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施用石灰与钙蒙脱石对酸性土壤硝化动力学过程的影响

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摘要: 土壤酸化是粘土矿物缓慢风化的自然过程,但近年来随着人类高强度的农业利用,土壤酸化现象逐渐加剧,而铵态氮肥的硝化作用是土壤酸化的主要贡献者之一。传统的施用石灰改良酸性土壤,常常会有反酸现象,并可能导致土壤板结。蒙脱石是碱性或中性土壤的主要粘土矿物组分,而在土壤酸化的过程中,蒙脱石被进一步风化掉。本文通过室内模拟实验,采用硝化动力学拟合及对净硝化速率的计算,分别研究了蒙脱石(Ca-M)和石灰(Ca-OH)对酸性黄壤硝化作用的影响。结果表明:酸性黄壤添加石灰或蒙脱石后,土壤均发生了显著的硝化作用,且硝化过程符合一级动力学模型 $N_{NO_3} = N_0 + N_p(1 - \exp(-k_1 t))$ ($P < 0.001$)。Ca-OH 处理土壤样品的净硝化速率($3.429 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$)显著大于 Ca-M 处理($2.381 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$);Ca-OH 处理土壤样品的潜在硝化速率(V_p)和平均硝化速率(V_a)在 pH 值 5.7 和 6.2 时分别为 $6.42, 8.58 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ 和 $2.71, 3.87 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, 均显著大于钙基蒙脱石处理(pH 值 5.7 和 6.2 时分别为 $3.40, 4.56 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ 和 $2.36, 3.04 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{day}^{-1}$)。结果表明采用石灰改良酸性土壤发生复酸化现象的可能性及程度大于钙基蒙脱石,本研究为酸性土壤改良提供了新的参考。

关键词: 土壤酸化;净硝化速率;酸性土壤改良

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Effects of Applying Lime and Calcium Montmorillonite on Nitrification Dynamics in Acidic Soil

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Abstract: Soil acidification is known as a natural and slow process along with clay mineral weathering. In recent years however, with intensive soil utilization in agriculture, soil acidification has increased dramatically and nitrification of ammonium nitrogen fertilizer is one of the main contributors to soil acidification. Lime application is the traditional practice to improve acidic soils but it often makes the soil acidic again leading to soil compaction in most cases. Montmorillonite is the main clay mineral component of alkaline or neutral soils, more so it is known to undergo further weathering processes during soil acidification. The laboratory-based incubations were used in this study, and nitrification was measured while kinetic curves were fitted to check the effects of decreasing soil acidity by lime (Ca-OH) and montmorillonite (Ca-M) on nitrification of the acidic soil. The results showed that significant nitrification was observed both in Ca-OH and Ca-M treatments, and the nitrification process was fitted in the first-order kinetic model, $N_{NO_3} = N_0 + N_p(1 - \exp(-k_1 t))$ ($P < 0.001$). The net nitrification rate of Ca-OH treatment ($3.429 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$) was significantly higher than that of Ca-M treatment ($2.381 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$). The potential nitrification rate (V_p) were $6.42, 8.58 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ at pH 5.7 and 6.2 respectively, and the average nitrification rate (V_a) of Ca-OH treatment were $2.71, 3.88 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ respectively, which were significantly greater than those of Ca-M treatment (V_p were $3.40, 4.56 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ and V_a were $2.36, 3.04 \text{ mg N} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$ at pH 5.7 and 6.2 respectively). Therefore the net nitrification rate, potential nitrification rate (V_p)

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and average nitrification rate (V_a) of Ca-OH treatment were significantly higher than that of Ca-M treatment, suggesting that the possibility and degree of soil reacidification by using lime to improve acidic soil is greater than using calcium montmorillonite. This study will provide a new reference for the improvement of acid soils.

Keywords: soil acidification; net nitrification rates; acidic soil improvement

土壤酸化是粘土矿物自然风化的结果^[1-3],自然状况下土壤 pH 下降 1 个单位,需要上千年或万年^[4]。但在强烈的人为活动影响下,如酸沉降及不当的农业措施的影响下,该过程可能缩短至短短数十年。近年来,高强度的农业利用尤其是氮肥的大量施用加剧了土壤的酸化^[5]。在湖北地区,桑农为了提高桑叶的产量,其氮肥用量甚至达到了 $454 \text{ kg} \cdot \text{hm}^{-2} \cdot \text{a}^{-1}$,超出 N 肥适宜剂量($375 \text{ kg} \cdot \text{hm}^{-2} \cdot \text{a}^{-1}$)21%^[6]。不同种类化肥对酸化影响不一,佟德利等^[7]发现 $\text{CO}(\text{NH}_2)_2$ 、 $(\text{NH}_4)_2\text{SO}_4$ 随施用量越大,pH 值下降越大,分别较对照处理下降 $0.11 \sim 0.43$ 、 $0.02 \sim 0.36$ 个 pH 单位,而 NH_4HCO_3 则是在 N $150 \text{ mg} \cdot \text{kg}^{-1}$ 时 pH 下降最大,N $300 \text{ mg} \cdot \text{kg}^{-1}$ 时 pH 下降最小。化肥施用导致土壤酸化的原因除氮素形态外,主要是促进了硝化作用的发生及硝酸盐的淋溶^[8],因为氧化 1 mol NH_4^+ 为 NO_3^- 会向环境中释放双倍的 H^+ ^[9]。土壤酸化不仅造成农产品减产^[10],还会导致钙、镁、钾等营养元素淋失而出现相关缺素病症,使得农产品质量下降,并威胁农业生态环境和全球生态环境。

土壤中发生的硝化过程是对 pH 高度敏感的典型过程^[11-12]。20 世纪初以来,关于酸性土壤中的硝化过程的认识随着研究的深入发生了显著变化。由开始的认为不存在^[13-14]到证明存在硝化作用^[15-19],至 20 世纪末,证明了酸性土壤中也可能有强烈的硝化作用存在^[20-21]。硝化作用会引起净质子的积累,进而造成土壤的酸化程度加深,还会增加植物根系金属中毒和缺乏某种元素导致机能不全的风险。因此,了解酸性土壤中氮循环过程的动力学及其原理机制是十分必要的,能够为控制微生物和植物合理利用氮源、减轻温室气体(N_2O)排放和硝酸盐淋溶提供理论方面的有效策略。

对酸化土壤的改良研究已有很多,如石灰、石膏及其相关工业副产品等^[22-23],包括近年来研究较多的生物质炭^[24-25]以及猪粪等有机肥^[26],但畜禽粪肥^[27]、部分工业副产品^[28]有引入重金属的潜在危险。多数工业副产品单施并不能达到较好改良效果^[22,29],如粉煤灰、碱渣、赤泥和磷石膏在与石灰混合施用时效

果最佳,既能提供足够的交换性钙,也能防止交换性镁、钾、钠等的流失^[23];而有机粪肥、生物质炭等则更多的是对土壤养分的改善。蒙脱石作为风化很弱的土壤次生粘土矿物,理论上能达到从根本上改善和缓解土壤酸化的目的。本文研究了石灰和蒙脱石对酸性黄壤硝化作用的影响,比较了两者对酸性土壤的改良效果,为以后的研究提供参考。

1 材料与方法

1.1 供试样品

1.1.1 供试土壤及蒙脱石

本实验所用黄壤采自重庆市北碚区缙云山自然保护区,该区为典型的酸性黄壤^[30],土壤质地为沙质壤土,成土母质为中生代三叠纪须家河组(T_3X_i)长石石英砂^[31]。供试土壤的基本性质:pH 值 5.2,有机质含量 $38.65 \text{ g} \cdot \text{kg}^{-1}$,全 N 含量 $1.66 \text{ g} \cdot \text{kg}^{-1}$,铵态氮含量为 $8.94 \text{ mg} \cdot \text{kg}^{-1}$,硝态氮为 $6.52 \text{ mg} \cdot \text{kg}^{-1}$,亚硝态氮为 $5.43 \text{ mg} \cdot \text{kg}^{-1}$ 。

研究对象为缙云山区约 800 m 处的土壤。该区人为影响因素较少,为减轻地形、母质等因素的影响,采样时选取了较平整的 3 个地点的土壤并充分混合,这 3 点处土壤地貌、颜色、质地均基本一致。

本实验所使用蒙脱石为提纯 Ca-蒙脱石,含量 98%,pH 值 7.58;氢氧化钙为 AR 试剂。

1.1.2 土壤样品制备及处理

土壤混合均匀风干后,去除植物落叶、根系等杂质后分别过 1、2 mm 筛备用。保留原样土壤,另取 4 份土壤样品分别用氢氧化钙(Ca-OH)和蒙脱石(Ca-M)将其 pH 值调至 5.7 和 6.2,分别记录其用量;而后根据调至一定 pH 值时氢氧化钙及蒙脱石添加量做培养实验,试验处理为:(1) 土壤原样 pH 值 5.2;(2)氢氧化钙调土壤 pH 值至 5.7;(3)氢氧化钙调土壤 pH 值至 6.2;(4)蒙脱石调土壤 pH 值至 5.7;(5)蒙脱石调土壤 pH 值至 6.2;每个处理 3 次重复。

试验土壤经 28 °C、20%含水量下暗箱预培养 7 d 后,添加氮源硫酸铵,添加量 $100 \text{ mg NH}_4^+ \cdot \text{N} \cdot \text{kg}^{-1}$ 干土,于 20%持水量、28 °C 下培养 28 d,每 7 d 采样 1

次,测定土壤pH值、硝态氮、氨氮。

1.2 测定方法

土壤pH值用1:2.5土水比电位法测定,总氮使用开氏消煮法测定,有机质使用重铬酸钾容量法测定^[32];氨氮使用靛酚蓝比色法测定,硝态氮用紫外分光光度法测定,亚硝酸盐使用氨基苯磺酸-(L-萘)乙二胺比色法测定^[33]。

1.3 土壤净硝化率的测定

培养期间,土壤的净硝化率通过每周测定土壤样品中NO₃-N浓度随培养时间变化的速率来表示,即净硝化速率=(培养第n d时硝态氮含量-第0 d时硝态氮含量)/培养天数。

1.4 数据处理

实验基础数据采用Microsoft Excel 2016进行处理后,利用Origin 8.6、SigmaPlot12.0进行数据分析、作图。

2 结果与分析

2.1 培养期间硝化动力学

不同处理土壤样品在20%含水量、28℃下培养28 d的净NO₃-N累积变化情况如图1所示。0~28 d,对照组和所有处理组的NO₃-N累积量均增加,且从同一时间来看,pH值越高,累积量越高。相同pH(pH值5.7和6.2)的土壤样品,不同处理其NO₃-N累积有差异,Ca-OH处理均比Ca-M处理累积量高;且其差异随pH值升高而增大,即高pH值6.2处理较低pH值5.7差异明显。土壤pH值提高后,第0~21 d NO₃-N累积增长迅速,且在21 d时pH 6.2(Ca-OH)处理观察到最大净硝化量(107.5 mg·kg⁻¹),最低为对照组土壤(54.5 mg·kg⁻¹)。从培养第0 d开始pH 6.2(Ca-OH)NO₃-N累积量较高,且这种趋势一直保持到培养结束($P<0.05$)。培养结束时,对照组和Ca-OH处理组土壤中的NO₃-N积累量不再增加,而Ca-M

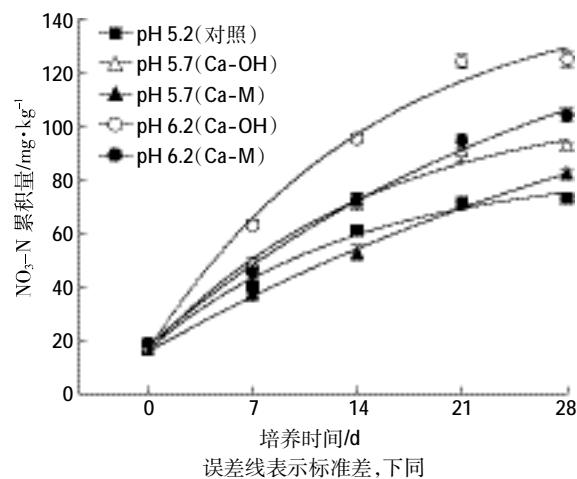


图1 培养期间NO₃累积与动力学拟合曲线

Figure 1 NO₃ accumulation and kinetic curve fitting during 28 days incubation at 28 °C with soil moisture of 20%

处理组土壤样品仍保持一定的增长趋势。

所有Ca-OH和Ca-M处理及对照组土壤,硝化模式均相似。回归分析表明,所有的处理其NO₃积累量随时间变化关系,符合一级动力学模型:

$$N_{NO_3} = N_0 + N_p (1 - \exp(-k_1 t)) \quad (P < 0.001)$$

硝化作用的模拟参数列于表1中。模拟硝化潜势介于72~184 mg·kg⁻¹之间,且最高值在pH 6.2(Ca-M)处理时达到。相同处理样品,硝化潜势(N_p)、潜在硝化速率(V_p)和平均硝化速率(V_a)均随pH升高而增大;相同pH(pH同为5.7和6.2时)不同处理土壤样品, Ca-M处理硝化潜势(N_p)均比Ca-OH处理高,但潜在硝化速率(V_p)和平均硝化速率(V_a),Ca-OH处理均比Ca-M处理高。

2.2 净硝化速率

整个培养过程中,前21 d的净NO₃-N积累增长显著,21~28 d积累不显著,表明硝化反应主要集中于前21 d,图2所示为培养21 d时的净硝化速率。当将酸性土壤调至同一pH值(5.7)时,Ca-OH处理组净

表1 培养期间硝态氮累积的一级动力学参数

Table 1 Parameters of first-order kinetics fitting nitrate-N accumulation during 28 days of incubation

处理	pH值	N _p /mg·kg ⁻¹	k ₁ /d ⁻¹	R ²	V _p /mg N·kg ⁻¹ ·d ⁻¹	V _a /mg N·kg ⁻¹ ·d ⁻¹
对照	5.2	72.0±6.3	0.068 6±0.014	0.991*	4.94	2.00
Ca-OH	5.7	97.2±7.0	0.066±0.011	0.994*	6.42	2.71
Ca-M	5.7	136±2.79	0.025±0.007	0.997*	3.40	2.36
Ca-OH	6.2	149±1.76	0.057 6±0.014	0.990*	8.58	3.87
Ca-M	6.2	184±7.43	0.024 8±0.009	0.995*	4.56	3.04

注:N_p硝化潜势;k₁一级反应速率常数;V_p由一级动力学V_p=k₁×N_p得到的潜在硝化速率;V_a平均净硝化率。数值为平均值±标准差;“*”表示P<0.001。

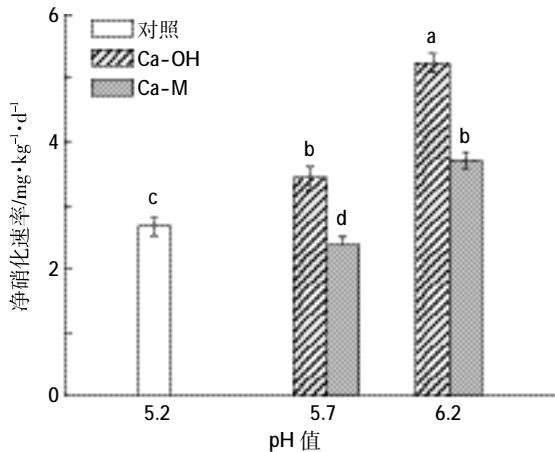
误差线表示标准差,不同字母代表显著性差异($n=3, P<0.05$)

图 2 培养 21 d 时 Ca-OH、Ca-M 处理

净硝化速率变化情况

Figure 2 Net nitrification rate changes with Ca-OH and Ca-M treated when 21 days incubated at 28 °C with soil moisture of 20%

硝化速率 ($3.429 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$) 明显大于 Ca-M 处理 ($2.381 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$) 组, 且 pH 值越高净硝化速率差异越明显 (pH 6.2 时, Ca-OH 处理为 $5.238 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$, Ca-M 为 $3.714 \text{ mg} \cdot \text{kg}^{-1} \cdot \text{d}^{-1}$); 对于蒙脱石提高酸性土壤的 pH 值后, 较空白对照相比, 其净硝化速率升高不明显, 甚至在 pH 5.7 时略低于空白对照组。净硝化速率越大说明复酸化现象越有可能发生。因此实验结果表明, 氢氧化钙处理复酸化可能性较钙蒙脱石大, 即用石灰等作改良剂施加到酸性土壤中后, 土壤很大程度上会发生复酸化, 这与孟赐福等^[34]研究结果一致。另外, 随着 pH 值的升高, 净硝化速率增加, 表明净硝化速率与土壤 pH 值正相关。

2.3 pH 值变化情况

图 3 为 pH 值随培养时间的变化情况。随着培养的进行, pH 值均下降, 且 pH 越高, 下降速度越快, 表明 pH 值越高, 越有利于硝化作用的进行, 与前人^[35-36]研究结果一致; 第二周至第三周下降显著, 尤其是 Ca-M 处理, 比 Ca-OH 处理更显著, 但最终均趋于一定值, 表明酸性土壤中酸度有底值, 土壤酸度降低至一定程度, 其 pH 值稳定。但总体上, Ca-M 处理较 Ca-OH 处理土壤酸度略低。

3 讨论

土壤发育程度不同, 则形成的土壤年龄、粘土矿物含量不同, 土壤粘土矿物受成土因素的影响, 是风化和成土作用的产物。幼年土一般风化程度较弱, 主

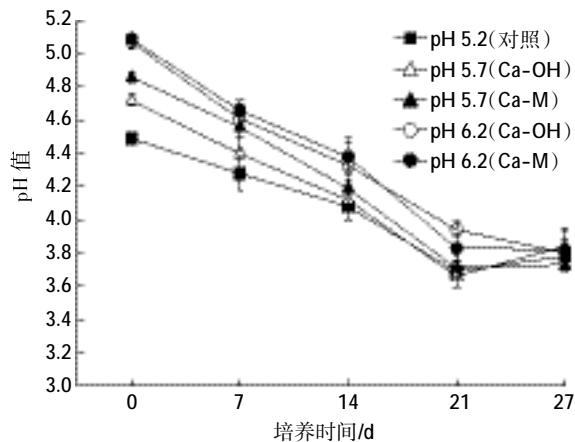


图 3 培养期间 Ca-OH、Ca-M 处理 pH 值变化情况

Figure 3 pH changes during 28 days incubation with Ca-OH, Ca-M treated

要为荒漠区土壤, 受水分限制而不易风化, 因此含较多矿物母质; 成年土或壮年土风化程度中等, 如黑土、褐土等, 常含较多的石英、伊利石、蛭石、蒙脱石; 而老年土为湿热区高风化强淋溶的土壤, 包括红壤、砖红壤, 原生矿物基本风化消失, 土壤粘粒矿物则主要为高岭石、氧化铁矿物等。长期的高风化强淋溶, 导致土壤酸化加速, 早期形成的碱性钙质土渐渐演变成中性土, 甚至酸性或强酸性土壤。酸化是土壤退化重要的一方面。土壤酸化是指土壤中氢离子增加或是土壤酸度由低变高的过程, 它是一个持续不断发生的自然过程, 但自然条件下的酸化进程非常缓慢, 需要上万年, 而在人为活动的影响下, 酸化进程大大加剧。土壤酸化会导致土壤中营养元素流失, 威胁农业生态环境, 降低土壤肥力水平, 造成农作物减产以及农产品品质下降; 同时还会增加金属、重金属化合物在土壤中的溶解度^[37-38], 从而加大重金属元素对环境的危害。

与中性或碱性土壤相比, 酸性土壤更不利于微生物的生长, 有些微生物在酸性土壤中甚至停止增长, 但仍有耐酸或嗜酸菌在其中具有生物活性, 如化能无机营养 AOA^[39-40], 它们在酸性土壤中能进行强烈的硝化作用。硝化作用本身就会产生氢离子, 而在酸性土壤中, 产生的氢离子不能被及时且很好地中和, 导致土壤酸度加大, 同时表现出更大的铝毒危害, 因而酸性土壤中的硝化作用的发生可能会产生比中性和碱性土壤更大的危害。从作物生产、食品安全、环境健康等方面考虑, 酸性土壤的改善都极为迫切。

传统上, 常用石灰来改良酸性土壤, 但石灰只能从表面上中和酸性土壤中的氢离子, 且石灰的长期

施用会使土壤发生复酸化、板结等现象^[34,41]。土壤酸化会促使矿物风化加速,促进土壤 2:1 型粘土矿物向 1:1 型高岭石转化,导致土壤阳离子交换量(CEC)减小^[2,42],而蒙脱石是粘土矿物的一种,其含量与土壤性能有关且呈正相关^[43],具有特定的 2:1 型晶体结构,pH 在 8 左右,能提高酸性土壤 pH 值,降低酸度。其次,蒙脱石不仅有较好的阳离子交换性、强吸附性(因其巨大的比表面积)、分散性等,还保水保肥,施入到土壤中,能很好地保持土壤中的氮素养分,减少氮素淋溶损失^[44],从而减缓土壤酸化。粘土矿物作为土壤的“骨架”,因此,通过直接改善该“骨架”,能改善土壤的表面电化学性质^[45-46],达到从根本上改良酸性土壤的目的,故从长远考虑,施加蒙脱石效果比石灰好。从本实验的硝化动力学模拟来看,Ca-M 处理的硝化潜势较 Ca-OH 处理强(表 1),也表明钙基蒙脱石较氢氧化钙对 pH 值提高更持久有效;就净硝化速率而言,Ca-M 处理明显比 Ca-OH 处理低(图 2)。因此用蒙脱石改良酸性土壤不但具有可行性且无复酸化现象发生或无明显复酸化现象发生,能持续有效改善酸性土壤。

本实验研究结果显示,两处理的潜在硝化(V_p)及平均硝化速率(V_a)都随 pH 值的升高而升高(表 1),也表明 pH 值过低不利于硝化微生物的生长及代谢;Ca-M,Ca-OH 处理都表现为 pH 值高(6.2)的土壤样品其净 NO₃⁻积累量较低 pH 值(5.7)的高(图 1),说明酸性土壤酸度降低,pH 值升高,对硝化作用有促进作用。其原因在于酸性土壤 pH 值升高后,土壤的理化性质及电化学性质发生改变,从而影响到其中微生物数量、种类和微生物酶活性的变化,包括硝化微生物^[46-48]。氨氧化古菌(AOA)和氨氧化细菌(AOB)是具有硝化作用的主要微生物,大多数研究表明酸性环境较中性和碱性环境更不利于硝化微生物维持较高数量和活性,土壤 pH 值升高后,AOA、AOB 的数量和活性均显著增加^[48-51],从而导致土壤中硝化作用显著增强。

酸性土壤改良的理论实验及田间试验并不缺乏,但利用蒙脱石改良酸性土壤的研究较少,缺乏其应用到实际的作物生产效果,以及土壤改良效果如土壤结构、交换性酸、硝化作用等的研究,尤其欠缺长期的田间试验的研究。因此蒙脱石改良的后期实际效果及与土壤充分结合的机理将是今后研究的重点,从而为作物生产及酸性土壤改良提供更切实有效的依据,尤其对从本质上改良酸性土壤提出重要参考。

4 结论

(1)酸性黄壤添加石灰或蒙脱石改良后的土壤均发生了显著的硝化作用,且硝化过程符合一级动力学模型:

$$N_{NO_3} = N_0 + N_p (1 - \exp(-k_1 t)) \quad (P < 0.001)$$

(2)石灰处理土壤样品的净硝化速率、潜在硝化速率(V_p)和平均硝化速率(V_a)均显著大于钙基蒙脱石处理,说明采用石灰改良酸性土壤发生复酸化现象的可能性及程度大于钙基蒙脱石。

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