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酸雨区不同用地类型土壤有效态 Cd 含量季节变化及关键影响因子

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摘要: 探明污染土壤重金属有效态含量的季节变化特征及其敏感影响因子, 对农业生产过程中减低重金属生态风险具有重要的参考价值. 研究于湘江中下游典型 Cd 超标农业小流域中选取稻田、旱作蔬菜地、丘陵林地这 3 类主要用地类型, 分析不同用地类型 Cd 活性的季节变化特征及其与土壤基本理化参数的关联. 为期 1 a 的原位监测结果显示, 研究区为典型酸雨区, 雨水 pH 值呈现冬、春季节低于夏、秋季. 稻田土壤总 Cd 含量显著高于旱作蔬菜地, 菜地显著高于林地, 3 种用地类型土壤总 Cd 含量季节特征相似, 均为夏秋季节略低于冬春两季. 3 种用地类型 Cd 有效态季节变化与总 Cd 含量无明显的相关性, 稻田土壤有效态 Cd 含量在 5~9 月的作物生长季明显低于其他月份, 而菜地和林地则恰好相反. 稻田土壤 Cd 有效性的最关键影响因子为 E_h , 呈显著正相关, 与土壤 pH 负相关, 菜地土壤与土壤 TOC 明显负相关, 而林地土壤 Cd 有效性与水溶性有机碳、TOC 呈现明显的正相关关系. 研究可为 Cd 超标土壤污染阻控与农业安全生产提供一定的数据参考.

关键词: 酸雨区; Cd 污染; 有效态 Cd; 季节变化; 关键影响因子

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Major Factors Influencing the Cd Content and Seasonal Dynamics in Different Land Cover Soils in a Typical Acid Rain Region

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Abstract: Understanding the seasonal variations in active heavy metal components and sensitive impact factors is of importance for the ecological risk reduction during the agricultural production processes. Paddy fields, vegetable lands, and hilly forests were selected as three main land cover types to assess the seasonal characteristics of Cd bioavailability and reveal how or to what extent it was affected by the physiochemical parameters of soils, under different land-use types in a typical Cd-contaminated watershed in the middle and lower reaches of Xiangjiang River. One-year *in situ* monitoring results showed that natural rainfall pH in winter and spring was lower than in summer and autumn in the study region. The total Cd content of paddy soils was significantly higher than that of the vegetable soil, while the hilly forest soil showed the lowest total Cd value. Similar seasonal variations in total Cd content were found in three soil types with slightly lower summer and autumn concentrations than spring and winter values, but no obvious correlation was detected between the total and the available Cd components. The paddy soil available Cd concentration during the 5-9-month crop growth season was significantly lower than the other months of the year, while vegetable cultivation and hilly forest soils showed the opposite trend. E_h was the key factor that had a positive influence on the Cd activity in paddy soil. Soil TOC concentration was negatively correlated with soil activity in vegetable soil. TOC, water soluble organic carbon, showed a significant positive correlation with Cd effectiveness. The results provide scientific references for Cd contamination control and safe agricultural production.

Key words: acid rainfall region; Cd contamination; available Cd; seasonal variation; key influencing factor

土壤重金属超标已成为当代社会最为关注的全球性环境污染问题之一^[1,2], 我国近 2×10^7 hm^2 农田存在不同程度的重金属超标问题, 其中以西南地区的重金属超标问题尤为严重^[1,3,4]. 湖南是我国著名的有色金属之乡, 多数有色以及稀有金属矿产的采冶均集中分布在湘江流域内, 导致湘江流域成为我国重金属超标最突出的流域之一, 耕地土壤中 Cd、Pb、As、Cu 等超标明显, 其中 Cd 超标率最高^[5,6]. 湖南省同属典型酸雨区, 降雨及土壤 pH 值

的双重酸性致使该区土壤金属离子活性较高, 具有较强的生物有效性和迁移性^[7], 易随降雨迁移到水环境中, 重金属随径流向水体迁移已成为受纳水体及其底泥沉积物重金属超标问题的主要诱因^[8-10].

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近年来,有关河、湖、海等水体与底泥沉积物、及其水生生物体内重金属污染风险来源及评价研究日益得到国内外研究学者的关注^[10~12],土壤遭受重金属污染后易随产汇流过程迁移到水体,进而通过生物富集及生物放大进入食物链危害人类健康^[7,10,13].在诸多重金属污染物中,Cd 是生物毒性最强的重金属元素之一,毒性仅次于汞^[14],是目前我国农田土壤超标率最高的重金属元素,其中尤以湖南农田土壤最为典型^[6],已有研究表明外源重金属污染物进入土壤后短期内有效态 Cd(水溶态和可交换态)所占比例较高,且具有较强的生物活性和移动性^[15].污染土壤重金属生物有效性及毒性主要表现为自由离子的活性,即可溶态的含量而非总量,土壤 pH 是影响 Cd 有效性所有参数中的最重要因子,环境 pH 值越低,Cd 等重金属离子可迁移性越高、毒性也就越大^[9,16],同时土壤中重金属会随环境因子变化在不同赋存形态间互相转化^[15].为减轻典型毒性重金属对生态系统与人类健康的危害,国内开展了大量的土壤重金属污染修复技术研究^[17],但却较少关注污染土壤中重金属活性的季节动态及其关键影响因子,因农田土壤的理化性质非常复杂,导致农田土壤重金属活性的季节性研究尤其缺乏,探明湘中典型酸雨区污染土壤重金属 Cd 有效态含量的季节变化及其关键影响因子对该区农业生产关键生态过程中重金属污染阻控与重金属面源污染定量估算与防治具有重要的科学价值和数据意义.

基于以上研究背景,本研究于典型重金属 Cd 超标农业小流域选取主要用地类型,分析土壤有效态 Cd 含量的季节变化与土壤重要理化及微生物影响因子关联,旨在探明不同用地类型有效态 Cd 的季节变化特征,并探索其关键影响因子,以为农业生产关键过程中重金属 Cd 污染阻控提供数据支持及科学参考.

1 材料与方法

1.1 区域概况

本研究选取湖南省株洲市马家河镇新马村(N27°50'1.3", E113°02'8.4")农业小流域,因附近一家小型电镀厂废水的无序排放而导致周围农田受到严重污染(电镀厂已于 2007 年关闭,该区已被国家环保部和湖南省确定为“重金属污染综合治理技术示范区”),研究区属亚热带季风性湿润气候,年均降雨量 1 300 mm,年均气温 17.6℃,属典型丘陵地貌,土壤为酸性红壤,且同属典型酸雨区,雨水样

品采集分析结果显示该区雨水呈现典型酸雨特征,雨水 pH 值为冬、春季节高于夏、秋季节,实验期雨水 pH 值季节动态见图 1.

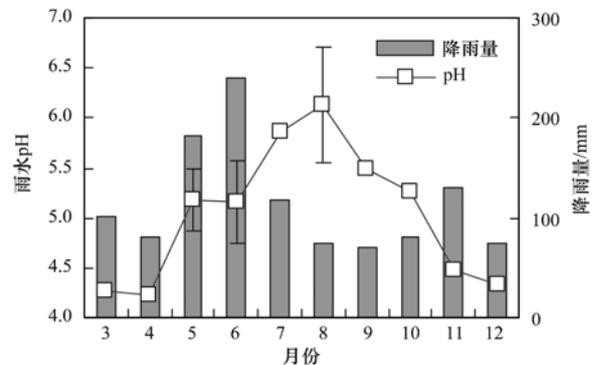


图1 研究区降雨与雨水 pH 季节变化特征

Fig. 1 Seasonal variations in precipitation and rain pH value in the study region

1.2 实验设计与样品采集

基于流域土地利用现状调研,研究选取流域内最主要的 3 种土地利用类型(丘陵林地、菜地、稻田)作为研究对象,并对采样地点进行 GPS 定位确定样地范围,样地基本概况见表 1. 结果可见,稻田、菜地土壤可交换态 Cd 含量显著高于丘陵林地,而稻田总 Cd 含量显著高于菜地,菜地显著高于丘陵林地(表 1).

本研究于 2015 年 3 ~ 12 月每月中旬用手持式土钻(Φ 8 cm)按三角形“三点法”采集 3 种不同用地类型的 0 ~ 15 cm 土层的土壤样品,并于三角形的每个样点区域用土钻随机平行采集 3 个重复样品混成一个土样,并装入布袋带回实验室放入冰柜保存待测.

1.3 分析方法与数据处理

文中土壤总 Cd、可交换态 Cd 含量测定均采用混匀后的新鲜土壤样品,同时测定土壤水分含量,文中显示的 Cd 含量均为换算后的烘干土壤含量.土壤总 Cd 含量用 HF-HClO₄-HNO₃ 消煮后用原子吸收分光光度计(TAS-990)测定.土壤可交换态 Cd 含量以水土比为 10:1 的 0.01 mol·L CaCl₂ 溶液浸提后用石墨炉分光光度计(GTA120,美国 Varrian)测定.数据质量控制方法为:每个土壤样品平行消解 3 份,同时设置空白消解对比,并均采用优级纯酸试剂,土壤、水质样品均测试了标准溶液、空白和平行样.土壤 Cd 分析过程中土壤成分标准物质 GSS-10 的回收率为 95.7% ~ 109.3%,分析结果与标准差范围为 3.9% ~ 7.2%,加标回收率 93.7% ~ 106.6%.其中空白对照均低于仪器检测限,平行样误差在 \pm 6.0%.

表 1 实验样地基本情况

Table 1 Basic characteristics of experimental parameters

| 参数 | 土地利用类型 | | |
|---|--------------|--------------|--------------|
| | 稻田 | 菜地 | 丘陵林地 |
| 平均坡度/(°) | 0 | 0 | 29 |
| 样地面积/m ² | 335 | 78 | 200 |
| 土壤 pH 值 | 4.57 ± 0.07 | 4.65 ± 0.09 | 5.31 ± 0.11 |
| 表层(0~5 cm)土壤容重/g·cm ⁻³ | 1.29 ± 0.06 | 1.14 ± 0.07 | 1.31 ± 0.10 |
| 土壤有机碳/% | 3.38 ± 0.63 | 2.97 ± 0.39 | 2.58 ± 0.81 |
| 有效态氮/mg·kg ⁻¹ | 115.7 ± 20.1 | 96.8 ± 13.0 | 78.8 ± 33.0 |
| 有效态磷/mg·kg ⁻¹ | 9.97 ± 1.39 | 8.12 ± 0.68 | 5.05 ± 0.93 |
| 土壤总 Cd ¹⁾ /mg·kg ⁻¹ | 4.16 ± 0.51a | 2.35 ± 0.57b | 1.39 ± 0.20c |
| 可交换态 Cd/mg·kg ⁻¹ | 0.24 ± 0.06a | 0.21 ± 0.09a | 0.09 ± 0.02b |

1) 同行相同字母表示差异不显著 ($P \leq 0.05$)

土壤 pH, 自然降雨雨水样品在无过滤情况下用密封干净的聚乙烯塑料瓶带回实验室及时用精密酸度计 (PHS3C 型, 测量精度 ± 0.01) 进行 pH 值的测定. 土壤距土面 5 cm 处的 Eh 值采用智能便携式氧化还原电位仪 (QX6530) 测定, 以铂电极为指示电极, 饱和甘汞电极为参比电极, 当 1 min 电位值变化不超过 1 mV 时读数, 每个处理重复测定 3 次. 土壤 TOC、速效氮、速效磷、土壤容重等基本理化参数采用土壤农化分析方法测定^[18], 土壤微生物量碳 (MBC) 与土壤水溶性有机碳 (WSOC) 采用土壤农化分析中的氯仿熏蒸-K₂SO₄ 提取法测定^[18,19].

实验数据采用 Microsoft Excel 2003 进行描述统计分析, 选取 SPSS 16.0 统计软件进行相关分析及方差与显著性分析 (显著性在 $\alpha = 0.05$ 水平下进行). 同时, 研究采用 CCA 方法进行 Cd 有效态含量影响因子关系分析, 其中 pH 值取当月多次观测的平均值.

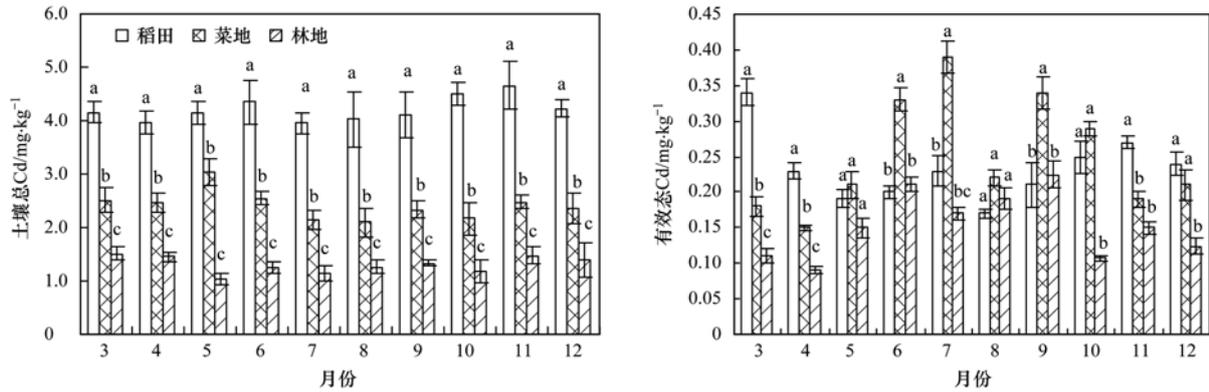
2 结果与讨论

2.1 不同用地类型土壤总量与可交换态 Cd 含量季节动态

研究结果显示, 稻田土壤总 Cd 含量明显高于旱作菜地, 菜地则明显高于丘陵林地, 可能因农业用地长期的肥料与农药施入、灌溉等因素导致稻田、菜地的土壤 Cd 累积效应明显, 而丘陵林地土壤 Cd 累积则主要来源于干湿沉降 (图 2), 3 种用地类型土壤总 Cd 含量的季节变化呈现一致性规律, 均为冬春季节略高于夏秋季节 (图 2). 已有研究表明大气中灰尘的沉降呈现明显的季节化运动特征, 冬季灰尘及其中 Cd 等重金属的沉降量最大, 春季次之, 而夏秋季则最低^[20,21], 加上南方降雨多分布在春夏

交际, 因此降雨淋洗与干沉降的季节特征双重影响导致了土壤总 Cd 含量呈现冬春季节略高于夏秋的特征^[22] (图 1 和图 2).

重金属的毒性不仅与其总量有关, 更大程度上由其形态分布决定, 国内外诸多研究指出土壤可交换态重金属浓度可有效反映其生物有效性、移动性与生态毒性^[23], 因此探明土壤可交换态 Cd 含量季节变化对于农业生产过程中阻控与减低 Cd 的生物累积性和生态毒性具有重要的指导价值. 农田土壤重金属污染修复措施的最终目标是通过各种技术方法阻隔 Cd 的生物有效性及其在作物体内的累积, 进而降低其农产品重金属 Cd 的超标风险. 研究分析了流域内旱作菜地与稻田 2 种主要的农业生产用地耕层 0~15 cm 土壤可交换态 Cd 的季节变化 (图 2), 稻田可交换态 Cd 含量与旱作菜地、丘陵林地的季节变化特征呈相反特征, 稻田土壤可交换态 Cd 含量为 5~9 月低于其他月份, 已有研究表明, 土壤水分变化可显著影响土壤重金属的有效性, 淹水土壤样品风干后交换态 Cd 明显提升^[24], 而 5~9 月为南方稻田生长季, 已有研究显示, 在淹水条件下, 土壤 Eh 会迅速降低, 土壤中 S 被还原为 S²⁻, S²⁻ 进而会与 Cd 离子形成 CdS 沉淀, 可大大降低土壤中 Cd 的生物有效性和毒性, 是导致稻田土壤可交换态 Cd 明显低于其他用地类型的主要原因^[25] (图 2). 研究显示, 土壤微生物及根系分泌物等生化作用强烈会导致土壤 pH 降低, 进而影响土壤中对 pH 值变化敏感的重金属 Cd 的生物活性, 尤其在南方酸性土壤中微生物和溶解性有机碳 DOC 含量增加对 Cd 的活化具有一定的促进作用^[4], 从而可能导致旱作菜地与丘陵林地土壤可交换态 Cd 含量均呈现夏季明显高于冬春季节 (图 2). 蔬菜地与林地土壤总 Cd 含



同时间系列不同小写字母表示差异显著, $P < 0.05$

图2 不同用地类型土壤总 Cd 及可交换态 Cd 含量季节动态

Fig. 2 Seasonal variation in total and available Cd concentrations for different land cover types

量虽然显著低于稻田,然而蔬菜地土壤有效态 Cd 含量显著高于淹水生长季的稻田土壤(图2),可见 Cd 污染农田进行水稻生产相比旱作农业具有较低的生态风险.

2.2 不同用地类型土壤有效态 Cd 及其关键影响因子

结果显示,3种用地类型土壤有效态 Cd 含量均与总 Cd 含量无明显相关性(图3),已有研究显示,淹水环境 Cd 的有效态所占比例与有机质含量呈显著正相关^[26],而稻田土壤 Cd 有效态与有机质含量则无明显正相关关系,可能由于稻田土壤的淹水、干燥环境的交替变化影响所致(图3).稻田土壤有效态 Cd 含量主要受 Eh 影响,与土壤 MBC、TOC、雨水 pH、土壤 pH、土壤温度等因子负关联,这一特征明显不同于菜地和林地土壤(图3).土壤微生物及根系分泌物等生化作用会导致土壤 pH 值呈现明显的季节特征^[27],会直接影响土壤中对 pH 值变化敏感的重金属 Cd 的生物活性,同时有报道称南方酸性土壤中溶解性有机碳 DOC 对 Cd 的活化具有一定的促进作用,在碱性土中则呈相反规律^[4],研究中影响旱作菜地土壤有效性 Cd 成分的因子比较复杂,与以往理解不同,菜地土壤 pH 值与 Cd 有效性呈现一定的正相关关系,与 MBC、Eh 等指标呈现弱的正相关,与雨水 pH、土壤 TOC、总 Cd 含量呈现较强的负相关关系,而林地土壤有效态 Cd 含量则主要受到土壤 TOC、WSOC 等有机质指标影响,大于雨水 pH、土壤 pH 等指标对 Cd 有效态的影响(图3).然而,由于实际生产环境年际变化大,因此研究尚需开展长时间序列的数据监测与收集工作,以便更好地为当地农田镉污染防治提供理论依据.

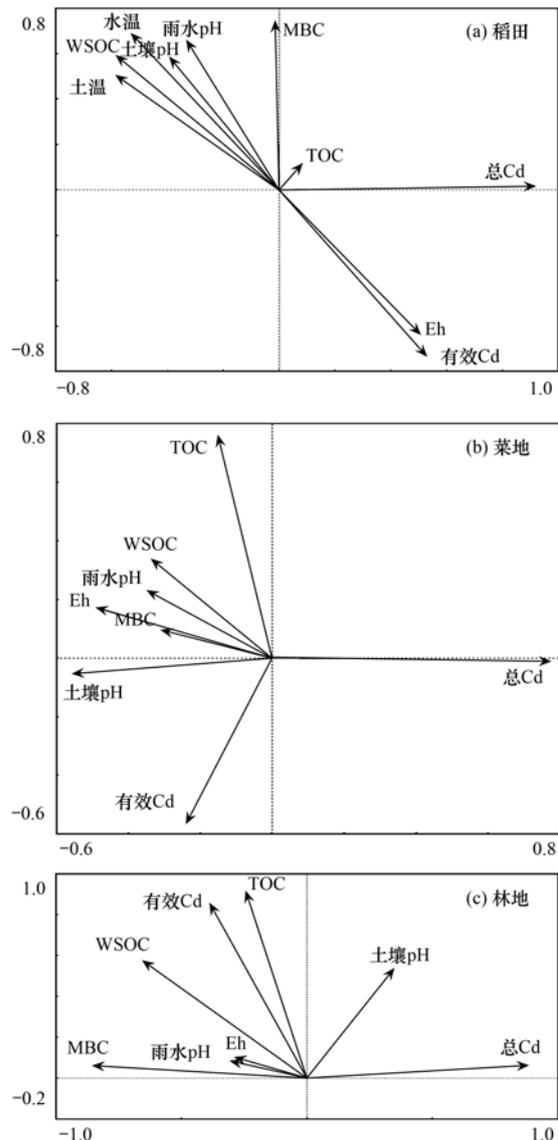


图3 不同用地类型土壤 Cd 有效性及其影响因子

Fig. 3 Cd activity and the sensitive impact factors of different land cover types

3 结论

不同用地类型土壤有效态 Cd 含量及其季节特征具有显著差异,稻田保持淹水是减低 Cd 生物有效性的有效措施。旱作蔬菜地的 Cd 总量低于稻田,但 5~9 月的生长季旱作蔬菜土壤有效态 Cd 含量显著高于稻田和林地,因此 Cd 污染土壤进行旱作农业生产可能具有更高的生态风险。林地土壤有效态 Cd 含量与可溶态有机质显著正相关。

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