Lead Isotopic Tracing of Coal-Based Anthropogenic Pollution in Agricultural Soils in Jianghan Plain, China

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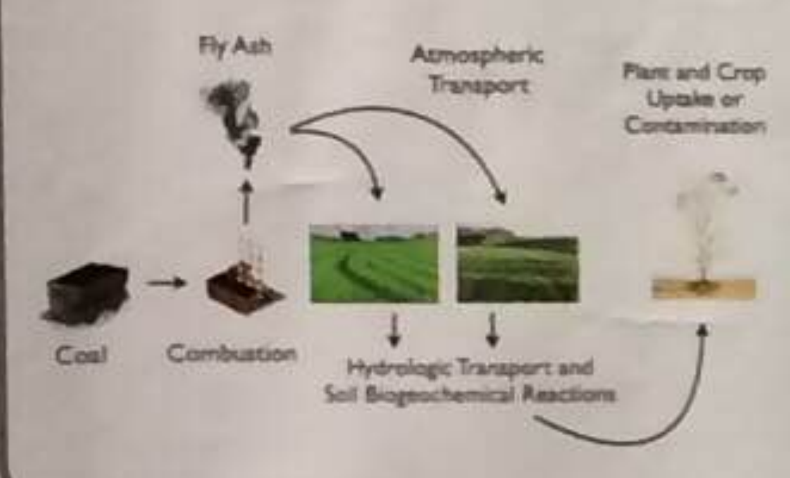
Coal and Agriculture in China

China's population demands large amounts of both electricity and food. China burns four billion tons of coal per year - half the world's total - and its 2,300 coal power plants release particulates containing high concentrations of heavy metals. These toxins then land on agricultural areas and may be incorporated into plants, posing a danger to human health. **The objectives of this study are to determine the extent to which soils are enriched in heavy metals, and to use lead isotope ratios to trace the sources of enrichment - both natural and anthropogenic.**

Chinese urban and agricultural areas have unusually strong effects on one another, as illustrated by the maps at left. Unlike many countries, where population centers and agricultural production occur in different areas, **China's areas of highest population density are in close proximity to its areas of high cultivation.** This juxtaposition of industry, energy production, and farmland creates unique environmental and human health challenges. Of particular concern are the effects of unmitigated large-scale coal combustion on the agricultural sector. Soil pollution may cause reduced crop yields and contamination of the food supply, leading to human health risks and economic difficulties. **It is thus a pressing issue to understand and quantify the extent to which industrial pollution is affecting soil chemistry, in order to pursue effective actions and policies for a healthier environment.**



The process by which coal-based pollutants contaminate agriculture is well-understood, and is illustrated in the diagram at left. Coal, which naturally contains traces of lead and other heavy metals, is burned in a power plant to generate electricity. The coal undergoes chemical changes and is released as fly ash, which contains many of those heavy metals. The airborne fly ash is deposited on cultivated areas, both around the power plant and many miles away. The fly ash and its contaminants enter the soil, where they participate in chemical reactions which render them available for uptake, and they accumulate in the crops.

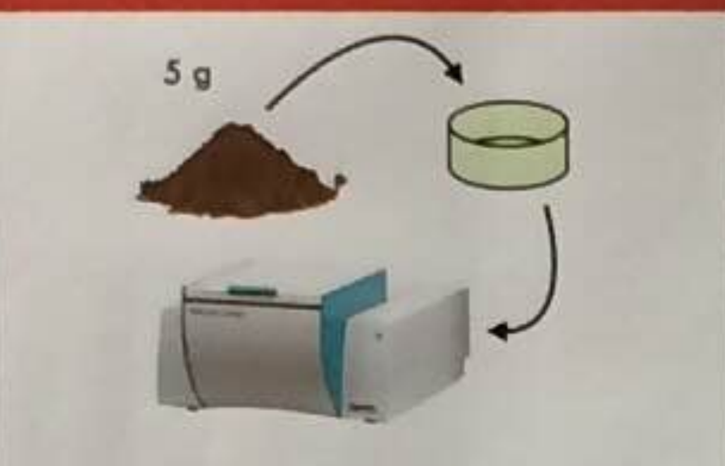
Field Site: Jianghan Plain

The Jianghan Plain is located in central China, approximately 800 kilometers west of Shanghai. Wuhan, the capital of Hubei province, lies in its center along the Yangtze River. **With a population of over ten million people, Wuhan is a prime example of the proximity of an industrial urban center to expansive agricultural land.** The Jianghan Plain is a cultivation area for many diverse crops, including beans, rice, squash, lotus, peppers, eggplant, cotton, and peaches. It is also an active industrial area, demanding enormous amounts of energy to support its growing biotechnology, telecommunications, automobile and equipment manufacture, and raw goods processing sectors.

Our field site is located north of the Yangtze River and the city of Wuhan, at the border of agricultural and industrial land, as the land-use map in the upper right shows. **The Huaneng Yangluo coal-fired power plant, shown in the photo below and indicated in red above, is a major source of pollution in the area, releasing 1.7 tons of fly ash into the atmosphere every 12 hours** (Ying et al., 2012). Our group visited the site in 2012 and again in 2014 to collect soil and fly ash samples.

The 2014 sampling area contains 69 field sites within a 25-kilometer radius of the Yangluo power plant (see middle map above). Soil samples were collected at each site at depths of 0-1 cm ('shallow') and >50 cm ('deep') below the surface. The shallow samples contain recently-deposited airborne particles, including fly ash pollutants. The deeper samples were taken from below the tillage depth, with a potential for containing fewer airborne pollutants. The soil samples were dried, transferred to the Stanford laboratory, ground into powder, and processed within two weeks of collection.

Lead Extraction and Data Collection Process



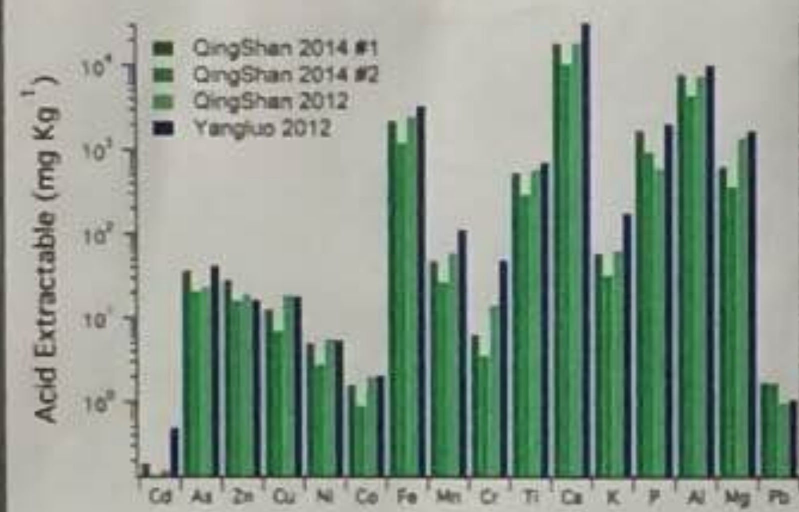
The X-Ray Fluorescence machine measures the total concentrations of a variety of metals in a single dry soil sample.

Inductively Coupled Plasma Mass Spectroscopy records the relative abundances of lead isotopes.

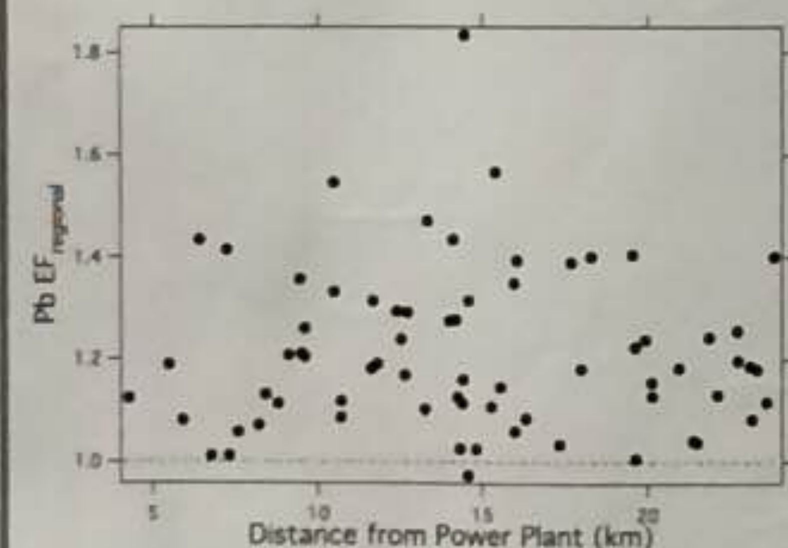
1 gram of dried, ground soil is mixed with 10 mL nitric acid and shaken at room temperature for 24 hours at 20°C to allow the lead to enter solution.

The mixture is then filtered and diluted, and placed in the ICP-MS machine for analysis. The ICP-MS's information about relative abundances of Pb isotopes can be used to determine the origins of the lead in the sample.

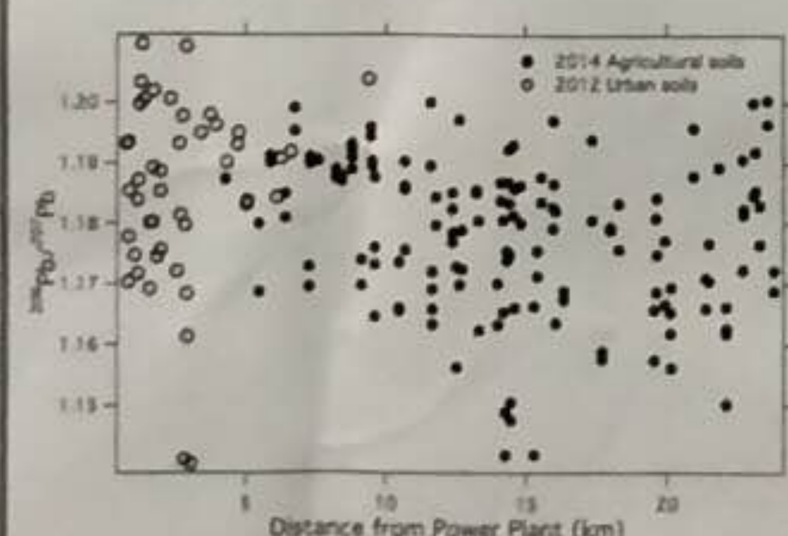
Results



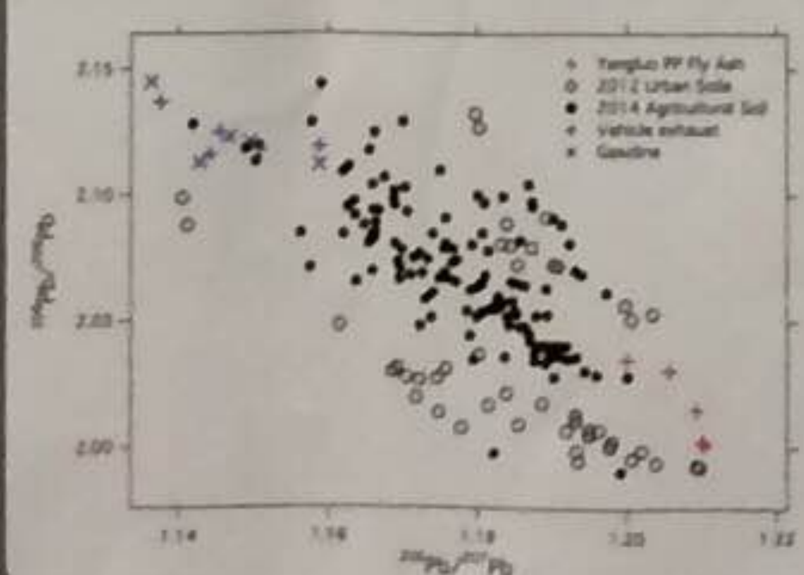
The data at left show the results of acid extractions performed on four different fly ash samples from two separate coal-fired power plants in 2012 and 2014. 1 gram of fly ash was mixed with 10 mL 0.5 M nitric acid. The data illustrate the amounts of **diverse, acid-extractable heavy metals in all samples of fly ash.** The two QingShan samples are replicates taken within an hour from the power plant stack.



The data at left show the enrichment factor of lead in our soil samples as a function of the sample site's distance from the Yangluo coal-fired power plant. The results show that **all but one soil sample have enriched lead amounts,** but a narrow range of enrichment factor values (all less than 1.9). The regional enrichment factor is calculated by normalizing $[Pb]_{surface}/[Ti]_{surface}$ with [Pb]/[Ti] relative to deep cores, 30 meters below surface level, from a geologically similar area approximately 60 kilometers from the Yangluo plant.



The data to the left show the ratio of ^{206}Pb to ^{207}Pb as a function of the sample site's distance from the Yangluo power plant. Both urban sites sampled in 2012 (within 10 km of the urban center) and agricultural sites sampled in 2014 (5 km to 20 km from the urban center) are shown. **The presence of radiogenic Pb, indicated by the concentration of ^{206}Pb, tends to decrease slightly with distance away from the urban center.**



The data to the left show the lead isotopic composition for the 2012 urban and 2014 agricultural soil samples within 20 km of the Yangluo power plant. **The origin of the lead found in the soils is dominated by two end-members: fly ash from the power plant and lead from vehicle exhaust and historically-used leaded gasoline.** Leaded gasoline used in China was primarily produced from imported Australian lead ores, which typically exhibit a low ^{206}Pb to ^{207}Pb ratio relative to other lead sources used for gasoline production (such as the United States and Europe).

Conclusions

The significant concentrations of heavy metals in all samples of fly ash indicate that fly ash particulates are a likely contributor of heavy metal contaminants to the soils and surfaces on which they deposit (graph 1). The soils, meanwhile, nearly all exhibit enriched Pb relative to local geogenic Pb values, indicating an anthropogenic Pb contaminant source (graph 2). Furthermore, the slight decrease in radiogenic Pb with increased distance from the Yangluo power plant (graph 3) serves as a further indicator of the power plant as a point source for airborne contaminants. **The combined evidence for significant quantities of heavy metals in various fly ashes along with enriched surface Pb amounts and higher radiogenic Pb concentrations near power plants all support the hypothesis that coal power plants such as Yangluo are contributing to the Pb enrichment of surrounding agricultural soils.** Furthermore, the isotopic signature of the lead found in the Jianghan Plain soils seems to be a combination of the known isotopic signatures of leaded gasoline and automobile exhaust from the Wuhan area, and fly ash from power plants such as Yangluo (graph 4). **Isotopic analysis of the Pb showing that the two main contributors to the elevated Pb are automobile exhaust and power plant fly ash lends further credence to our hypothesis that coal combustion is a significant contributor to the heavy metals present in the agricultural soils of the Jianghan Plain.**

It is clear that anthropogenic pollutants from coal power plants like the Huaneng Yangluo power plant in Wuhan have a significant effect on the composition of nearby agricultural soils. **Important further studies must be done, however, to determine the exact mechanisms by which these pollutants interact with the soil biogeochemistry and potentially become available for uptake by planted crops.** Understanding these processes is essential knowledge for further exploration of the direct agricultural effects of severe airborne pollution. **It will be of utmost concern to build off of these findings and determine the extent to which plant uptake of these contaminants affects both crop yield and the health of the humans or livestock who consume these contaminated agricultural products.**

While the Jianghan Plain is an ideal place to study these effects of industrial activity on agriculture, the concerns with airborne pollutants are relevant throughout China. **China currently operates over 620 coal combustion power plants,** mainly located in the areas of both high population density and high cultivation. The potential effects on human health and the agricultural economy due to these vast quantities of toxic pollutants are of urgent concern. Prudent policy decisions regarding the future of coal combustion in China require a thorough understanding of the mechanisms, processes, and effects related to coal fly ash and its deposition on agricultural areas.

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