

# 添加污泥对尾矿砂理化性质及香樟生理特性的影响

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**摘要:**以香樟作为指示植物,选取黄岩、临海和路桥地区污水处理厂污泥,将污泥与尾矿砂按(污泥质量比例为0%对照、25%、50%和75%)配比进行栽培试验。测定添加污泥对尾矿砂理化性质以及香樟生理特性的影响。结果表明:随着污泥比例的增加,混合基质中的有机质、全氮、全磷明显增加,pH值明显降低,离子交换量明显减少,Cu和Cd总量明显增加,而Pb总量明显减少,Zn总量没有明显变化,Cu、Cd和Zn的DTPA提取量明显增加,而Pb的DTPA提取量明显减少。黄岩和临海污泥在25%和50%比例时,香樟叶和茎的生物量和叶绿素含量明显增加,而根的生物量没有明显变化,在75%比例时,生物量和叶绿素含量均明显减少;而添加路桥污泥使香樟叶、茎和根的生物量和叶绿素含量明显减少。丙二醛含量则与生物量和叶绿素含量呈现相反的变化特征。黄岩和临海污泥在25%和50%比例时,根和叶的Cu、Cd、Pb和Zn含量明显减少,在75%比例时,Cu、Cd和Zn含量则明显增加;而添加路桥污泥使叶和根的Cu、Cd和Zn含量明显增加,Pb含量明显减少。研究表明添加污泥提高了尾矿砂的养分含量,同时改变了其重金属组成,对污泥重金属含量和有机质组成的监控可以准确地预测污泥改良后尾矿砂对植物毒性的变化。

**关键词:**污泥;尾矿砂;香樟;理化性质;生理特性

## Effects of sewage sludge amendment on physico-chemical properties of mine tailings and physiological responses of *Cinnamomum camphora*

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**Abstract:** In order to investigate the effects of sewage sludge amendment on physico-chemical properties of mine tailings and physiological responses of *Cinnamomum camphora* to the mixture, three different sites of sewage sludge (Huangyan (HY), Linhai (LH) and Luqiao (LQ)) were collected, and then mixed with mine tailings in which the mass percentage of sewage sludge was 0%, 25%, 50% and 75%, respectively. Results showed that sewage sludge amendment increased the nutrient of mine tailings and changed the heavy metal composition of mine tailings. With the increase of sludge amendment ratios, organic matter, total N and total P were significantly increased, while pH and cation exchange capacity were significantly decreased; total and DTPA-extractable contents of Cu, Cd and Zn except for total Zn were significantly increased, while total and DTPA-extractable contents of Pb were significantly decreased. It can be inferred that the composition of heavy metals in the mixture was related with the original heavy metal characteristics of both mine tailings and sewage sludge. The biomass of leaves, stems and roots, and chlorophyll contents consistently decreased with the increase of LQ sewage sludge amendment ratios, which may be related with the high Cu contents in LQ sludge. The biomass of leaves and stems, and chlorophyll contents of *C. camphora* grown at 25% and 50% sewage sludge in HY and LH were more than those at control (0), whereas they were significantly decreased at 75% sewage sludge content. The change of MDA contents, the indicator of biotoxicity, was reverse to those of biomass and chlorophyll. Therefore, the 50% amendment ratios of HY and LH sludge was assumed the best remediate measures due to the least biotoxicity to *C. camphora*. The biotoxicity had a closely relation with the accumulation of heavy metals in *C. camphora*. Cu, Cd, Pb and Zn contents of

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leaves and roots at 25% and 50% ratios of HY and LH sewage sludge were more than those of control (0%), but Cu, Cd and Zn contents of leaves and roots greatly increased at 75% ratios of HY and LH sewage sludges. However, with the increase of LQ sewage sludge amendment ratios, Cu, Cd and Zn contents of leaves and roots consistently increased, and were more than those of HY and LH sludge treatments. Notably, Pb in the leaves and roots consistently decreased with the increase of LQ sewage sludge amendment ratios, which may be related to the decrease of total and DTPA-extractable contents of Pb. Only in LQ sludge treatments, the Cu, Cd and Zn contents of leaves and roots had positive correlation with their DTPA-extractable contents, respectively, which may be related to different organic matter composition between LQ and other two sludge types. Therefore, heavy metal contents and organic matter composition in the sewage sludge could help to predict the bio-toxicity to plants when mine tailings were amended with sludge.

**Key Words:** sewage sludge; mine tailing sand; *Cinnamomum camphora*; physico-chemical properties; physiological properties

城市污泥是指城市生活污水、工业废水处理过程中产生的固体废弃物。因人口快速增长和城市化,污泥废弃物的不合理贮存对城市环境构成威胁,并日益成为世界许多城市的主要环境问题<sup>[1]</sup>。目前,我国年废水排放总量约在400多亿t,每年排放干污泥约为550—600万t,且在不断增加之中<sup>[1]</sup>。到2010年,中国城市化率将达40%,城镇人口总量将增至6.7亿人。按每人每天产生50g(干物质)污泥计算,城市污泥排放量每年将达1200万t(干物质)以上<sup>[2]</sup>。尾矿是在矿石选取精矿石后外排的脉石废渣。据中国环境年鉴资料,尾矿是我国排放量最高的工业固体废弃物,2006年我国尾矿排放量达265.4Mt,占总工业固体废弃物排放量的28.1%<sup>[3]</sup>。至今我国尾矿占地面积已高达90多万hm<sup>2</sup>,由于尾矿养分贫瘠、水流失较快,以及高的重金属毒性,使得植物难以在尾矿上生长<sup>[4]</sup>,其复垦率只有10%左右。因此,如何合理、经济、有效地处置数量巨大的污泥和尾矿等工业固体废弃物已成为非常紧迫的任务。

城市污泥处理与处置方法主要包括土地利用、填埋、焚烧和投海,但是容易污染地下水、大气等环境,且焚烧成本较高<sup>[5]</sup>。城市污泥也可被施加到农田、草地、森林和园林生态系统,虽然可增加植物产量<sup>[6-9]</sup>,但同时导致重金属和病原物污染,氮磷过剩,从而引起一系列水污染环境问题<sup>[10]</sup>。最近研究表明在尾矿上施用污泥(城市污泥一般含有较高的有机质和矿质营养(有机质300—600 g·kg<sup>-1</sup>、氮10—40 g·kg<sup>-1</sup>和磷6—15 g·kg<sup>-1</sup>)<sup>[11]</sup>)可提供有机碳并改善土壤肥力,改良尾矿理化性质和防治水土流失,提高微生物的活性和迅速有效地恢复植被<sup>[12-13]</sup>,而且可以避开食物链,减少对人类健康的威胁。但是,过多添加污泥会增加对植物的毒性和重金属的溶出<sup>[14]</sup>。

香樟(*Cinnamomum camphora*)属于国家二级保护物种,是我国南方珍贵的用材和重要的城市绿化树种之一,具有重要的经济和观赏价值。在南方尾矿调查过程中,已发现香樟能够生长在一些重金属尾矿上,但有关香樟对重金属的耐性生理国内外鲜有报道。本研究以浙江省台州地区3个污水处理厂脱水后污泥和三门Pb/Zn尾矿砂为材料进行不同比例配比,并栽种香樟幼苗,研究添加不同比例污泥后尾矿砂理化特性的变化以及对香樟生理特性的影响,为今后利用污泥进行尾矿基质改良及其相应的香樟植物修复提供理论依据和参考。

## 1 材料与方法

### 1.1 试验材料

试验污泥取自黄岩、临海和路桥3个地区污水处理厂脱水后的污泥,在实验室风干、研磨,过2mm筛,标记后,分别装入塑料袋备用。尾矿砂取自三门Pb/Zn尾矿砂堆积地。2007年秋季,香樟种子收集于三门Pb/Zn尾矿砂堆积地边缘林下,并于2008年3月将种子播种于校园内废弃空旷草地,定时浇水,等两片真叶长出,选择生长特征相似的幼苗移栽供试验使用。

## 1.2 研究方法

将黄岩、临海和路桥地区污水处理厂污泥粉末分别与尾矿砂按照质量比进行配比,最终形成4个污泥-尾矿砂混合系列,每个系列的污泥所占质量比例为0% (尾矿砂,作为对照)、25%、50% 和 75%。不同比例混合基质分别放于5个(总共45个)塑料盆中( $30\text{cm} \times 20\text{cm} \times 20\text{cm}$ )混合均匀,然后浇水浸泡,风干后,再次浇水浸泡,重复3次。等污泥-尾矿砂混合基质充分混合稳定后,分别从每个塑料盆中取100g混合基质,风干用于理化指标的测定,剩余部分分别装于塑料花盆中(直径18cm、高25cm),每盆装2.0 kg混合基质,每个处理5个重复。每个花盆种4棵香樟幼苗,定期浇水(每次200 mL),水缓慢浸满花盆基质而未流出。生长90d后取第3—5叶(从上至下)进行叶绿素和丙二醛生理指标测定,整株收获,将根、茎和叶分开,分别称量鲜重。

## 1.3 指标测定

### 1.3.1 植物生理指标

叶绿素和丙二醛含量测定参照章家恩等的方法<sup>[15]</sup>。

### 1.3.2 植物与土壤重金属

准确称取植物体样品干重0.2—0.5g,加5mL浓硝酸浸泡过夜,放于红外线消化炉中,依次在90℃、140℃和180℃下分别消化30min,冷却后,加1mL高氯酸,然后在180℃下再消化120min,然后将消化液冷却、定容,过滤;准确称取0.5g风干土(过1mm筛)放于消化管中,然后加4mL的浓硝酸和1mL的浓盐酸过夜浸泡。第2天将过夜浸泡的消化管放在红外线消化炉中,先在90℃下煮30min,再升温到140℃,并保持30min,再升温到180℃,并保持1h。取出消化管,加1mL高氯酸,160℃下保持20min。再升温到180℃,并保持2h以上,直到消化液变清,土变白为止,然后将消化液冷却、定容,过滤;准确称取10g风干土在150mL锥形瓶中,加20mLDTPA提取液( $0.005\text{mol}\cdot\text{L}^{-1}$  DTPA,  $0.01\text{mol}\cdot\text{L}^{-1}$   $\text{CaCl}_2\cdot 2\text{H}_2\text{O}$ ,  $0.1\text{mol}\cdot\text{L}^{-1}$  TEA, pH = 7.3),密封放在回旋震荡器中,按 $120\text{r}\cdot\text{min}^{-1}$ 提取2h,同时设对照,用滤纸过滤。滤液重金属浓度均用ICP/OES(Optima 2100DV, Perkin Elmer, USA)测定。

### 1.3.3 尾矿砂理化指标

土壤有机质、总氮、总磷、pH和离子交换量,按常规的土壤化学分析方法进行测定<sup>[16]</sup>。

## 1.4 数据分析

采用SPSS 10.0统计软件对数据进行单因素方差分析(one-way analysis of variance, ANOVA),即在平均值比较基础上,采用LSD方法,在 $P = 0.05$ 或 $0.01$ 水平进行数据差异显著性检验,以比较不同地区污泥和不同比例混合基质间各指标的差异显著性。采用Canoco 4.15多元统计软件,利用其中的冗余分析(redundancy discriminant analysis, RDA),将香樟生长生理指标定义为反应变量,土壤理化性质和重金属含量定义为解释变量,首先以回归分析方法将所有解释变量降维,建立若干个新的线性组合,并以方差贡献率最大的两个线性组合得分为轴作二元回归图。图中各解释变量与横坐标轴夹角大小反映该变量与该线性组合的关系密切程度,反应变量与解释变量的重要程度分别用箭头的长短来表示,通过蒙特卡罗检验,反应变量与解释变量的关系用二者之间夹角来表示,夹角在0—90°之间表明二者呈正相关,等于90°表明不相关,大于90°表明呈负相关<sup>[17]</sup>。

## 2 结果与分析

### 2.1 污泥理化特性

黄岩、临海和路桥污水处理厂污泥均为酸性污泥( $\text{pH} < 6.5$ ,表1)。3种污泥与其他城市污泥理化特性相似,均含有丰富的有机质、总氮和总磷,而且具有较高的离子交换量。高的重金属含量是3个地区污泥的重要特点,尤其铜含量较高,路桥地区铜含量超过 $1000\text{ mg kg}^{-1}$ 。

### 2.2 不同污泥配比对尾矿砂理化性质的影响

随着污泥比例的增加,混合基质的pH值逐渐降低,有机质、总氮和总磷含量则逐渐增加(表2)。路桥污泥混合基质的有机质增加最多,添加75%比例时,有机质增加至 $372.19\text{ g kg}^{-1}$ 。但是,混合基质的离子交换







已有研究表明,施用污泥有利于提高土壤有机质、全N及土壤理化性状如土壤电导率和速效N、P、K含量<sup>[6, 19-20]</sup>,并可增加土壤有效N、土壤全C含量及提高土壤pH<sup>[21]</sup>。此研究同样发现随着污泥比例的增加,混合基质中有机质、全氮和全磷呈明显的上升趋势。但是,随着污泥比例的增加,混合基质的pH值和离子交换量呈明显的下降趋势,这与三门Pb/Zn尾矿砂具有高pH值和离子交换量有关,污泥添加起到了稀释作用,降低了pH值和离子交换量。Ye等<sup>[22]</sup>研究表明将污泥应用于Pb/Zn尾矿的垦植,不仅增加全碳、N、P和K含量,而且降低尾矿基质的Zn、Pb、Cd全量,继而降低了植物对Zn、Pb、Cd的吸收和积累。研究发现混合基质的重金属含量取决于污泥和尾矿砂两个组成部分的重金属含量特征。与尾矿砂Cu、Cd和Pb含量相比,3种污泥的Pb含量明显偏低,Cu和Cd含量明显偏高,因此随着污泥比例增加,混合基质的Pb全量明显减少,Cu和Cd全量明显增加。DTPA可提取量是植物可获得重金属含量的重要指标<sup>[23]</sup>。Cu、Cd和Zn的DTPA可提取量随污泥比例的增加呈明显增加的趋势,而Pb的DTPA可提取量则明显减少,这与混合基质中Cu、Cd和Zn总量明显增加,Pb总量明显减少有关,冗余分析结果显示其全量和可提取量均呈不同程度的正相关。而且,有机质含量的DTPA明显提高也可导致Cu、Cd和Zn的DTPA可提取量的增加<sup>[24]</sup>,冗余分析结果显示混合基质的有机质含量与Cu、Cd和Zn的DTPA可提取量均呈不同程度的正相关。

目前,添加污泥后混合基质理化性质变化的监控和植物毒性测定是对污泥改良措施风险评估的主要方法<sup>[25]</sup>。本研究以香樟为指示植物来测定不同比例配置的污泥/尾矿砂混合基质对生物的毒性特征。随着黄岩和临海污泥比例增加,香樟叶和茎的生物量均呈明显增加趋势,这与Morera等<sup>[26]</sup>研究混合基质对向日葵毒性特征的结果相似。但是,添加路桥污泥对香樟生长产生明显的抑制作用,叶片过高的Cu含量可能是对生长抑制的主要原因。相比叶片,3种污泥对根生长均表现出较强的抑制作用,这可能与根中高的重金属含量有关。叶绿素a、b含量和叶绿素全量的变化与叶片生物量变化呈现相似的规律,其与混合基质中Cu、Cd和Zn的DTPA可提取量呈现不同程度的负相关,由于DTPA可提取量决定了植物体吸收重金属的含量,Cu、Cd和Zn对叶绿素合成的抑制作用<sup>[27]</sup>导致了负相关关系的产生。类胡萝卜素含量随着污泥比例的增加均呈稳定增加趋势,其具有较强的抗氧化作用<sup>[28]</sup>,因此类胡萝卜素含量的增加可能是香樟对重金属的耐性特征。丙二醛是反映植物体内自由基的积累导致细胞膜质过氧化程度的重要指标<sup>[29]</sup>,冗余分析结果显示香樟叶和根的丙二醛含量与Cu、Cd和Zn的DTPA可提取量均呈不同程度的正相关,因此丙二醛含量可作为尾矿污泥改良和植被修复过程中重金属植物有效性和毒性的重要检测指标。黄岩和临海污泥在25%和50%比例时,香樟叶和根的SOD、POD和CAT酶活性均明显低于对照,表明添加污泥减轻了香樟的氧化损害,其抗氧化酶系统并没有完全启动,香樟生物量和叶绿素含量的增加证明了这一点。但是,路桥污泥在25%比例时使得SOD、POD和CAT酶活性迅速增强,大于50%比例后,酶活性迅速

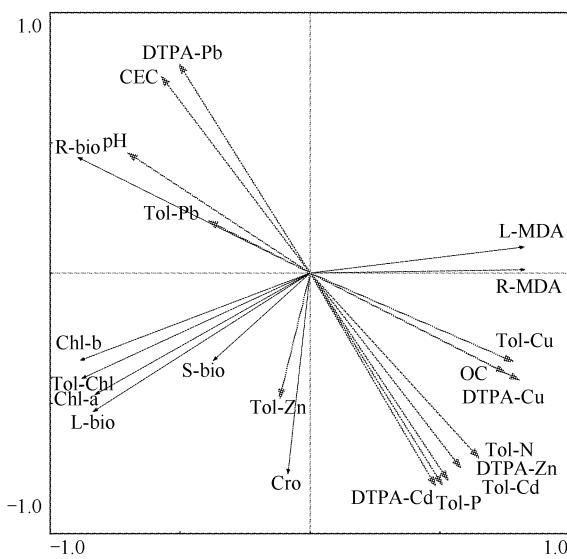


图1 香樟生长生理指标和与混合基质理化性质的关系

Fig. 1 Relationships between physiological and growth parameters of *C. camphora* and physical-chemical variables in sludge-mine tailing composites

L-bio, S-bio, R-bio: 叶、茎和根生物量 The biomass of leaves, stems and roots, respectively; Chl-a, Chl-b, Tol-Chl, Cro: 叶绿素a,b,叶绿素全量和类胡萝卜素 Chlorophyll a, Chlorophyll b, Total Chlorophyll and Crotonoid; L-MDA, R-MDA: 叶和根的丙二醛含量 The MDA contents of leaves and Roots, respectively; OC: 有机碳 Organic C; TN: 全氮 Total nitrogen; TP: 全磷 Total phosphorus; CEC: 阳离子交换量 Cation exchangeable capacity; DTPA-Cu, Pb, Cd 和 Zn 分别为 Cu、Pb、Cd 和 Zn 的 DTPA 可提取量 The soil content of Cu, Pb, Cd and Zn extracted with the diethylenetriaminepentaacetic acid, respectively; Tol-Cu、Tol-Pb、Tol-Cd、Tol-Zn 分别为 Cu、Pb、Cd 和 Zn 总含量 The total contents of Cu, Pb, Cd and Zn, respectively

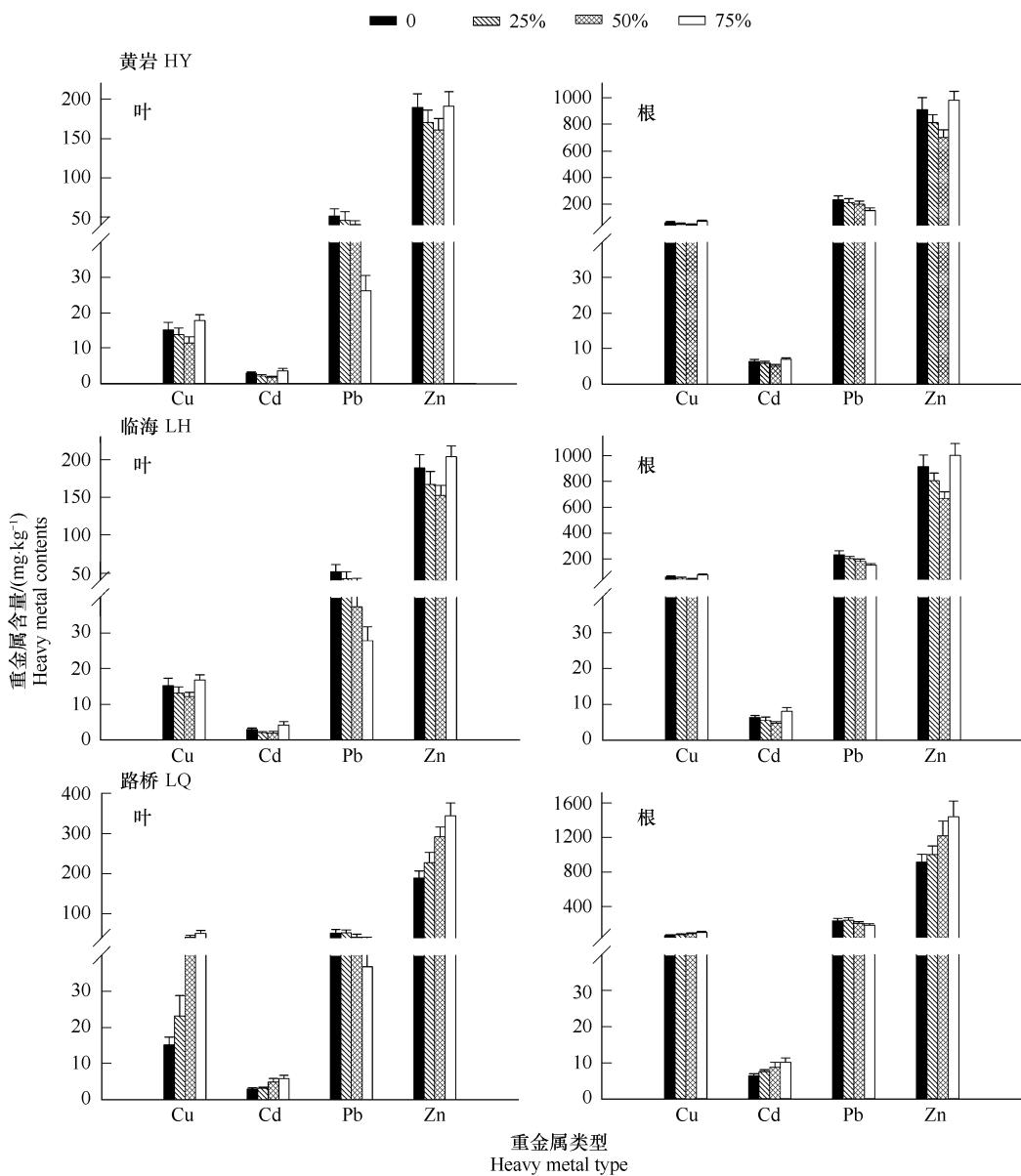


图2 不同污泥配比处理下香樟根和叶的重金属含量

Fig. 2 The heavy metal contents of leaves and roots of *C. camphora* at different ratios of adding sludges

下降并且显著低于对照,这表明高强度的氧化损害破坏了香樟的抗氧化酶系统<sup>[30]</sup>。

土壤中重金属的植物有效性是评估其对植物毒性的重要指标,而DTPA可提取量是反映土壤中植物的重金属可吸收总量的重要指标。3种污泥处理中,香樟根和叶的Pb含量均与混合基质中Pb的DTPA可提取量呈现明显的正相关( $P < 0.05$ ),但是,只有路桥污泥处理中,香樟根和叶的Cu、Cd和Zn含量与混合基质中Cu、Cd和Zn的DTPA可提取量呈现明显的正相关( $P < 0.05$ )。这可能由于添加污泥不仅增加了Cu、Cd和Zn含量,而且积累了大量的有机质,其对重金属的有效性同样有着重要的影响<sup>[31-32]</sup>。重金属的植物有效性受到有机质类型的影响,如果属于可溶性有机物质则可增加重金属植物有效性<sup>[31, 33]</sup>;如果属于有机酸等物质,则与重金属形成稳定的结合物<sup>[34-35]</sup>,明显抑制重金属植物有效性,而且有机酸与重金属结合并不影响重金属的DTPA可提取性<sup>[35]</sup>。因此,黄岩和临海污泥的有机质可能属于有机酸类有机物质,从而减少了Cu、Cd和Zn的生物有效性,导致香樟Cu、Cd和Zn含量的减少,但是Cu、Cd和Zn在混合基质中的DTPA可提取量并没有减少,导致其没有呈现正相关。香樟根积累了大量的重金属,同时限制其向地上部分运输,这与其他重金属

耐性植物的吸收转运特征相似<sup>[36]</sup>。

我国分布着许多由于采矿和冶炼而产生的废弃尾矿砂堆积地,用污泥对尾矿砂进行改良并加以植被修复,是一条经济可行的解决污泥处置难题的措施。本研究表明添加污泥提高了尾矿砂的有机质含量,同时改变了重金属的含量及其生物有效性。以香樟作为修复植物,黄岩和临海污泥在50%比例时,其对香樟的生物毒性最低,可以作为该地区主要的污泥改良修复技术。同时,污泥的有机物质组成可能对改良过程中重金属的生物有效性有着重要的影响,在未来的修复改良过程中应给予充分重视。

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