



蚯蚓粘液-秸秆炭共同作用对生活污泥堆肥中重金属的影响

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蚯蚓粘液-秸秆炭共同作用对生活污泥堆肥中重金属的影响^{*}

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摘 要: 生活污泥中富含的重金属限制其资源化利用, 为钝化重金属活性, 降低污泥毒害效果和提高其利用价值, 以 40 mL 蚯蚓粘液和 2%、4%、6%、8% 秸秆炭为添加剂对 2 kg 污泥进行堆肥, 研究粘液、粘液协同秸秆炭添加对污泥堆肥后重金属变化的影响。结果显示, 与对照组污泥堆肥相比, 粘液堆肥污泥后 pH 升高 1.42%, 总氮、总磷含量降低 7.87%、14.18% ($P<0.05$); 而粘液协同秸秆炭堆肥污泥后, 污泥逐渐呈碱性, 电导率提升 5.71%–9.58% ($P<0.05$), 有机质含量升高 7.71%–24.60% ($P<0.05$), 丰富了堆体中可溶性离子和有机物含量, 但总氮、总钾含量分别降低 19.10%–30.95%、7.87%–14.31%。在添加粘液对污泥堆肥后, 重金属总量均表现出下降趋势, Ni、Zn、Pb 的较活液形态向难以降解的残渣态转化, 使残渣态所占比例较 CK 处理分别升高 61.81%、120.19%、72.51%; 当添加粘液和秸秆炭对污泥堆肥后, 重金属总量继续表现出下降趋势, 碳酸盐结合态 Ni 和 Pb、铁锰结合态 Pb、可交换态 Zn 逐步向稳定的残渣态转化, 而有机结合态 Cu 却向可交换态和残渣态转化, 钝化了堆肥污泥中 Ni、Zn、Pb, 活化了 Cu。最后根据分析得出结论, 粘液协同秸秆炭改变污泥中 pH 来影响重金属 Ni、Zn、Pb、Cu 有效态, 粘液+8% 秸秆炭处理对污泥重金属的影响较为理想。

关键词: 蚯蚓粘液; 秸秆炭; 生活污泥堆肥; 重金属含量; 重金属形态

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Effects of earthworm mucus and straw charcoal on heavy metals during domestic sludge co-composting*

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Abstract: Heavy metals restrict the reuse of municipal sludge. To passivate the activity of heavy metals, reduce sludge toxicity, and create new value, 2 kg of sludge was composted with 40 mL earthworm mucus and 2%, 4%, 6%, and 8% straw charcoal, to investigate changes in the heavy metal mobility in sewage sludge. The results showed that, compared with the control sludge compost (CK), the pH increased by 1.42% ($P<0.05$) and the total nitrogen and total phosphorus decreased by 7.87% and 14.18%, respectively ($P<0.05$), after the addition of the mucus to the sludge. After adding both the mucus and straw charcoal to the sludge compost, the sludge gradually became alkaline; furthermore, its electrical conductivity value increased by 5.71%–9.58% ($P<0.05$), and organic matter content increased by 7.71%–24.60% ($P<0.05$). Although this enriched the content of soluble ions and available organic matter in the compost, the total nitrogen and potassium contents decreased by 19.10%–30.95% and 7.87%–14.31%, respectively, resulting in the loss of plant nutrients. By adding mucus to the sludge compost, different total heavy metal contents showed different declining trends; these included Cd, Cu, Ni, Zn, and Pb, which decreased by 3.59%, 7.03%, 10.93%, 8.39%, and 5.11% ($P<0.05$, except Ni), compared to the CK treatment group. The more active forms of Ni, Zn, and Pb were transformed into an unavailable residue form that was difficult to degrade; therefore, the proportion of residual forms increased by 61.81%, 120.19%, and 72.51%, respectively, compared with the CK treatment. When the mucus and different proportions of straw charcoal were added to the sludge, the total heavy metal contents decreased further. The total amount of Cd, Pb, Cu, Ni, and Zn decreased by 37.18%, 67.36%, 6.07%, 59.59%, and 31.82%, respectively, in the mucus plus 8% straw charcoal treatment group ($P<0.05$). The Ni and Pb associated with the carbonate, Pb associated with iron-manganese, and exchangeable Zn were gradually shifted to the residue form, so that the available contents of Ni, Pb, and Zn were significantly decreased by 28.08%, 42.00%, and 28.31%, respectively, in the mucus plus 8% straw charcoal treatment group, which passivated Ni, Zn, and Pb in the composted sludge. In contrast, organic form of Cu was converted into exchangeable and residual forms. Its available content was increased by 89.82% ($P<0.05$) in the mucus plus 8% straw charcoal treatment group, and Cu was activated in the sludge during composting. In the analysis of the effect of mucus and different ratios of straw charcoal on the availability of heavy metals after composting, it was found that the correlation coefficients of straw charcoal addition with the available forms of heavy metals Cu, Zn, Pb, and Ni reached significant levels of 0.906, -0.909, -0.847, and -0.639 ($P<0.05$), respectively, while the correlation coefficients with Cd were lower. Finally, based on the principal component analysis and stepwise regression equations, mucus in combination with straw charcoal influenced the pH of the sludge compost, affecting the mobility of Ni, Zn, Pb, and Cu. Therefore, mucus plus 8% straw charcoal is an effective approach for treating the heavy metals in the sludge.

Keywords: Earthworm mucus; Straw charcoal; Municipal sludge compost; Heavy metal content; Heavy metal speciation

近些年,随着更多污水处理厂的投入运用,污泥产生量也快速提高,根据我国住房和城乡建设部公布的《2020年城乡建设统计年鉴》^[1],截至2020年年底干污泥产量已达1333万t,较高的产量会使污泥大量暴露于露天环境中,产生较多挥发性有机化合物、温室气体、具有异味的气体(H_2S 、 NH_3)和渗滤液等危害环境的有毒有害污染物^[2]。而生活污水中存在重金属的现象较为普遍,如若无法妥善解决,也会破坏环境并酿成二次污染^[3]。目前污泥的常规处置仍是以填埋和焚烧为主,但前者会占用大量土地

资源,后者不仅成本高,还会排放大量尾气,在未来很可能被堆肥及土地利用等具有良好发展的资源化技术所代替^[4]。污泥堆肥对降低污泥中重金属含量和削弱重金属毒性有重要作用。研究发现,在污泥中添加不同改良剂,可使重金属有效态和形态发生不同变化。如:堆肥中添加木屑和蘑菇渣能改变重金属浓度和存在形态,增强重金属稳定性^[5];添加蒙脱石可显著降低重金属Pb、Cu、Zn活性,钝化重金属有效性^[6],磷矿粉能钝化堆肥中Cu的活性^[7]。而生物炭因其比表面积大,富含多种能与离子进行配

位反应和交换反应的官能团,可降低重金属移动性^[8],如 Liu 等^[9]发现生物炭对污泥堆肥时,能滞留养分和钝化重金属 Cu、Zn、Pb、Ni 和 Cr; Malinowski 等^[10]采用木炭对污泥堆肥时发现重金属毒性显著降低,显著加快堆肥化进程。因此,生物炭常用作添加剂改良污泥中重金属,效果甚佳。

蚯蚓粘液是蚯蚓从体表排出的一种组分较为复杂的淡黄色粘状液体,主要由电解质、糖类化合物、脂质、氨基酸、微量营养素、有益微生物等混合物组成,在蚯蚓的生命活动中具有重要作用^[11-13]。研究发现,蚯蚓粘液具有启动和促进植物残体矿化、腐殖化的作用,是堆肥微生物生长发育的重要营养源^[14],能改变重金属在污染土壤中的存在状态^[15],也可提高番茄 (*Solanum lycopersicum*) 幼苗中叶绿素含量,促进其对微量金属元素的吸收^[16]。Sizmur 等^[15]还发现蚯蚓粘液中富含溶解性碳、溶解性氮和能产生两性离子的氨基酸,可以与重金属等污染物发生络合反应,从而改变重金属 As 的存在形态和降低其流动性。因此,粘液对重金属在环境中的行为变化起着重要作用。

生物炭和粘液均具有调控环境中重金属活性的作用^[9,15-16],前者广泛作为堆肥和污染土壤的改良剂,后者在土壤中有一定研究,而在堆肥中研究较为鲜见,对堆肥中重金属影响更为鲜见。鉴于此,本试验

研究蚯蚓粘液及其与生物炭协同作用,并通过设置粘液协同不同比例秸秆炭对污泥堆肥,探究粘液单独作用及两者共同作用后能否调节污泥中重金属活性,从而对重金属形态和有效态产生怎样影响,以为更好地处理污泥中重金属活性提出新方案。

1 材料和方法

1.1 试验材料

选取安徽省淮南市某生活污水处理厂新鲜污泥,该污泥通过污泥浓缩池和板框深浓度脱水工艺获得。秸秆炭选取在实验室切成段状后自然风干的玉米 (*Zea mays*) 秸秆,将其置于升温速度为 $20\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ 的马弗炉中 $500\text{ }^{\circ}\text{C}$ 热解 2 h,于干燥器中冷却后研磨,过 100 目筛,获得材料比表面积 $45.05\text{ m}^2\cdot\text{g}^{-1}$,平均孔径 5~5.5 nm。蚯蚓为江苏省某蚯蚓养殖基地的赤子爱胜蚓 (*Eisenia fetida*),蚯蚓粘液采用不损伤蚯蚓的低电压电流 (5 V、10 mA,电击后蚯蚓可正常存活) 刺激法获取^[17]:将蚯蚓于实验室驯化 5~10 d,筛选活性强、个体大且含有环带的蚯蚓 600 g (约 1200 条),用蒸馏水冲洗后放入排粪盒中排粪 24 h,取出后用超纯水冲洗蚯蚓 2~3 次,放置在粘液提取装置中^[14],通电 3 次,每次持续 1 min,电击间隙 1 min,共获取 50 mL 粘液,将其稀释 10 倍^[18],贮存于 $-20\text{ }^{\circ}\text{C}$ 冰箱中。污泥、粘液和秸秆炭的理化性质如表 1 所示。

表 1 供试材料的基本理化性质
Table 1 Basic physiochemical properties of the testes raw materials

性质 Property	污泥 Sludge	蚯蚓粘液 Earthworm mucus	秸秆炭 Straw charcoal
电导率 Electrical conductivity ($\text{ms}\cdot\text{cm}^{-1}$)	1.11±0.03	0.05±0.00	0.86±0.05
pH	6.70±0.02	6.89±0.20	8.87±0.09
含水率 Moisture content (%)	76.67±0.05	—	8.42±0.02
有机质 Organic matter (%)	16.65±0.89	0.37±0.03	17.61±0.63
总氮 Total nitrogen ($\text{g}\cdot\text{kg}^{-1}$)	27.82±0.82	0.15±0.01	0.63±0.05
总磷 Total phosphorus ($\text{g}\cdot\text{kg}^{-1}$)	8.88±0.32	0.007±0.00	2.82±0.10
总钾 Total potassium ($\text{g}\cdot\text{kg}^{-1}$)	13.24±1.55	0.023±0.00	15.90±0.70
总锌 Total Zn ($\text{mg}\cdot\text{kg}^{-1}$)	737.00±44.55	—	79.50±2.12
总铅 Total Pb ($\text{mg}\cdot\text{kg}^{-1}$)	14.73±2.09	—	—
总铜 Total Cu ($\text{mg}\cdot\text{kg}^{-1}$)	60.43±5.27	72.90±1.78	19.37±0.66
总镍 Total Ni ($\text{mg}\cdot\text{kg}^{-1}$)	32.40±5.06	—	2.45±0.07
总镉 Total Cd ($\text{mg}\cdot\text{kg}^{-1}$)	1.62±0.11	—	0.05±0.00

“—”为未检出。“—” means not detected.

1.2 试验方法

为研究粘液、粘液+秸秆炭对污泥堆肥后重金属含量的影响,设置污泥堆肥 (CK) 对照组,试验组 5 个:污泥+粘液堆肥 (S0),污泥+粘液+2%、4%、6% 和 8% 秸秆炭堆肥 (S1、S2、S3 和 S4,污泥干重比);所有堆肥组均取 2 kg 新鲜污泥,采用 Huang 等^[14]试验方案添加 40 mL 粘液,每组分别设置 3 个平行

样品,控制堆体周边环境温度在 $25\text{ }^{\circ}\text{C}$ 左右,含水率保持在 60%~70%,堆肥时间选择能让粘液充分利用并稳定堆体的 35 d^[14,19]。堆肥试验结束后,将堆肥后的污泥自然风干后进行研磨过 100 目筛,制得分析样品。

1.3 分析方法

取适量过筛污泥于纯水中磁力搅拌 1 h 后离心,

通过电位法测定 pH 和电导率(EC)^[19];总氮(TN)、总磷(TP)在灭菌锅中处理 30 min 后采用碱性过硫酸钾法测定^[20];有机质(OM)通过重铬酸钾于 150 ℃ 恒温箱氧化消解 30 min 后冷却测定^[21];重金属总量通过氢氟酸、硝酸和高氯酸联合消解法,有效态通过 DTPA-2Na 溶液浸提法,形态通过 Tessier 五步连续提取法,并将获取的重金属溶液使用 AAS 进行测定^[20]。试验均加入国家标准物质进行质量控制,回收率和相对标准偏差符合要求。

1.4 数据处理与分析

使用 Excel 2021 对数据进行整理和计算,用 SPSS 26 对数据进行单因素方差分析(ANOVA)、多重比较(LSD)、相关性和逐步回归方程分析(P 为 0.05 或 0.01),用 ORIGIN 2022 对数据进行主成分分析(PCA)和作图。

2 结果与分析

2.1 堆肥后污泥理化性质

蚯蚓粘液-秸秆炭共同作用对污泥堆肥后,在不同试验组间的理化性质和营养物质含量间表现出不

同差异(表 2)。与污泥堆肥对照组(CK)相比,单独添加粘液对污泥堆肥后(S0)的 pH、EC 均升高,OM、TN、和 TP 均降低,其中 pH 升高显著($P<0.05$),TN、TP 降低显著($P<0.05$)。当粘液-秸秆炭共同作用对污泥堆肥后(S1-S4),与 CK 相比,pH、EC 和 OM 在粘液+8% 秸秆炭(S4)时分别升高 9.97%、19.40% 和 24.60% ($P<0.05$),TN 和 TP 分别降低 30.95% 和 19.84% ($P<0.05$);粘液+4% 秸秆炭(S2)时,pH、EC 和 OM 分别升高 5.55%、12.68% 和 12.01% ($P<0.05$),而 TK 和 TP 与对照无显著差异。这表明添加粘液和秸秆炭,能促进有机质的矿化、腐质化和脱氨基作用,释放碱性物质,使污泥呈弱碱性;而生物炭残留较多的难降解碳,也会提升 OM 含量;堆体中氨气的释放和磷、钾的滤出,促使污泥中营养物质逐渐产生损失^[9,14,22-23]。由此,所有试验组中粘液联合 4% 秸秆炭(S2)处理的污泥效果较好,堆体呈弱碱性,OM 含量较丰富,养分损失少。

2.2 堆肥后污泥中重金属含量变化

由表 3 可知,采用粘液-秸秆炭共同作用对污泥

表 2 蚯蚓粘液-秸秆炭共同作用对污泥堆肥理化性质和营养物质的影响

Table 2 Effects of applying earthworm mucus and straw charcoal on the selected physical and chemical properties and nutrients of sludge compost

处理 Treatment	pH	电导率 Electrical conductivity (mS·cm ⁻¹)	有机质 Organic matter (%)	总氮 Total nitrogen	总磷 Total phosphorus (g·kg ⁻¹)	总钾 Total potassium
CK	6.92±0.04e	1.34±0.01d	15.65±0.50d	32.31±0.31a	21.37±0.31a	15.14±0.16a
S0	7.02±0.06d	1.38±0.03d	15.45±0.15d	29.77±0.70b	18.34±0.12b	15.31±0.32a
S1	7.25±0.02c	1.46±0.02c	16.85±0.30c	25.56±0.32c	23.05±1.80a	13.95±0.96a
S2	7.30±0.04c	1.51±0.02bc	17.53±0.28b	26.14±0.26c	22.81±0.50a	12.98±2.53a
S3	7.47±0.08b	1.54±0.08ab	17.22±0.27bc	21.64±0.65d	17.57±1.90b	13.66±1.40a
S4	7.61±0.05a	1.60±0.01a	19.50±0.26a	22.31±0.60d	17.13±1.15b	13.32±1.65a

CK: 单独污泥堆肥; S0: 污泥+粘液堆肥; S1: 污泥+粘液+2% 秸秆炭堆肥; S2: 污泥+粘液+4% 秸秆炭堆肥; S3: 污泥+粘液+6% 秸秆炭堆肥; S4: 污泥+粘液+8% 秸秆炭堆肥; 同列不同字母表示不同处理间差异显著($P<0.05$)。CK: sludge composting; S0: sludge + mucus composting; S1: sludge + mucus + 2% straw charcoal composting; S2: sludge + mucus + 4% straw charcoal composting; S3: sludge + mucus + 6% straw charcoal composting; S4: sludge + mucus + 8% straw charcoal composting. Different letters in the same column show significant differences among different treatments ($P<0.05$).

表 3 蚯蚓粘液-秸秆炭共同作用的污泥堆肥后重金属含量

Table 3 Effect of applying earthworm mucus and straw charcoal on heavy metal contents of sludge compost mg·kg⁻¹

处理 Treatment	镉 Cd	铅 Pb	铜 Cu	镍 Ni	锌 Zn
CK	1.95±0.02a	18.00±0.05a	65.85±3.90a	82.78±4.15a	858.00±39.09a
S0	1.88±0.08a	17.08±4.25a	61.22±4.79ab	73.73±6.38b	786.00±0.30a
S1	1.65±0.05b	11.78±1.48b	59.67±0.98ab	44.40±1.98c	783.00±7.50a
S2	1.53±0.08b	8.83±1.45bc	61.68±1.78ab	41.91±0.44cd	676.50±18.00b
S3	1.15±0.15c	6.37±1.70c	61.75±0.43ab	35.05±1.33de	645.00±7.50bc
S4	1.23±0.23c	5.88±1.48c	61.85±1.95ab	33.45±5.96e	585.00±29.25c
GB 4284—2018 (A)	<3	<300	<500	<100	<1200
欧盟 European Union	20~40	750~1200	1000~1750	300~400	2500~4000

CK: 单独污泥堆肥; S0: 污泥+粘液堆肥; S1: 污泥+粘液+2% 秸秆炭堆肥; S2: 污泥+粘液+4% 秸秆炭堆肥; S3: 污泥+粘液+6% 秸秆炭堆肥; S4: 污泥+粘液+8% 秸秆炭堆肥; 同列不同字母表示不同处理间差异显著($P<0.05$)。CK: sludge composting; S0: sludge + mucus composting; S1: sludge + mucus + 2% straw charcoal composting; S2: sludge + mucus + 4% straw charcoal composting; S3: sludge + mucus + 6% straw charcoal composting; S4: sludge + mucus + 8% straw charcoal composting. Different letters in the same column show significant differences among different treatments ($P<0.05$).

堆肥后, 重金属总量均表现出降低趋势。与 CK 相比, 仅添加粘液的试验组 (S0) 中重金属 Cd、Pd、Cu、Zn 有降低趋势, 但差异不显著, 而 Ni 含量显著降低 10.93% ($P<0.05$); 当粘液-秸秆炭共同作用对污泥堆肥后, 重金属总量的降低幅度随秸秆炭含量加大而增加, 在粘液+8% 秸秆炭试验组 (S4) 重金属总量降低幅度达最大, Cd、Pb、Cu、Ni 和 Zn 含量分别降低 36.92%、67.33%、6.07%、59.59% 和 31.82% ($P<0.05$)。综上所述, 粘液协同秸秆炭堆肥后对降低污泥中多数重金属效果较好, 而对 Cu 效果一般, 堆肥后重金属总量低于《农用污泥污染物控制标准》(GB 4284—2018) A 级标准, 更远低于欧盟污泥农用标准^[24], 此后可斟酌用作园林公路绿化、草坪基质及农作。

2.3 堆肥后对污泥中重金属有效态的影响

堆肥体系中有效态重金属是指能被植物体直接汲取、富集的重金属, 包含水溶态、酸溶态、螯合态和吸附态等多种形态, 具有较强毒害作用, 而本试验中有效态包含可交换态和碳酸盐结合态^[25]。如表 4

所示, 单独添加粘液污泥堆肥 (S0) 后, 重金属有效态均有降低趋势, 其中有效态镉含量显著降低 ($P<0.05$), 可能粘液中富含的氨基酸和蛋白质提供与重金属络合的有机官能团, 降低其有效性。当粘液-秸秆炭共同作用对污泥堆肥后, Cu 有效态含量逐步上升, Ni、Pb、Zn 有效态含量表现为下降趋势。与 CK 相比, 粘液+8% 秸秆炭处理 (S4) 变化幅度最大, 有效态铜含量升高 89.68%, 有效态镍、铅、锌含量分别下降 28.05%、41.99%、28.30% ($P<0.05$); 而有效态镉含量在粘液+2% 秸秆炭处理组 (S1) 较 CK 降幅最大, 为 38.46% ($P<0.05$), 随后开始升高, 但始终低于 CK。这是粘液和秸秆炭的共同作用, 产生能固化重金属的负电荷基团, 降低多数重金属有效性, 而腐殖质中溶解性有机碳活化了 Cu, 提升了其有效性^[26-27]。此外, 利用相关性分析粘液+不同比例秸秆炭对堆肥后 (S0-S4) 重金属有效态的影响发现, 秸秆炭添加量与重金属 Cu、Zn、Pb、Ni 有效态的相关系数分别为 0.906、-0.909、-0.847、-0.639, 均达显著水平 ($P<0.05$), 而与有效态镉含量相关系数较低, 为 0.392。

表 4 蚯蚓粘液-秸秆炭共同作用对污泥堆肥后重金属有效态含量的影响

Table 4 Effects of applying earthworm mucus and straw charcoal on contents of available heavy metals of sludge compost $\text{mg}\cdot\text{kg}^{-1}$

处理 Treatment	有效态镉 Available Cd	有效态铅 Available Pb	有效态铜 Available Cu	有效态镍 Available Ni	有效态锌 Available Zn
CK	0.39±0.05a	7.43±0.00a	3.10±0.24d	5.17±0.10a	8.41±0.10a
S0	0.28±0.01d	6.60±0.29a	3.05±0.47d	4.58±0.09ab	8.29±0.14a
S1	0.24±0.04d	5.56±0.82b	4.70±0.10c	4.63±0.68ab	7.67±0.45b
S2	0.34±0.03bc	5.21±0.31bc	5.19±0.30bc	4.17±0.37bc	6.74±0.31c
S3	0.36±0.01ab	4.85±0.54bc	5.69±0.29ab	4.14±0.18bc	5.79±0.31d
S4	0.29±0.02cd	4.31±0.54c	5.88±0.10a	3.72±0.59c	6.03±0.46d

CK: 单独污泥堆肥; S0: 污泥+粘液堆肥; S1: 污泥+粘液+2%秸秆炭堆肥; S2: 污泥+粘液+4%秸秆炭堆肥; S3: 污泥+粘液+6%秸秆炭堆肥; S4: 污泥+粘液+8%秸秆炭堆肥, 同列不同字母表示不同处理间差异显著 ($P<0.05$)。CK: sludge composting; S0: sludge + mucus composting; S1: sludge + mucus + 2% straw charcoal composting; S2: sludge + mucus + 4% straw charcoal composting; S3: sludge + mucus + 6% straw charcoal composting; S4: sludge + mucus + 8% straw charcoal composting. Different letters in the same column show significant differences among different treatments ($P<0.05$).

为了深入解析不同处理组与堆肥后重金属有效态关系, 利用主成分分析法 (PCA) 得到如下结果: 两种主成分 (PC1=76.2%, PC2=20.8%) 的累积方差贡献率达 97.0%, 符合要求 (图 1), 表示不同处理组对重金属有效态影响, 污泥堆肥对照组 (CK) 和粘液+8% 秸秆炭堆肥处理组 (S4) 在 PC1 方向上分别表现出明显正和负效应, 粘液+2% 秸秆炭堆肥处理组 (S1) 在 PC2 方向上表现出明显负效应。由此可知, CK 对降低污泥中重金属有效态含量效果较差, 而 S4 和 S1 效果较好。此外, 通过重金属有效态的因子加载发现, Zn、Pb、Ni 和 Cd 分别在 PC1 和 PC2 正方向上作用强, 而 Cu 在 PC1 负方向上作用强。结果表明, 粘液+2% 秸秆炭 (S1) 对钝化污泥中重金属 Cd 效果较好, 而粘液+8% 秸秆炭 (S4) 能更好地钝化污泥中

重金属 Ni、Pb、Zn 活性, 却活化了重金属 Cu, 这与图 2 所示结果一致。

2.4 堆肥后污泥中重金属形态变化

重金属在污泥中以可交换态、碳酸盐结合态、铁锰结合态、有机结合态和残渣态 5 种形态存在, 而堆肥能改变重金属在污泥中的化学形态分布, 从而钝化污泥中重金属的生物活性。其中可交换态和碳酸盐结合态在 5 种形态中活性较强, 易于被植物体所富集, 是影响污泥中重金属有效性的最主要形态, 铁锰结合态较为稳定, 易受酸碱度所影响, 有机结合态与残渣态化学性质最为稳定, 较难被植物吸收富集。

如图 2 所示, 各处理组堆肥后重金属 Cu 以有机结合态为主, Zn 以铁锰结合态为主, Ni 和 Pb 以残渣态为主, 而 Cd 以可交换态为主。单独添加粘液对污

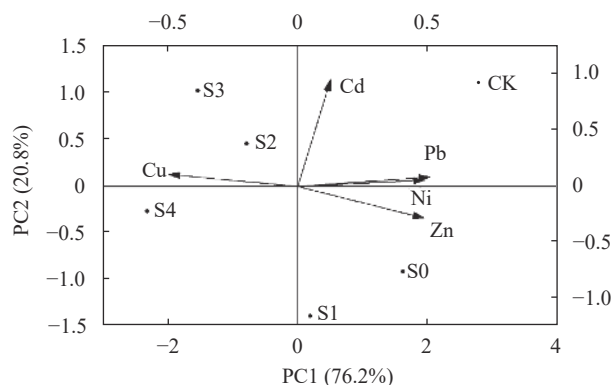


图 1 粘液和秸秆炭添加与污泥堆肥重金属含量的关系
Fig. 1 Relationship between applying earthworm mucus and straw charcoal and heavy metal contents of sludge compost

CK: 单独污泥堆肥; S0: 污泥+粘液堆肥; S1: 污泥+粘液+2% 秸秆炭堆肥; S2: 污泥+粘液+4% 秸秆炭堆肥; S3: 污泥+粘液+6% 秸秆炭堆肥; S4: 污泥+粘液+8% 秸秆炭堆肥。CK: sludge composting; S0: sludge + mucus composting; S1: sludge + mucus + 2% straw charcoal composting; S2: sludge + mucus + 4% straw charcoal composting; S3: sludge + mucus + 6% straw charcoal composting; S4: sludge + mucus + 8% straw charcoal composting.

泥堆肥后, 重金属均由较为活泼形态朝着残渣态转化, 较 CK 的残渣态所占比例提升了 33.19%~120.19%。当粘液-秸秆炭共同作用对污泥堆肥后, Ni 和 Pb 的碳酸盐结合态、Pb 的铁锰结合态、Zn 的可交换态逐步向稳定难降解的残渣态转化, 在添加 8% 秸秆炭时, 重金属 Ni、Pb、Zn 的残渣态所占比例较 CK 分别提高 40.26%、102.74% 和 208.01%; 而 Cu 的有机结合态却同时向可交换态和残渣态这两种活性相异的形态转化, 加入 8% 秸秆炭时可交换态和残渣态所占比例较 CK 试验组分别升高 65.20% 和 50.82%; 残渣态 Cd 也向可交换态和碳酸盐结合态两种活性较强形态转化, 但这两种形态比例均未超过 CK。这表明粘液协同秸秆炭堆肥后, 有机质的加速矿化和腐质化, 使产生的多种活性基团络合重金属, 降低其迁移和转化能力, 从而钝化部分重金属活性, 使其形态变的更为稳定; 而 Cu 在矿化和腐质化下发生游离, 与溶解性有机碳等物质结合, 降低了其固化效果, 从而朝着相反形态变化^[28-30]。

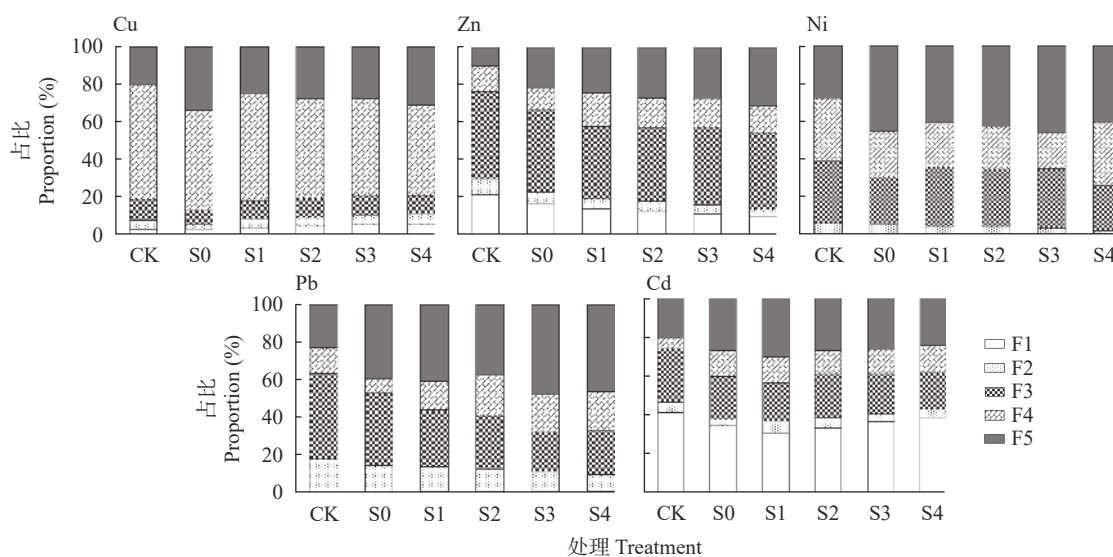


图 2 蚯蚓粘液-秸秆炭共同作用对污泥堆肥后重金属有效态分布的影响

Fig. 2 Effects of applying earthworm mucus and straw charcoal on distribution of available forms of heavy metals during sludge composting

CK: 单独污泥堆肥; S0: 污泥+粘液堆肥; S1: 污泥+粘液+2% 秸秆炭堆肥; S2: 污泥+粘液+4% 秸秆炭堆肥; S3: 污泥+粘液+6% 秸秆炭堆肥; S4: 污泥+粘液+8% 秸秆炭堆肥。F1: 可交换态; F2: 碳酸盐结合态; F3: 铁锰结合态; F4: 有机结合态; F5: 残渣态。CK: sludge composting; S0: sludge + mucus composting; S1: sludge + mucus + 2% straw charcoal composting; S2: sludge + mucus + 4% straw charcoal composting; S3: sludge + mucus + 6% straw charcoal composting; S4: sludge + mucus + 8% straw charcoal composting. F1: exchange form; F2: carbonate bound form; F3: iron-manganese bound form; F4: organic form; F5: residual.

3 讨论

当粘液-秸秆炭共同作用对污泥堆肥后, pH 显著升高, 一方面原因是粘液和秸秆炭增加了堆肥中微生物丰度, 促进了氨基酸脱氨基作用, 大量氨气释放到堆肥中, 使 pH 逐渐升高; 另一方面原因是秸秆炭

释放自身的弱碱性物质, 也可提升堆肥体系中酸碱度^[9,31-32]。EC 与污泥中可溶性矿物盐浓度密切相关。Pan 等^[12]发现蚯蚓粘液可作为诱导有机质矿化的启动因子, 提高矿化速率; 此外, Huang 等^[14]还发现添加剂能提高堆肥体系中微生物活性, 提升有机质分

解速率,产生大量 HCO_3^- 、 NH_4^+ 和其他无机盐等,使 EC 值逐渐提升,这也与本研究结果相类似。粘液协同秸秆炭堆肥污泥后还能促进污泥中有机质分解成矿物盐,使有机质含量在单一粘液堆肥的处理组下降,不断添加秸秆炭后,残留较多结构稳定的难溶性有机碳,使堆肥体系中有有机质含量增加^[33]。

Guo 等^[33] 研究发现,生物炭能促进堆肥体系中氨气的释放,而其较高的比表面积和多孔结构对 NH_3 和 NH_4^+ 的吸附作用,又会减小氮损失;生物炭也能提高堆肥中氧气含量,减少 N_2O 和 CH_4 排放,进一步降低氮损失^[34]。本研究结果则不同,单独添加粘液对污泥堆肥后,因粘液能刺激堆肥体系中微生物活性和提高细菌丰度,可将大量氮元素通过氨化、反硝化作用以 NH_3 、 N_2 、 N_2O 形式损失^[22];当粘液与不同比例秸秆炭共同作用对污泥堆肥后,粘液协同秸秆炭为参与氮反应的微生物提供更加有利的生存环境,使氨挥发能力远超过生物炭的吸附能力^[35],伴随着秸秆炭产生的稀释作用,造成总氮含量降低;Wei 等^[23] 发现,磷元素常以固态或液态存在于堆体中,当有机物快速降解产生的有机酸会溶解部分难溶性磷到滤液中,导致磷流失,这也可能与生物炭稀释作用和阳离子交换量有关,污泥中产生的有机酸也是 K 溶解的主要机制,从而部分溶解性钾随滤液溶出,再加上生物炭的稀释作用,造成 K 含量略降低^[36]。

Duan 等^[32] 观察到,有机废物中添加生物炭能促进堆肥基质快速降解,使活性有机物向稳定的腐殖质转化并络合重金属,提高重金属在堆肥中的水溶性,导致重金属总量因滤出而损失。这与本研究结果相类似,在本试验中添加粘液能提高堆肥污泥中有机质矿化和腐殖化速率,使腐殖质中富里酸吸附重金属成为结合体随滤液浸出,造成重金属总量降低^[14];当粘液与不同比例秸秆炭共同作用对污泥堆肥后,又对堆肥产生了较强的稀释效果,进一步降低重金属总量并减少重金属对堆肥的毒害作用,提高

堆肥质量^[10,37-38]。

堆肥过程中生物炭通过物理吸附、氧化还原反应、沉淀络合、静电作用和离子交换反应等机制固定重金属,钝化堆肥中重金属活性^[26]。在本试验中,粘液-秸秆炭共同作用对污泥堆肥后降低了重金属 Cd、Ni、Pb、Zn 有效态含量,却提高了 Cu 有效态含量,由此对各处理组堆肥后的重金属有效态和理化性质进行相关性分析得出(表 5),重金属 Ni、Pb、Zn 有效态含量与 pH 表现为负相关 ($P<0.01$)。随着秸秆炭添加比例提高, pH 也呈上升趋势并促使重金属 Ni、Pb、Zn 有效态含量逐渐降低。这是因为粘液中富含游离氨基酸和蛋白质^[16],为堆体提供较多氨基、羧基、羟基等官能团,伴随着污泥中秸秆炭的添加,秸秆炭自身碱性及其易降解物质(乙酸、丁酸等)分解,碱度也逐渐提高,促进了官能团脱质子化反应,产生较多带负电荷的基团于堆体中并与重金属发生络合反应,钝化重金属 Ni、Pb 和 Zn^[26,39-40]。本试验结果与 Li 等^[26] 证明的堆肥体系 pH 维持在 5.5~12.5 范围内, Pb 主要以 $\text{Pb}(\text{OH})_2$ 形式存在类似;也与 Liu 等^[9] 发现的添加生物炭到污泥中进行好氧堆肥后,堆体 pH 升高,对污泥中 Ni 有效性产生钝化效果相一致。重金属 Ni、Pb、Zn 含量与有机质含量和 EC 也表现出负相关 ($P<0.01$),可能生物炭对微生物的存活环境具有调节作用,粘液也可提升污泥中细菌丰度,进而促进粘液秸秆炭堆肥体系中 OM 分解成腐殖质,使重金属被堆肥过程中形成的能够吸附和固定重金属离子的胡敏酸所络合,钝化重金属活性^[33,41]。Cu 有效态与 pH、EC 和 OM 表现出显著正相关 ($P<0.01$),因为粘液-秸秆炭共同作用污泥,能使堆体腐殖化产生较多溶解性有机碳(包括富里酸和类黄腐殖酸等),促进溶解性有机碳对 Cu 的结合,提高了 Cu 在污泥堆肥中的可移动性,进而提高 Cu 有效态含量^[28-29,32,42],这也与 Zhao 等^[27] 发现 Cu 易与有机质中溶解性有机碳结合,提高 Cu 的迁移率相一

表 5 污泥堆肥中重金属有效态含量与理化性质的相关关系

Table 5 Correlations between contents of available forms of heavy metals with physical and chemical properties of sludge compost

重金属 Heavy metal	理化性质 Physicochemical property					
	pH	电导率 Electrical conductivity	有机质 Organic matter	总氮 Total nitrogen	总磷 Total phosphorus	总钾 Total potassium
镍 Ni	-0.776**	-0.734**	-0.666**	0.727**	0.551*	0.238
铅 Pb	-0.906**	-0.872**	-0.820**	0.903**	0.337	0.465
锌 Zn	-0.906**	-0.854**	-0.796**	0.905**	0.431	0.388
镉 Cd	-0.229	-0.202	-0.194	0.271	0.049	-0.044
铜 Cu	0.944**	0.932**	0.866**	-0.937**	-0.239	-0.565*

和*分别表示在 $P<0.01$ 和 $P<0.05$ 水平显著相关。 and * indicate significant correlation at $P<0.01$ and $P<0.05$ levels, respectively.

致。Cd 有效态含量受堆体理化性质影响较小, 可能生物炭表面的阳离子交换对 Cd 具有吸收作用, 从而降低了 Cd 活性^[26,33]。为了深入了解污泥中理化性质

对重金属有效性的直接影响, 对其进行回归方程分析发现(表 6), pH 是影响堆肥污泥中重金属 Ni、Pb、Zn、Cu 有效态的最关键因素。

表 6 污泥堆肥中重金属有效态含量与理化性质的回归方程

Table 6 Regression equations between contents of available forms of heavy metals with physical and chemical properties of sludge compost

重金属 Heavy metal	逐步回归方程 Stepwise regression analysis	拟合指数 R^2	显著性 P
锌 Zn	$y=35.939-3.965x_{\text{pH}}$	0.81	<0.01
镍 Ni	$y=17.474-1.801x_{\text{pH}}$	0.58	<0.01
铜 Cu	$y=-32.766+4.901x_{\text{pH}}+0.089x_{\text{TP}}$	0.92	<0.01
铅 Pb	$y=36.106-4.194x_{\text{pH}}$	0.81	<0.01
镉 Cd	—	—	—

x_{pH} 为污泥堆肥的 pH, x_{TP} 为污泥堆肥的总磷含量。“—”为未检出。 x_{pH} is the pH of the sludge compost, x_{TP} is the total phosphorus content of the sludge compost. “—” means not detected.

单独粘液添加到污泥中堆肥后降低了重金属不稳定性, 这可能是粘液中含有的电解质和较多糖蛋白、粘多糖、凝集素、血蓝蛋白等大分子混合物^[12], 为微生物代谢创造适宜微环境, 改变堆肥中微生物分布, 促进堆肥基质的矿化和腐殖化, 使矿化后的有机质及粘液中原本富含的多种活性基团与重金属形成络合物, 钝化污泥中重金属活性^[14,30]。当粘液-秸秆炭共同作用对污泥堆肥后, 重金属 Pb、Ni 和 Zn 的有效态所占比例降低, 可能是粘液和秸秆炭协同作用提升了堆肥 pH, 增强堆体中腐殖质官能团(羧基等)脱质子化, 产生较多能与重金属有效络合的负电荷, 并形成表面沉淀物, 固定重金属^[43-44], Zheng 等^[45]也发现重金属 Ni 的前 3 种形态与 pH 呈负相关, 这也与 Liu 等^[9]研究发现生物炭对污泥堆肥降低了重金属 Pb、Zn 和 Ni 的生物可利用度相一致; 污泥中残渣态所占比例随生物炭比例增加也逐渐提升, 这时堆体基质的矿化能力增强, 促进了有效硅产生, 产生较多可与重金属络合成极难溶沉淀的硅酸根, 从而提升残渣态所占比例^[46]。随着粘液和秸秆炭的添加, 有机结合态 Cu 向可交换态和残渣态转化, Cd 的交换态所占比例也逐渐提高, 这时有有机结合态 Cu 在污泥中含量丰富, 在粘液和秸秆炭的作用下促进有机质矿化和腐殖化, 使更多有机 Cu 游离, 并与溶解性有机碳等物质结合, 降低了 Cu 的固化效果^[27-29,42], 而试验中 Cd 的变化与 Liang 等^[37]试验结果相类似。

4 结论

1) 粘液-秸秆炭共同作用对污泥堆肥后能提高污泥碱度和有机质, 却导致养分损失; 当粘液+4% 秸秆炭时, 堆肥后污泥呈碱性, 有机质含量丰富, 养分损失少。

2) 粘液-秸秆炭共同作用对污泥堆肥后能降低污泥中重金属总量, 钝化了 Ni、Pb、Zn, 而活化 Cu; 进一步对污泥中重金属有效态分析得出, pH 是影响重金属有效态的最关键因素, 而粘液+8% 秸秆炭钝化重金属 Ni、Zn、Pb 效果甚佳, 对重金属 Cu 具有活化效果。

3) 单独添加粘液对污泥堆肥后的重金属活性降低, 残渣态所占比例提高; 而粘液-秸秆炭共同作用对污泥堆肥后, 碳酸盐结合态 Ni 和 Pb、铁锰结合态 Pb、可交换态 Zn 逐步朝着稳定的残渣态转化, 而 Cd 和 Cu 变化相异, 前者朝着可交换态转化, 后者朝着可交换态和残渣态转化。

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