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Tracing metal sources in core sediments of the artificial lake An-Dong, Korea: Concentration and metal association

Mansik Choi [a,*], Jongkyu Park [a], Dongjin Cho [a], Dongjun Jang [a], Miseon Kim [a], Jongwoo Choi [b]

[a] Department of Ocean Environmental Sciences, Chungnam National University, Daejeon 305-764, Republic of Korea
[b] Environmental Measurement & Analysis Center, National Institute of Environmental Research, Incheon 404-708, Republic of Korea

HIGHLIGHTS

- Highly enriched metals were found in environmental media near the Zn smelter.
- Metal concentrations in lake sediments were the highest at the core bottom (1980).
- Metal concentrations decreased to the background level from 1980 to 2000.
- Cd and Zn concentrations increased again from 2000 to the present.
- Ore mining and Zn smelter contributed sequentially to lake sediments.

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ABSTRACT

The concentration and source of trace metals in the artificial lake An-Dong, which has widespread abandoned mines and a Zn smelter upstream of the drainage basin, were investigated. Soils (18 ea), stream waters (15 ea) and sediments (15 ea) in the main channel and five tributaries downstream of the Zn smelter towards the lake (~50 km downstream) were collected. And two core sediments were also taken from the middle of the lake. All samples were analyzed for trace metals in bulk and in a 1 N HCl-leached fraction.

Although the soil and stream sediments consisted mostly of sand-sized grains, concentrations of metals (Cu, Zn, Cd and Pb) were very high in all samples, including soils, stream waters and sediments at sites near the Zn smelter. However the metal concentrations decreased rapidly downstream, suggesting that the area of impact of the smelter lies within 5 km. Highly enriched metal concentrations were also found in dated core sediments from the lake; while the highest concentrations of Co, Ni, As, Cu, Zn, Cd and Pb were detected in the bottom of the sediment core (dated 1980) they decreased towards 2000, and only Cu, Zn and Cd concentrations increased again in present-day samples. Since the temporal variation in metal concentrations appeared consistent with historical variation in ore mining and Zn smelter production rates, a model combining the production rates of each was developed, which estimated 3%, 12% and 7% contributions from Zn smelter compared to ore mining production rate to levels of Cu, Cd and Zn, respectively, suggesting the different pathways by different sources. In addition, analysis of Cd/Zn and Cu/Zn ratios showed that contamination from ore mining decreased from 1980 to 2000, and smelting processes were most likely responsible for metal enrichment (Cu, Cd and Zn) from 2000 to the present.

1. Introduction

Trace metals released into aquatic environments, whether originating from natural weathering or from human activities, may harm human health and ecosystems, through bio-accumulation and bio-magnification (Oehme, 1978; Vernet, 1991). Individual countries have developed environmental conservation protocols to decrease metal concentrations in the environment, and remediation works are often undertaken in metal-contaminated areas. Before the implementation of regulations and the onset of remediation works, investigation of the behavior and fate of metals in a target environment should be undertaken.

Metals released into the environment through atmospheric and/or riverine pathways are eventually deposited in sediments within an aquatic reservoir, hence the sediment layer may preserve the history of metal contamination in the drainage basin (Monna et al., 2000; Choi et al., 2007; Cheng and Hu, 2010).

Lake An-Dong is an artificial lake constructed in 1977 by damming to reserve agricultural and industrial water and to generate hydroelectric

* Corresponding author.
E-mail address: mschoi@cnu.ac.kr (M. Choi).

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power. It has a surface area of around 51.5 km^2 and a drainage area of about 1584 km^2. In the northern upstream area, there are widespread abandoned mines. These include Yonhwa mine (1961–1987 yr), Keumho mine (previously Janggun, 1976–present), and the Sukpo Zinc (Zn) smelter, producing Zn products from 1973 to the present, on the site of Yonhwa mine. Ore mining contributes mine wastes and tailings to the environment, while metal-smelting processes are inevitably accompanied by gaseous and particulate emissions, sewage waters and solid wastes (Dudka and Adriano, 1997).

Soils around the mining tails and waste ore piles have been found to be contaminated with Zn and Pb (KRETC, 2005) and sediments in lake An-Dong showed higher concentrations of Cu, Zn, Cd, Pb and Hg than the effective range low (ERL) level of the National Oceanic and Atmospheric Administration (NOAA) (Park et al., 2012a). Furthermore, some fish in this lake, such as bluegill and bass, were found to harbor higher Hg concentrations than those in other rivers and lakes in Korea (Byeon et al., 2010).

It is therefore necessary to identify sources of metal deposits in lake An-Dong, to prevent further contamination and develop remediation protocols. Although Pb isotope characterization in some lake sediments indicated that mining tails and ore wastes contributed to Pb contamination (Park et al., 2012b), more detailed investigation of the extent of smelter waste contamination in the lake is required. Sampling of various monitoring media such as soil, stream waters, sediments in the main stream and tributaries, and temporal variation analysis using core sediments is necessary to develop a comprehensive recovery plan.

This study investigated metal concentrations and metal enrichment in soils downwind of the smelter, in stream waters and sediment downstream of the smelter but upstream of the lake, and in dated core sediments taken within the lake. In addition, the contamination contribution from the smelter as well as the mining was assessed by comparing the contamination history in sediments with the production history of mining and smelting activities.

2. Samples and method

2.1. Samples

A synopsis of metal concentrations throughout environmental media including soils, stream water and sediments sampled along the main stream and in five tributaries between the smelter and the lake in June 2012 is shown in Fig. 1. Two gravity core sediments were collected to investigate the temporal variation of metal contamination in July 2013 (Fig. 1).

Firstly, 18 soil samples at 10 sites were collected at two depths (upper: 2–3 cm from the surface, lower: 10–15 cm). As the average wind direction in this area (An-Dong) over the past 10 years was predominantly northwest and southeast (KMA, 2003–2012), sample sites were chosen downwind of the smelter (up to 16.5 km from the smelter). The five sites closest to the smelter were located at an elevation around 50 m higher than the smelter, placing them above the top of the smelter chimney. All soil samples were largely coarse-grained sand with small amounts of fine-grained materials. The soil samples were collected in vinyl bags and delivered to a laboratory. A total of 15 stream water and sediment samples were collected from sites located approximately midway between the smelter and the lake, from both the main river channel and tributaries. Surface sediments submerged below the water level (2–3-cm depth), were collected from the channel bank by hand using a plastic scoop, and stream waters from the middle of the stream were collected by hand using a pole sampler. All stream sediments, including those from the main channel and tributaries (with the exception of sample SS 11),

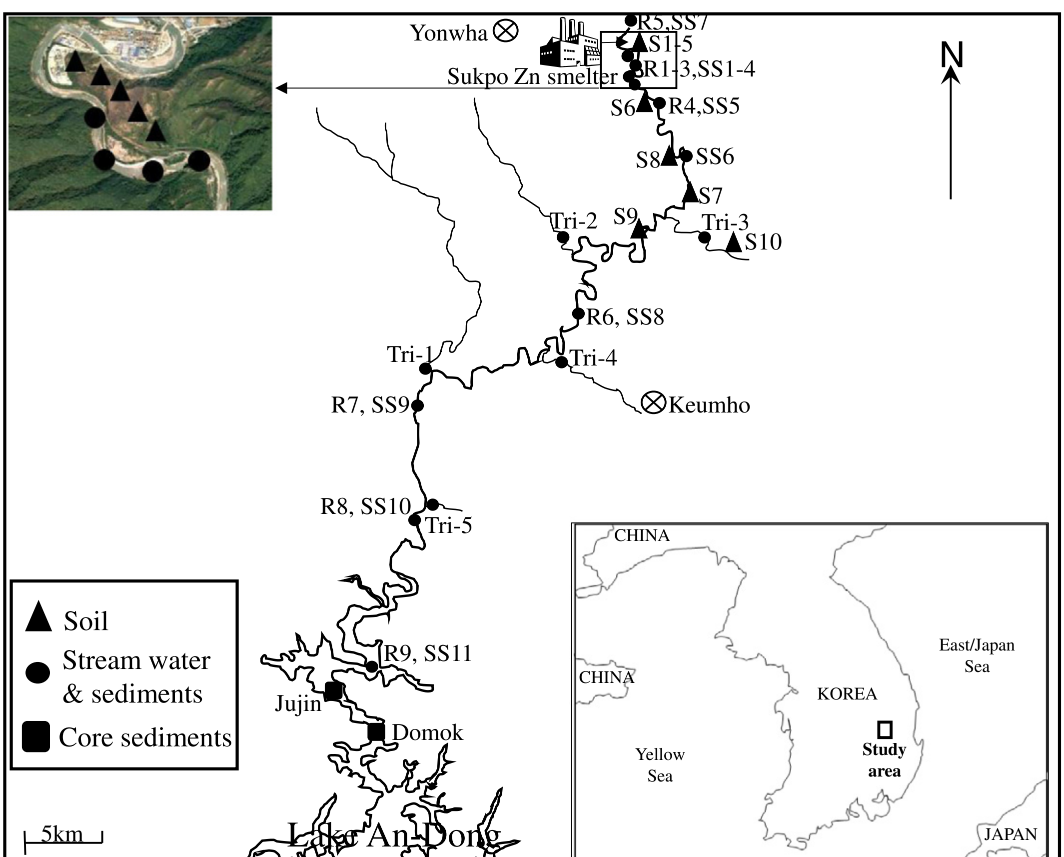

Fig. 1. Sampling sites for soils (S1–S10), stream waters (R1–R9) and stream sediments (SS1–SS11) in the main channel, those in tributaries (Tri1–Tri4) and core sediments (Jujin and Domok) in lake An-Dong.