining district, which encompasses multiple small-size open-pit mining sites, is located in the remote mountainous part of northern Guangdong province in China (Fig. 1). The production scale of this mine was set at 0.05 Mt/yr of raw iron ore and 0.03 Mt/yr raw lead zinc ore, which merely meet the minimum thresholds for establishing a mining operation. Although the planned production period was 10 years, the actual production lasted no more than 5 years at the mining district. A cluster of mining sites (mostly abandoned) are scattered on the small hills with good vegetation coverage. Except for the mining and farming activities, and very limited vehicle traffic, surface soils in this area are not affected by other obvious anthropogenic sources. Thus, this area is ideal for examining the impact of small-scale mining activities on the surrounding environment. The total contents of heavy metals in the mine tailings, surface soils, and water samples collected in the area were analyzed in this study, and the chemical speciation of heavy metals in selected surface soil samples were also characterized. Sources of the heavy metals in the surface soils of the local farmlands was identified by Pearson's correlation matrix and multivariate statistical analysis, including principal component analysis (PCA) and cluster analysis (CA). Pollution indices were used to assess the overall pollution status of the surface soils, and the potential ecological risk posed by the heavy metals in the farmland soils was also evaluated. Finally, the mass fluxes of heavy metals released into the surface soils from the mining sites were estimated.

2. Materials and methods

2.1. Study area and sample collection

Yaoposhan polymetallic mine (24°31'16", 113°04'29") is located in Shaoguan, close to the border between Guangdong and Hunan provinces of south China (Fig. 1). The primary minerals of this mine include limonite, pyrite, lead zinc ore, and some rare earth minerals. A cluster of open-pit mining sites are situated on the top of the hills. Relatively large-scale exploitation activities started in 2012, while most of the production sites were abandoned in 2014. To date, primitive ore mining still takes place at several sites. The tailings have been piled up on the hillside with no tailing dams established or other protective measures implemented. This area has a subtropical humid monsoon climate, with long-term annual average temperature and average precipitation of 20.2 °C and 1817 mm, respectively. With essentially complete stripping of the surface vegetation, the heavy rainfall in the area, and the resulting surface runoff, caused significant surface erosion within the mining district (Fig. S1). Fine grains of the mine tailings are easily

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