学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（工学）　氏名　MUFALO Walubita

学 位 論 文 題 名

The extent of pollution and bioaccessibility of lead and zinc from a legacy mine in Kabwe, Zambia, and immobilization of toxic elements by half-burnt dolomite
(ザンビアカブウェ鉱山の鉛、亜鉛による汚染、バイオアクセシビリティと半焼成ドロマイトによる不溶化)

Kabwe is a town in the central province of Zambia that has been a global site of concern due to the historic mining of lead (Pb) and Zinc (Zn). The production of the mineral resources ceased after the depletion of the ore body in 1994. The by-products from the mining activities were left unattended; consequently, pollution of the surrounding environment was inevitable. The mine wastes (MWs) from the legacy mine include slag heaps, mining residue materials (MRM), and waste rocks. These MWs contain potentially toxic elements (PTEs) such as lead (Pb) and Zinc (Zn). Exposure to PTEs such as Pb may affect multiple organ systems resulting in biological and physiological changes in the human body, causing anemia, weakness, and kidney and brain damage.
The children are particularly the most vulnerable to Pb exposure. This is because they frequently explore their environment through hand-to-mouth activities, subsequently may increase their Pb uptake from the polluted environment through accidental ingestion or inhalation. Since various researchers have previously reported high blood Pb levels in the residents of Kabwe, it is imperative to understand the sources of Pb contamination, the extent of pollution, bioaccessibility of PTEs, and possible remediation technology to reduce the contamination levels PTEs from the historic mine. Of all the MWs in Kabwe, the MRM occupies a more extensive area; on the other hand, surrounding soils such as playground soils (SPs) are a potential source of Pb intoxication for the children of Kabwe.
Therefore, this study focussed on the MRM and SPs near a legacy mine in Kabwe, Zambia. The leaching behavior, the extent of pollution in SPs, solid-phase partitioning, and bioaccessible fractions of Pb and Zn in various particle sizes, coupled with the solubility of the elements from MRM and SPs in simulated fluids, were analyzed. The study is significant for bridging the gap between previously reported high blood Pb levels and the bioaccessible Pb and Zn in the MRM and SPs. Finally, countermeasures against reducing Pb and Zn in MRM by half-burnt dolomite (HBD) were also investigated.
The dissertation contains five chapters.
In chapter 1, the general introduction of Pb contamination in Kabwe is briefly reviewed and introduced. The necessities and objectives of the present study are then depicted.
In chapter 2, the extent of pollution and leaching behavior of the elements from SPs were investigated. Kabwe mine wastes are responsible for contaminating the surrounding soil and dust in the Kabwe district. Unfortunately, these wastes arise from the historical mining activities of Pb and Zn, which lacked adequate waste management strategies. As a result, the contaminants like Pb and Zn have spread across the Kabwe district. To assess the soil pollution derived from the previous mining activities, we characterized topsoil samples (n = 8) from the SPs. In this study, the degree of contamination, geochemical partitioning, and leachability, coupled with the release and retention of Pb and Zn, were studied. The SPs were classified as extremely enriched (enrichment factor > 40) and contaminated with Pb (geo-accumulation index > 5). On average, Pb (up to 89 %) and Zn (up to 69 %) were

bound with exchangeable, weak acid-soluble, reducible and oxidizable phases, which are considered 'geochemically mobile' phases in the environment. The leachates from the soils (n = 5) exceeded the Zambian standard for Pb in potable drinking water (Pb < 0.01 mg/L). Furthermore, the spatial distribution of Pb and Zn showed a significant reduction in contents of Pb and Zn with the distance from the mine area.

In chapter 3, sequential extraction of Pb and Zn in MRM with different particle sizes and their solubility in simulated fluids were investigated. Based on the previous studies showing that some of the residents of Kabwe had high blood Pb levels above recommended value, simulated fluids representing lung conditions (artificial lysosomal fluid (ALF), gamble solution (GS), and Phosphate-buffered saline (PBS) were used to assess the solubility of Pb and Zn in the dust (< 30 μ m) of MRM samples. Both Pb and Zn were soluble with ALF. However, only Zn was detected with GS and PBS. About 2.9 % of Pb was bioaccessible with ALF in MRM and up to 37 % of Pb was bioaccessible in the SPs samples. Compared with solubility by oral intake using HCl, higher bioaccessible fractions of Pb and Zn were recorded in both MRM and SPs. Different particle size distributions of MRM did not significantly affect the solid-phase partitioning of Pb and Zn in the residues. However, in case of accidental ingestion, substantial amounts of Pb and Zn from the exchangeable phase, carbonates, and Fe/Mn oxides could be available for adsorption in the bloodstream from the MRM and SPs samples. The results suggest that oral intake of contaminated wastes could be considered a more significant route for Pb uptake than inhalation. These findings may explain the high blood Pb levels previously reported in Kabwe.

In chapter 4, the countermeasures for reducing Pb and Zn in MRM were elucidated by using HBD in column and pilot-scale experiments. In this study, the efficacy of the HBD was evaluated by mixing MRM with 1, 5, and 10 % ratios of the HBD in columns. The leaching of Pb and Zn was compared with the addition of 10 % HBD to MRM in pilot-scale experiments. Measurements of mineralogical and chemical constituents of the MRM, sequential extraction, and Phreeqc modeling of the leachates were carried out to determine the attenuation mechanisms of Pb and Zn. The sources of Pb and Zn in the MRM were mainly from the exchangeable and carbonate fractions. The SEM-EDS observations suggest that Pb and Zn were primarily associated with sulfide minerals although XRD did not detect the sulfide minerals in MRM. Thus, the dissolution of secondary soluble sulfate minerals contributed to the leaching of Pb and Zn. The introduction of HBD into the MRM increased effluent pH and decreased the leaching concentrations of Pb and Zn. The addition of 10 % HBD was enough to suppress the leaching of Pb and Zn in both the column and pilot-scale experiments. The saturation indices and SEM-EDS observations suggest that the retention mechanisms of Pb and Zn by HBD accredit to precipitation, co-precipitation, and adsorption reactions. These results present an effective, inexpensive, and long-term immobilization technology for toxic elements in hazardous wastes from legacy mines.

Chapter 5 summarizes all general conclusions of the present study. The results could provide the fundamental knowledge of leachability and geochemical partitioning and the bioaccessibility of Pb and Zn in Kabwe, Zambia. Furthermore, long-term remediation technology of chemical immobilization by HBD should be evaluated for the legacy mine.