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铜尾矿坝不同恢复年限土壤理化性质和酶活性的特征

王瑞宏, 贾彤*, 曹苗文, 柴宝峰

(山西大学黄土高原研究所, 太原 030006)

摘要: 金属矿产资源开采使尾矿坝迅速升高, 矿区生态环境遭到严重破坏. 土壤理化性质和酶活性是衡量生态系统功能的重要指标, 可作为评价土壤恢复质量的重要因子. 以山西省垣曲县十八河铜尾矿区 9 个尾矿子坝作为研究对象, 分析不同恢复年限子坝土壤理化性质与土壤酶活性之间的关系. 结果表明, 不同恢复年限子坝的土壤理化性质差异较大, 随着恢复年限的增加, 土壤养分含量显著升高. 过氧化氢酶与碳氮比显著负相关, 脲酶与总氮含量和土壤含水量均显著正相关, 磷酸酶和蔗糖酶与土壤理化因子之间无显著关系. 土壤中铜含量随子坝恢复年限增加而逐渐累积, 砷和镉含量先增加后降低, 恢复后期逐渐达到稳定水平, 各子坝的土壤锌含量无显著差异. 为铜尾矿区的土壤生态系统恢复及退化机制的研究提供一定的生态学依据.

关键词: 恢复年限; 土壤酶; 土壤理化性质; 生态系统; 铜尾矿

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Characteristics of Soil Physicochemical Properties and Enzyme Activities over Different Reclaimed Years in a Copper Tailings Dam

WANG Rui-hong, JIA Tong*, CAO Miao-wen, CHAI Bao-feng

(Institute of Loess Plateau, Shanxi University, Taiyuan 030006, China)

Abstract: Mining for metal and mineral resources lead to the rapid rise of tailings dams and caused serious damage to the ecological environment of the mining area. Soil physicochemical characteristics and enzyme activities were important indexes for ecosystem functions, and they were also important factors in evaluating soil restoration qualities. We selected nine sub-dams of the Eighteen River copper tailings in Yuanqu County, and analyzed the relationship between soil physicochemical properties and soil enzyme activities. The results showed that there were great differences in soil physicochemical properties over different reclaimed years, and as the reclaimed years passed, soil nutrient contents significantly increased. There were significant negative correlations between catalase and the ratio of soil carbon and nitrogen, and urease was positively correlated to total nitrogen and soil moisture. Phosphatase and sucrose demonstrated no significant relationships with soil physicochemical factors. Copper content gradually accumulated in soil as the restoration period of sub-dams increased. Arsenic and cadmium content increased initially and then decreased before they gradually reached a stable level. In addition, there was no significant difference in zinc content among different sub-dams. Together, these results provide the ecological basis for further studies in soil ecosystem restoration and degradation mechanisms in copper tailings.

Key words: phytoremediation years; soil enzymes; soil physicochemical properties; ecosystem; copper tailing

金属矿产资源开发活动过程中, 大量重金属元素随着废石、尾砂、矿尘等直接进入矿区及其周边地区的土壤中, 成为矿区环境污染的主要来源^[1]. 开采形成的废弃物占用大量土地资源, 使原有的生态系统严重退化和恶化, 同时, 矿产资源的开发导致土壤团聚体破坏, 土壤肥力降低, 以及土壤理化特性和生物特性的恶化, 对矿区生态环境造成严重破坏^[2-4]. 中条山铜矿是北方最大的铜业生产基地, 年产矿量 400 万 t 以上, 为全国最大的非煤地下开采矿山, 该区域矿产资源以铜为主, 伴生有钴、钼、金、银等多种金属^[5]. 山西省垣曲县北方铜业铜矿峪矿十八河尾矿坝随排砂量的增加, 尾矿坝抬升速度逐渐加快, 尾矿的大量堆积对当地生态环境造成严重污染和破坏^[6], 因此, 对铜矿尾矿区进行合理且高效的生态修复具有重要意义. 土壤生态功能恢复是退化的陆地生态系统恢复和持续发展的关

键^[7,8], 因此, 生态修复既要考虑到植被类型, 也要考虑到恢复期间土壤肥力及微生物群落的动态变化特征. 目前主要通过植物-微生物协同修复模式^[9]及植被演替特征^[10]来评价矿区修复效果, 而针对修复年限长达 45 a 以上的尾矿坝土壤生态功能恢复的研究还相对较少.

土壤酶是高分子有机物催化分解的一类具有蛋白性质的生物催化剂, 它是评价土壤质量的一个重要指标, 其主要来源是植物根系分泌物、土壤微生物活动和动植物残体, 参与土壤中各种有机质的分解、合

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作者简介: 王瑞宏(1991~), 女, 硕士研究生, 主要研究方向为微生物生态学, E-mail: 190809451@qq.com

* 通信作者, E-mail: jiatong@sxu.edu.cn

成、转化及与无机物质的氧化还原等过程,其活性与土壤理化特性及土壤类型等关系密切,能很好地指示土壤有机物分解和元素循环等土壤生态变化^[11-14]. 土壤酶作为土壤生态系统中最活跃的有机成分之一,几乎参与了土壤中所有的生物化学过程,能够反映土壤生物化学过程的强度与方向. 同时,土壤酶活性与土壤理化性质密切相关,二者均为衡量生态系统功能的重要指标^[7],土壤酶能够活化土壤中各类元素的化合物,进而提升土壤有效养分、改善土壤质量;同时土壤理化性质提供了酶促反应的底物和环境,直接影响着酶活性^[7]. 关于土壤理化特征和酶活性的研究表明,山西煤矿修复区,不同植被方式对土壤理化特征和四种酶活性(蔗糖酶、过氧化氢酶、脲酶和多酚氧化酶)均有显著影响^[7]. 黄土丘陵区不同人工林型对土壤脲酶、蔗糖酶活性有明显的影响,土壤碱解氮、含水率和速效磷是影响脲酶活性的主要因素,而蔗糖酶活性主要受土壤有机碳含量的影响^[15]. 然而,除生态修复中不同植被类型对土壤会产生影响外,不同恢复年限也是影响矿区修复土壤环境的重要因素,土壤养分的积累和土壤酶活性的提高,对于土壤肥力的恢复具有重要意义.

因此,本文以北方铜业铜矿峪矿十八河铜尾矿坝作为研究对象,对重金属污染环境9个不同恢复年限尾矿坝的土壤进行研究:①探讨随子坝恢复年限的增加,土壤理化性质与酶活性的动态变化;②土壤各重金属含量随尾矿坝恢复年限的增加有何变化;③各个恢复年限子坝的土壤理化特征与酶活性的相互关系. 通过阐明9个不同恢复年限铜尾矿子坝土壤特性与酶活性发生变化的主要驱动因子,以为铜尾矿区的土壤生态系统恢复及退化机制提供生态学依据.



(a) 研究区全景; (b) 尾矿子坝分布和建坝时间

图 1 研究区样方设置

Fig. 1 Profile of the sampling plots of study area

1 材料与方法

1.1 试验地概况

十八河尾矿库(35°15'~35°17'N, 111°38'~111°39'E)于1969年建坝,位于选矿厂西南6 km的河谷中,由上游拦洪坝和下游尾矿坝及两侧山梁围成. 在矿山采选、矿石冶炼过程中有大量的含有多种重金属的尾矿废水和废渣产生,由于悬浮剂硫化钠、碱石灰以及其它有机试剂在浮选过程中的使用,使得尾矿中有机化合物、硫化物含量过高且呈现碱性. 选矿产生的废弃物以矿砂的形式逐年堆积于十八河尾矿坝,每3~5 a在原子坝基础上堆积形成一个新的子坝,并在矿砂表层覆盖30 cm厚客土,尾矿坝初期坝底标高486 m,坝顶标高509 m,现已堆筑14道子坝,堆积高度84 m,总坡比1:6. 该区域属大陆性季风气候,四季分明,春季干旱多风,夏季雨量集中,冬季少雪干燥. 年均降雨量为780 mm,年均气温14℃,无霜期大于200 d^[5].

1.2 研究方法

1.2.1 土壤样品采集

2015年7月,采用空间代替时间序列的方法,在恢复年限分别为4、8、15、19、23、27、31、35、46 a的9个不同子坝范围内设置样地(图1和表1),以子坝外围没有自然植被生长的裸地土壤为对照. 每个样地内以S型曲线在无植被覆盖区域进行五点取样,带回实验室,供室内分析测定土壤理化性质.

1.2.2 土壤理化性质和酶活性的测定

(1)土壤理化性质 土壤含水量%(SWC)用烘干法测定^[16],将盛有新鲜土样的铝盒置于105℃的烘箱中过夜烘干至恒质量;土壤pH(土水比为1:2.5)用点位法测定,称取10 g风干土样加入25 mL

无 CO₂ 蒸馏水, 搅拌均匀, 静置 30 min 后测定; 土壤全氮(TN)、全碳(TC)、全硫(TS)用 vario MACRO cube 元素分析仪(德国)测定^[17]; 硝态氮(NO₃⁻-N)、铵态氮(NH₄⁺-N)和亚硝态氮(NO₂⁻-N)含量用全自动间断化学分析仪(CleverChem 380, Germany)测定; 土壤粒度(PS)用激光衍射粒度分析仪(Matersizer 3000 Laser Diffraction Particle Size Analyzer, UK)测定; 样品重金属[砷(As)、铬(Cd)、铜(Cu)、铅(Pb)和锌(Zn)]含量用 ICP-AES(Icap6000, Thermo Fisher, UK)测定^[16]。

表 1 尾矿坝恢复年限概况

Table 1 Tailings dam and the major vegetation

| 子坝编号 | 建坝年份 | 恢复年限/a |
|------|------|--------|
| 516 | 1969 | 46 |
| 523 | 1981 | 35 |
| 525 | 1985 | 31 |
| 529 | 1989 | 27 |
| 531 | 1993 | 23 |
| 536 | 1997 | 19 |
| 540 | 2001 | 15 |
| 550 | 2009 | 8 |
| 560 | 2014 | 4 |

(2) 土壤酶活性测定 脲酶采用苯酚钠-次氯酸钠比色法, 其活性以 37℃ 下培养 24 h 后 1 g 土壤产生的 NH₃-N 毫克数表示; 蔗糖酶采用 3,5-二硝基水杨酸比色法, 其活性以 37℃ 下培养 24 h 后 1 g 土壤产生的葡萄糖的毫克数表示; H₂O₂ 酶采用 KMnO₄ 滴定法, 其活性以 20 min 内 1 g 土壤分解的过氧化氢的毫克数表示; 磷酸酶采用磷酸苯二钠比色法, 其活性以 37℃ 下培养 24 h 后 1 g 土壤中释放出的酚的质量表示^[17]。

1.3 数据处理

各样地变量之间的显著性差异均采用单因素方差分析(One-way analysis)和 Duncan 分析, 土壤理化性质与酶活性之间的关系采用 Pearson 相关分析, 数据统计分析在 Excel 2003、SPSS 13.0、R × 64 3.3.1、SigmaPlot 12.5 和 Canoco 5.0 软件上完成。

2 结果与分析

2.1 不同恢复年限中土壤理化性质变化

2.1.1 土壤 TC、TN、TS、C/N、pH、SWC 及 PS 的变化

从图 2 看出, 随恢复年限增加, 土壤 TC、TN、

TS 含量总体呈现上升趋势, 516 号(46 a)土壤 TC、TN、TS 含量均高于 CK。其中, 516 号子坝 TC、TN 和 TS 含量均高于其他各子坝[图 2(a)~2(c)], 表明随着子坝恢复年限增加, 可以逐渐提高土壤养分含量。不同恢复年限各子坝土壤 C/N 之间没有显著差异, 但均显著高于 CK[图 2(d)]。而土壤 pH 在不同子坝中变化不明显, 531 号(23 a)和 550 号(8 a)子坝土壤 pH 显著高于 525 号(31 a)子坝[图 2(e)]。除对照土壤外, 其他各子坝之间土壤含水量的差异不显著[图 2(f)]。540 号(15 a)子坝土壤粒径显著高于其他各个子坝[图 2(g)]。

2.1.2 土壤重金属含量的变化

土壤 As 和 Cd 含量随着恢复年限的增加呈现先增加后降低的趋势, 其中, 560 号(4 a)显著低于 536 号(19 a)子坝土壤 As 含量[图 3(a)]。类似地, 除 550 号子坝以外, 560 号(4 a)子坝土壤 Cd 含量显著低于其他各子坝土壤 Cd 含量[图 3(b)]。垣曲属大陆性季风气候, 四季分明, 春季干旱多风, 会将恢复初期子坝上的部分矿砂吹散至中间层各级子坝导致 As 和 Cd 较高。不同子坝中 Cu 的含量均高于 CK, 516 号子坝土壤中 Cu 含量达到最大(553.53 mg·L⁻¹), 且显著高于其他各个子坝[图 3(c)]。Pb 的含量在 560 号(4 a)中最低, 随着恢复时间的增加 Pb 含量也增加, 其值逐渐接近 CK 中 Pb 含量, 540 号(15 a)与 560 号(4 a)之间存在显著差异[图 3(d)]。CK 中 Zn 含量显著高于各不同恢复年限的子坝, 但各子坝之间 Zn 含量无显著差异[图 3(e)]。这表明恢复年限对土壤 Pb 影响不大, 而随着恢复年限的增加, 土壤中 Cu 含量却逐渐累积。

2.1.3 不同恢复年限中土壤铵态氮、硝态氮和亚硝态氮的变化

从图 4 可知, 在不同子坝中土壤铵态氮和硝态氮值均低于 CK, 各子坝之间, 516 号子坝土壤铵态氮含量最高(8.48 mg·kg⁻¹)。531 号子坝土壤硝态氮含量最大(6.65 mg·kg⁻¹), CK 土壤硝态氮含量显著高于各个子坝, 而亚硝态氮总体变化不明显, 且不同恢复年限子坝之间, 铵态氮、硝态氮和亚硝态氮均无显著差异。

2.2 土壤酶活性与土壤理化因子之间的相关性分析

通过对土壤中过氧化氢酶、磷酸酶、脲酶和蔗糖酶与土壤理化因子做相关分析, 从图 5 可以看出, 过氧化氢酶与 C/N 呈极显著负相关关系, 与其它理化因子之间均未达到显著水平; 脲酶与 TN 呈显著

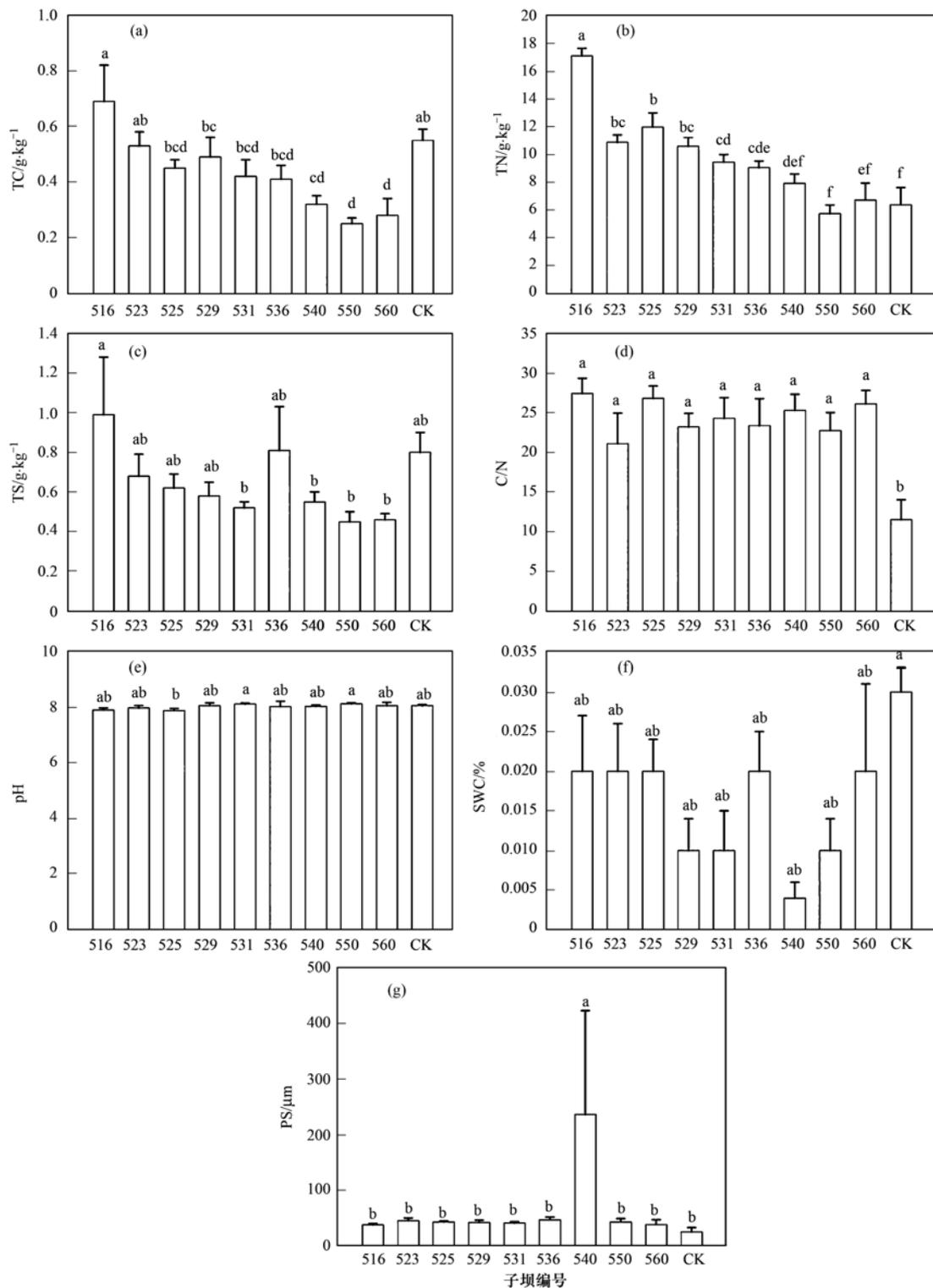


图2 不同恢复年限子坝土壤理化性质的变化

Fig. 2 Soil physical and chemical properties of sub-dams with different reclaimed years

正相关关系,与土壤含水量呈极显著正相关关系;磷酸酶和蔗糖酶与所有理化因子之间均无显著关系;4种酶活性与土壤各个重金属含量均无显著相关(表2).

2.3 土壤性能分析

由图6(a)可得,550、540和531号的土壤性能比较接近,且pH是影响土壤性能的主要因素;而523号主要受到TC影响,525号和536号土壤性能

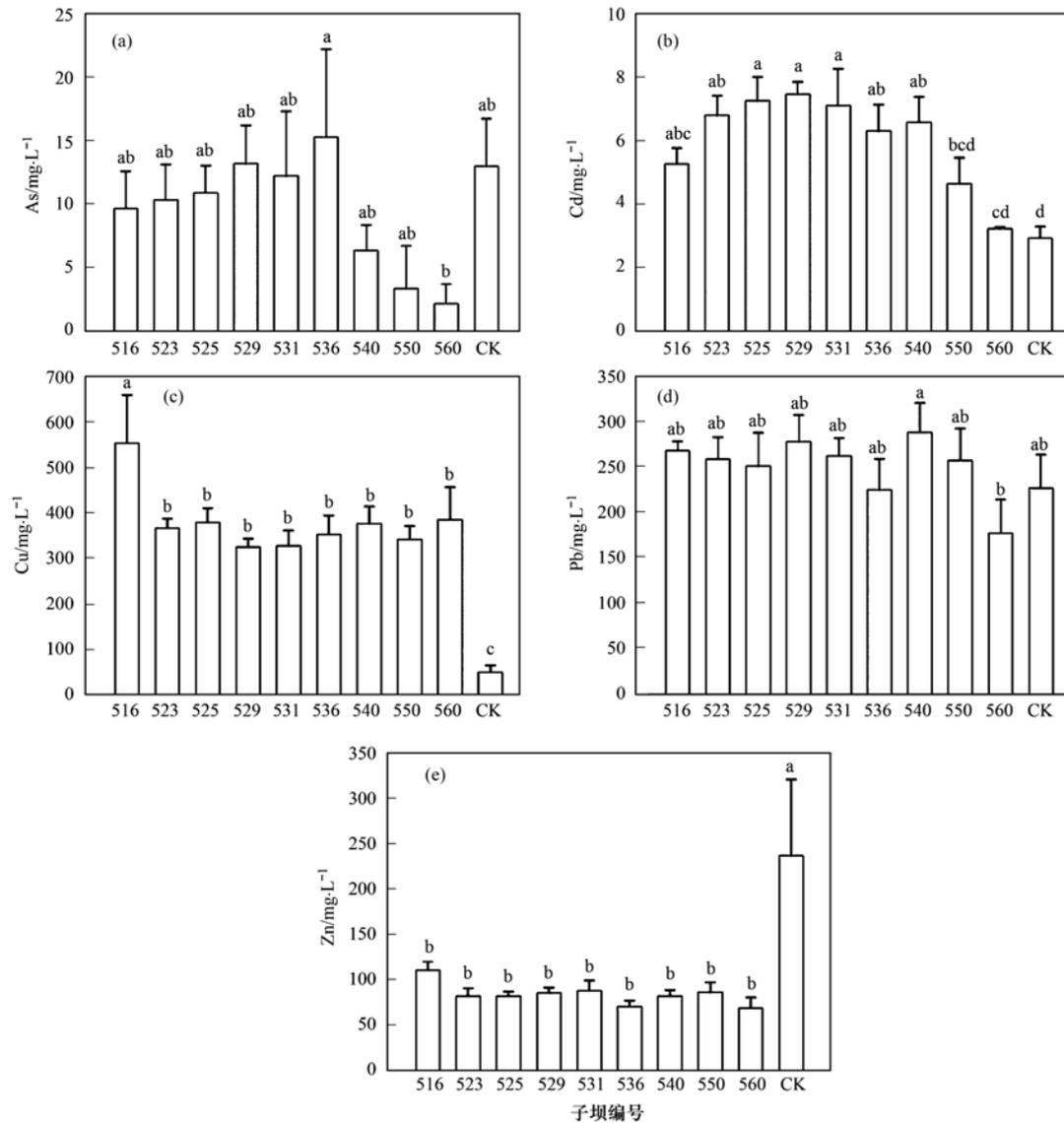


图3 不同恢复年限中土壤重金属含量的变化

Fig. 3 Changes in soil heavy metal contents in different reclaimed years

表2 土壤酶活性与理化因子之间的相关性

Table 2 Correlation between soil enzyme activity and physical and chemical factors

| 理化因子 | 过氧化氢酶 /mL·(g·20 min) ⁻¹ | 磷酸酶 /mg·(g·24 h) ⁻¹ | 脲酶 /mg·(g·24 h) ⁻¹ | 蔗糖酶 /mg·(g·24 h) ⁻¹ |
|---------------------------------|---------------------------------------|-----------------------------------|----------------------------------|-----------------------------------|
| NH ₄ ⁺ -N | -0.003 | 0.092 | -0.212 | 0.146 |
| NO ₃ ⁻ -N | 0.164 | 0.122 | 0.091 | -0.149 |
| NO ₂ ⁻ -N | -0.080 | -0.175 | -0.132 | -0.156 |
| TC | -0.082 | 0.04 | 0.148 | 0.046 |
| TN | 0.224 | 0.098 | 0.305* | -0.114 |
| TS | 0.015 | 0.030 | -0.091 | 0.127 |
| C/N | -0.423** | -0.090 | -0.162 | 0.210 |
| SWC | 0.224 | -0.172 | 0.442** | -0.151 |
| pH | 0.119 | 0.092 | 0.118 | -0.152 |
| PS | -0.175 | -0.014 | -0.174 | 0.099 |
| As | 0.078 | 0.028 | 0.095 | -0.152 |
| Cd | 0.033 | 0.245 | 0.059 | -0.125 |
| Cu | -0.278 | -0.079 | -0.167 | 0.198 |
| Pb | -0.027 | -0.084 | 0.105 | -0.234 |
| Zn | -0.021 | -0.021 | -0.157 | 0.090 |

1) * $P < 0.05$ 表示差异显著, ** $P < 0.01$ 表示差异极显著

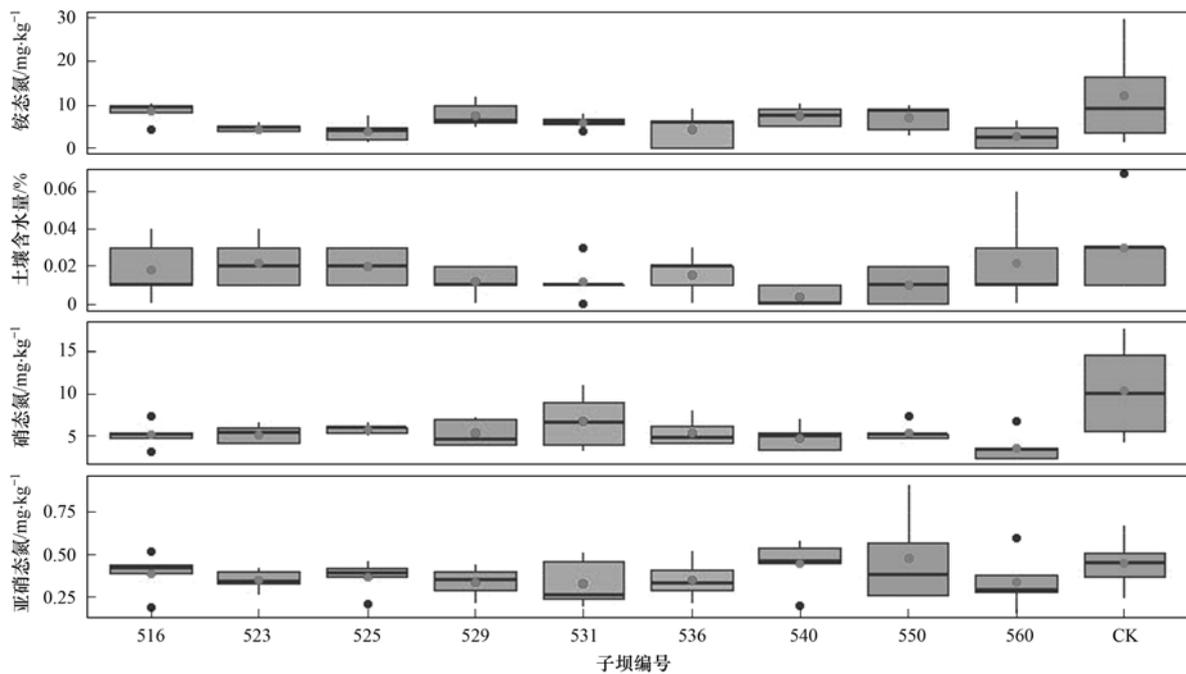


图 4 不同恢复年限中土壤铵态氮、硝态氮和亚硝态氮的变化

Fig. 4 Ammonium nitrogen, nitrate nitrogen, and nitrite nitrogen changes in soil over different reclamation years

较接近,土壤中 C/N 对其影响较大;但 CK 和 560 号子坝受土壤理化因子的影响不大;由图 6(b) 可得,516、523、525、529、531 和 536 号子坝受到重金属 As、Cd、Cu 和 Pb 影响较大,而 CK 主要受重金属 Zn 影响,这表明随着子坝恢复年限增加,土壤重金属含量受到的影响逐渐变小。

3 讨论

土壤理化性质是表征土壤质量的重要指标,本研究结果表明,土壤养分随着尾矿恢复年限的增加逐渐升高,这与 Zhao 等^[18]的研究结果一致,主要由于土壤恢复过程中,地表枯落物的增加可以使土壤中腐殖质增加,从而有效改善土壤理化性质^[19]。有研究表明,在煤矿塌陷区随着复垦年限的增加,土壤孔隙度和颗粒分形维数增大^[20]。本研究中,恢复 15 a 子坝土壤粒径达到最大值,而其他各个年限土壤粒径之间没有显著差异,这可能由于具体恢复年限不同,且铜尾矿坝土壤组成以矿砂为主,因而除 540 号子坝外,各梯度的土壤粒径并没有明显差异。该研究中土壤呈碱性,随恢复年限增加,土壤 pH 有下降趋势,这与杨艳芳等^[21]的研究结果一致,可能原因是地表凋落物增多及根部微生物活动加强,导致腐殖酸和有机酸增加^[16]。重金属在土壤中的存在形态

并不是一成不变,而是随着土壤环境的变化(如土壤酸碱度、有机质含量、颗粒组分、氧化还原电位等)而改变^[22]。本研究中,土壤中 As 和 Cd 的含量随着恢复年限的增加呈现先增加后降低的趋势,这可能与重金属的存在形态有关。有研究表明,大多数元素总量的增加主要引起稳定态(如残渣态)含量增加,而活动态(水溶态和离子交换态)受总量的影响较小^[23, 24]。

本研究中,脲酶和过氧化氢酶因土壤理化因子不同而有显著变化(图 5)。土壤脲酶是好气性水解酶,其活性与土壤氮素相关^[25],且脲酶与土壤含水量及氮含量显著正相关,这与李君剑等研究结果一致^[19],可能原因是脲酶将酰胺态有机氮化物转化为植物可以直接吸收利用的无机氮化物,其活性反映土壤供氮能力与水平,因此,当土壤 TN 含量升高时,土壤中脲酶活性也提高^[26, 27]。有研究发现,在 pH 为 3.1 ~ 7.1 的土壤中,脲酶活性与 pH 间呈显著的正相关^[28],但也有研究表明 pH 与脲酶活性之间并不相关^[2],这与本研究结果一致,可能由于铜尾矿子坝土壤 pH 大于 7.9 的原因^[29]。此外,脲酶与土壤含水量呈极显著正相关,这与牛世全等^[27]的研究结果一致,土壤水分增加为各种酶促反应提供反应条件与场所,使土壤酶活性升高,其活性随土壤含水量升高而增强^[30~32]。过氧化氢酶用作表示土

壤氧化强度,它广泛存在于土壤和生物体内,本研究
发现土壤过氧化氢酶与土壤 C/N 有显著负相关性,
可能原因是在恢复过程中植被通过根系分泌物和残

体向土壤提供碳和氮,影响土壤有机质的输入,从而
影响土壤结构和理化性质,使过氧化氢酶的活性降
低^[32~34]。

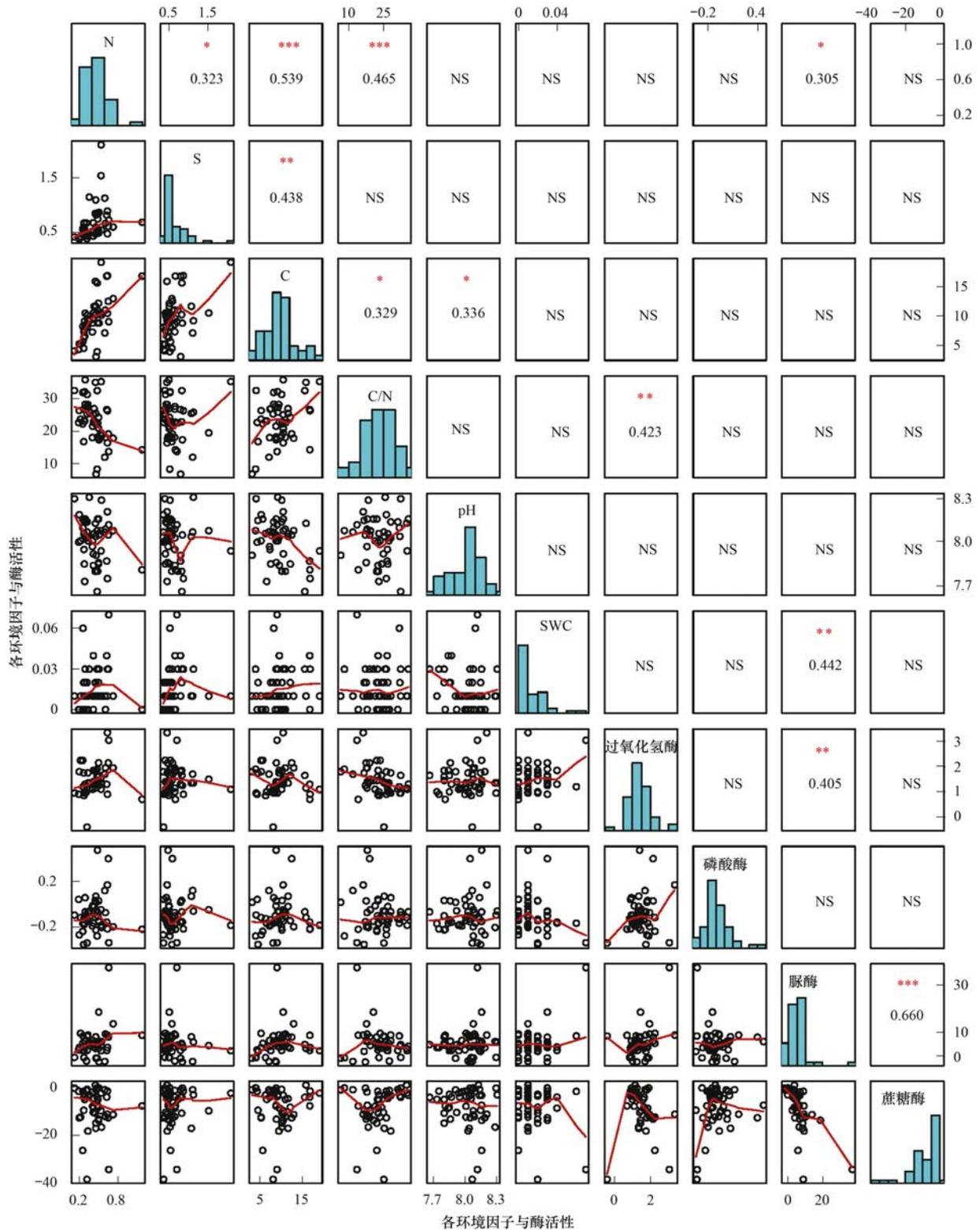
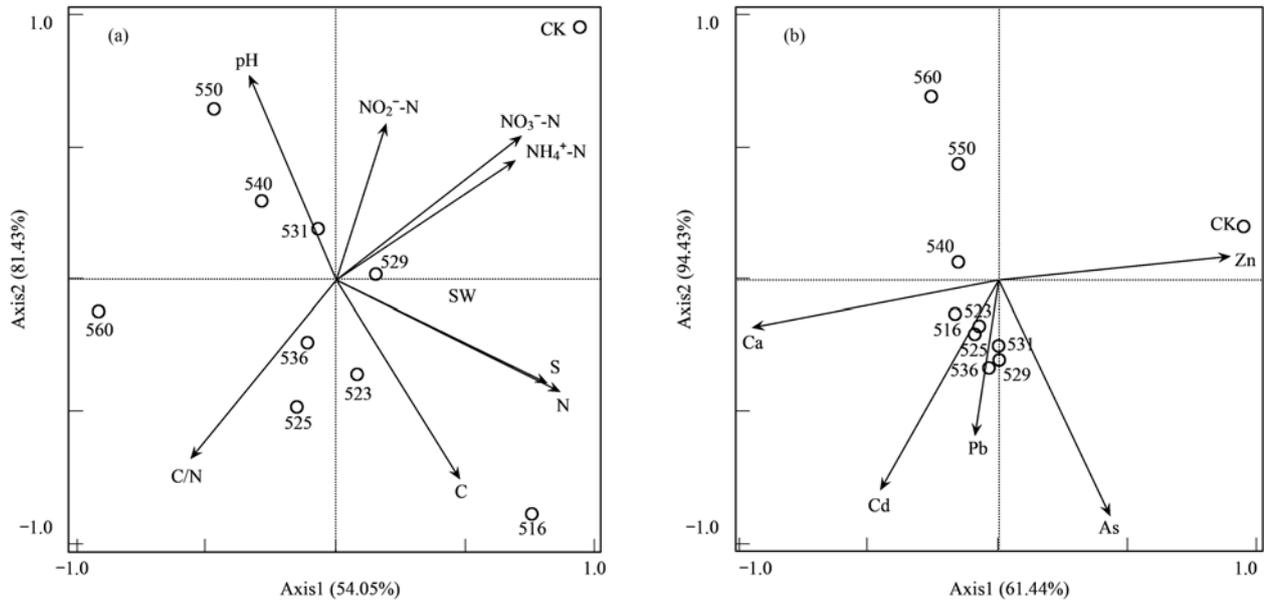


图5 土壤酶活性与环境因子之间的相关性

Fig. 5 Correlation between soil enzyme activities and environmental factors



(a) 不同子坝与环境因子; (b) 不同子坝与重金属

图 6 不同恢复年限的子坝与土壤理化因子之间的 PCA 分析

Fig. 6 Principal component analysis (PCA) between different dams and physicochemical factors

4 结论

(1) 不同恢复年限子坝土壤理化性质差异较大,随着恢复年限增加,土壤养分含量显著升高。

(2) 土壤 Cu 含量随着子坝恢复年限增加而逐渐累积,土壤中 As 和 Cd 含量先增加后降低,恢复后期逐渐达到稳定水平,各子坝土壤 Zn 含量无显著差异。

(3) 过氧化氢酶与碳氮比为极显著负相关关系,脲酶与总氮含量显著正相关关系,脲酶与土壤含水量为极显著正相关关系,磷酸酶和蔗糖酶与土壤理化因子之间无显著关系。

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