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外源铵态氮对典型耕作土壤冻结过程中 N_2O 排放的影响

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摘要:【目的】农业土壤是 N_2O 的主要排放源,国内以往研究多集中在外源养分对作物生长季 N_2O 排放的影响,而对冬季 N_2O 排放特征和影响因素缺少系统研究。为明确添加外源铵态氮对典型耕作土壤冻结过程中 N_2O 排放特征的影响,本文应用冰柜模拟冬季土壤冻结过程,研究室温—冻结过程不同铵态氮浓度对 3 种典型地带性耕作土壤 N_2O 排放的影响,以期为调控农田氮肥管理控制土壤 N_2O 排放提供理论依据。**【方法】**试验设 12 个处理,包括 3 种土壤类型(黑土、潮土、黄土),4 个外源 NH_4^+-N 浓度梯度(0、80、200、500 mg/kg 土,分别以 N_0、N_{80}、N_{200}、N_{500} 表示)。具体方法是将 3 种土壤的风干土样 150 g 分别装入广口瓶中,加入 NH_4^+-N 溶液,使土壤湿度达到田间持水量,置于 25℃恒温环境中培养, 24 h 后分别于 0、10、20、30 min 时采集气体,再放到 -10℃的冰柜中,分别在冷冻 0.5 h、2.5 h、6.5 h、13.5 h、23.5 h、43.5 h 时采集气体,用气相色谱检测样品的 N_2O 气体浓度。**【结果】**室温条件下在一定范围内增加外源铵态氮施用量能够促进黑土和潮土 N_2O 的排放,添加 80 mg/kg 铵态氮的黑土和潮土的 N_2O 排放通量分别比各自对照增加 2854.7% 和 192.1%,均达 5% 显著水平,但铵态氮浓度过高会抑制黑土和潮土的 N_2O 排放;黄土在室温培养条件下 N_2O 排放通量接近零。随冻结时间的延长,黑土和潮土的 N_2O 排放通量逐渐降低,其降低速度均呈现 $N_{80} > N_{200}$、$N_{500} > N_0$ 的趋势,且两种土壤的 N_{80} 处理分别与各自其他处理的差异达 5% 显著性水平;冻结 0.5 h 的黑土添加外源 NH_4^+-N 处理的 N_2O 排放通量比对应初始值(冻结 0 h)降低了 64.95%～72.46%,冻结 2.5 h 后比初始值降低 79.1%～89.29%,在冻结 6.5 h 时接近零排放,黑土的 N_0 处理在冻结过程中 N_2O 排放通量基本无变化,数值始终较小;潮土各处理在冻结 0～6.5 h 内 N_2O 排放通量逐渐降低,且处理间差异减少,冻结 0.5 h 时潮土 4 个处理的 N_2O 排放通量比对应初始值(0 h)降低了 47.25%～58.34%,冻结 2.5 h 降低了 84.35%～94.99%,其中 N_0 处理的 N_2O 排放通量在冻结 2.5 h 后达到稳定的零排放状态,而 3 个添加外源 NH_4^+-N 的处理在冻结 6.5 h 后达到稳定的零排放状态;黄土各处理在室温和冻结过程中 N_2O 排放通量始终处于较低的水平,且变化范围较小,处理间无显著差异。室温—冻结全过程黑土和潮土的 N_2O 累计排放量均呈 $N_{80} > N_{200}$、$N_{500} > N_0$,且黑土的 N_{80} 处理与 N_0 处理间均达到 5% 显著水平;潮土不同铵态氮浓度处理间无显著性差异;黄土 N_2O 累计排放量处于较低水平或呈负排放状态,其中 N_{500} 处理 N_2O 的累计负排放量最大。方差分析结果表明,外源铵态氮对 3 种土壤 N_2O 累计排放量均有显著影响。**【结论】**室温条件下,适量的外源铵态氮可促进黑土和潮土的 N_2O 排放,但外源铵态氮浓度过高则可抑制 N_2O 的排放;冻结过程中添加外源铵态氮黑土和潮土的 N_2O 排放通量逐渐降低,且降低速度逐渐变缓最终接近零排放;室温—冻结过程添加外源铵态氮黄土的 N_2O 排放通量始终处于极低水平,甚至出现负排放现象;添加外源铵态氮对室温—冻结过程不同土壤类型 N_2O 累计排放量有显著影响。建议在潮土和黑土上降低冻前土壤的铵态氮含量从而减少 N_2O 的排放。

关键词: 耕作土壤;冻结过程;铵态氮;N_2O;排放通量
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Effects of ammonium-N application on N_2O emission from three types of soils in freezing process

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Abstract:【**Objectives**】Agricultural soil is a main N_2O emission source, which was proposed as one of the six greenhouse gases in the Kyoto Protocol. Previous studies have focused on effects of exogenous available nutrients on soil N_2O emission during crop growing seasons. However, mechanism on N_2O emission in winter is barely reported. In farming fields, lower temperature could block N uptake by plant and leave more N substrate for microbes to produce N_2O. In order to clarify influence of NH_4^+-N application on N_2O emission in the freezing process, a lab-scale experiment was conducted to investigate the effects of NH_4^+-N concentrations on N_2O emission from 3 types of soils, black soil, fluvo-aquic soil and loess soil.【**Methods**】The experiment involved 3 types of soils, and each one with 4 NH_4^+-N concentrations, 0, 80, 200 and 500 mg/kg soil (abbreviated N_0, N_{80}, N_{200} and N_{500}), and three replicates. Each incubator was bottled with 150g soil sample which was sprayed with the concentration NH_4^+-N solution into the incubator to reach soil WFC (black soil WFC was 40%, fluvo-aquic soil WFC was 35%, and loess soil WFC was 22%). Put the incubators under 25℃ for microorganisms cultivation lasting for 24 h, then removed the gas samples from the headspace of incubators by the syringes every 10 minutes (0, 10, 20 and 30 min) 4 times in 30 minutes. After collecting the gas samples, we put the incubators in freezer under −10℃ temperature condition. Respectively, after 0.5 h, 2.5 h, 6.5 h, 13.5 h, 23.5 h and 43.5 h, gas samples were removed from incubators' headspace at 0, 10, 20 and 30 min and analyzed using a gas chromatograph.【**Results**】In this study, the appropriate NH_4^+-N addition would increase N_2O fluxes of the black soil and fluvo-aquic soil compared with their corresponding controls, and the N_2O fluxes of the 80 mg/kg soil treatment are increased by 2854.7% and 192.1% ($P<0.05$) compared with the control. While the excessive NH_4^+-N addition attenuates the promotion, the N_2O emission from loess soil fluctuates around zero in the room temperature incubation. With the freezing proceeding, the N_2O fluxes of two types of soil (black soil and fluvo-aquic soil) added with NH_4^+-N reduced gradually, and the reduction rates are in the order: $N_{80}>N_{200}$, $N_{500}>N_0$, in addition, the rates of N_{80} from the two soils have significant differences ($P<0.05$) with other treatments respectively in the first 0.5 h. For black soil, the N_2O fluxes of N_{80}, $N_{200}$the N_{500} are decreased by 64.95% −72.46% compared with initial values in 0.5 h, and in 2.5 h, the fluxes decreased by 79.1% −89.29% and are 0 emission by the time of 6.5 h (consumed over 98.4%). However, the control remains small fluxes and slight changes in the duration. The N_2O fluxes of the fluvo-aquic soil treatments decreased gradually at the freezing stage, the differences among treatments are lower gradually, the soils N_2O emissions are dropped by 47.25% −58.34% in the first 0.5 hour, and decreased by 84.35% −94.99% in 2.5 h. For the treatments with NH_4^+-N addition, N_2O emissions are different from that of the control, and achieve stable state (0 emission) in 2.5 h. The loess soil N_2O emission shows a low level of fluxes throughout the room temperature and freezing incubation process, and the change range is small. In addition, there are no significant differences among the treatments. The overall process of experiment, rank four treatments ordering to total N_2O emissions of black soil and fluvo-aquic soil: $N_{80}>N_{200}$, $N_{500}>N_0$, the black soil N_{80} differs from N_0 significantly ($P<0.05$), and no significant differences are found among the fluvo-aquic soil treatments. The loess soil treatments have low emission amount, and negative emissions and there are no significant differences among the loess soil treatments. According to the multi-factor analysis of variance, the different levels of NH_4^+-N application affect the total N_2O emission significantly.【**Conclusions**】Under room temperature, the N_2O emissions from black soil and fluvo-aquic soil are promoted by the appropriate concentration of NH_4^+-N addition, however the N_2O emissions are inhibited by the high concentration addition. In the freeze process, the N_2O fluxes from the two types of soil with the NH_4^+-N addition are gradually decreased as the time passed, and the N_2O fluxes fall to 0 eventually. N_2O flux of loess soil shows low level throughout room-temperature incubation and freezing process. The total N_2O emissions are affected significantly by different concentrations of NH_4^+-N application. In the respect of

agricultural control measures in N_2O emissions, massive NH_4^+-N fertilizer application should be avoided in black soil and fluvo-aquic soil prior to soil freezing period, however, no significant effect is observed in loess soil N_2O emission during various concentration of NH_4^+-N application.

Key words: cultivated soil; freezing process; ammonium nitrogen; N_2O; flux

氧化亚氮(N_2O)是重要的温室气体之一,N_2O对大气增温潜势大,在大气中存留的时间长[1-2],在短时期内很难降低其浓度[3]。人类活动是N_2O排放的最直接原因,而农业排放占人类活动总贡献量的81%[4-5]。我国46.3%的国土面积经历着季节性冻结过程,其中包括我国东北、西北及华北多数平原耕作区[6-7],冻结作用作为中高纬度地区的普遍现象,深刻影响着土壤生物地球化学循环过程和强度[8]。Teep等[9]研究发现,冬季土壤N_2O排放量甚至占全年总排放量的40%～50%,因此有必要加强冻结过程中土壤N_2O排放特征的研究。耕作土壤通常受到强烈的人为干预和调控,如追施铵态氮肥或尿素,这部分氮除部分被利用和损失外很多将会残留在土壤中,从而影响冻结过程土壤N_2O的排放[10]。为了揭示添加铵态氮肥如何影响冻结过程中土壤N_2O的排放特征,不同土壤类型特别是土壤理化性状在冻结过程中对土壤N_2O排放影响的主要因素是否一致,本文以黑土、潮土和黄土3种典型耕作土壤为研究对象,研究不同铵态氮浓度对冻结过程中土壤N_2O的排放通量的影响,以期为调控农田氮肥管理,控制土壤N_2O排放提供理论依据和技术支撑。

1 材料与方法

1.1 试验材料

供试土壤为黑土、潮土和黄土3种典型耕作土壤,黑土取自黑龙江省海伦市海伦农场大豆种植区,当地年均气温2.0℃(最高温37.7℃,最低温-40.3℃),年均降水量500～600 mm,无霜期120 d,种植制度为一年一熟。潮土取自天津市杨柳青镇大柳滩玉米种植区,该区年均气温11.6℃(最高温40.9℃,最低温-22.5℃),年均降水量577.8 mm,无霜期203 d,种植制度为冬小麦-夏玉米轮作。黄土采自陕西安塞地区,本区年均气温8.8℃(最高温36.8℃,最低温-23.6℃),年均降水量296.6～645.0 mm,无霜期157 d,种植制度为一年一熟。采样时间均为2012年6月,采样深度0—20 cm,每个采样区面积约2000 m^2,采用蛇形布点每区采集10个点,每个采样点采集2 kg土样,混匀后,用四分法取约10 kg土壤样品。土样经室内风干,研磨过0.25 mm筛,备用。3种土壤的基本理化性状见表1。

1.2 试验设计

试验设12个处理,包括黑土,潮土和黄土3种土壤类型,每种土壤设4个外源NH_4^+-N浓度,分别为0、80、200、500 mg/kg土(以N_0、N_{80}、N_{200}、N_{500}表示),每个处理重复3次。NH_4^+-N溶液用分析纯氯化铵试剂配制。

1.3 试验方法

准备36个PE塑料广口瓶,容积为840 mL(底面Φ10 cm),将顶盖钻直径约为2 mm的洞,用气相色谱进样垫和密封胶粘合后备用,并测试广口瓶密封性良好。广口瓶装入风干土样150 g,用注射器沿广口瓶四壁缓慢均匀加入不同浓度的NH_4^+-N溶液以驱赶封闭在土体内部的气体,使土样湿度达到田间持水量(黑土、潮土和黄土分别注入60、52.5和33 mL)。同时准备若干1 mL注射器。

1.4 样品采集及分析

将加入氯化铵溶液的广口瓶开盖置于恒温25℃环境中培养24 h,使水分和氮素在土样中分布达到平衡,并恢复微生物活性。采集气体时将瓶盖盖严后开始计时,分别在0、10、20、30分钟用注射器采集1 mL气体样品,把采集好的注射器针头扎入橡胶垫中以保持气体密封。准备与冰柜内部底面积相同大小的苯板,在苯板上挖出与广口瓶底面积相同的孔洞,将广口瓶口放入孔洞内,并使广口瓶四周与苯板以及苯板四周与冰柜紧密接触以维持广口瓶内土样的冷冻状态。根据3种土壤所在地区的气温调节冰柜为-10℃。分别在冷冻0.5 h、2.5 h、6.5 h、13.5 h、23.5 h、43.5 h时盖上广口瓶盖子取样,气体采集方法同上。气体采集完成后立即用安捷伦5890气相色谱仪检测N_2O气体浓度。测定条件为:柱温40℃,转化器温度300℃,^{63}Ni电子捕获器(ECD)检测器温度为320℃,进样量为1 mL,载气为95% Ar与5% CH_4混合气,流速为20 mL/min。色谱柱为80/100目PorapakQ的填充柱。

表 1　3 种土壤的基本理化性状

Table 1　Physical and chemical characteristics of three selected soils

土壤类型 Soil type	pH	有机质 OM (g/kg)	全磷 Tot. P (g/kg)	全氮 Tot. N (g/kg)	铵态氮 NH_4^+-N (mg/kg)	硝态氮 NO_3^--N (mg/kg)	C/N	田间持水量 WFC (%)	质地 Texture
黑土 Black soil	5.31	55.9	1.07	2.13	9.34	28.54	15.22	40	粘壤 Clay loam
潮土 Fluvo-aquic soil	7.81	21.5	0.57	0.67	4.98	22.13	18.61	35	砂壤 Sandy loam
黄土 Loess soil	7.73	8.11	0.46	0.79	6.31	17.94	5.95	22	轻壤 Light loam

N_2O 气体排放通量 Flux[11] 的计算公式为

$$Flux = \rho \times V \times \Delta C/\Delta t \times 273/[A \times (273 + T)]$$

式中:$\Delta C/\Delta t$ 表示时间间隔为 Δt 时的 N_2O 浓度变化量;ρ 为标准状态下 N_2O 气体密度;V 和 A 分别为广口瓶中空气体积和底面积;T 为采样时广口瓶内的温度。

试验数据用 Excel 2003 进行处理,用 SAS 9.0 软件进行方差分析和 Duncan's 新复极差比较。

2　结果与分析

2.1　外源铵态氮对冻结过程中黑土 N_2O 排放通量的影响

室温条件下添加 NH_4^+-N 的黑土 N_2O 排放通量总体呈现 $N_{80} > N_{200}$、$N_{500} > N_0$ 趋势,分别比 N_0 增加了 2854.7%、1293.4% 和 1243.1%(图 1),且 N_{80} 处理 N_2O 排放通量比 N_0 显著提高,说明在一定范围内增加铵态氮可促进黑土 N_2O 的排放。0～6.5 h 的冻结过程各处理 N_2O 排放通量逐渐降低,且处理间差异逐渐减少,N_2O 排放通量降低的速度为 $N_{80} > N_{200}$、$N_{500} > N_0$,且 N_{80} 与其他处理间差异显著,表明冻结过程显著降低了 NH_4^+-N 对土壤 N_2O 排放的促进作用。N_0 处理的 N_2O 排放通量在冻结过程中基本无变化,数值始终较小。冻结 0.5 h 添加外源 NH_4^+-N 处理的 N_2O 排放通量分别比对应的初始值(0 h)降低了 64.95%～72.46%,冻结 2.5 h 后比初始值(0 h)降低 79.1%～89.29%,冻结 6.5 h 后添加外源 NH_4^+-N 处理的 N_2O 排放通量降低至零,随后所有处理维持零排放状态。

2.2　外源铵态氮对冻结过程潮土 N_2O 排放通量的影响

室温条件下添加 NH_4^+-N 的潮土 N_2O 排放通量各处理总体呈 $N_{80} > N_{200} > N_0$、N_{500} 趋势(图 2),N_{80} 和 N_{200} 处理的 N_2O 排放通量分别比 N_0 增加了 192.05% 和 99.05%,且 N_{80} 处理 N_2O 排放通量与

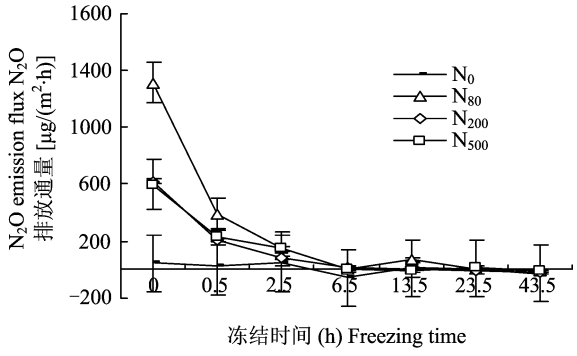

图 1　不同浓度外源 NH_4^+-N 对黑土冻结过程中 N_2O 的排放通量的影响

Fig. 1　N_2O fluxes from the black soil amended with NH_4^+-N during the freezing process

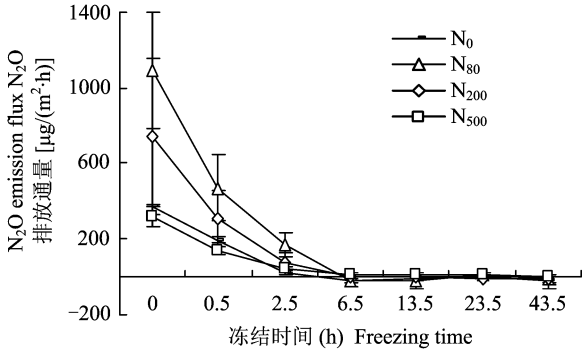

图 2　不同浓度外源 NH_4^+-N 对潮土冻结过程中 N_2O 排放通量的影响

Fig. 2　N_2O fluxes from the Fluvo-aquic soil amended with NH_4^+-N during the freezing process

N_0 和 N_{500} 处理间差异达 5% 显著水平。0～6.5 h 的冻结过程各处理 N_2O 排放通量逐渐降低,且处理间差异减少;N_2O 排放通量降低速率各处理呈 $N_{80} > N_{200} > N_0$、N_{500} 的趋势;冻结 0.5 h 时 4 个处理潮土的 N_2O 排放通量分别比对应的初始值(0 h)降低了

47.25%～58.34%，冻结2.5 h降低了84.35%～94.99%。N_0处理N_2O排放通量在冻结2.5 h后达到稳定的零排放状态，而3个添加外源NH_4^+-N的处理在冻结6.5 h后达到稳定的零排放状态，随后所有处理维持零排放状态。

2.3 外源铵态氮对冻结过程中黄土N_2O排放通量的影响

室温和冻结过程中4个处理黄土的N_2O排放通量变化范围较小，始终处于－48.59～20.62 μg/(m²·h)范围内(图3)，且各处理间无显著性差异。N_{80}和N_{500}分别在6.5 h和23.5 h出现负排放现象，但与其他处理的差异也不显著。

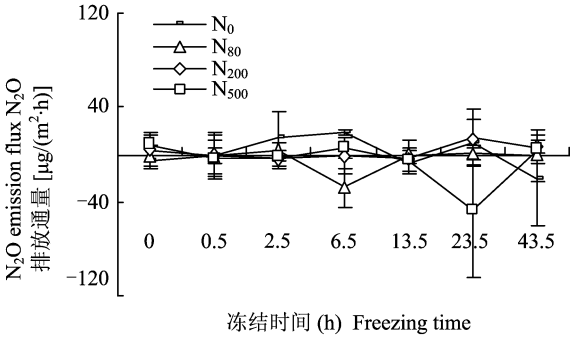

图3 不同浓度外源NH_4^+-N对黄土冻结过程中N_2O排放通量的影响

Fig.3 N_2O fluxes from the Loess soil amended with NH_4^+-N during the freezing process

2.4 外源铵态氮对冻结过程中3种土壤N_2O累计排放量的影响

从图4可以看出，室温—冻结全过程黑土和潮土N_2O累计排放量均呈N_{80} > N_{200}、N_{500} > N_0。黑土的N_{80}处理N_2O累计排放量与N_0处理间有显著差异，潮土N_{80}处理的N_2O累计排放量高于其它处理，但差异不显著，表明一定含量范围内添加外源铵态氮能够促进黑土和潮土N_2O的累计排放量，而外源铵态氮浓度过高会导致N_2O累计排放量降低，因此N_{500}处理的N_2O累计排放量与N_{200}处理之间均无显著差异。添加外源铵态氮黄土的N_2O累计排放量处于较低水平或呈负排放状态，其中N_{500}处理的N_2O累计负排放量最大，但与其他处理间的差异并不显著。

3 讨论

室温条件下，在一定范围内增加外源铵态氮浓度，会促进黑土和潮土N_2O的排放(图1～图2)。其可能的原因是，第一，这两种土壤的pH基本处于土壤硝化和反硝化微生物生长繁殖适宜的范围[12]；第二，这两种土壤的C/N相对比较合适，能够为微生物生长提供碳源[13]；第三，土壤本底铵态氮含量较低，外源铵态氮的添加为黑土和潮土提供了参与硝化和反硝化反应的电子供体，有利于提高土壤硝化和反硝化细菌的活性[14]，也说明氮素可能是限制黑土和潮土N_2O排放的重要因素之一，与杨黎等利用模型分析的结果一致[15]。但是，添加过高浓度的铵态氮却对土壤N_2O的排放产生抑制作用，如图1和图2中表现出的N_{200}和N_{500}处理土壤N_2O排放通量显著低于N_{80}处理，可能是因为过量的外源铵态氮抑制了微生物的活性[16-18]。室温条件下添加外源铵态氮对黄土N_2O的排放无显著影响，且各处理N_2O排放通量均维持在零排放左右(图3)，说明土壤有机碳含量很低的条件(如黄土有机质含量仅为8.11 g/kg)很难提供给细菌，特别是异养细菌活动所需的足够的能源物质，此时即便增加外源用于反硝化和硝化的底物浓度也很难激发土壤微生物活性而形成较高的N_2O排放通量[19]。

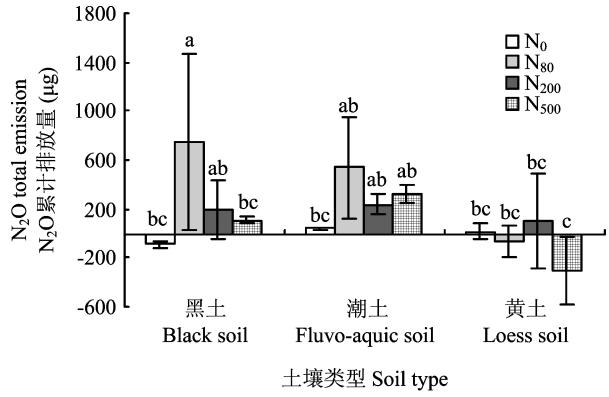

图4 外源NH_4^+-N对冻结过程中3种土壤N_2O累计排放量的影响

Fig.4 Total N_2O emissions from Black, Fluvo-aquic and Loess soils amended with NH_4^+-N during the freezing process

[注(Note)：柱上不同字母表示处理间差异达5% Different letters above the bars mean significant among treatments at the 5% levels.]

随冻结时间的延长，黑土和潮土N_2O排放通量逐渐降低，因为本来受外源铵态氮添加而激发的硝化反硝化微生物活性在持续低温胁迫下会逐渐降低[20]。N_2O排放通量降低速度随冻结的持续进行而逐渐变缓也表明了是一个生物学主导过程。有研

究表明，黑土微生物经过长期低温环境的选择，微生物细胞对低温产生的适应机理能够快速完成细胞质浓缩，使细胞处于休眠状态，降低冷冻危害[8]，在室温条件下较高的 N_2O 排放通量（N_{80}处理）较其他处理排放时间持续更长，如潮土的 N_{80}处理 N_2O 排放比 N_{500}处理晚 4 h（分别在冻结 6.5 h 和 2.5 h）达到稳定状态（0 排放），这与王风等[21]的研究结果一致，表明微生物活性在逆境胁迫下是逐渐降低的，需要一个长时间过程才能够达到新的稳定平衡状态[22-23]。黄土 N_2O 排放通量在冻结过程中始终维持较低的或零排状态，仍然是由土壤本身的理化性质所决定的，此外添加外源铵态氮可能会更加剧土壤有机碳不足的矛盾，甚至在冻结过程中出现 N_2O 负排放现象[24-25]。

室温—冻结过程中黑土和潮土的 N_2O 累计排放量趋势一致，在一定范围内增加外源铵态氮能够促进 N_2O 的累计排放量，过量的外源铵态氮又抑制了土壤 N_2O 累计排放量，可能与铵态氮对土壤微生物活性的抑制作用有关[16]。黄土 N_2O 累计排放量处于较低水平或负排放状态，主要是由土壤理化性质所决定，特别是微生物活性在较低有机碳供给条件下明显地会受到抑制[26-27]。

4　结论

1）室温条件下，在一定范围内添加外源铵态氮使黑土和潮土的 N_2O 的排放通量显著增加，但过高浓度的外源铵态氮会抑制 N_2O 的排放；随冻结的持续进行添加外源铵态氮黑土和潮土的 N_2O 排放通量逐渐降低，且降低的速度逐渐变缓；随冻结的持续进行 N_2O 排放通量最终接近零排放，但室温时 N_2O 的排放通量高的处理达到稳定零排放速率的时间有向后延长的趋势。

2）室温—冻结过程添加外源铵态氮的黄土 N_2O 排放通量始终处于极低水平，甚至出现负排放现象，处于 $-48.59 \sim 20.62$ μg/(m^2 · h)范围内。

3）添加外源铵态氮对室温—冻结过程不同土壤类型 N_2O 累计排放量的影响显著。从控制土壤 N_2O 排放的角度来讲，潮土和黑土在越冬前应尽量降低土壤铵态氮肥的浓度，黄土无需考虑铵态氮肥的施用浓度。

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