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### 引用本文:

赵迪, 张理胜, 罗元, 等. 养殖肥液灌溉土壤磷淋失阻控: 镧改性生物炭[J]. 农业环境科学学报, 2021, 40(11): 2574–2581.

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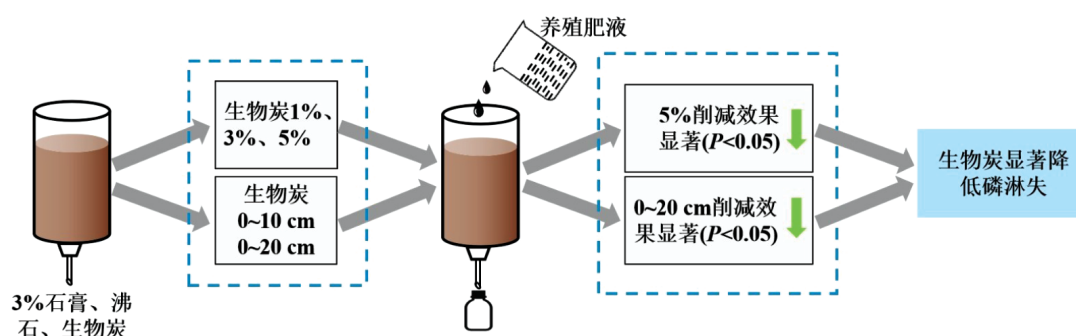


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# 养殖肥液灌溉土壤磷淋失阻控: 镧改性生物炭

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**摘要:**为探究镧改性生物炭伴施养殖肥液灌溉条件下土壤磷素的淋溶特征和阻控效果,采用土柱模拟试验对比镧改性生物炭和石膏、沸石传统阻控剂对磷淋溶的阻控特征,及镧改性生物炭不同添加量(1%、3%和5%)和不同施用方式(0~20 cm和0~10 cm混合)的影响效果。结果表明:磷淋失阻控效果呈现镧改性生物炭>石膏>沸石,且镧改性生物炭处理阻控效果最好( $P<0.05$ );镧改性生物炭施加处理总磷淋失量比对照处理削减了16.3%~58.3%,随镧改性生物炭施加量增加削减效果显著升高,这主要是因为生物炭对磷酸盐和有机磷发生了吸附;镧改性生物炭0~20 cm和0~10 cm施用方式总磷淋失量分别削减23.4%~58.3%和16.3%~45.0%,全土层混合削减效果更显著( $P<0.05$ );镧改性生物炭对正磷酸盐和有机磷的吸附机制主要是沉淀作用与络合作用。研究表明,在有机无机复合磷素输入条件下,镧改性生物炭的应用能够明显阻控磷淋失。

**关键词:** 镧改性生物炭; 阻控; 养殖肥液; 磷素; 淋失

中图分类号: X713; S141 文献标志码: A 文章编号: 1672-2043(2021)11-2574-08 doi:10.11654/jaes.2021-1027

## Prevention and control of phosphorus leaching in soil irrigated with biogas slurry by lanthanum modified biochar

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**Abstract:** To explore the leaching characteristics and control effects of phosphorus in soil under irrigation conditions of lanthanum-modified biochar combined with biogas slurry, the soil column simulation test was used to compare the resistance and control

收稿日期: 2021-09-06 录用日期: 2021-09-08

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基金项目: 国家重点研发计划项目(2017YFD0800403); 中央级公益性科研院所基本科研业务费专项(Y2021XC18); 云南省重大科技专项计划项目(202102AE090011)

Project supported: The National Key Research and Development Program of China(2017YFD0800403); The Central Public-interest Scientific Institution Basal Research Fund(Y2021XC18); The Major Science and Technology Special Plan of Yunnan Province(202102AE090011)

characteristics of lanthanum-modified biochar, gypsum, and zeolite traditional inhibitors on phosphorus leaching, and the effects of the addition amount (1%, 3%, and 5%) and application method (0~20 cm and 0~10 cm mixture) of lanthanum-modified biochar. The results showed that the control effect of phosphorus leaching loss was lanthanum-modified biochar > gypsum > zeolite, and the control effect of lanthanum-modified biochar was the best ( $P<0.05$ ). Compared with the control treatment, the total phosphorus leaching loss of lanthanum-modified biochar was reduced by 16.3%~58.3%, and the reduction effect was more evident with an increase in the amount of adsorbed phosphate and organophosphorus. The amount of leached total phosphorus was reduced by 23.4%~58.3% and 16.3%~45.0%, respectively, and the effect of mixed reduction of the whole soil layer was more significant ( $P<0.05$ ). The adsorption mechanism of lanthanum-modified biochar for orthophosphate and organophosphorus was primarily precipitation and complexation. Results indicate that under the organic-inorganic composite phosphorus input, the application of lanthanum-modified biochar can prevent and control phosphorus leaching loss.

**Keywords:** lanthanum modified biochar; inhibiting; biogas slurry; phosphorus; leaching loss

我国畜禽粪污年产量约为38亿t,但农田利用率不足60%<sup>[1]</sup>。将畜禽粪污经厌氧发酵后产生的养殖肥液应用于农田是资源化利用的重要途径。养殖肥液是一种富含丰富磷元素且可被二次利用的肥料<sup>[2]</sup>,比普通有机肥中磷含量高17.5%<sup>[3]</sup>,将其应用于农田不仅可以提高土壤养分供给能力,还可以降低由于直接排放造成的环境污染<sup>[4]</sup>。然而,养殖肥液属于高浓度有机废水<sup>[5]</sup>,长期大量施用造成了土壤磷素饱和程度增加,磷淋失风险加大,农业面源污染问题突出<sup>[6-9]</sup>,严重威胁水体安全。因此,采取有效措施降低养殖肥液还田过程中磷素淋失已成为亟待解决的重要问题。

目前,土壤磷淋失阻控多采用石膏、白云石、碳酸钙、沸石、硫酸铝、粉煤灰、聚丙烯酰胺等传统阻控剂<sup>[10]</sup>。然而,长期大量施用可能会对土壤质量产生负面影响<sup>[11]</sup>。一些研究表明生物炭在降低磷淋失的同时对土壤质量影响较小<sup>[12]</sup>。生物炭是在缺氧或无氧条件下热解产生的表面积大、稳定性强<sup>[13]</sup>、表面电荷密度高<sup>[14]</sup>、吸附性强、含碳丰富的多孔环境功能材料。大量报道指出普通生物炭对磷吸附能力较弱<sup>[15-16]</sup>,改性后吸附性能可明显提升<sup>[17-19]</sup>。而镧改性生物炭由于其具有高吸附量及受pH影响小等优点备受关注<sup>[20-21]</sup>,KOILRAJ等<sup>[22]</sup>报道镧改性蔗糖生物炭对磷酸盐吸附能力为 $13.3 \text{ mg} \cdot \text{g}^{-1}$ ;镧改性橡木生物炭和镧改性介孔稻壳生物炭吸附容量分别为 $46.37 \text{ mg} \cdot \text{g}^{-1}$ <sup>[23]</sup>和 $45.62 \text{ mg} \cdot \text{g}^{-1}$ <sup>[24]</sup>;罗元等<sup>[25]</sup>报道镧改性核桃壳生物炭对磷酸盐吸附能力为 $38.53 \text{ mg} \cdot \text{g}^{-1}$ 。

然而,国内外对镧改性生物炭的研究大多集中在水体磷酸盐,对其应用于土壤磷淋失阻控和养殖肥液这种有机无机复合体系磷吸附尚未有明确的研究结论,对土壤磷淋失阻控效果和机制需要进一步探究。本试验采用土柱淋溶模拟试验探究养殖肥液灌溉条件下,镧改性生物炭和石膏、沸石传统阻控剂对磷淋

溶特征的影响,并优化镧改性生物炭施加量和施用方式,为控制农田土壤磷素淋失和养殖肥液安全利用提供科学依据。

## 1 材料与方法

### 1.1 试验装置

土柱模拟淋溶装置如图1所示。装置材料为PVC,内径为10 cm,高度为25 cm;土壤经风干后与添加物混合,按照 $1.3 \text{ g} \cdot \text{cm}^{-3}$ 的容重模拟填装,分两层填装,保证土壤上下容重一致<sup>[26]</sup>。填装前在管壁内侧均匀涂抹一层凡士林以减小边缘效应。土柱装填完成后,各处理加蒸馏水600 mL使土壤水分接近饱和,室温下稳定2 d后开展淋溶试验。

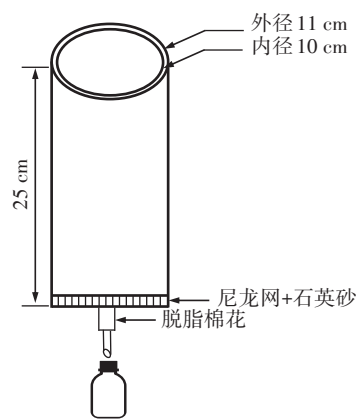


图1 淋溶装置图

Figure 1 Leaching simulator

### 1.2 试验材料

试验土壤采自天津市宁河区设施蔬菜大棚0~20 cm耕作层,土壤类型为壤土,将采集的土壤经自然风干后过2 mm筛备用。土壤pH值为8.23,有机质含量 $8.6 \text{ g} \cdot \text{kg}^{-1}$ ,全氮含量 $0.81 \text{ g} \cdot \text{kg}^{-1}$ ,速效磷含量 $1.35 \text{ mg} \cdot$

$\text{kg}^{-1}$ 。养殖肥液取自天津市西青区益利来养殖有限公司常年稳定运转的塞流式厌氧反应器, pH值为7.92, 总磷浓度  $75.17 \text{ mg} \cdot \text{L}^{-1}$ , 磷酸根浓度  $66.37 \text{ mg} \cdot \text{L}^{-1}$ 。石膏和沸石分别购自河北盛益矿产品有限公司和河南省巩义市恒鑫滤料厂, 纯度分别为96%和65%; 供试生物炭由核桃壳经镧改性制备, 热解温度为  $400^\circ\text{C}$ , 时间为2 h, 镧投加比率为1:5(核桃壳粉与  $\text{LaCl}_3$  溶液比值,  $m:V$ ), 过60目筛备用<sup>[25]</sup>。

### 1.3 试验设计与样品采集

试验设9个处理: 处理1, 养殖肥液(CK); 处理2, 养殖肥液+3%石膏(0~20 cm混合, 3%QC); 处理3, 养殖肥液+3%沸石(0~20 cm混合, 3%QF); 处理4, 养殖肥液+1%生物炭(0~20 cm混合, 1%QB); 处理5, 养殖肥液+3%生物炭(0~20 cm混合, 3%QB); 处理6, 养殖肥液+5%生物炭(0~20 cm混合, 5%QB); 处理7, 养殖肥液+1%生物炭(0~10 cm混合, 1%SB); 处理8, 养殖肥液+3%生物炭(0~10 cm混合, 3%SB); 处理9, 养殖肥液+5%生物炭(0~10 cm混合, 5%SB)。每个处理设置3个重复。其中, 石膏、沸石和生物炭施加量以土壤干质量为基数, 施磷总量设置为  $140 \text{ kg} \cdot \text{hm}^{-2}$ , 折算成养殖肥液共1400 mL, 每隔3 d灌施200 mL养殖肥液, 共连续试验7次。收集每次淋溶液并测定其体积、pH值、总磷浓度和正磷酸盐浓度。淋溶试验结束后, 陈化一周, 按照0~10 cm和10~20 cm分层采集土壤样品, 测定土壤速效磷含量。

### 1.4 测定方法与数据分析

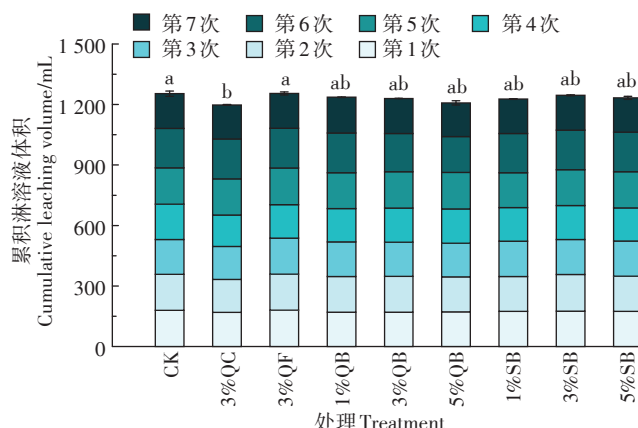
土壤及淋溶液pH采用pH计测定(水土比为2.5:1), 有机质采用重铬酸钾氧化-比色法测定, 土壤速效磷采用  $0.5 \text{ mol} \cdot \text{L}^{-1} \text{ NaHCO}_3$  浸提(液土比为20:1)<sup>[27]</sup>, 全自动流动注射分析仪(FIA-6000+)测定。总磷和正磷酸盐浓度采用全自动流动注射分析仪(FIA-6000+)测定。累积淋溶量为7次淋溶量之和。

试验数据采用Excel 2010和SPSS 22.0软件进行统计分析, 处理间差异采用单因素方差分析法(One-way ANOVA), 显著性水平为0.05。使用Pearson系数进行相关性分析( $P < 0.05$ ), 采用Origin 8.0软件进行绘图。

## 2 结果与分析

### 2.1 淋溶液体积和pH变化特征

阻控剂添加土壤淋溶液体积变化特征如图2所示。CK处理累积淋溶液体积为1258 mL, 占添加养殖肥液的89.86%。沸石和生物炭处理累积淋溶液体



不同小写字母表示处理间差异显著( $P < 0.05$ )。下同  
Different lowercase letters indicate significant differences among treatments( $P < 0.05$ ). The same below

图2 阻控剂添加后土壤淋溶液累积体积

Figure 2 Cumulative volumes in leaching water

积略低于CK处理, 说明二者均能起到保持土壤水分的作用, 但处理间差异不显著。不同生物炭施用量及施用方式处理下的累积淋溶液体积差异均不显著, 但随生物炭用量增加略有降低, 且5%QB和5%SB处理累积淋溶液体积最小, 分别比CK减少2.9%和2.4%。这说明生物炭添加提高了土壤持水性能。

阻控剂添加土壤淋溶液pH值变化特征如图3所示。在7次灌溉过程中, 淋溶液pH出现一定的波动, 但变幅较小。淋溶结束后, 各处理淋溶液pH值均处于7.20~7.70, 与CK相比, 石膏和沸石处理淋溶液pH分别升高了0.50和0.44, 生物炭两种施用方式(QB和SB)处理下淋溶液pH升高了0.05~0.19和0.20~0.37。表明各种阻控剂可降低淋溶液中 $\text{H}^+$ 浓度, 具有提高淋溶液pH的效果。

### 2.2 淋溶液中总磷浓度和累积淋失量变化特征

阻控剂添加土壤淋溶液总磷浓度及累积淋失量如图4所示。随灌溉次数的增加, 各处理土壤淋溶液总磷浓度呈升高趋势且增幅逐渐变缓。第7次灌溉后淋溶液总磷浓度呈现  $\text{CK} > 3\% \text{QF} > 3\% \text{QC} > 1\% \text{SB} > 1\% \text{QB} > 3\% \text{SB} > 3\% \text{QB} > 5\% \text{SB} > 5\% \text{QB}$ , 生物炭效果显著优于传统阻控剂。每次灌溉淋溶液总磷浓度均是CK处理最高, 浓度为  $10.7 \sim 152.2 \mu\text{g} \cdot \text{L}^{-1}$ 。石膏和沸石处理下总磷浓度分别比CK处理降低了19.9%和14.1%, 处理间差异并不显著( $P > 0.05$ )。淋溶液中总磷浓度随生物炭用量增加而降低, 添加量为5%水平时能够显著削减磷淋失量, 总磷浓度为  $3.0 \sim 88.4 \mu\text{g} \cdot \text{L}^{-1}$ 。两种生物炭施用方式(QB和SB)下, 总磷浓度分别比CK处理降低了23.4%~58.3%和16.3%~45.0%,

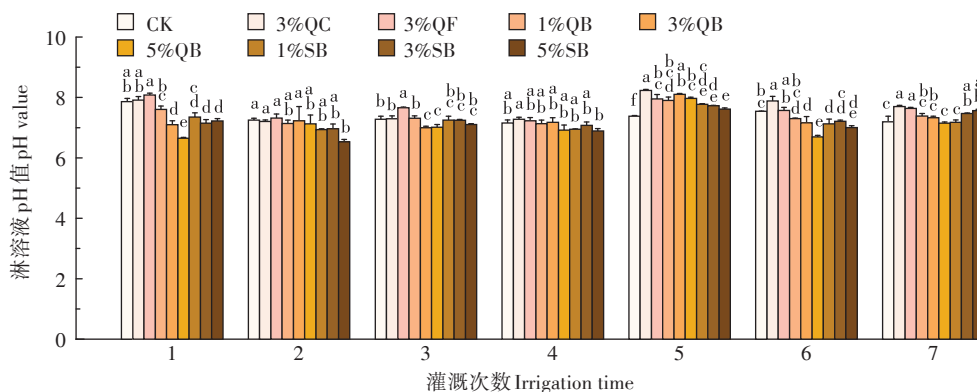


图3 阻控剂添加后淋溶液的pH值

Figure 3 pH values in leaching water of different treatments

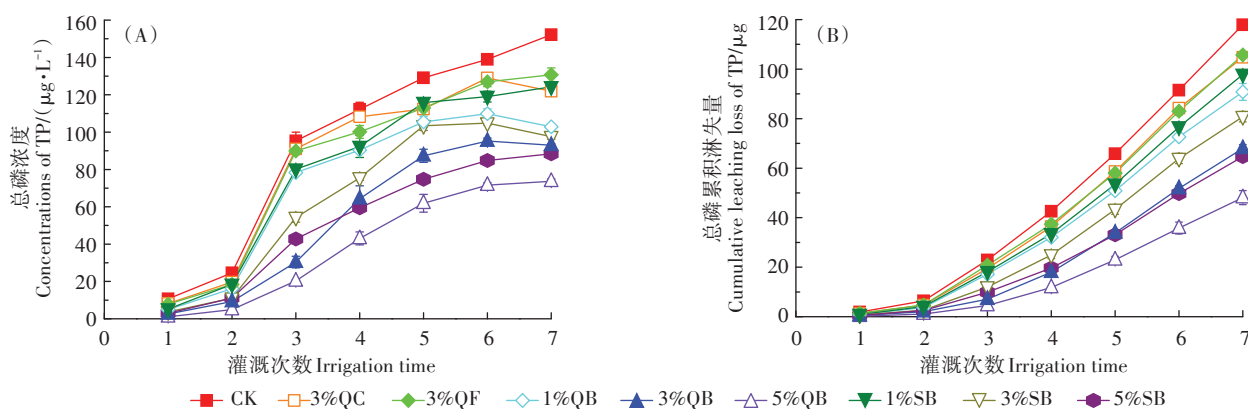


图4 阻控剂添加后总磷淋溶浓度与累积淋失量

Figure 4 Total phosphorus concentrations and cumulative leaching loss in different treatments

全土层混合比上层混合阻控效果更好。5%QB处理总磷累积淋失量比CK处理显著降低了59.7% ( $P < 0.05$ ), 全土层混合阻控效果优于上部土层混合。

### 2.3 淋溶液中正磷酸根浓度和累积淋失量变化特征

阻控剂添加土壤淋溶液正磷酸根浓度及淋失量如图5所示。随灌溉次数的增加,各处理土壤淋溶液正磷酸盐浓度呈现上升趋势。第7次灌溉后淋溶液正磷酸根削减呈现5%QB>5%SB>3%QB>3%SB>1%QB>1%SB>3%QF>3%QC>CK。其中,CK处理正磷酸根浓度最高,5%QB处理正磷酸根削减效果最好,浓度比CK处理显著降低了61.8% ( $P < 0.05$ )。石膏和沸石处理正磷酸盐浓度较CK处理分别降低了3.2%和13.6%。生物炭两种施用方式下(QB和SB)正磷酸根浓度分别比CK处理降低了15.9%~54.0%和18.7%~50.4%,且随生物炭用量增加而降低,全土层混合比上层混合对正磷酸根的阻控效果更好。各处理正磷酸根累积淋失量随灌溉次数增加而增加。由各处理淋溶液总磷及正磷酸根含量可判断出各处理淋溶液中均含有一定量的有机磷,镧改性生物炭对总

磷的阻控作用是通过对无机磷和有机磷共同吸附体现的。

### 2.4 磷淋失相关分析

淋溶液中总磷及正磷酸根相关分析如表1所示。淋溶液pH值与淋溶液总磷浓度和总磷淋失量呈显著正相关,与淋溶液正磷酸根浓度和正磷酸根淋失量呈极显著正相关。淋溶液总磷浓度、总磷淋失量分别与正磷酸根浓度、正磷酸根淋失量呈极显著正相关。表明总磷淋溶同正磷酸盐的淋溶是互相影响的过程。

### 2.5 土壤速效磷含量变化特征

阻控剂添加土壤速效磷含量如图6所示。各处理土壤速效磷含量均表现为0~10 cm高于10~20 cm土层。与CK相比,石膏和沸石处理分别使0~10 cm土层速效磷含量降低2.3%和1.0%,差异均不显著 ( $P > 0.05$ ),使10~20 cm土层速效磷含量分别升高18.5% ( $P < 0.05$ )和降低2.1% ( $P > 0.05$ )。两种生物炭施用方式(QB和SB)下的0~10 cm土层速效磷含量比CK处理分别显著降低了45.5%~88.8%和78.6%~89.3% ( $P < 0.05$ ),10~20 cm土层速效磷含量分别减少了

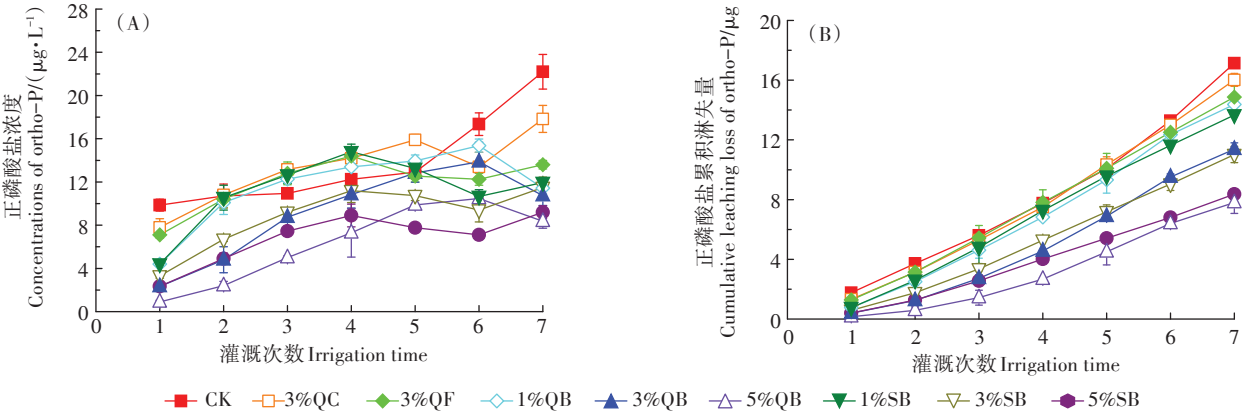


图5 不同处理下正磷酸根淋溶浓度与累积淋失量

Figure 5 Ortho-P concentrations and cumulative leaching loss in different treatments

表1 淋溶液中总磷及正磷酸根浓度及淋失量相关分析

Table 1 Correlation analysis of total phosphorus and ortho-P leaching with water parameters of leachate

| 指标 Index | 体积 Volume | pH 值 pH value | 总磷浓度 Total phosphorus concentration | 总磷淋失量 Total phosphorus leaching loss | 正磷酸根浓度 Ortho-P concentration | 正磷酸根淋失量 Ortho-P leaching loss |
|----------|-----------|---------------|-------------------------------------|--------------------------------------|------------------------------|-------------------------------|
| 体积       | 1         |               |                                     |                                      |                              |                               |
| pH 值     | 0.174     | 1             |                                     |                                      |                              |                               |
| 总磷浓度     | 0.415     | 0.739*        | 1                                   |                                      |                              |                               |
| 总磷淋失量    | 0.426     | 0.749*        | 1.000**                             | 1                                    |                              |                               |
| 正磷酸根浓度   | 0.307     | 0.801**       | 0.959**                             | 0.958**                              | 1                            |                               |
| 正磷酸根淋失量  | 0.356     | 0.798**       | 0.962**                             | 0.961**                              | 0.998**                      | 1                             |

注: \*在0.05水平上显著, \*\*在0.01水平上显著。淋溶液体积、pH值、总磷浓度、总磷淋失量、正磷酸盐浓度和正磷酸盐淋失量均为7次淋溶液的平均值。

Note: \*significant at 0.05 level, \*\*significant at 0.01 level. Values of leachate volume, pH, total phosphorus concentrations, the amount of total phosphorus, ortho-P concentrations, and the amount of ortho-P in leachate are the average of seven leaching tests.

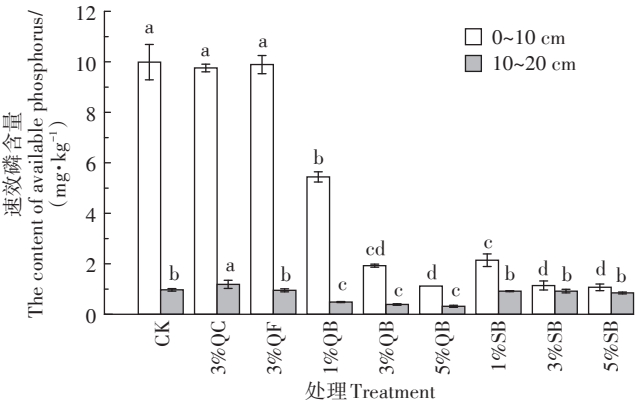


图6 不同处理土壤中速效磷含量

Figure 6 The content of available phosphorus in soil of different treatments

50.0%~67.4% ( $P<0.05$ )和5.1%~12.7%。生物炭相同施用方式3种水平下(1%、3%和5%)0~10 cm土层速效磷含量随生物炭添加量增加而降低,而10~20 cm土层速效磷含量均未达到显著差异( $P>0.05$ )。

3 讨论

3.1 传统阻控剂与镧改性生物炭对土壤磷素淋失的影响

镧改性生物炭对土壤总磷、正磷酸盐及有机磷淋失阻控效果显著优于石膏和沸石,主要是因为镧改性生物炭与石膏、沸石改性生物炭的抑制机理不同,发挥的作用也不同,使得阻控磷素淋失的效果更佳。石膏主要依靠其中的 $\text{Ca}^{2+}$ 把土壤中活性强的磷形态转化为稳定的磷形态<sup>[28]</sup>;沸石主要通过其吸附性能及增加土壤团聚体来控制磷的浸出<sup>[29-31]</sup>;而镧改性生物炭可通过多种机制阻控土壤磷素淋失:一是提高土壤有机碳含量,改善土壤持水能力,固持养分,减少磷素损失<sup>[32]</sup>;二是生物炭本身具有强烈的吸附固定作用<sup>[33]</sup>,经镧改性后对于阴离子的络合沉淀作用也会加强<sup>[34-35]</sup>;三是金属氢氧化物的羟基还可以被磷取代生成稳定的物质,有效降低磷的淋溶损失<sup>[23]</sup>。养殖肥液

中水溶性全磷主要以正磷酸盐形式存在,少量为有机磷<sup>[36]</sup>。本研究发现,淋溶液中正磷酸盐占总磷的比例相对较低,而有机磷占比较高。这主要是由于石膏、沸石和生物炭中的钙、铝和镧会分别与土壤或养殖肥液中的磷酸根离子形成 $\text{CaHPO}_4$ 、 $\text{AlPO}_4$ 和 $\text{LaPO}_4$ 等物质<sup>[37]</sup>,这些化合物在土壤中不易移动<sup>[38]</sup>,从而将磷酸盐保留在土壤中。有机磷因其移动性大而易流失<sup>[39]</sup>,这也是造成淋溶液中有机磷含量相对较高以及土壤磷淋失的重要因素。对于有机磷的淋失阻控,石膏、沸石中含有的钙和铝均可通过络合作用对有机磷进行固定,而镧改性生物炭固定有机磷的机制主要包括:一是生物炭中含有的镧与有机磷进行络合<sup>[40]</sup>;二是质子化带正电的基团可通过静电吸引作用固定有机磷<sup>[41]</sup>;三是生物炭上—OH、C=C、COO—等有机官能团可通过相似相溶的原理促进其对有机磷的固定作用<sup>[42]</sup>;四是生物炭上丰富的表面活性吸附位点为有机磷的吸附提供了一定的位点<sup>[43]</sup>。

### 3.2 生物炭施用方式与添加量对土壤磷素淋失的影响

相比生物炭上层混合方式,全土层混合的方式总磷累积淋失量降低了7.2%~25.7%,原因主要是施用深度的变化导致削减效果不同。生物炭全土层混合增加了其吸附的路径和时间,为磷素在生物炭上的吸附提供了更多的机会,赢得了扩散需要的时间<sup>[44]</sup>,从而最大限度地发挥了生物炭对磷素淋溶损失的阻控作用。此外,生物炭的添加量也会影响其对土壤中磷淋失的阻控效果。试验发现不同添加水平生物炭(1%、3%和5%)对淋溶液中总磷浓度削减显著,削减率变化范围在16.3%~58.3%,随生物炭添加量的增加,淋溶液中总磷的浓度及累积淋失量降低,均与生物炭添加量呈负相关关系,众多研究也得出了类似结论<sup>[45-46]</sup>。这说明生物炭添加量越多,其能够提供更多的表面活性吸附位点及金属(氧化物)越多,从而加强了对磷的吸附固定作用。

## 4 结论

(1)不同添加剂对土壤总磷和磷酸根淋溶阻控效果呈现镧改性生物炭>石膏>沸石,镧改性生物炭处理阻控作用显著高于传统阻控剂,特别是在有机无机复合磷素输入条件下。

(2)镧改性生物炭对总磷淋失的削减率达16.3%~58.3%,随镧改性生物炭添加量的增加而升高,镧改性生物炭对磷酸根和有机磷均有吸附效果。

(3)镧改性生物炭全土层混合处理效果优于上部

土层混合方式,总磷淋失削减率分别为23.4%~58.3%和16.3%~45.0%。

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