Fingerprints and Fluxes of Lead (Pb) from Coal Fly Ash Disposal in China, India, and the United States

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Coal combustion is one of the major anthropogenic activities that release lead (Pb) to the natural environment. Despite extensive research and technology to reduce the atmospheric emission of Pb from burning coals for electricity, there has been minimal research on the fluxes and impacts of Pb associated with coal ash disposal in the environment. This study provides insights into the isotopic fingerprints and quantities of Pb retained in coal fly ash in China, India, and the U.S., the world's top three coal producers. Pairwise comparison between feed coal and fly ash samples collected from coal-fired power plants from each country shows that Pb is markedly enriched in fly ash relative to feed coal and Pb isotope composition of fly ash largely resembles that of parent coal. The distinct Pb isotopic fingerprints of fly ash compared to that of ore deposits allow for tracing the release of coal fly ash Pb into the environment. Following the phase-out of leaded gasoline worldwide, the annual flux of Pb from coal fly ash disposal between 2000 and 2020 decreased slightly in the U.S. and India but increased notably in China and exceeded that of Pb atmospheric emission. In total, approximately 227, 56, and 50 Gg Pb have been disposed with fly ash in China, India, and the U.S., respectively, posing a significant environmental burden. Freshwater lakes in North Carolina have been receiving Pb from adjacent coal ash disposal facilities for the past 40 - 70 years. The results of a Bayesian Pb isotope mixing model show that the total Pb accumulation in the sediments of these lakes is greater than that in other lakes without coal ash impacts in the Southeast U.S., and the mean relative contribution of Pb from coal fly ash (~26%) is significantly higher than that of leaded gasoline (\sim 7%). This implies that, at local and regional scales, the input of coal fly ash Pb can outweigh that of other anthropogenic sources. Our study lays the groundwork for future research on the environmental fate and transport of released fly ash Pb and its potential risks to both ecological and human health.

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