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酸雨对含磷物质钝化修复的农田土壤磷流失的影响

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摘要:为探讨酸雨对经不同含磷物质钝化修复的镉污染农田土壤磷流失和磷有效性的影响,本研究选用磷酸二氢钙(MCP)、磷酸二氢铵(MAP)和磷酸二氢钾(MKP)3种典型含磷物质对镉污染农田土壤进行室内钝化试验,在室温下培养3个月。采用两种类型[硫酸型酸雨(SSAR)和混合型酸雨(SMAR)]、3种酸度(pH=3.0、4.0、5.6)的模拟酸雨(SAR)对钝化修复后土壤进行浸泡处理,并测定酸雨浸出液的总水溶性磷(TDP)和土壤的有效磷(AP)。结果表明:在各种酸雨条件下,MAP和MKP处理的土壤磷浸出率均显著高于对照组(CK),它们的磷浸出率分别达32.63%~38.57%和41.48%~49.29%,而MCP处理的磷浸出率与CK相比无显著差异。土壤磷浸出率随酸雨pH值的升高而降低,且SSAR处理的磷浸出率高于SMAR处理。3种含磷物质的添加均显著增加了土壤有效磷的含量,但在SAR处理后,土壤有效磷含量显著下降,且呈现MCP>MAP>MKP的规律。此外,SSAR处理的有效磷含量低于SMAR处理。因此,从土壤磷流失与磷有效性的角度考虑,MCP比MAP和MKP更适合于酸雨区镉污染农田土壤的钝化修复。

关键词:钝化修复;含磷物质;酸雨;磷流失;有效磷

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Effects of simulated acid rain on the leachability of phosphorus in agricultural soils amended with phosphorous compounds

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Abstract: This study evaluated the effects of simulated acid rain (SAR) on the leachability and availability of phosphorus (P) in cadmium (Cd)-contaminated agricultural soils amended with P compounds. Cd-contaminated soils were treated with calcium dihydrogen phosphate (MCP), ammonium dihydrogen phosphate (MAP), and potassium dihydrogen phosphate (MKP), and incubated for 3 months. These treatments were followed by two types of SAR [sulfuric acid rain (SSAR) and mixed acid rain (SMAR)] with three levels of acidity (pH = 3.0, 4.0, and 5.6). The total water-soluble P (TDP) of the leachate and available P (AP) in the soils were determined. The results showed that the leachability of P in MAP- and MKP-stabilized soils was significantly higher than that in controls (CK), being 32.63%~38.57% and 41.48%~49.29%, respectively. However, there was no significant difference in P leachability between the MCP-stabilized soils and the CK. Leachability of P decreased with increasing SAR pH. Additionally, the leachability of P in the SSAR treatments was higher than that in the SMAR treatments. The application of the three P compounds significantly increased AP concentration in soils. However, the AP concentration in the three stabilized soils significantly decreased after SAR soaking, in the order of MCP > MAP > MKP. Additionally, the AP concentration in the SSAR treatment was lower than that in the SMAR treatment. Therefore, MCP is more suitable for passivation in Cd-contaminated agricultural soils in acid rain areas than MAP and MKP, according to the leaching risk and availability of P in soils.

Keywords: passivation remediation; phosphorous compounds; acid rain; phosphorus leachability; phosphorus availability

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镉是我国农田土壤中常见的重金属污染物之一^[1-2]。作为我国重要的水稻种植区,南方地区农田土壤中的镉浓度高于我国其他地区^[3],该地区生产的水稻中有相当一部分超过了《食品安全国家标准 食品中污染物限量》(GB 2762—2017)中镉的限量值^[4]。大量研究表明,有毒金属在土壤中的积累会严重损害土壤的生产功能^[5-8],此外,镉在土壤-植物系统中具有较高的迁移性,会对农业生产、粮食安全以及人体健康构成严重威胁^[7,9-10]。此外,长期镉污染还可能降低土壤微生物的活性和多样性,进而影响土壤的养分循环^[11-13]。因此,如何降低农田土壤中镉的迁移性和生物有效性成为了国内外研究的热点^[14]。

钝化修复技术由于其修复效果好、成本低、适用性广等特点已被广泛应用于中轻度镉污染农田土壤修复中^[15-18]。含磷物质是常用的钝化剂之一,其不仅可有效降低农田土壤中镉的迁移性和生物有效性^[19-21],还可显著增加土壤磷含量,促进磷在土壤中的积累。大量研究表明磷在耕层土壤中的累积可能会增加土壤磷流失的风险^[22-23],并可能引起地表水富营养化^[24-25]。使用含磷物质钝化修复镉污染农田土壤,必将导致大量磷元素在耕层土壤中的积累,从而增加土壤磷流失的风险,而磷流失又可能导致被稳定的镉再度被活化^[14,26-27]。此外,我国南方地区是世界三大酸雨区之一,酸雨导致土壤酸化,进而加剧农田土壤氮、磷等养分的流失^[28-30]。现有研究表明,酸雨会加剧羟基磷灰石、磷酸二氢钾等含磷物质钝化修复的镉污染土壤中的磷流失,从而导致周边水体富营养化^[31-32]。因此,十分有必要研究酸雨对含磷物质钝化修复的镉污染农田土壤中磷流失及磷有效性的影响。

磷酸二氢钙(MCP)是一种略溶于水、水溶液呈酸性的无机磷酸盐,磷酸二氢铵(MAP)和磷酸二氢钾(MKP)是常用的可溶性磷肥,现有研究表明它们对土壤中的有效态镉均有良好的钝化效果^[14,33-34],而有关酸雨作用下其钝化修复的镉污染农田土壤中磷流失和磷有效性的研究少有报道。本研究以中轻度镉污

染农田土壤为研究对象,选择MCP、MAP和MKP 3种典型含磷物质作为供试钝化剂,通过室内钝化试验和模拟酸雨浸泡试验,探明酸雨对不同含磷物质钝化修复的镉污染农田土壤磷流失和磷有效性的影响,进而为酸雨区镉污染农田土壤含磷钝化剂筛选提供决策支持。

1 材料与方法

1.1 土壤样品准备

供试土壤为潴育型水稻土,采自江苏省扬州市某镉污染稻田,采样深度为0~20 cm。土壤样品经自然风干,剔除植物残体、石块等杂质后,过2 mm筛备用。供试土壤呈中性,pH值为7.25,土壤有机质含量为25.7 g·kg⁻¹,土壤总镉含量为1.55 mg·kg⁻¹。

1.2 钝化培养试验

称取12份供试土壤,每份100 g,按照3份1组将12份供试土壤随机分成4组,分别装入500 mL烧杯中;MCP、MAP和MKP 3种含磷物质分别按照表1所示的添加量各称取3份,分别添加到随机抽取的1组3个装有供试土壤的烧杯中,用玻璃棒搅拌并确保含磷物质与供试土壤混合均匀,并分别记为MCP组、MAP组和MKP组;剩下1组不添加任何物质作为对照组(标记为CK组),向每个烧杯中添加等量去离子水,确保土层上面有2~3 cm液面。为防止水分快速蒸发并允许一定程度的气体交换,烧杯口用带孔的保鲜膜封住,并置于恒温培养箱中在(25±3)℃下养护90 d,期间保持土壤中含水量为70%~80%。养护结束后将烧杯中土壤取出,自然风干,粉碎过2 mm筛,密封保存备用。处理后的各土壤总磷含量见表1。

1.3 模拟酸雨试验

为反映我国南方酸雨的特征,本研究分别制备了硫酸型酸雨(SSAR, SO₄²⁻/NO₃⁻=4:1)和混合型酸雨(SMAR, SO₄²⁻/NO₃⁻=2:1),每种酸雨配制3种酸度(pH=3.0、4.0和5.6)。这些模拟酸雨通过向去离子水添加适量的H₂SO₄和HNO₃的混合物进行制备。

表1 含磷物质添加量及处理后土壤总磷含量(n=3)

Table 1 The dosage of phosphorous compounds and total phosphorus concentration in treated soils(n=3)

处理 Treatment	含磷物质 Phosphorous compounds	分子式 Molecular formula	分子量 Molar mass/(g·mol ⁻¹)	每杯添加量 Addition amount each pot/g	土壤的总磷含量 Total phosphorus concentration in soils/(g·kg ⁻¹)
MCP	磷酸二氢钙	Ca(H ₂ PO ₄) ₂ ·H ₂ O	252.04	5.04	13.06±0.08
MAP	磷酸二氢铵	NH ₄ H ₂ PO ₄	115.03	4.60	13.76±0.05
MKP	磷酸二氢钾	KH ₂ PO ₄	136.09	5.44	13.42±0.20
CK	未添加	—	—	—	2.24±0.02

将2 g培养后土壤置入50 mL离心管中,加入10 mL模拟酸雨(SAR)浸泡24 h,再以4 000 r·min⁻¹的速率离心5 min,收集上清液;重复上述过程共3次,总共加入30 mL的SAR,来模拟酸雨的短期影响。将3次浸提的上清液混合,并用0.45 μm聚丙烯滤膜过滤。向离心管中加入10 mL去离子水,振荡混匀,离心5 min(4 000 r·min⁻¹),排出上清液,重复上述过程共3次,取出土壤样品留作下一步试验备用。

1.4 磷含量测定

分别采用HClO₄-H₂SO₄消化法和NH₄F-HCl法测定土壤总磷和有效磷含量^[35]。采用钼锑抗分光光度法测定SAR浸出液中水溶性磷(TDP)^[36]。按照不低于总样本数量的10%设置测定平行样,经分析测定平行样的相对标准偏差在4.2%~10.7%。

1.5 土壤磷浸出率的计算

本研究采用土壤磷浸出率评价不同含磷物质钝化修复的镉污染农田土壤的磷流失风险,具体计算公式如下:

$$\text{磷浸出率}(\%) = \frac{C_L \times V_L}{C_s \times m_s} \times 100\%$$

式中: C_L 为SAR浸提液中TDP的含量,mg·L⁻¹; V_L 为SAR浸提液的体积,L; C_s 为未经SAR处理土壤的总磷含量,mg·kg⁻¹; m_s 为模拟酸雨试验中每个处理所添加土壤的质量,kg。

1.6 统计分析

本研究所有处理均进行了3次重复,并计算了各处理的平均值和标准误差(SE)。使用SPSS 20.0进行单因素方差分析(ANOVA),并使用最小显著性差异法(LSD)进行多重比较。

2 结果与讨论

2.1 酸雨对土壤磷流失的影响

各处理的土壤磷流失量见图1。在相同SAR条件下,MCP处理的磷流失量显著低于MAP和MKP处理,且土壤的磷流失量呈现MKP>MAP>MCP的规律,这可能与MCP是一种微溶性的无机磷酸盐,其释放动力学过程比MAP和MKP等传统的水溶性磷肥慢有关。

如表2所示,添加MAP和MKP会使土壤磷浸出率显著增加,它们的磷浸出率分别为32.63%~38.57%和41.48%~49.29%,说明水溶性含磷物质的添加可加剧土壤磷流失,这与先前许多研究一致,即添加含磷物质可导致土壤中磷的累积,这些磷在酸雨作用下大量释放会增加富营养化的风险^[37-39]。此外,水溶性磷酸盐的快速溶解也是导致土壤磷浸出率增加的原因之一^[34]。然而,MCP处理与CK的磷浸出率无显著差异,这可能与MCP较低的溶解度有关。此外,MCP处理的磷浸出率显著低于MAP和MKP处理,表明在酸雨条件下,MCP钝化修复土壤的磷流失风险显著低于MAP和MKP钝化修复土壤。

如图2所示,在含磷物质与酸雨类型均相同的条件下,土壤磷的浸出率随酸雨pH值的降低而降低,且在相同pH条件下,SSAR处理的磷浸出率比SMAR处理高。该结果说明,酸雨的酸度和阴离子组成是影响含磷物质钝化修复的镉污染农田土壤磷浸出率的两个重要因素,且土壤的磷浸出率随酸雨的酸度和SO₄²⁻/NO₃⁻的增加而增加。许中坚等^[30]通过室内模拟酸雨淋溶试验研究了酸雨对土壤磷素释放的影响,结果表

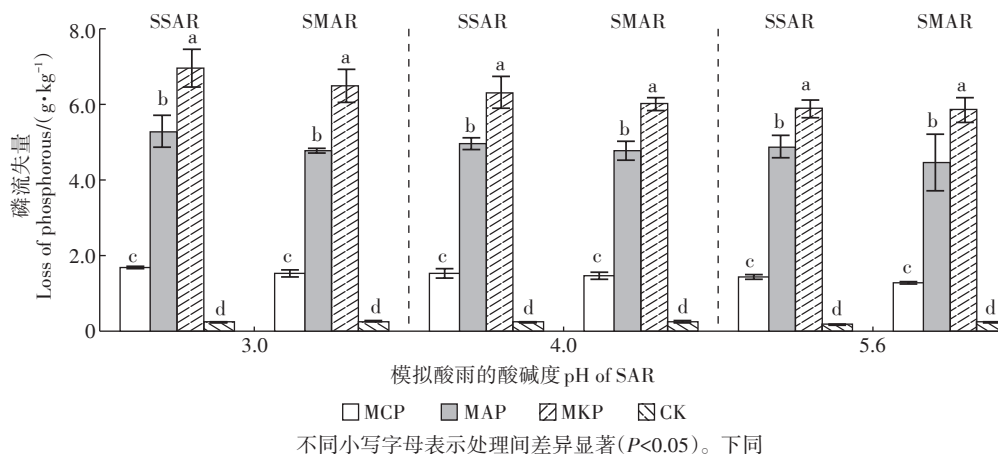


图1 不同含磷物质处理的土壤磷流失量

Figure 1 Loss of phosphorous in soils amended with different phosphorous compounds

明,酸雨酸度的升高使土壤磷的释放量增加,这可能是土壤磷浸出率随酸雨酸度的增加而增加的原因。有研究表明,NO₃会导致土壤中铁化合物的氧化,并促进氧化铁化合物[如Fe(Ⅲ)氧化物/氢氧化物]的形

成,从而增加了土壤对活性磷的吸附^[40-41]。此外,SMAR中的NO₃可以促进反硝化过程的发生,同时消耗H⁺^[44]。上述可部分解释在其他条件相同的情况下,SMAR处理土壤磷的浸出率较SSAR处理低这一现象。

2.2 酸雨对土壤有效磷的影响

各处理的土壤有效磷含量见表3和图3。如图3所示,在SAR处理前,3种含磷物质的添加均可增加土壤有效磷的含量,但SAR处理后各组土壤的有效磷含量均显著降低。该结果可能与土壤中可溶性磷以及部分解吸附磷的淋失有关,因为这两种磷是土壤有效磷的主要来源^[42-43]。此外,也有研究表明,酸雨可使土壤吸附的活性铝被大量释放,从而增加了土壤对磷的活性吸附点位,增强了土壤对磷酸根离子的配位吸附,进而导致了土壤有效磷含量的降低^[44]。

由图3可知,在SAR处理前,各处理的有效磷含量呈现MKP>MAP>MCP的规律,但在SAR处理后,各处理的有效磷含量则呈现MKP<MAP<MCP的规律。该结果表明,与MAP和MCP处理相比,MCP处理可更好地抵抗酸雨条件下土壤有效磷的降低,这可能是由

表2 不同酸雨类型对3种含磷物质钝化处理土壤磷的浸出率的影响(%)

Table 2 Effects of different types of SAR on the leachability of phosphorus in the soils amended with the three different phosphorous compounds(%)

处理 Treatment	SAR的类型 Type of SAR	pH=3.0	pH=4.0	pH=5.6
MKP	SSAR	49.29±3.78a	45.03±1.31a	44.02±1.83a
	SMAR	48.55±3.25a	44.74±3.21a	41.48±2.41a
MAP	SSAR	38.57±3.06b	36.15±1.15b	35.64±2.20b
	SMAR	34.80±0.47b	34.76±1.82b	32.63±5.44b
MCP	SSAR	13.09±0.26c	11.82±0.94c	11.10±0.46c
	SMAR	11.84±0.65c	10.53±0.75c	9.24±0.20c
CK	SSAR	12.19±0.74c	12.11±1.30c	11.74±1.67c
	SMAR	11.74±0.56c	11.56±0.74c	8.96±0.37c

注:同列不同小写字母表示处理间差异显著(P<0.05)。下同。
Note: Different lowercase letters in a column indicate significant differences among treatments at P<0.05. The same below.

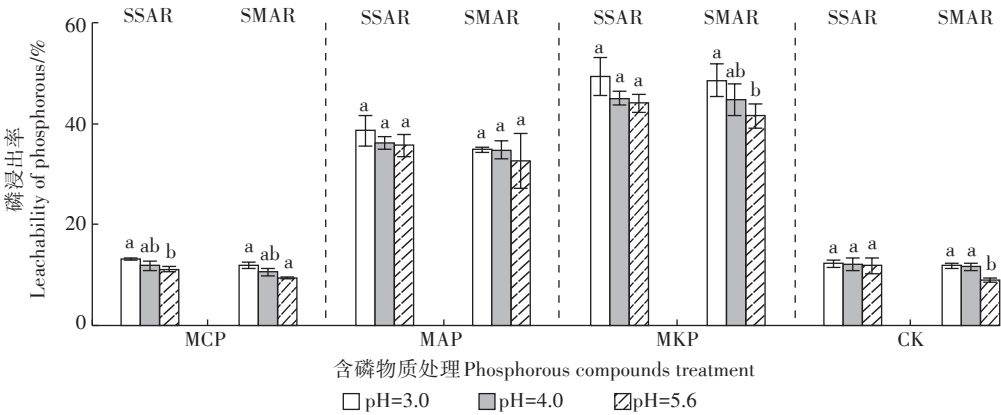


图2 不同pH值的模拟酸雨处理的土壤磷浸出率

Figure 2 Phosphorus leachability in soils treated with SAR with different pH

表3 不同模拟酸雨类型和pH对土壤有效磷含量的影响(mg·kg⁻¹)

Table 3 Effects of different types and pH of SAR on the available phosphorus concentration of soils(mg·kg⁻¹)

SAR类型Type of SAR	SAR的酸碱度pH of SAR	MCP	MAP	MKP	CK
SSAR	3.0	4.83±0.15c	4.69±0.16c	3.85±0.34c	0.16±0.01d
	4.0	5.20±0.24c	4.50±0.13cd	3.77±0.18c	0.22±0.01c
	5.6	4.83±0.23c	4.04±0.14d	3.72±0.13c	0.23±0.01c
SMAR	3.0	6.94±0.57b	6.14±0.28b	5.39±0.29b	0.31±0.02b
	4.0	7.23±0.59ab	6.01±0.48b	4.82±0.41b	0.32±0.02b
	5.6	7.37±0.61ab	6.07±0.10b	4.75±0.29b	0.32±0.03b
未经处理土壤		7.98±0.35a	9.36±0.27a	10.71±0.82a	0.41±0.01a

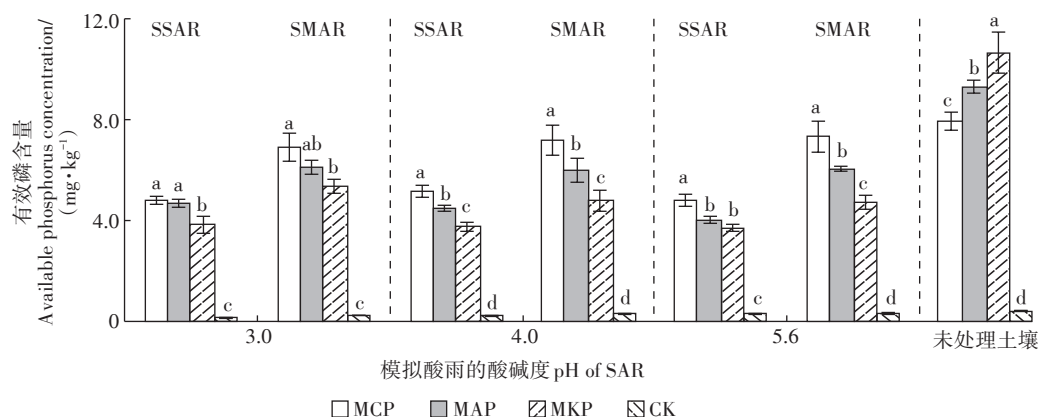


图3 不同含磷物质处理的土壤有效磷含量

Figure 3 Available phosphorus concentration in soils treated with phosphorus compounds

于MCP处理的磷浸出率显著低于MAP和MKP处理。此外,在其他条件均相同时,SSAR处理的有效磷含量低于SMAR处理,即土壤有效磷含量随酸雨 $\text{SO}_4^{2-}/\text{NO}_3^-$ 的增加而降低,这说明酸雨的阴离子组成是影响含磷物质钝化修复的镉污染农田土壤有效磷含量的重要因素。该结果可能是由于SMAR中较多的 NO_3^- 促进了土壤中的反硝化作用,同时反硝化作用消耗了部分 H^+ ,削弱了铁、铝、锰对土壤中活性磷的固定作用,从而减缓了土壤有效磷的降低^[14,45-46]。

3 结论

(1)酸雨浸泡导致含磷物质钝化修复的镉污染农田土壤磷浸出率显著增加,土壤有效磷含量显著降低;含磷物质钝化修复的镉污染农田土壤磷浸出率随着酸雨酸度和 $\text{SO}_4^{2-}/\text{NO}_3^-$ 的增加而增加,其有效磷含量随酸雨 $\text{SO}_4^{2-}/\text{NO}_3^-$ 的增加而降低。

(2)在同种酸雨浸泡条件下,磷酸二氢钙处理土壤的磷浸出率显著低于磷酸二氢铵和磷酸二氢钾处理,其有效磷含量高于磷酸二氢铵和磷酸二氢钾处理。因此,从土壤磷流失与磷有效性的角度考虑,磷酸二氢钙相比于磷酸二氢铵和磷酸二氢钾更适合于酸雨区镉污染农田土壤的钝化修复。

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