博士文库

城市表层土壤重金属富集淋滤特征与磁学响应

王学松　著

ACCUMULATION-LEACHING CHARACTERISTICS AND MAGNETIC PROXIES OF HEAVY METALS IN URBAN TOPSOIL

中国环境科学出版社

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摘 要

近年来，重金属对人类和其他生物的危害受到了广泛的关注。城市土壤由于能够作为接纳多种途径释放的重金属（如工业废弃物、汽车尾气、燃煤废弃物以及其他活动产生的废物等）的汇，因此，城市土壤中重金属含量的高低常常可以作为指示城市环境污染强弱的“示踪物”。徐州作为一座资源型城市和重工业城市，煤炭开采和利用、重型机械、化工、电力等工业极为发达，重金属污染源多且较为复杂。为此，本书选择徐州城市土壤为研究对象，重点考察城市土壤中重金属的富集与淋滤特征，探讨徐州城市土壤磁学特征与重金属污染强度之间的相关关系，建立相应的磁学诊断模型，取得了如下主要研究成果。

1．城市土壤重金属的地球化学的基本特征

按照重点区域与面上均匀分布相结合的原则，系统地采集了表层土壤样品 21 件和柱状样品 10 件（0～50 cm，间隔为 5 cm）。利用 XRF、ICP-MS、ICP-AES 等方法分别测定了土壤样品中 Sn、Li、Ga、Ba、Fe、Mn、Co、Be、Ti、Al、Hg、Cr、Sb、As、Bi、Pd、Pt、Au、Ni、Cd、Br、Zn、Cu、S、Pb、Se、Mo、Sc、V 和 Ag 共 30 种常量元素和重金属元素的含量；同时测定了土壤的基本理化参数和进行了某些典型土壤样品的矿物分析。研究结果表明，土壤样品中 Ag、Se、Pb、Cu、Zn、Cd、Hg 和 Sb 重金属元素含量的中值分别为 0.28、0.42、36、32、102、0.42、0.18 和 3.46 mg/kg，高于我国土壤对应元素的背景值（0.13、0.29、26、22.6、72.4、0.097、0.065 和 1.21 mg/kg）。

多元统计分析（聚类分析和因子分析）的结果显示，徐州城市土壤重金属元素可以分为三类，其中 Sn、Li、Ga、Ba、Fe、Mn、

Co、Be、Ti 和 Al 等主要来源于土壤母质，土壤中的 Hg、Cr、Sb、As、Bi 等元素主要与燃煤相关，而土壤中的 Pd、Pt、Au、Ni、Cd、Br、Zn、Cu、S、Pb、Se、Mo、Sc 和 Ag 等重金属元素则主要来源于交通运输过程。利用 GIS 软件 s 构建的重金属元素空间分布特征显示了徐州城市土壤重金属同时存在线源（来自交通运输过程）和点源（主要分布在工业区）的贡献。

采自徐州钢铁厂附近的柱状样品中重金属的分布特征显示了土壤中人为发生源的重金属元素如 Ag、Bi、Pb、Cd、Zn 等主要分布在 0～30 cm，而来自土壤母质的元素如 Al、Ti、Sc、Ga、V、Cr 和 Mn 等在 0～50 cm 分布比较均匀。

了解研究区域土壤中重金属元素的背景值对于准确判断重金属的富集特征具有重要作用。本书利用元素的对数正态分布图粗略地估算了研究区域土壤中重金属元素的背景值，结果表明，除了土壤样品中 Hg、Cd 两种元素外，其他元素的估算数值与我国土壤背景值相当。

2. 城市土壤重金属的富集与淋滤特征

土壤的理化性质影响土壤中重金属元素的迁移过程。土壤样品的基本理化参数（pH，EC，CEC，TOC，TIC，土壤质地，主要的无机矿物 Fe、Al、Mn 等）与重金属的相关分析表明，两者的相关关系比较复杂，其中与土壤样品中 Al 元素含量呈显著正相关的元素有 Ti、Ga、Li、V、Co、Be、Mn 和 Pt；与 Fe 元素含量呈显著正相关的元素有 Se、Ba、Pb、Cu、Zn、Cr、As、Sb、Li、Ga 和 V；与土壤样品中 Mn 元素含量呈显著正相关的有 Ti、Ga、Ba、Li、V、Co、Be 和 Sn。

土壤样品中有机质和腐植酸与重金属元素的相关关系不甚显著，但是土壤样品中富里酸（fulvic acid，FA）与重金属元素 Fe、Ag、Ba、Li、Pb、Cu、Zn、Ni、Cr、As、Sb 和 Mo 呈显著正相关；土壤样品中碳酸盐含量与 Ti、Li、V、Mn、Be 等元素呈显著负相关。

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土壤中矿物颗粒表面积的差别导致其对重金属元素的富集特征不同。总的来说，土壤样品中黏土和粉砂的含量与重金属元素含量呈正相关，而土壤中砂粒由于其表面积较大，一般与重金属元素的含量呈负相关。典型样品（1#和 12#）中不同重金属元素的质量载荷差异明显，其中 Pb 元素一般在 45～125 μm 质量载荷占有优势，而元素 Cu 的质量载荷表现为在不同样品中分布不同。

淋滤实验能够阐明土壤中重金属元素的键合状态和淋滤特征。模拟极端条件下，土壤样品的浸出毒性淋滤实验（TCLP）结果表明，所分析的元素（Pb、Cu、Zn、Ni、Cr、Co、As、Sb、Mn）的淋滤浓度都在规定的范围之内，表明这些元素在土壤中主要是以稳定的状态存在，而以离子结合态形式存在较少。为了进一步详细地了解土壤中重金属元素的赋存形态，分别对 1#和 12#样品进行了系列提取实验（BCR），土壤样品中 Pb、Cu、Zn、Ni、Cr、Co、As、Sb、Mn 主要是以铁-锰氧化态和有机结合态存在，而酸可提取态所占比重较小。

0.05 mol/L Na_2EDTA 对 1#和 12#样品的淋滤效率明显地大于水的淋滤，而且 EDTA 淋滤前后土壤样品中重金属元素的赋存形态都出现了不同程度的重新分布，表现为不同重金属在不同样品中分布特征不同。

为了能够准确地判断土壤样品中重金属元素对人体健康危害的影响程度，对 12 个土壤样品进行了模拟胃肠吸收的淋滤实验（SBET）。总的来说，所研究的重金属元素的淋滤浓度低于规定的最大允许值；逐步回归分析显著淋滤浓度一般只与土壤样品中重金属元素的总浓度有关，与基本理化参数关系基本无关。

3. 城市土壤的磁学特征以及磁学参数与重金属元素的关系

利用磁学参数作为工业污染区域土壤中重金属元素含量的代用指标逐渐受到重视。测定了土壤样品的一些磁学参数（χ、*SIRM*、*SOFT*、*ARM*），并且计算了一些常用的比值参数如χ_{fd}、χ_{ARM}、F_{300}、S_{-100}、χ_{ARM}/χ 和$\chi_{ARM}/SIRM$；对典型样品进行了热磁实验和扫描电

镜分析（SEM-EDX）。徐州城市土壤的磁学特征主要为多畴和假单畴的亚铁磁性矿物主导，不完整的反铁磁性矿物贡献较小；钢铁厂附近的柱状样品的磁性特征显示人为产生的磁性矿物主要集中在0～30cm，这与人为输入的重金属的分布区域一致。

5 个基本的磁学参数（χ_{fd}、χ_{ARM}、χ、*SIRM*、*SOFT*）与土壤样品的主要理化特征的相关分析表明，土壤样品中的 Fe 元素的含量主要与χ_{ARM}、χ、*SIRM*、*SOFT* 呈显著正相关；土壤样品中黏土矿物的含量主要与指示细粒子的参数如χ_{fd}、χ_{ARM}/χ 和$\chi_{ARM}/SIRM$ 呈显著正相关，与χ、*SIRM*、*SOFT*、χ_{ARM}、F_{300}、S_{-100} 等磁学参数相关性较差；土壤样品中粉砂含量与这些磁学参数的相关性与黏土的趋势一致；样品中砂粒含量与磁学参数χ_{fd}、χ_{ARM}/χ 和$\chi_{ARM}/SIRM$ 呈显著负相关，与χ、*SIRM*、*SOFT*、χ_{ARM}、F_{300}、S_{-100}、$\chi/SIRM$ 等不具有相关性。土壤中有机质能够与磁性矿物形成络合物导致磁性矿物的溶解，所以土壤样品中有机质含量与指示磁性矿物数量的磁学参数χ、*SIRM*、*SOFT*、S_{-100} 呈显著的负相关，与χ_{ARM}/χ 和$\chi_{ARM}/SIRM$ 呈显著正相关，与χ_{ARM}、F_{300}和$\chi/SIRM$没有相关性。

土壤样品中磁学参数的数值与重金属元素的含量之间的相关关系分析表明，与磁化率（χ）呈显著正相关的元素有 Fe（0.486）、Se（0.507）、Sc（0.539）、Mo（0.440）、S（0.692）；与频率磁化率（χ_{fd}）呈显著正相关的元素有 Al（0.704）、Ti（0.669）、Ga（0.612）、Li（0.717）、V（0.731）、Co（0.489）、Mn（0.575）和 Be（0.696）；与软剩磁（*SOFT*）呈显著正相关的元素为 Fe（0.468）、Se（0.624）、Pb（0.561）、Cu（0.440）、Mo（0.520）、S（0.607）；与非磁滞剩余磁化率（χ_{ARM}）呈显著正相关的元素为 Ag（0.465）、Se（0.802）、Pb（0.847）、Cu（0.810）、Zn（0.749）、Cr（0.609）、Mo（0.692）和 Br（0.556）；与饱和等温剩磁（SIRM）呈显著正相关的元素包括 Fe（0.594）、Se（0.725）、Pb（0.742）、Cu（0.544）、Zn（0.496）、Cr（0.527）和 Mo（0.563）。污染负荷因子（pollution load index，PLI）指示重金属复合污染程度的大小，PLI 与磁学参数χ、χ_{fd}、*SOFT*、*SIRM*、χ_{ARM}的相关系数分别为 0.784、0.027、0.438、0.551

和 0.761。

在分析了许多重金属污染的样品的磁学特征的基础上，提出了土壤样品中重金属污染的磁学诊断模型：

{重金属污染的土壤样品}={土壤样品的磁化率大于该区域磁化率的背景值且频率磁化率的数值小于 3%}

徐州钢铁厂附近的土壤样芯磁学参数与元素浓度之间的关系表明，土壤样芯中 Pb、Cu、Zn、Cd、Bi、Se、Hg、Fe、Mn 的含量与磁学参数χ、*SOFT*、*SIRM*、χ_{ARM} 的数值之间存在着十分相似的垂向变化关系，指示了土壤样芯中高浓度的磁性矿物和重金属系由外源输入引起的；重金属元素与磁学参数的显著相关表明这些磁学参数可以作为土壤中重金属迁移的代用指标。土壤样芯中元素 S 与磁学参数的相关性非常显著，指示土壤中 S 与磁性矿物来源相同，根据表层土壤中 S 元素的含量可以评估大气中 SO_2 的污染程度，但是，是否可以利用土壤的磁学参数作为大气中 SO_2 污染程度代用指标仍需进一步探讨。

土壤中铁锰氧化物结合态在一定程度上控制了土壤中 Pb、Cu、Zn、Cd、Bi、Se、Hg 等重金属元素的生物地球化学循环。

关键词：城市土壤；重金属；环境磁学；替代指标；淋滤；富集；徐州

Summary

In recent years public attention has been focused increasingly on heavy metal pollution and its effect on man and other creatures. Heavy metals in urban soils have been shown to very useful tracers of environmental pollution because urban soils are the "recipients" of large amount of heavy metals from a variety of sources including industrial waste, vehicle emissions, coal burning waste and other activities.

Starting with the investigation of sediments in the 1980s, the research area of environmental magnetism has grown steadily in the last two decades. The main advantages of magnetic measurements in the field are their simplicity, speed and low costs. Large data-sets can be acquired in short time and repeated measurements easily performed. Presently, one of the main topics is the use of magnetic measurements as a proxy in heavy metal pollution studies. Up to now, the relationship between heavy metals and magnetic parameters has been exhaustively studied. Nevertheless, the relationships between magnetic particles and pollutants (heavy metals) are complex and they are different for each industrial process. In cement production, for example, additives turned out to influence the amount of magnetically susceptible material in the dusts. The complexities of the relations between pollutants and magnetic particles, and the fact that have a common source but are usually separate particles make it impossible to derive a single function to calculate pollutant concentrations from one of the magnetic parameters.

Xuzhou (China) is an important centre of modern day heavily industrial activities where thick coal measures provided the fuel for

manufacture. The city also has abundant industrial activities including a coal-fired power plant and companies that produced organic and inorganic chemicals. The main wind direction is from the northwest, although in general, wind velocities are low, even approaching zero at times. This enhances the deposition of particulates within the city. Around the world a great many studies have evaluated the heavy metal concentrations in urban soils. Nevertheless, in Xuzhou, such studies are non-existent to date. For this reason, this study was aimed at providing a contribution to a database on the heavy metal status in the city as a basis for a wide variety of environmental applications as well as an approach to assess the relationship between geochemistry and the health of ecosystems. In this study, some innovative achievements have been acquired as follows:

1. The main geochemical characteristics of heavy metals in Xuzhou urban soils

Concentrations of Sn, Li, Ga, Ba, Fe, Mn, Co, Be, Ti, Al, Hg, Cr, Sb, As, Bi, Pd, Pt, Au, Ni, Cd, Br, Zn, Cu, S, Pb, Se, Mo, Sc, V and Ag were measured on 21 topsoil samples collected from the city of Xuzhou in order to: assess the distribution of these elements in the urban environment; discriminate natural and anthropogenic contributions; and identify possible sources of pollution. Mineralogy, physico- chemical parameters and major element contents of the soils were also determined to highlight the influence of "natural" features on the heavy metal concentrations and their distribution.

Medians of Ag, Se, Pb, Cu, Zn, Cd, Hg and Sb concentrations of the investigated urban soils are 0.28, 0.42, 36, 32, 102, 0.42, 0.18 and 3.46 mg/kg, respectively. These values are higher, in some case by different order of size, than those of unpolluted soils in China that average 0.13, 0.29, 26, 22.6, 72.4, 0.097, 0.065 and 1.21 mg/kg. An ensemble of basic and multivariate statistical analysis (cluster analysis and factor analysis) was performed to reduce the multidimensional space of variables, thus defining three sets of heavy metals as tracers of natural and anthropogenic influences. Results show that Sn, Li, Ga, Ba, Fe, Mn, Co, Be, Ti and Al are interpreted to be mainly inherited from parent materials, whereas Hg, Cr, Sb, As, Bi can be inferred to be tracers of anthropogenic pollution associated with coal burning and Pd, Pt, Au, Ni, Cd, Br, Zn, Cu, S, Pb, Se, Mo, Sc and Ag can be inferred to be tracers of anthropogenic pollution linked with traffic transport. Maps of heavy metal distributions were constructed for the whole urban area pointing to vehicle traffic as the main source of diffuse pollution and also showing the contribution of point sources of pollution to urban topsoils. The core (located from near iron-steel plant) displayed a systematic drop in concentrations of heavy metals (Ag, Bi, Pb, Cd, Zn, etc) at depths of 0-30 cm, indicating anthropogenic inputs, whereas concentrations of Al, Ti, Sc, Ga, V, Cr and Mn are almost consistent at depths of 0-50 cm.

The total concentrations of heavy metals are not the most important information for assessing heavy metal pollution of certain areas. The determination of natural levels of these metals is of the same importance. For the background levels of Xuzhou urban soils we have selected the method used by Acero et al. (2003). Our background values of Hg and Cd are much higher than those of unpolluted soils of China while the other values are insignificantly different.

2. Characteristics of accumulation and leaching of heavy metals in Xuzhou urban soils

As is widely recognized in the literature there are a number of physical and chemical properties of soils affecting metal mobilization and immobilization processes. To analyze the relationship between these soils parameters (pH, EC, CEC, TOC, TIC, major mineral and grain

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size) and the heavy metal concentrations, a Pearson R correlation analysis has been applied to the 21 soil samples. Results of the statistical analysis have shown a large variety and complexity in these relationships. Between the contents of Al and the other metal concentrations there are significant correlations with Ti, Ga, Li, V, Co, Be, Mn and Pt; between the Fe contents and the other metal concentrations there are significant correlations with Se, Ba, Pb, Cu, Zn, Cr, As, Sb, Li, Ga and V; between the Mn contents and the other metal concentrations there are significant negative correlations with Ti, Ga, Ba, Li, V, Co, Be and Sn.

Between the fulvic acid percentage and the metal concentrations there are significant and positive correlations with Fe, Ag, Ba, Li, Pb, Cu, Zn, Ni, Cr, As, Sb and Mo. Between the inorganic matter percentage and the metal concentrations there are significant and negative correlations with Ti, Li, V, Mn, and Be.

The relations of metal concentrations to grain size were also determined to evaluate the influence that the surface area had on controlling the metal concentrations. Totally speaking, clay and silt percentages have a positive correlation while the sand percentages have a negative relationship. For Pb, the mass loading is mainly contributed from the grain size of 45-125 μm while the contribution of Cu mass loading is much different from the different samples.

Following the gross assessment of heavy metals in the urban soils, leaching tests were performed on specific soils to elucidate heavy metal associated mineral fractions and general leach-ability. Extraction tests illustrated the low leachability of the soils. The Toxicity Characteristic Leaching Procedure (TCLP) was performed on two different soils and it was shown that all metal leachates analyzed were below regulatory limits. Sequential extraction (BCR) was performed on two soils. The leached acid-extractable fractions were low for the analyzed metals. Also the importance of the iron-manganese (reductive), organic-sulfide (oxidative) and residual fractions was identified. Next, the distribution of Pb, Cu, Zn, Ni, Co, Cr, As and Mn in two contaminated soils were determined before and after treating the soils with an 0.05 mol/L Na_2EDTA solution. The proportions of different metals accumulated in four fractions are very complex.

In order to estimate the human bioavailability quotients for heavy metals(Pb, Cu, Zn, Ni, Co, Cr, As and Mn), the Simple Bioavailability Extraction Test(SBET) was conducted on 12 soil samples. The leachate concentrations were low for all analyzed metals and the results were modeled using stepwise multiple regression analysis.

3. Magnetic properties and relationships between magnetic parameters and heavy metals in Xuzhou urban soils

Magnetic measurements were increasingly used as a proxy for heavy metal concentrations in soils influenced by industrial emissions in recent years. The values of magnetic parameters (χ, *SIRM*, *SOFT*, *ARM*) were determined from the soil samples and the values of χ_{fd}, χ_{ARM}, F_{300}, S_{-100}, χ_{ARM}/χ and $\chi_{ARM}/SIRM$ were calculated. Thermomagnetic analysis and SEM-EDX were used to identify ferrimagneitc fractions. The results indicat the magnetic characteristics are dominated by the multi-domain/pseudo-single domain ferri-magnetic particles. Anti-ferro-magnetic minerals has a little contribution to the magnetic properties of Xuzhou urban soils. The distributions of values of different magnetic parameters in profiles also reveal that anthropogenic magnetic minerals are at the depths of 0-30 cm.

To analyze the relationship between soil parameters and the 5 parameters (χ, *SIRM*, *SOFT*, χ_{fd}, χ_{ARM}) which indicate the contents of magnetic mineral, a Pearson R correlation analysis was conducted to 21 soil samples. Between the Fe contents and the above magnetic parameters there are significant and positive correlations with χ, *SIRM*, *SOFT*, and χ_{ARM}. Between the clay percentages and the magnetic parameters there are significant and positive correlations with χ_{fd}, χ_{ARM}/χ and $\chi_{ARM}/SIRM$, no significant correlations with χ, *SIRM*, *SOFT*, χ_{ARM}, F_{300}, S_{-100}. Between the silt percentages and the magnetic parameters there are the same trends similar to that of clay in soils. Between the fine sand percentages and the magnetic parameters there exist significant and negative correlations with χ_{fd}, χ_{ARM}/χ and $\chi_{ARM}/SIRM$; no significant correlations with χ, *SIRM*, *SOFT*, χ_{ARM}, F_{300}, S_{-100}. Between the organic matter percentages and the parameters, there are significant and negative correlations with χ, *SIRM*, *SOFT*, S_{-100}; no significant correlations with χ_{ARM}, F_{300}, $\chi/SIRM$; significant and positive correlations with χ_{ARM}/χ and $\chi_{ARM}/SIRM$.

The relationships between heavy metal concentrations and the values of magnetic parameters were also performed. Between values of χ and heavy metal concentrations there are significant and positive correlations with Fe (0.486), Se (0.507), Sc (0.539), Mo (0.440), and S (0.692). Between values of χ_{fd} and heavy metal concentrations there are significant and positive correlations with Al (0.704), Ti (0.669), Ga (0.612), Li (0.717), V (0.731), Co (0.489), Mn (0.575), and Be (0.696). Between values of *SOFT* and the heavy metal concentrations there are significant and positive correlations with Fe (0.468), Se (0.624), Pb (0.561), Cu (0.440), Mo (0.520), and S (0.607).

Between the values of χ_{ARM} and heavy metal concentrations there are significant and positive correlations with Ag (0.465), Se (0.802), Pb (0.847), Cu (0.810), Zn (0.749), Cr (0.609), Mo (0.692), Br (0.556). Between the values of *SIRM* and the heavy metal concentrations there are significant and positive correlations with Fe (0.594), Se (0.725), Pb (0.742), Cu (0.544), Zn (0.496), Cr (0.527), and Mo (0.563). The correlation coefficients between the magnetic parameters（χ，χ_{fd}，*SOFT*，*SIRM*，χ_{ARM}） and PLI（Pollution Load Index） are 0.784，0.027，0.438，0.551 and 0.761，respectively.

Soils containing significantly elevated heavy metals were also identified using two criteria：χ（values > background values） and χ_{fd}（values < 3%）.

Magnetic measurements and chemical elemental analysis were conducted on soil core from near Xuzhou iron-steel plant. The relation that vertical variations of values of some magnetic parameters（χ，χ_{ARM}，*SOFT*，*SIRM*） are in good agreement with that of concentrations of heavy metals（Pb，Cu，Zn，Cd，Se，Fe，Mn，Hg） reveals magnetic minerals and heavy metals in soils are due mainly to anthropogenic inputs. Significant correlations between magnetic parameters and heavy metals also show these magnetic parameters（χ，χ_{ARM}，*SOFT*，*SIRM*） could be used as proxies of concentrations of heavy metals at varying depth. The relationship that there exists very significant correlation between magnetic parameters and elevated concentrations of element S indicates there is common source between these magnetic minerals and S. According to the concentrations of S in the soil profile，the concentrations of SO_2 in the air near Xuzhou iron-steel plant may be assessed，but the relation whether the concentrations of SO_2 are conjectured by determining the corresponding these values of magnetic parameters is still discussed further. The biogeochemical cycles of Pb，Cu，Zn，Cd，Se，Fe，Mn and Hg in the soils are associated with the concentrations of Fe-Mn oxides to some extent.

Keywords：urban soils；heavy metals；environmental magnetism；proxy；leaching；accumulation；Xuzhou

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