Science of the Total Environment 613–614 (2018) 331–341

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

# Reconstruction of atmospheric trace metals pollution in Southwest China using sediments from a large and deep alpine lake: Historical trends, sources and sediment focusing

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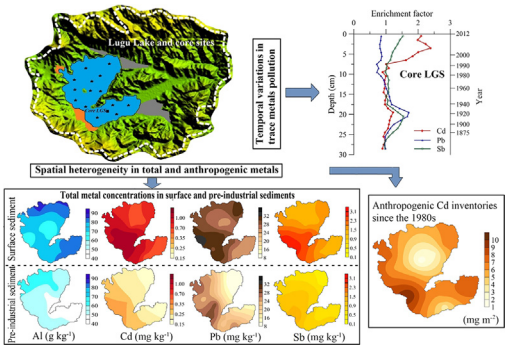
## HIGHLIGHTS

- Atmospheric trace metals pollution over 150 years in Southwest China is reconstructed.
- Cd and Sb were typical pollutants since the mid-1980s while Pb pollution peaked in the 1920s.
- Metals pollution was primarily associated with non-ferrous metal smelting emissions.
- Spatial heterogeneity of metals was less influenced by in-lake sediment focusing.
- Spatial accumulation of anthropogenic Cd and Sb was affected by in-watershed transport.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 13 June 2017
Received in revised form 24 July 2017
Accepted 8 September 2017
Available online xxxx

Editor: D. Barcelo

*Keywords:*
Trace metal pollution
Temporal patterns
Sources
Sediment focusing
Lugu Lake
Southwest China

## ABSTRACT

Atmospheric pollution, one of the leading environmental problems in South and East Asia, and its impact on the terrestrial environmental quality remain poorly understood particularly in alpine areas where both historical and present-day mining and smelting operations might leave an imprint. Here, we reconstructed atmospheric trace metals pollution during the past century using core sediments from a large and deep alpine lake in Southwest China. The implication of in lake and/or in watershed sediment focusing in pollution quantification is discussed by analyzing 15 sediment cores. Factor analysis and enrichment factor indicated Cd, Pb and Sb as the typical pollutants. Distinct peaks of Pb and Sb pollution were observed around the 1920s, but little Pb pollution was detected in recent decades, different from other studies in similar regions. Cadmium pollution was observed until the mid-1980s synchronized with Sb. The distinctive variations in atmospheric trace metal pollution process in Southwest China highlight the regional and sub-regional sources of metal pollutants, which should be primarily attributed to non-ferrous metal smelting emissions. Both natural and anthropogenic metals showed wide concentration ranges though exhibited similar temporal trends in the 15 cores. Spatial variations of anthropogenic metals were influenced by the in-watershed pollutants remobilization, whereas, natural metals were regulated by the detrital materials in the sub-basin. In-lake sediment focusing had little influence on the spatial distributions of all metals, different from the traditional sediment focusing pattern observed in small lakes. Anthropogenic Cd accumulation

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http://dx.doi.org/10.1016/j.scitotenv.2017.09.073
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in sediments ranged from 1.5 to 10.1 mg $m^{-2}$ in a specific core with an average of 6.5 mg $m^{-2}$ for the entire lake, highlighting that a reliable whole-lake pollutant budget requires an analysis of multiple cores. Our study suggests that the management of aquatic ecosystem health should take the remobilization of in-watershed stored pollutants into consideration especially under increasing human perturbation.

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## 1. Introduction

Trace metals such as Cd, Cu, Pb, Sb and Zn, also called potentially harmful elements, are common contaminants of concern in ecosystems due to their persistence, toxicity, bioaccumulation and biomagnification in food chains (Vicente-Martorell et al., 2009; Birch and Apostolatos, 2013). These metals if released into the atmosphere can be transported over long distances with aerosols and/or particulate forms (Hur et al., 2007; Colbeck, 2008; Spiro et al., 2013). Because of these reasons the contaminants can accumulate in remote environments not directly related to local human activities, such as in oceans (Birch, 2017), alpine areas (Yang et al., 2010; Bacardit et al., 2012; Rose et al., 2012; Bing et al., 2016), and polar regions (Shotyk et al., 2003; Hur et al., 2007). Emissions from fossil fuel combustion, use of leaded gasoline and smelting are considered as the primary sources of atmospheric trace metals pollution. Diversity in sources and process of economic development affects the temporal and spatial patterns of local, regional and even global atmospheric trace metals pollution (Klaminder et al., 2008; Bindler et al., 2011; Liu et al., 2013). In Europe and North America, as former emission and pollution hotspots, atmospheric trace metals pollution decreased remarkably in recent decades coinciding with the implementation of strict environmental protection measures (Bindler et al., 2004; Sarkar et al., 2015). However, in South and East Asia, including China, which is a major non-ferrous metal production center and is the largest consumer of fossil fuels in the world (Tian et al., 2015), serious atmospheric pollution from trace metals has occurred in recent years coinciding with rapid economic development (Yang et al., 2010; Liu et al., 2013; Bing et al., 2016). But the knowledge of temporal trends, deposition fluxes and spatial characteristics of atmospheric metals is still limited, especially in remote regions that were previously considered pristine (Yang et al., 2010; Wang et al., 2010; Bing et al., 2016). This makes the understanding of the influence of anthropogenic activities on regional and continental environmental quality unclear.

Lake sediment is an important sink of atmospheric trace metals (Bindler et al., 2011). With proper correction for geochemical background, vertical variations of sedimentary trace metals can be used to reconstruct the atmospheric pollution process and to decipher the anthropogenic emission characteristics (Shotyk et al., 2003; Yang et al., 2010; Liu et al., 2013). Nevertheless, recent studies have indicated that the quantitative reconstruction of atmospheric trace metals pollution using sediment cores can be complicated by in-lake and/or in-watershed storage and transport of metal-enriched sediments, termed as "sediment focusing" (Bindler et al., 2001; Rose et al., 2012; Engstrom and Rose, 2013; Yang, 2015). These researchers gave a few examples of such effect on pollutants distribution in lakes elsewhere, such as Härsvatten in Sweden (Bindler et al., 2001), Lochnagar in Scotland (Engstrom and Rose, 2013), Légunabens in the French Pyrenees, Plan and Vidal d'Amunt in the Spanish Pyrenees (Bacardit et al., 2012). Multicore analysis was considered as an effective solution in improving the understanding of pollution dynamics in lakes and overcoming the influence of sediment focusing towards quantitative reconstruction of trace metals pollution (Rippey et al., 2008; Engstrom and Rose, 2013). Such analyses have been performed in small and shallow lakes referred to above. In contrast, widespread large, deep and/or morphometrically complex lakes throughout the world and in China (Cael and Seekell, 2016) may also constitute an important archive in interpreting regional environmental evolution including atmospheric trace metals pollution (Wang et al., 2010; Vogel et al., 2010). Nevertheless, the regulating factors of sediment focusing and its influence on the reconstruction of metal pollution record are seldom studied in this type of lakes.

The Yunnan-Guizhou Plateau, located in the Southwest China, is one of the important non-ferrous metal production centers in China (ECYCNMI, 2007). The production of non-ferrous metals in Yunnan Province reached over $2.0 \times 10^6$ tons $yr^{-1}$ in recent years from $<1 \times 10^{-4}$ tons $yr^{-1}$ in the early 1950s (ECYCNMI, 2007). Previous studies have indicated that the non-ferrous metal ore mining and smelting have been the most important source of trace metals (e.g., Cd, Pb and Zn) pollution in Southwest China according to sedimentary records from lakes Fuxian (Liu et al., 2013), Yangzong (Zhang et al., 2012), Erhai (Li et al., 2016) and Hongfeng (Wang et al., 2015). However, additional sources of pollution, e.g., industrial and municipal discharges, to the previously studied lakes in this region compromises the sedimentary records and makes it difficult to distinguish the contribution from atmospheric deposition. The broader understanding of historical process, sources of atmospheric trace metals in alpine regions of Southwest China and their similarity and/or dissimilarity to other regions remains limited (Liu et al., 2013; Bing et al., 2016), which may hamper understanding the influence from this emission hotspot on the regional, continental and even global atmosphere quality and terrestrial environmental quality.

The objectives of this study are to: (1) reconstruct historical trends of atmospheric trace metals pollution in alpine region of the Southwest China, (2) identify potential sources of anthropogenic metals, and (3) explore the spatial heterogeneity of metals and response to in-lake and in-watershed processes and its influence on quantitative reconstruction of pollutants. To investigate this, we collected 15 sediment cores from a large and deep alpine lake (Lugu Lake) and measured trace (Cd, Cr, Cu, Ni, Pb, Sb and Zn) and major metals (Al, Fe and Ti) in the cores.

## 2. Materials and methods

### 2.1. Study area and sampling

Lugu Lake (27°41′–27°45′N, 100°45′–100°50′E), a typical alpine lake, is located at an altitude of 2685 m on the southeast margin of the Qinghai-Tibet Plateau, Southwest China (Fig. 1a, b). This is one of the deepest freshwater lakes in China with a maximum water depth of ~94 m (mean ~ 40 m) and surface area of ~50.5 $km^2$ (catchment area of ~171.4 $km^2$) (Wang and Dou, 1998). The lake is located in an inter-mountain basin with a maximum altitude of 3870 m. The basin is dominated by limestone, mudstone, sandstone and siltstone (Wang and Dou, 1998; Gao et al., 2010). The Lugu Lake region is mainly affected by the Indian monsoon, and the climate is temperate with distinct dry and wet seasons with a mean annual precipitation of ~1000 mm (Wang and Dou, 1998; Wang et al., 2016). Approximately 80–90% precipitation occurs between May and September. The mean annual air temperature is approximately 12.8 °C with a maximum monthly mean of 19.4 °C in July and a minimum of 5.6 °C in January (Wang and Dou, 1998; Wang et al., 2016). The lake is mainly fed from precipitation and ephemeral runoff. It has one outlet, the Gaizu River, which drains the lake via the Caohai wetland to the southeast into the Yalong River (a tributary of the Yangtze River).

Before the 1900s, the Lugu Lake catchment was characterized by a low population density (<5000 people) (Wan and Guo, 1997). Since the first half of the 20th century, increasing human activities, including deforestation and farming, have been recorded in the catchment

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