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不同污水处理工艺非二氧化碳温室气体的释放

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摘要:甲烷和氧化亚氮是两种重要的非二氧化碳温室气体.城市污水处理厂是甲烷和氧化亚氮的重要释放源.因此,为探究不同污水处理工艺甲烷和氧化亚氮的释放现状和变化规律,通过浮流式表面集气罩对西安市第三污水处理厂(Orbal 氧化沟工艺)和第四污水处理厂(A/A/O工艺)生物处理过程中甲烷和氧化亚氮的排放情况进行测定,比较不同污水处理工艺中非二氧化碳温室气体的释放情况,并以第四污水处理厂为例研究溶解氧、温度对非二氧化碳温室气体释放量的影响.结果表明,西安市第三污水处理厂每 m^3 进水释放甲烷1 $181 \, mg($ 以 CH_4 计) 、氧化亚氮 $36.20 \, mg($ 以 N_2O 计) ,西安市第四污水处理厂每 m^3 进水释放甲烷209 mg(以 CH_4 计) 、氧化亚氮 $54.64 \, mg($ 以 N_2O 计) .温度、曝气方式、溶解氧浓度、亚硝酸盐氧化速率和最大产甲烷活性是甲烷和氧化亚氮释放量的重要影响因素.

关键词:甲烷;氧化亚氮;A/A/O;Orbal 氧化沟;污水生物处理

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Non-CO₂ Greenhouse Gas Release from Different Biological Wastewater Treatment Processes

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Abstract: Methane (CH_4) and nitrous oxide (N_2O) are two of the most important non- CO_2 greenhouse gases. And municipal sewage treatment plant is an important anthropogenic source of CH_4 and N_2O . Therefore, it is essential to measure the production and emission of CH_4 and N_2O during biological wastewater treatment process. The surface emission flux isolation chamber was employed to collect the gas sample from the Xi'an No. 3 WWTP (the Orbal oxidation ditch process) and Xi'an No. 4 WWTP (A/A/O process) to determine the contents of methane and nitrous oxide. And the effects of temperature and dissolved oxygen concentration on non- CO_2 greenhouse gases emission from Xi'an No. 4 WWTP were discussed. The results showed that methane and nitrous oxide emission factors from No. 3 WWTP were 1 181 mg CH_4 per m³ influent and 36. 20 mg N_2O per m³ influent, respectively while those from the No. 4 WWTP were 209 mg CH_4 per m³ influent and 54. 64 mg N_2O per m³ influent. In addition, the important influencing factors which affected methane and nitrous oxide emission were temperature, aeration strategy, DO, nitrite oxidation rate and specific methanogenic activity.

Key words: methane; nitrous oxide; A/A/O; Orbal oxidation ditch; biological treatment of wastewater

甲烷(CH₄)和氧化亚氮(N₂O)是两种常见的非二氧化碳温室气体,其温室效应分别是二氧化碳的25 倍和298 倍^[1],即在低排放量的情况下也会对大气环境造成较大威胁. 城市污水生物处理过程中会产生甲烷和氧化亚氮. 根据调查,2000~2009 年我国氧化亚氮的排放量逐年增加^[2],污水厂进水中总氮负荷中约有7%转化为一氧化氮和氧化亚氮^[3]. 随着各国日益严格的环境标准,污水处理量逐年增加,因此调查和控制污水处理厂生物处理过程中温室气体的释放具有重要意义.

甲烷主要在有机物的去除过程中产生,主要发生在氧化还原电位较低的区域(缺氧区或厌氧区). 经过发酵产酸菌、产氢产乙酸菌和产甲烷菌等一系列微生物依次作用,复杂有机物最终被氧化成甲烷和二氧化碳.氧化亚氮主要在生物脱氮过程中产生,主要发生在硝化阶段和反硝化阶段.在硝化阶 段,氧化亚氮主要是来源于羟胺(NH₂OH)氧化和硝化菌的反硝化过程,此外硝酰基(NOH)的非生物反应、好氧反硝化菌的存在与异养菌的硝化等几个过程也会造成氧化亚氮的产生^[4~7].氧化亚氮是反硝化阶段的中间产物,其进一步还原受阻时就会造成氧化亚氮的逸出排放,可能的原因有两种:①氧化亚氮还原酶(Nos)受到抑制,如pH、溶解氧等因素均会限制其活性,造成氧化亚氮的积累;②某些反硝化系统中没有 Nos,如荧光假单胞菌(Pseudomonas fluorescens)等,对于这些反硝化菌,其最终产物就是氧化亚氮^[8,9].根据报道,硝化阶段是氧化亚氮产生和释放的主要阶段;在反硝化阶段,只要保证反硝化

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正常进行的条件,氧化亚氮的产生和释放是可以完全避免的.由于污水处理工艺的不同,如曝气方式、水力停留时间等运行参数不同,脱氮过程中优势微生物种群不同,这就造成了氧化亚氮的产生和释放量的不同.

近年来各国学者对污水处理过成中温室气体产生的相关问题进行了大量研究^[10~13],但这些报道主要集中研究单一污水处理工艺温室气体释放情况,对不同污水处理工艺温室气体排放差异及其原因的研究有限.本研究选取我国污水处理常见工艺——氧化沟工艺(西安市第三污水处理厂,Orbal氧化沟工艺)和A/A/O工艺(西安市第四污水处理厂,A/A/O工艺)对其甲烷和氧化亚氮的释放量进行测定,比较不同污水生物处理工艺对非二氧化碳温室气体释放的影响,并探讨了溶解氧、温度与温室气体释放量之间的关系,为污水处理厂温室气体产生和释放的研究提供基础资料,对研究污水处理厂温

室气体减排措施,并对研发新的污水处理工艺具有 重要的实践意义.

1 材料与方法

1.1 污水处理厂概况和试验布置

西安市第三污水处理厂(三污)位于西安市东部,总投资 2.62 亿元,服务面积约为 21 km²,日处理能力为 15 万 m³(二期工程部分运行). 进水主要是来自纺织、印染、机械加工等行业的工业废水(占55.3%)和生活污水,进水水质复杂,各项指标波动范围大[14,15]. 西安市第四污水处理厂(四污)位于西安市北郊,总投资 4.19 亿元,服务面积约为 45 km²,日处理量为 25 万 m³(一期工程)[16],污水来源主要为生活污水和工业废水,生活污水与工业废水的比例约为 4:6. 两个污水处理厂采用的处理工艺是目前我国比较常见的处理工艺,其基本情况见表 1.

表 1 两个污水处理厂基本情况/mg·L-1

Table 1 Basic information of the two wwills mig-L							
污水厂	处理工艺		COD	$\mathrm{NH_4^+}$ -N	TN	曝气方式	
三污	Orbal 氧化沟	进水	390 ~ 825	15. 9 ~ 47. 5	23. 45 ~ 98. 06	表面转盘曝气	
		出水	10. 93 ~ 50. 79	1. 57 ~ 18. 82	5. 42 ~ 25. 53	A Print A IIII WAY	
四污	A/A/O	进水	187 ~651	16. 8 ~ 44. 3	39. 47 ~ 58. 72	底部微孔曝气	
		出水	16. 6 ~ 51. 4	0. 45 ~ 14. 71	2. 38 ~ 20. 47		

西安市第三污水处理厂采用 Orbal 氧化沟工艺,相当于一系列完全混合反应器的串联,根据其特点,共布设采样点 4 个[图 1(a)],其中厌氧选择池中采样点1个,氧化沟外沟、中沟、内沟各设采样点1个,采样点位于曝气转盘上游. 西安市第四污水处理厂主要生物处理工艺为 A/A/O 工艺,属于推流式反应系统,因此取样点应按照水流方向布设. 缺氧段、厌氧段各布设1个,好氧段布设3个,共5个采样点[图 1(b)].

1.2 采样方法

气体样品采用浮流式表面集气罩收集,气相色谱测定. 浮流式表面集气罩类似静态箱^[17,18],为PVC 材质的圆柱体(ϕ 15 cm,h = 10 cm),顶端有采样口和温度计,使用前检查其气密性. 采样时将集气罩沉没在液面下,随着污水生物处理过程中不断产生气体,集气罩中充满气体后,气体样品通过取样口用气体采样袋收集. 气体样品在24 h 内利用气相色谱仪分析甲烷和氧化亚氮气体浓度^[19-22],同时在采样点周围取水样,根据标准方法测定其中NH₄⁺-N、NO₂⁻-N、NO₃⁻-N、COD、SS 和 VSS 等.

1.3 释放速率和释放因子计算

温室气体释放速率定义为单位时间内释放的温室气体量,是表征温室气体释放的重要指标. 在曝气区,释放速率($E_{GHG,aem}$)的表达式如下:

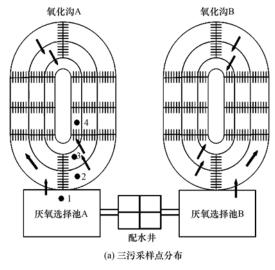
$$E_{\text{GHG, aero}} = Q_{\text{air}} \cdot c_{\text{GHG, aero}}$$
 (1)
式中, $E_{\text{GHG, aero}}$ 为曝气区温室气体释放速率 $(\text{kg}\cdot\text{d}^{-1}); Q_{\text{air}}$ 为生物曝气池曝气量 $(\text{m}^3\cdot\text{d}^{-1}); c_{\text{GHG, aero}}$ 为温室气体样品浓度 $(\text{g}\cdot\text{m}^{-3}).$

非曝气区(三污氧化沟各沟道及四污缺氧区和 厌氧区)的温室气体排放速率($E_{\mathrm{GHG,non-aero}}$)计算方法 如下:

$$E_{\rm GHG,non-aero} = \frac{V_{\rm gas} \cdot c_{\rm GHG}}{\Delta t} \cdot \frac{A_{\rm non-aero}}{A_{\rm SEFIC}} \tag{2}$$

式中, $E_{GHG,non-aero}$ 为非曝气区温室气体释放速率 $(kg \cdot d^{-1}); V_{gas}$ 为一定时间内浮流式表面集气罩中收集的气体总体积 $(m^3); \Delta t$ 为浮流式表面集气罩进行气体收集的时间 $(d); A_{non-aero}$ 为非曝气区表面积 $(m^2); A_{SEFIC}$ 为浮流式表面集气罩横截面 (m^2) .

排放因子(w_{CHG})定义为单位进水排放非二氧化碳温室气体的量,是对比不同工艺温室气体排放



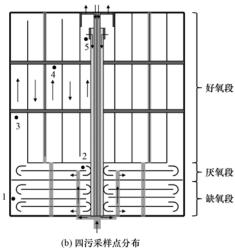


图 1 现场采样点分布示意

Fig. 1 Distribution of sampling points in the two WWTPs

的重要指标. 其计算方法如下:

$$w_{\text{GHG,influent}} = \frac{E_{\text{GHG}}}{Q_{\text{influent}}}$$
 (3)

式中, $w_{\text{GHG,influent}}$ 为按污水厂每天进水量计算的温室气体排放因子(kg·m^{-3}); Q_{influent} 为污水处理厂每天进水量($\text{m}^3 \cdot \text{d}^{-1}$); E_{GHG} 为温室气体释放速率(kg·d^{-1}).

1.4 最大比产甲烷活性(SMA)与辅酶 F₄₂₀

污泥最大比产甲烷活性(specific methanogenic activities, SMA)采用血清瓶试验法^[23]. 试验期间每隔2h测定气体中甲烷浓度,根据式(4)计算SMA.

$$\mu_{\text{max CH}_4} = 4 \times \frac{\text{d}c_{\text{CH}_4} \times V_{\text{CH}_4}}{\text{d}t} \times \frac{1}{X}$$
 (4)

式中,4 为甲烷的 COD 当量; $\mu_{\text{max CH}_4}$ 为最大比产甲烷活性[$\text{mg}\cdot(\text{g}\cdot\text{d})^{-1}$](以 COD 计); $\frac{\text{d}c_{\text{CH}_4}\times V_{\text{CH}_4}}{\text{d}t}$ 为甲

烷的产率 $(mg \cdot d^{-1}); X$ 为血清瓶中污泥浓度(g). 污泥中辅酶 F_{420} 采用紫外分光光度法 $[^{24}]$.

1.5 氨氧化速率和亚硝酸盐氧化速率

氨氧化速率(AUR)、亚硝酸盐氧化速率(NUR)采用间歇试验法,测定起始浓度为 40 mg·L⁻¹,温度为 20℃.

2 结果与分析

2.1 污水厂非二氧化碳温室气体排放

为了获得数据能较准确地反映两个污水厂非二氧化碳温室气体的排放情况,对两个污水处理厂非二氧化碳温室气体排放进行一年的监测.

两个污水厂甲烷的排放情况见图 2. 三污甲烷的释放总量(以 CH_4 计,下同)为26 542 ~ 66 518 kg·a⁻¹,释放因子为每 m³ 进水 717 ~ 1 822 mg. 四污甲烷的释放总量为11 746 ~ 30 540 kg·a⁻¹,释放因子为每 m³ 进水 129 ~ 335 mg. 根据西安市人均综合用水定额[150 $L\cdot(\text{人·a})^{-1}$]和两个污水厂的处理量,两个污水厂甲烷的人均释放因子分别为39.81 ~ 99.77 g·(人·a)⁻¹ 和 7.05 ~ 18.33 g·(人·a)⁻¹. 对比两个污水厂甲烷的释放情况可以看出,夏季(6~8 月)甲烷释放量高于冬季(12 月~次年2 月),而三污的甲烷释放量高于四污.

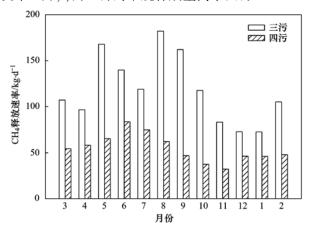


图 2 甲烷释放速率历时(逐月)变化

Fig. 2 Monthly variation of CH₄ release rate of the two WWTPs

两个污水厂氧化亚氮的释放情况见图 3. 三污氧化亚氮释放总量(以 N_2O 计,下同)为1613~28063 kg·a⁻¹,释放因子为每 m³ 进水 29.47~51.99 mg,氧化亚氮人均释放因子为 1.61~2.85 g·(人·a)⁻¹;四污的氧化亚氮释放总量为3425~5953 kg·a⁻¹,释放因子为每 m³ 进水 37.54~65.23 mg,人均释放因子为 2.06~3.57 g·(人·a)⁻¹. 对比两个污水处理厂氧化亚氮的释放情况,夏季(6~8

月)温度高时氧化亚氮的释放量要高于冬季(12月~次年2月)温度低时的释放量,且四污氧化亚氮的释放量高于三污.

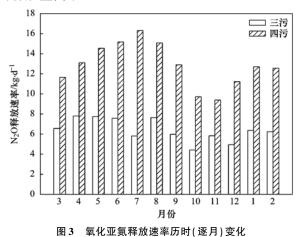


Fig. 3 Monthly variation of N2O release rate of the two WWTPs

两个污水处理厂非二氧化碳温室气体的二氧化碳当量比较情况如图 4 所示. 从中可以看出,三污的二氧化碳当量[每 m^3 进水 40. 31 $\mathrm{g}(以\ \mathrm{CO}_2\ \mathrm{H})]$ 高于四污[每 m^3 进水 21. 51 $\mathrm{g}(以\ \mathrm{CO}_2\ \mathrm{H})]$,表明从非二氧化碳温室气体释放角度考虑, $\mathrm{A/A/O}$ 工艺更具优势.

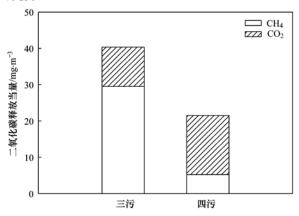


图 4 两个污水处理厂二氧化碳排放当量比较 Fig. 4 CO₂ equivalent of the two WWTPs

2.2 最大产甲烷活性 (specific methanogenic activity, SMA)与辅酶 F₄₂₀

通过测定两个污水处理厂活性污泥的最大产甲烷活性及辅酶 F_{420} 的浓度发现,三污活性污泥在 3 种不同基质下的最大产甲烷活性均比四污活性污泥 高(图 5). 而三污活性污泥的辅酶 F_{420} 浓度为 0.019 2 μ mol·g⁻¹,四污为0.013 4 μ mol·g⁻¹.最大产甲烷活性与辅酶 F_{420} 的结果均可说明三污活性污泥的产甲烷活性高于四污,因此可解释三污甲烷排放量高于四污.

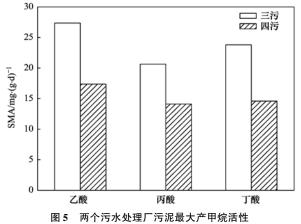


Fig. 5 SMA of the two WWTPs

2.3 氨氧化速率和亚硝酸盐氧化速率

对比两个污水处理厂活性污泥的氨氧化速率和亚硝酸盐氧化速率(见表2)可以发现两个污水处理厂的氨氧化速率基本接近而亚硝酸盐氧化速率相差较大. 三污的亚硝酸盐氧化速率接近氨氧化速率,这表明亚硝酸盐可以被快速氧化,处理系统不会发生亚硝氮积累. 四污亚硝酸盐氧化速率仅是按氧化速率的一半,说明四污的处理系统容易发生亚硝氮积累. 而亚硝酸盐的浓度与氧化亚氮的产生有着密切的联系^[25]. 亚硝酸盐的累积不但造成硝化阶段的反硝化的增强^[26],还会降低反硝化速率,造成一氧化氮和氧化亚氮的积累^[27],进而造成氧化亚氮释放量升高.

表 2 两个污水处理厂的硝化速率

Table 2 Nitrification rate of the two WWTPs

名称	单位	三污	四污
氨氧化速率(以NH ₄ + N计)	$mg \cdot (g \cdot h)^{-1}$	2. 43	2.58
亚硝酸盐氧化速率(以NO ₂ -N计)	$mg \cdot (g \cdot h)^{-1}$	2. 74	1.46

2.4 温室气体释放的影响因素

试验过程中发现温室气体的排放量随温度和曝气区溶解氧的变化而变化. 为探究温室气体释放量与溶解氧、温度的变化关系,以 A/A/O 工艺为例进行研究.

2.4.1 溶解氧的影响

试验期间四污厌氧区和缺氧区的溶解氧浓度相对稳定,因此这两个区域甲烷与氧化亚氮的释放量相对稳定.在好氧区,随着溶解氧浓度的变化,甲烷、氧化亚氮的释放量变化范围较大(图 6).甲烷释放量随着溶解氧浓度的升高而增大,线性相关系数为 0.70.因为水中溶解性甲烷会随着曝气溢出,溶解氧越高曝气强度越大,溢出的溶解性甲烷越多.

而氧化亚氮释放量随溶解氧浓度的升高减小,相关 系数为0.65. 这是由于当好氧池中溶解氧浓度较低 时($< 2 \text{ mg·L}^{-1}$),活性污泥中部分硝化菌 (Nitrosomonas europaea)会进行反硝化反应导致氧 化亚氮释放量增加[4];并且较高的溶解氧可抑制亚 硝氮的积累,进而减小氧化亚氮的释放量. 本研究 得出的溶解氧浓度与甲烷、氧化亚氮释放量的关系 趋势与王金鹤[5]的研究一致.

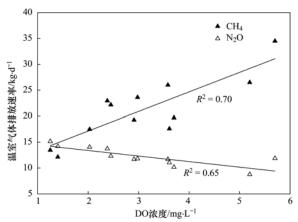
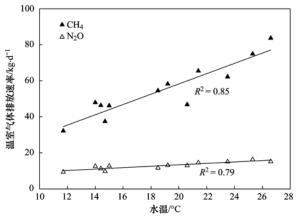


图 6 好氧区甲烷、氧化亚氮释放速率与溶解氧的关系 Fig. 6 Relationship between CH_4 , N_2O release rate and dissolved oxygen concentration in oxic tanks

2.4.2 温度的影响

甲烷与氧化亚氮的释放量随水温的升高而增 加,出现夏季的释放量明显大于冬季的情况,因此有 必要探讨温度对污水处理过程中温室气体释放量的 影响. 由图 7 可知甲烷、氧化亚氮释放速率和污水 温度有较显著的相关性($R^2 = 0.85, R^2 = 0.79$). 甲 烷释放量随温度升高而升高是因为产甲烷菌的活性 随着基质温度的升高而增加[28]. 这一变化关系与 王金鹤[5]和亓鹏玉等[29]的研究结果相同. 氧化亚



甲烷、氧化亚氮释释放速率与水温的关系 Relationship between CH₄, N₂O release rate and wastewater temperature

氮释放量随水温升高而增加是由于一定温度范围内 温度越高氨氧化速率越快,亚硝氮积累率也越 高[30]. 但氧化亚氮释放量与水温的变化关系与王 金鹤[5]和张婷婷等[31]的研究结果相反,与郭宁 等[30]和王亚官等[32]的研究结果相同. 这说明温度 对氧化亚氮释放量的影响机制较为复杂,还需进一 步的研究.

3 讨论

影响污水处理过程中非二氧化碳温室气体释放 的因素有很多,从试验数据来看,主要是温度、曝气 方式和溶解氧浓度. 此外,微生物的种群结构、水 力停留时间、C/N 等工艺参数也会影响温室气体的 释放.

从两个污水处理厂甲烷和氧化亚氮释放的历时 变化中可以看出,在夏季温度较高时,温室气体释放 量比较大,而在冬季,由于温度下降,温室气体释放 量明显降低. 这是由于水温影响产甲烷菌及硝化菌 的活性,从而影响温室气体的产生和释放.

不同的曝气方式会对生物处理单元中溶解氧的 浓度造成影响,而溶解氧的浓度也会影响甲烷和氧化 亚氮的释放[33,34].溶解氧对甲烷的产生具有明显的 抑制作用,低溶解氧浓度有利于甲烷的产生、释放, 因此厌氧区是甲烷的主要释放源. 硝化阶段的溶解 氧不足会引起硝化菌的反硝化,造成氧化亚氮的产生 和释放[4,35]. 若溶解氧浓度过高,反硝化过程可能受 到抑制,也会造成氧化亚氮的产生和释放[4,36].

亚硝酸盐浓度对温室气体的释放也有影响,亚 硝酸盐浓度较高时,污水处理系统中 ORP 值上升, 抑制甲烷的产生. 有研究表明,抑制甲烷产生的亚 硝酸盐浓度阈值为 8 mg·L-1[37,38]. 而亚硝酸盐的 浓度与氧化亚氮的产生和释放有直接联系. 亚硝酸 盐浓度的增加会导致硝化过程中氨氧化菌反硝化作 用的增强,该过程会将亚硝酸盐还原为 N,O^[26];在 反硝化期间高浓度的亚硝酸盐会导致反硝化速率降 低并造成一氧化氮和氧化亚氮的积累[27],进而引起 氧化亚氮释放量的升高.

系统中亚硝酸盐的浓度还与污泥中微生物种群 结构直接相关,由于 AOB 和 NOB 数量比的差异,造 成了两种工艺中氨氧化速率和亚硝酸盐氧化速率的 不同,造成了不同系统中亚硝酸盐的浓度差异. 已 经有研究指出[39],在 Orbal 氧化沟污泥中,参与硝化 过程的两类主要细菌氨氧化细菌(AOB)和亚硝酸 盐氧化细菌(NOB)的数量相当,而在 A/A/O 工艺 中, AOB 的数量约为 NOB 的两倍, 且优势菌属 Nitrosomonas europaea 已经被证实能够进行反硝化, 而硝化菌的反硝化过程会产生氧化亚氮.

4 结论

西安市第三污水处理厂(Orbal 氧化沟工艺)每m³进水释放甲烷1181 mg(以 CH4 计)、氧化亚氮36.20 mg(以 N2O 计),西安市第四污水处理厂(A/A/O 工艺)每m³进水释放甲烷209 mg(以 CH4计)、氧化亚氮54.64 mg(以 N2O 计).两个污水处理厂非二氧化碳温室气体的二氧化碳当量分别为每m³进水40.31 g(以 CO2 计,三污)和21.51 g(以 CO2 计,四污).对比两个污水处理工艺,Orbal 氧化沟工艺的温室气体释放量高于A/A/O 工艺,因此从非二氧化碳温室气体释放角度考虑,A/A/O 工艺更具优势.影响污水厂非二氧化碳温室气体产生释放的主要因素是温度、曝气方式和溶解氧浓度.此外,活性污泥的产甲烷活性和亚硝酸盐氧化速率也是影响甲烷及氧化亚氮释放的重要原因.

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