



# Distributions and pools of lead (Pb) in a terrestrial forest ecosystem with highly elevated atmospheric Pb deposition and ecological risks to insects

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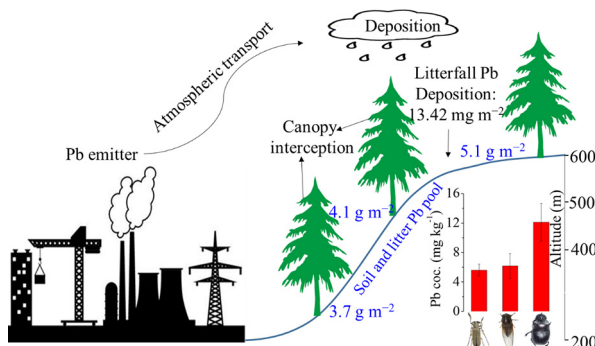
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## HIGHLIGHTS

- Lead concentrations and pools were elevated in soils and vegetation of the subtropical forest.
- Lead concentrations and pools were deepened on TOM pools and altitude.
- Atmospheric deposition was the major source of lead in forest.
- Lead in dung beetle would potentially pose negative influence to predators along food chains.

## GRAPHICAL ABSTRACT



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## ABSTRACT

There is growing interest in how heavy metals in remote ecosystems are elevated and affect environmental health. However, no studies have investigated atmospheric lead (Pb) deposition influences on the Pb bioaccumulation in insects in forests. Here we measure Pb concentrations and pools in forest vegetation, litterfall, organic soil, mineral soil, as well as litterfall deposition fluxes in a region severely affected by atmospheric deposition. We also analyzed Pb in insects which feed in the polluted forest vegetation and litter. Assessment of high Pb loads causing potential ecological risk to insects was also studied. Total Pb pool in the vegetation was 0.12 g m<sup>-2</sup> and annual litterfall deposition flux of Pb was 13.42 mg m<sup>-2</sup>, which was much higher than those in the background areas. Pools of Pb from litter to mineral topsoil averaged 4.3 g m<sup>-2</sup>, which accounted for 97.3% of total pools (biomass + soil) in the forest ecosystem. Pools of Pb in surface soils were correlated significantly with the pools of total organic matter and elevation. Atmospheric deposition was inferred the major source of Pb in the forest ecosystem, which can be supported by the highest Pb concentrations in the moss and overstory foliage. The maximum Pb concentration was showed in the dung beetle (12.1 mg kg<sup>-1</sup>) residing in the soils compared that in the longicorn and of cicada, which would potentially pose negatively influence to predators along food chains.

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## 1. Introduction

Lead (Pb) in terrestrial environments is the result of both natural and anthropogenic activities and high Pb concentrations can affect the environment and ecological functioning. Human activities, including the production and use of Pb batteries, motor vehicle gasoline combustions, coal combustions, sintering and smelting of minerals are the principal source of Pb emissions (Csavina et al., 2014; Duan and Tan, 2013). Previous research indicated that anthropogenic emissions of Pb to the environment have caused regional contamination around the world (Huang et al., 2008; Klaminder et al., 2005, 2006; Miller and Friedland, 1994). The Pb is easily transported to remote ecosystems by emissions from the transportation and industrial sectors (Gross et al., 2012; Li et al., 2017). A large proportion of heavy metal in atmospheric particulates is deposited on foliage and conveyed to the soil by rain wash off and shedding of the foliage (Bringmark et al., 2013; Richardson and Friedland, 2016), because the forest canopy acts as a receptor for different atmospheric contaminants (Richardson et al., 2015b; Zhou et al., 2015a, 2016). In particular, this phenomenon is important in mountain forests, as high mountains can act as orographic barriers against coming air masses (Szopka et al., 2013). Steinnes et al. (2005) suggested that >90% of O horizon Pb was polluted from atmospheric deposition in Norway, even at remote sites in the far north, and their work suggested that similar studies should be done in other parts of the world to objectively assess the anthropogenic contribution of Pb to surface soil. Since the precise information of Pb concentrations in forest components is lacking, our understanding of the impact of anthropogenic activities on the environment is uncertain.

Investigations in terrestrial systems have identified the forest as large sinks of atmospheric Pb, so organisms inhabiting the forest ecosystem are usually exposed to high Pb concentrations (Šerić et al., 2007). Organisms have a demonstrated ability to effectively bio-accumulate Pb from environmental exposure (Aissaoui et al., 2017; Conti et al., 2017; Ernst et al., 2008; Lindqvist and Block, 1997; Mensens et al., 2017). Knowledge of the Pb concentrations in the different species of insects is very useful for ecological risk assessment because Pb in insects pose serious risks of secondary poisoning of vertebrate predators (Conti et al., 2017; Šerić et al., 2007). Insects are important components of food chain in the forest ecosystem and have been proposed as bioindicators of heavy metal contaminations (Conti et al., 2017). Moreover, these insects are situated at the lower trophic levels of food chains and preyed upon by several vertebrate animals, resulting in a migratory route for the bioaccumulation of pollutants in food chains (Andre et al., 2010; Conti et al., 2017; Dai et al., 2004; Ernst et al., 2008; Hsu et al., 2006; Pižl and Josens, 1995). Previous studies showed that several invertebrates accumulated large amounts of Pb and resulted in ecological poisoning (Conti et al., 2017; Šerić et al., 2007), although the synthesis of metallothioneins in some animal tissues was usually induced to achieve Pb homeostasis under Pb stress due to the detoxification of Pb (Zhou et al., 2015b).

China is the largest developing country worldwide. With the rapid development of economy during the past four decades, large amounts of Pb has emitted to the atmosphere (Duan and Tan, 2013). According to the study by Tian et al. (2015), the proportion of Pb in leaded gasoline emitted to the air is estimated at about 76% of total atmospheric Pb emissions for the period before 2000. Based on the Pb concentration in the gasoline, the 64 years between 1949 (the founding of PR China) to 2012 can be divided into two phases: the leaded gasoline period (1949 to 1990: gasoline with high Pb concentration ( $0.64 \text{ g L}^{-1}$ ); 1991–2000: gasoline with low Pb concentration ( $0.35 \text{ g L}^{-1}$ )), and the unleaded gasoline period (2001 to 2012,  $0.005 \text{ g L}^{-1}$ ). Therefore, the anthropogenic emission of Pb experienced two fluctuations from 1949 to 2012, which occurred in 1991 with the decreasing rate of 26% and occurred in 2001 with the decreasing rate of 61.6% in 2001. However, along with the rapid increase of vehicle volume and oil consumption as well as the rapid industrial development, a substantial increase was

once again experienced from 7747.2 t in 2001 to 14,397.6 t in 2012, at an annual average growth rate of about 5.8% (Tian et al., 2015).

In this study, the Pb concentrations in the litterfall, litter, soils, vegetation tissues and insects were investigated at Tieshanping Forest Park, southwestern China. Our previous studies showed that the forest has received elevated atmospheric mercury deposition (Zhou et al., 2015a, 2016), but no studies attempt to study other pollutants in this area. To the best of our knowledge, the current study is the first to investigate atmospheric Pb in foliage and soil compartments in combination with measurements of Pb in insects. The objectives are (1) to characterize the possible source of forest Pb distribution in the litter, organic soil and mineral topsoil horizons; (2) to explore the Pb of litterfall depositions and stocks in the vegetation; (3) to assess the potential ecological risk to insects. We hypothesized that the atmospheric Pb depositions were the main source of Pb contamination in the forest catchment and higher Pb depositions has resulted in ecological risks to local animals.

## 2. Materials and methods

### 2.1. Study area

Our study was carried out in an experimental forest catchment (44 ha) within Tieshanping Forest Park ( $29^{\circ}38'N$ ,  $104^{\circ}41'E$ ), a protected area situated at about 20 km to the northeast of the Chongqing City center (Fig. S1). The catchment is one of Sino-Norwegian multidisciplinary Integrated Monitoring Program on Acidification of Chinese Terrestrial Systems (IMPACTS) project study sites. The rainy season extends from May to October (75% of the rainfall) and mainly controlled by the Indian monsoon. The mean annual temperature is  $18.2^{\circ}C$  and the annual rainfall is 1028 mm (Zhou et al., 2017). The primary soil type in the catchment is Haplic Acrisol according to FAO. The catchment is mainly dominated by the tree species of Masson pine (*Pinus massoniana* Lamb.) and China fir (*Cunninghamia lanceolata* Lamb.), camphor (*Cinnamomum camphora* (L.) Presl) and Schima (*Schima superba* Gardn. et Champ.) were the non-dominant tree species. Chongqing is an important industrial region in southwest China and consumes large amounts of fossil fuels. The mean concentration of Pb in atmosphere was  $149 \text{ ng m}^{-3}$  in 2012 (Tan, 2013) and Pb contaminations were regarded as serious environmental burdens (Duan and Tan, 2013; Tao et al., 2003; Wu et al., 2011).

### 2.2. Vegetation

To characterize the Pb distribution in the vegetation (overstory, shrub and herb layer), three sample plots of  $10 \times 10 \text{ m}^2$  were established in January 2015. The plots were located on the top of the mountain with the elevation of 500 m a.s.l. and located at a flat terrain with slope  $< 10^{\circ}$ . Samples of overstory stand were collected from Masson pine, and samples of shrub were collected from Schima. Samples from the two species included root, bole wood, branch, bark and foliage in each subplot. Briefly, foliage and branch were collected in the middle canopy, 5–7 m and 2–3 m above the ground for overstory and shrub, respectively, using a stainless steel pole saw. Bole wood and bark were sampled at breast height using a 4 mm increment corer and clean knife blade, respectively. Roots were collected by spade at the 30–50 cm of soil profile. All the vegetation samples were collected in triplicate per plot.

In January, biomass of understory including those of herbs, shrubs and saplings (diameter at breast height  $< 3 \text{ cm}$ ), were completely harvested using  $1 \text{ m}^2$  quadrates in triplicate at each of the three plots and then weighed, when the biomass of the understory were the maximum in the year. The representative understory species of Old World forkedfern (*Dicranopteris dichotoma* Bernh.) and chain fern (*Woodwardia japonica* Lectin.) were sampled to determine the Pb pools, which were the most abundant in the study area. Additionally,

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