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典型锑冶炼地块不同功能区土壤中锑、砷污染分布特征及风险评估

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摘要:【研究目的】我国拥有世界上最丰富的锑(Sb)资源, 近些年来锑及其伴生的砷污染已成为我国的典型环境问题。本文聚焦研究锑冶炼地块污染物的垂向以及不同功能区分布特征, 试图对阐明锑冶炼行业的土壤污染风险提供帮助。【研究方法】本文通过采集四处典型的锑冶炼厂各重点功能区不同深度的土壤样品, 对比分析了土壤 Sb、As 在不同功能区污染水平的差异化特征以及垂向上污染物含量的累积迁移情况。并采用单项污染指数法探究表层土壤 Sb 及 As 在不同功能区受污染程度, 同时根据重金属 Sb、As 的毒性响应系数, 对潜在生态风险的分级标准进行合理调整, 分析表层土壤 Sb、As 的生态风险。【研究结果】土壤 Sb 和 As 含量在垂向上均表现为表层>中层>深层, 并且表层土壤 Sb 和 As 含量均属于强变异等级。不同功能区 Sb、As 含量的差异明显, 总体表现为储存区>废水治理区>生产区, 随着深度的增加, Sb 含量均值在三大功能区均超标, 但是 As 含量均值到中层仅在储存区超标, 到深层便不存在超标。Sb 和 As 在三大功能区的污染水平及生态风险均为储存区>废水治理区>生产区, 且污染风险最大值点位均位于储存区, Sb 在三大功能区均是重度污染点位占比最高处于极强的生态风险, As 在三大功能区除储存区外, 废水治理区和生产区均是清洁、尚清洁点位占比大处于中强度生态风险。【结论】锑冶炼地块多年的人为生产经营活动, 引起了表层土壤 Sb 和 As 的严重污染富集。垂向上来看 Sb 是首要污染物, 并且污染超标已迁移至中深层, 其伴生元素 As 的污染水平尚可接受主要集中在表层; 三大功能区中储存区是污染水平、生态风险最高的区域, 但若一旦发生污染, 同时也需要关注废水治理区。

关键词: 锑污染; 砷污染; 垂向分布; 不同功能区; 风险评价; 环境地质调查工程

创 新 点: (1)分析锑冶炼地块主要污染物的垂向以及不同功能区分布情况, 首次确定了 Sb 和 As 在土壤垂向上的累积和不同功能区的分布差异。(2)对潜在生态风险评价中土壤重金属污染毒性响应系数和分级标准根据实际情况进行优化调整, 合理给出了锑冶炼行业对土壤产生的潜在生态风险。

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Distribution characteristics and risk assessment of antimony and arsenic contamination in soils of different functional areas of typical antimony smelting sites

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Abstract: This paper is the result of environmental geological survey engineering.

[Objective] China has the richest antimony (Sb) resources all over the world. In recent years, antimony and its associated arsenic pollution have become a typical environmental problem in our country. This paper focuses on the vertical distribution and the distribution characteristics in different functional areas of pollutants in the antimony smelting site, aiming to provide assistance in clarifying the soil pollution risks of the antimony smelting industry. **[Methods]** This paper presents the results of a study conducted to investigate the differentiated characteristics of soil antimony (Sb) and arsenic (As) pollution levels in different functional areas and the cumulative migration of pollutant contents in the vertical direction. Soil samples were collected at different depths from four typical antimony smelting plants in different key functional areas. The single pollution index method was employed to ascertain the pollution degree of surface soil Sb and As in different functional areas. In order to analyse the ecological risk of surface soil Sb and As, the grading standard for potential ecological risk was adjusted in a reasonable manner according to the toxicity response coefficients of heavy metals Sb and As. **[Results]** The soil Sb and As contents exhibited a vertical distribution, with the highest concentrations observed in the surface layer, followed by the middle layer and finally the deep layer. Furthermore, the Sb and As contents in the surface layer exhibited high variability. The varies of Sb and As contents in different functional areas is obvious, and the overall performance is storage area > wastewater treatment area > production area. As the depth increased, the average Sb content exceeded the standard in all three functional zones. However, the average As content exceeded the standard only in the storage area of the middle layer and did not exceed the standard in the deep layer. The pollution level and ecological risk of Sb and As in the three functional zones were as follows: storage area > wastewater treatment area > production area, moreover, the maximum value of pollution risk was in the storage area. As for Sb, heavy pollution points accounted for the highest ration in three functional zones, and the ecological risk is “extremely strong”. As for As, the proportion of “clean” / “not yet clean” is considerable in three functional zones exception for storage area, and the ecological risk in the wastewater treatment area and the production area is “medium”. **[Conclusions]** Years of anthropogenic production and operation activities at the antimony smelting site have resulted in significant pollution and enrichment of Sb and As in the surface layer of soil. A vertical view of the data reveals that Sb is the primary pollutant, with pollution levels exceeding the standard migrating to the middle and deep layers. The pollution level of its accompanying element, As, is still acceptable, with the majority concentrated in the surface layer. In the three functional areas, the storage area owns the highest level of pollution and ecological risk. Additionally, the wastewater treatment area should be concerned in case of the pollution occurs.

Key words: antimony pollution; arsenic pollution; vertical distribution; different functional areas; risk assessment; environmental geological survey engineering

Highlights: (1) The vertical distribution of major pollutants in antimony smelting plots, as well as their distribution in different functional areas, are firstly analyzed in this paper. This study also determines the accumulation of Sb and As stains in the vertical direction of the soil and the differences in their distribution in different functional areas. (2) The toxicity response coefficients and grading standards for soil heavy metal pollution in the evaluation of potential ecological risk are optimized and adjusted according to the actual situation in this study. This allows for a reasonable estimation of the potential ecological risk of the antimony smelting industry on soil.

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1 引言

工业场地受污染是一个全球性的环境问题(尧一骏, 2016), 尤其是冶炼场地。生产过程中衍生的各类型污染物对土壤、地下水等环境介质甚至人体

健康产生严重危害(李强等, 2021; 孙厚云等, 2023; 任宇等, 2024)。众多学者对铅锌冶炼、铝冶炼等行业的风险进行评估研究, 但对锑冶炼行业关注的较少, 锑冶炼的特征污染物主要是锑及伴生的砷元素(库文珍等, 2012; 袁程等, 2015)。锑(Sb)和砷(As)

是一种具有金属光泽的类金属元素,二者化学结构类似,且均具有毒性或者潜在毒性,高含量的 Sb 会让人呕吐、腹泻,严重的会造成心血管疾病、心脏病和呼吸疾病等,同时长期暴露在 As 污染的环境中会增加糖尿病、心血管疾病、神经功能障碍患病风险,还会导致皮肤病变等(Oorts, 2009; Takahashi and Shozugawa, 2010; Okkenhaug et al., 2013)。综合其潜在毒性和致癌性,欧盟和美国环境保护协会分别于 1976 年和 1979 年把 As、Sb 列为优先控制污染物(He et al., 2019; Zhang et al., 2020)。

我国拥有世界上最丰富的锑(Sb)资源,已探明储量的矿区有近 200 处,近年来,锑矿探明储量仍在不断增加,由于锑矿采冶以及含锑制品的广泛使用,大量的锑及所伴生的砷元素被释放进入土壤等表生环境介质中,这使得锑、砷污染已成为我国的典型环境问题(Wang et al., 2011; Ning et al., 2015)。目前,对锑矿区污染特征的研究多集中在表层土壤的横向污染分布特征以及赋存相态方面的研究,对污染物在工业场地不同功能区含量的差异性以及在土壤中纵向分布的规律研究较少。实际上,各功能区无论在生产环节承担的角色还是现场环境状况都不尽相同,污染状况自然也不该以整个场地一概而论,找到不同功能区污染程度的差异化,对精准管理企业可提供技术支撑,其次重金属污染会随时间垂直向下累积迁移,最终污染到地下水,为了实现土壤污染物对地下水影响的风险管控,研究土壤污染物在垂直方向上的分布特征是非常有必要的。

本文通过采集四处典型的锑冶炼厂各重点功能区不同深度的土壤样品,采用横向比对、纵向分析等多维角度对数据特征进行了剖析,对比分析了土壤 Sb、As 在不同功能区污染水平的差异化特征以及垂向上污染物含量的累积迁移情况,采用单项污染指数法探究 Sb 及 As 在不同功能区的含量和受污染水平,同时根据重金属 Sb、As 的毒性响应系数,合理地调整潜在生态风险的分级标准,分析各功能区的潜在生态风险,以期工业场地环境管理提供科学依据。

2 材料与方法

2.1 研究区概况

研究区位于陕西省的南部,陕南地区矿产资源

丰富,是陕西省有色金属储量富集区之一,已探明的矿区资源有汞、锑、铅、锌、金、银、钡、铜等 39 种。其中锑矿的品味高、储量大,矿石主要由辉锑矿和石英组成。本次选取了四处典型且生产时间大于 20 年的锑矿冶炼厂,结合实际情况共布设 25 个采样点,依厂区来看,在 A 厂区内布点 4 个, B 厂区内布点 6 个, C 厂区内布点 6 个, D 厂区内布点 9 个;依功能区来看,在生产区布点 10 个,废水治理区 7 个,储存区 8 个。具体见图 1。

2.2 布点、采样及检测

综合考虑污染物分布及类型、迁移途径以及地块水文地质等情况,识别疑似污染区域并进行布点,采用 Geoprobe 原状土壤钻机高频冲击方式进行钻探取样,采集各布点区域内土壤点位表层(0~0.5 m)、中层(0.5 m 至地下水位线)和深层($\geq$ 地下水位线)土壤样品,采样时应避开杂填土及管线等,中层土壤样品在存在污染痕迹或现场快速检测设备识别污染相对较重的深度采集,同时每个地块需按照不少于总样品数的 10% 采集土壤密码平行样,其中密码样仅作为质量控制,不纳入后续统计分析。样品装于土壤样品袋中编号、记录并保存,样品总数量为 75 个。

采集的土壤样品均室温避光风干,去除动植物残体、碎石等杂物,研磨过 100 目尼龙筛后置于牛皮纸袋中备用。本研究中, Sb 和 As 含量的测定方法参照《土壤和沉积物 汞砷硒铋锑的测定》(HJ 680—2013),采用微波消解/原子荧光法对土壤样品进行前处理与检测,检出限均为 0.01 mg/kg。整个测试过程中实验室严格按照质量体系运行,通过进行土壤标准物质和密码平行样的对比,确保测定结果精密度和准确度率均达到或优于设计要求,测试质量精准可靠。

2.3 风险评估

2.3.1 单项污染指数法

单项污染指数是指土壤重金属含量的实测结果与评价标准的比值,能直接地反映出其污染程度(郭畔等, 2019; 张江华等, 2020; 周伟等, 2021)。计算公式具体为:

$$P_i = C_s^i / C_n^i \quad (1)$$

式中, P_i 为土壤中重金属的单项污染指数; C_s^i 为 i 元素实测结果, mg/kg; C_n^i 为其评价标准, mg/kg,

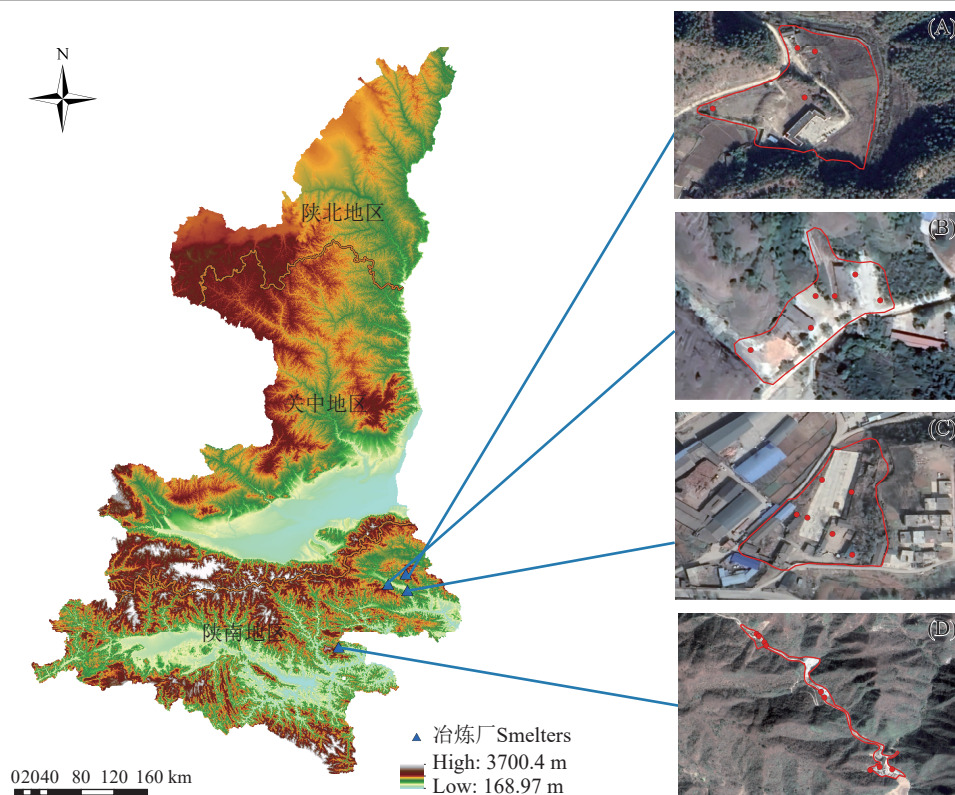


图1 研究区示意图

Fig.1 Schematic diagram of the study area

本研究的评价标准以《土壤环境质量 建设用地土壤污染风险管控标准(试行)》(GB36600—2018)中的第二类用地(工业用地)风险筛选值为准,其中Sb和As的风险筛选值为180、60。污染等级划分具体为: $P_i \leq 1$ 为清洁; $1 < P_i \leq 2$ 为尚清洁; $2 < P_i \leq 3$ 为轻度污染; $3 < P_i \leq 5$ 为中度污染; $P_i > 5$ 为重度污染(郭伟等,2011;崔罗肖等,2022)。

2.3.2 潜在生态指数法

潜在生态指数法(Potential Ecological Risk Index, PERI),既充分考虑了重金属的含量,又结合了重金属的毒性水平,能够综合反映重金属对生态环境的影响能力(Chen et al., 2023),是目前最为常用的评价土壤重金属污染程度和生态风险的方法(陈佳林等,2021;林苙等,2021)。计算方法如下:

$$E_r^i = T_r^i \times \frac{C_i}{C_n} \quad (2)$$

式中: E_r^i 为重金属*i*的潜在生态危害系数; C_i 为重金属*i*的实测值,mg/kg; C_n 为重金属*i*的风险筛选值,mg/kg; T_r^i 为重金属*i*的毒性响应系数。Sb和As的毒性响应系数与毒性系数相同(Yao et al.,

2019),毒性响应系数分别为10(Wang et al., 2018)、7(徐争启等,2022)。该方法的分级标准需要根据实际情况进行调整,根据Hakanson对 E_r^i 第一级上限值由非污染系数($C=1$)与参评污染物毒性响应系数最大值相乘得到,其他风险级别的上限值分别用上一级值乘以2得到。本研究所有污染物中的最大毒性系数为10,故 E_r^i 第一级界限值为10,调整后的潜在生态风险指数分级标准(马建华等,2020;秦元礼等,2022)见表1。

3 结果与讨论

3.1 土壤金属锑、砷含量垂向分布特征

对四处典型的锑矿冶炼厂各重点功能区不同深度的75个土壤样品检测分析,结果如表2和图2所示。锑冶炼厂土壤Sb的平均值为5420.63 mg/kg。在不同深度土壤中均有Sb检出,且随着土壤深度的增加,Sb含量逐渐降低,此结果与国内外许多学者的研究结果一致(刘庚等,2018;林苙等,2021);土壤中Sb的平均值为表层(13027.44 mg/kg) > 中层(2581.48 mg/kg) > 深层(652.96 mg/kg),且表层土

表 1 潜在生态风险指数分级标准
Table 1 Grading criteria for the potential ecological risk index

潜在生态风险	轻度	中度	较强	很强	极强
E_r^i	<10	10~20	20~40	40~60	≥60

壤 Sb 的最大值(122471 mg/kg)同样高于中层(17500 mg/kg)和深层(9070 mg/kg)。根据差异显著性分析,表层土壤 Sb 含量与中层和深层相比差异显著($P<0.05$),表明 Sb 在表层土壤中存在明显富集。总体上,土壤 Sb 含量的变异程度非常高,变异系数高达 288%,其中深层土壤 Sb 含量的变异系数(278%)大于表层和中层(194% 和 167%),均属于强变异等级,表明 Sb 含量在土壤分布中不均匀,其含量受锑冶炼的人为活动影响较大(管孝艳等,2012)。

As 的含量呈现出来与 Sb 类似的规律。同 Sb 一样,在不同深度土壤中均有 As 检出,且随着土壤深度的增加,As 含量逐渐降低,表层土壤 As 的平均值为 246.9 mg/kg>中层(89.61 mg/kg)>深层(33.6 mg/kg),且表层土壤 As 的最大值(2195 mg/kg)同样高于中层(653 mg/kg)和深层(197 mg/kg)。总体上,土壤 As 含量的变异程度非常高,变异系数高达 227%,其中表层和中层土壤 As 含量的变异系数(180% 和 145%)大于深层(117%),由于深层土壤 As 含量的变化也属于强变异等级,表明土壤母质对 As 的空间分布同样具有显著影响,而表层变异系数大于深层,说明人类活动对土壤 As 的分布造成了一定的干扰。

与《土壤环境质量 建设用地土壤污染风险管控标准(试行)》(GB 36600—2018)对比,Sb、As 在表层土壤的点位超标率最高,其中 Sb 的点位超标率(超筛选值、超管制值)均达到了 96%,As 的点位超标率为 64%(超筛选值)和 44%(超管制值),明显低

于 Sb 的超标率。随着深度的加深,两种元素的超标率虽显著下降,但在中层和深层依旧存在超筛选值和超管制值现象,可见污染主要发生在表层土壤但同时也存在向下累积的风险。

3.2 不同功能区土壤锑、砷含量垂向分布特征

不同功能区 Sb、As 含量的差异明显,总体表现为储存区>废水治理区>生产区(图 3)。Sb 含量在表层土壤表现为储存区(22272.63 mg/kg)>废水治理区(10731.71 mg/kg)>生产区(7238.3 mg/kg),As 含量在表层土壤表现为储存区(525.38 mg/kg)>废水治理区(126.6 mg/kg)>生产区(108.34 mg/kg),主要是因为储存区用于堆存锑矿石或者冶炼渣,且多属于地面硬化破损或未硬化、没有防雨棚,长期接受雨淋,表层土壤易发生污染物渗透扩散,而其他两类功能区多属于硬化完整地面,所以在表层土壤储存区 Sb、As 含量明显高于其他两类功能区。中层土壤 Sb 含量表现为生产区>储存区>废水治理区,As 含量表现为储存区>废水治理区>生产区,但三大功能区的含量差异性相较于表层土壤明显减小,并且 As 含量(均值)在废水治理区和生产区已不存在超标。深层土壤 Sb 表现为废水治理区>生产区>储存区,As 表现为储存区>生产区>废水治理区,其中 As 含量(均值)在三大功能区均不超标,但 Sb 的含量在废水治理区的含量明显高于其他两类功能区,说明在废水治理区,一旦发生污染,污染物更易随着水体介质的下渗迁移至深层。

总体来看,锑冶炼厂的三大功能区发生污染风险最高的区域是储存区,一旦发生污染,向深层迁移风险最高的则是废水治理区,因而在捕捉污染时,需重点关注储存区的表层土壤及废水治理区的池体以下深层土壤。

表 2 土壤 Sb、As 描述性统计分析
Table 2 Descriptive statistical analysis of Sb and As in soil

项目	层位	均值	中位数	标准差	范围	变异系数	筛选值	超标率%	管制值	超标率%
垂向	Sb	总体	5420.63	892	15599.48	2.88		73		68
		表层	13027.44	2920	25266.12	1.94	180	96	360	96
		中层	2581.48	967	4306.71	1.67		80		80
		深层	652.96	63.9	1813.72	2.78		44		22
	As	总体	123.37	36.3	280.04	2.27		60		22
		表层	246.9	106	444.96	1.80	60	64	140	44
		中层	89.61	32.9	129.58	1.45		44		20
		深层	33.6	18.7	39.36	1.17		12		4

注: Sb和As的筛选值、管制值参考《土壤环境质量 建设用地土壤污染风险管控标准(试行)》(GB 36600—2018)中的第二类用地(工业用地)标准值,其均值、中位数、标准差、范围、筛选值、管制值的单位均为mg/kg。

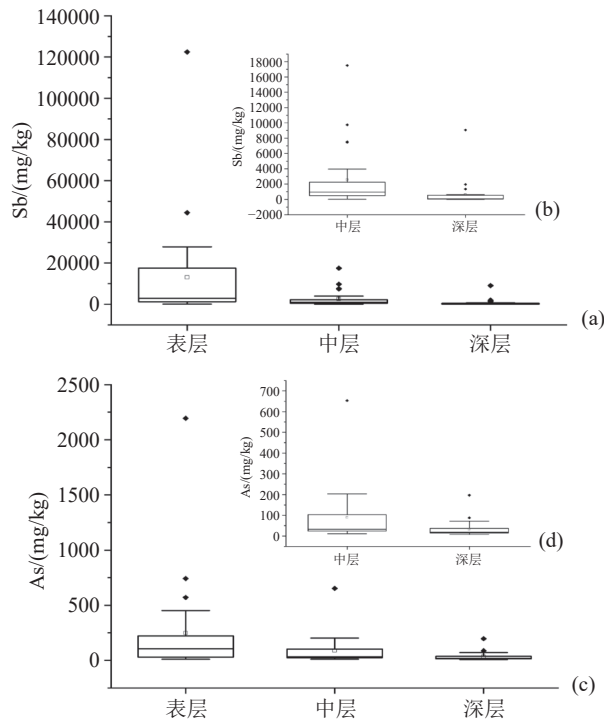


图2 土壤 Sb、As 垂向分布特征
Fig.2 Vertical distribution characteristics of Sb and As in soil

3.3 表层土壤金属锑、砷潜在生态风险评价

以《土壤环境质量 建设用地土壤污染风险管控标准(试行)》(GB36600—2018)中的第二类用地(工业用地)风险筛选值为准,采用单因子污染指数对不同功能区表层土壤 Sb、As 污染等级进行划分(表3)。结果显示:表层土壤 Sb 的单项污染指数范围在 0.24~680.39,最小值点位位于生产区、最大值点位位于储存区,平均值为 72.37,有 84% 的点位处于重度污染水平,仅 1 个点位处于清洁水平;Sb 在三大功能区均是重度污染点位占比最高,从单项污染指数均值来看,储存区最高(123.74),其次为废水治理区(59.62)和生产区(40.21)。表层土壤 As 的

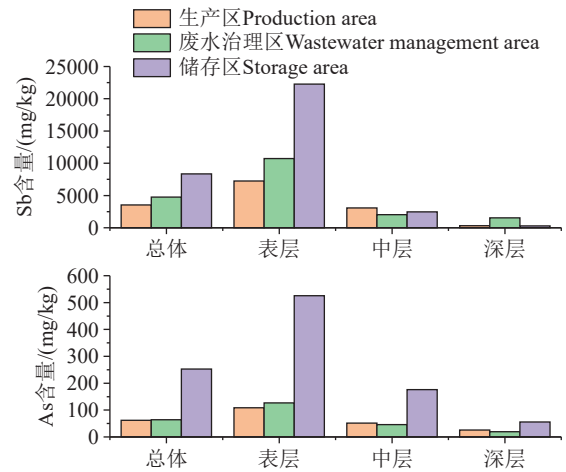


图3 不同功能区土壤 Sb、As 含量对比
Fig.3 Comparison of Sb and As content in soils of different functional areas

单项污染指数范围在 0.2~36.58,平均值为 4.12,最小值点位位于废水处理区、最大值点位同样位于储存区,有 40% 的点位处于中度、重度污染水平,同时,可以看出 As 比 Sb 的污染指数要低很多,可见在锑冶炼场地, Sb 是首要污染物,其伴生的元素 As 污染水平尚可接受,从单项污染指数均值来看, As 在三大功能区除储存区外,废水处理区和生产区均是清洁、尚清洁点位占比大,从单项污染指数均值来看,储存区最高(8.76),其次为废水治理区(2.11)和生产区(1.81)。总体而言,表层土壤 Sb 和 As 污染程度高低顺序为: 储存区>废水治理区>生产区。

表层土壤 Sb、As 的潜在生态风险指数计算结果(表4),冶炼地块表层土壤 Sb 的潜在生态风险指数(E_i^p)范围在 1.71~4762.76,最大值位于储存区,有 72% 的点位处于极强风险,使得 Sb 整体表现为极强的潜在生态风险,三大功能区也都表现为极强的潜在生态风险,其中储存区有 87.5% 的点位处于极强风险,废水处理区有 85.7% 的点位处于极强风

表3 不同功能区表层土壤 Sb、As 单因子污染指数统计

Table 3 Statistics of single-factor pollution index of Sb and As in surface soil of different functional areas								
项目	功能区(点位数)	单因子污染指数	范围	清洁	尚清洁	轻度污染	中度污染	重度污染
Sb	总体(25)	72.37	0.24~680.39	1	1	1	1	21
	储存区(8)	123.74	2.89~680.39	/	/	1	/	7
	废水治理区(7)	59.62	5.78~246.87	/	/	/	/	7
	生产区(10)	40.21	0.24~154.44	1	1	/	1	7
As	总体(25)	4.12	0.2~36.58	9	5	1	6	4
	储存区(8)	8.76	2.47~36.58	/	/	1	5	2
	废水治理区(7)	2.11	0.2~7.52	4	1	/	1	1
	生产区(10)	1.81	0.32~9.52	5	4	/	/	1

表 4 不同功能区表层土壤 Sb、As 生态风险指数统计

Table 4 Ecological risk index statistics of Sb and As in surface soil of different functional areas								
项目	功能区 (点位数)	均值	范围	表层土壤样品各级生态风险样品数				
				轻度	中度	较强	很强	极强
Sb	总体 (25)	506.62	1.71~4762.76	2		2	3	18
	储存区 (8)	866.16	20.18~4762.76			1		7
	废水治理区 (7)	417.34	40.44~1728.07				1	6
	生产区 (10)	281.49	1.71~1081.11	2		1	2	5
As	总体 (25)	41.15	1.97~365.83	9	5	5	2	4
	储存区 (8)	87.56	24.67~365.83			4	2	2
	废水治理区 (7)	21.1	1.97~75.17	4	1	1		1
	生产区 (10)	18.06	3.22~95.17	5	4			1

险,生产区有 50% 的点位于极强风险。Sb 在三大功能区均为极强的生态风险且储存区>废水治理区>生产区(图 4)。As 的潜在生态风险指数(E_r^i)范围在 1.97~365.83,最大值同样位于储存区,处于很强、极强的点位仅占 24%,三大功能区其中储存区的生态风险最高,有 50% 的点位于很强、极强风险,废水治理区和生产区均只有 1 个点位于极强风险,风险主要集中在轻度和中度,其中废水治理区有 71.42% 的点为轻中度风险,生产区有 90% 的点位于轻中度风险 As 仅在储存区表现为极强的生态风险,在废水治理区表现为很强的生态风险,在生产区表现为中度生态风险(图 4)。

4 结论

(1)锑冶炼地块 Sb 含量及其伴生的 As 的在表层土壤含量均明显高于《土壤环境质量 建设用地土壤污染风险管控标准(试行)》(GB 36600—2018)的

第二类用地(工业用地)筛选值甚至管制值,说明冶炼厂的生产活动对土壤 Sb 和 As 的分布造成了一定的干扰,引起了表层土壤的严重污染富集。Sb 和 As 含量在垂向上均表现为表层>中层>深层,随着深度的增加,两种元素均存在超标点位,但超标率在逐渐下降。

(2)通过对表层土壤含量的分析,Sb 和 As 在三大功能区中均是储存区的含量最高,随着深度的增加,Sb 含量均值在三大功能区均超标,并且在废水治理区的含量明显高于其他两类功能区,垂向上来看 Sb 是首要污染物,并且污染超标已迁移至中深层,今后类似场地一旦发现污染物超标,需关注废水治理区的中深层。As 含量均值到中层仅在储存区超标,到深层便不存在超标。

(3)Sb 和 As 在三大功能区的污染水平及生态风险均为储存区>废水治理区>生产区,且污染风险最大值点位均位于储存区,Sb 在三大功能区均是重度污染点位占比最高处于极强的生态风险,As 在三大功能区除储存区外,废水治理区和生产区均是清洁、尚清洁点位占比大处于中强度生态风险,表明污染风险等级最高的是储存区。

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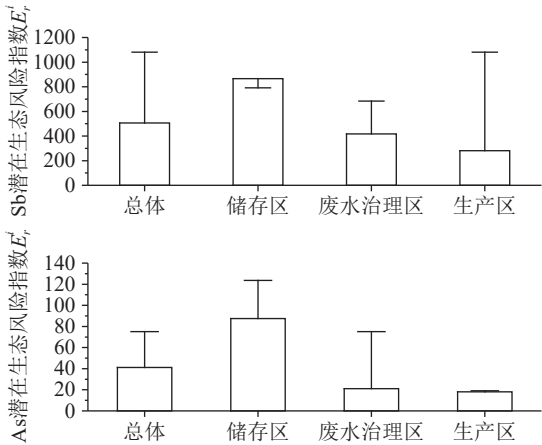


图 4 不同功能区表层土壤 Sb、As 生态风险指数对比
Fig.4 Comparison of ecological risk index of Sb and As in surface soil of different functional areas

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