

論文 / 著書情報
Article / Book Information

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| 題目(和文) | 安定同位体比を用いた下水処理過程における一酸化二窒素の生成・消滅機構の研究 |
| Title(English) | A study of nitrous oxide production and consumption mechanisms in wastewater treatment system using stable isotopic analysis |
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| 出典(和文) | 学位:博士(理学), 学位授与機関:東京工業大学, 報告番号:甲第9611号, 授与年月日:2014年9月25日, 学位の種別:課程博士, 審査員:吉田 尚弘,小島 英理,山田 桂太,豊田 栄,丹治 保典 |
| Citation(English) | Degree:Doctor (Science), Conferring organization: Tokyo Institute of Technology, Report number:甲第9611号, Conferred date:2014/9/25, Degree Type:Course doctor, Examiner:,,,, |
| 学位種別(和文) | 博士論文 |
| Category(English) | Doctoral Thesis |
| 種別(和文) | 要約 |
| Type(English) | Outline |

Thesis Outline

Nitrous oxide (N_2O), a potent greenhouse gas that is important in exacerbating climate change, is also a stratospheric ozone depleting substance. It is producible in substantial amounts during biological nitrogen removal processes in wastewater treatment plants where inorganic nitrogen compounds are converted to NO_3^- and N_2 gas by bacterial nitrification and denitrification. Moreover, N_2O can also be produced by partial nitrification (PN) during PN process followed by an anaerobic ammonium oxidation (anammox) process (PN-anammox process), which has several advantages such as no need for external carbon addition and less energy and oxygen requirement. To identify the key pathways of N_2O production, the nitrogen and oxygen stable isotope ratios are useful tools for the analysis of N_2O production and consumption pathways because they can be determined according to isotope ratios of the precursor materials. After the development of analytical methods for intramolecular distribution of ^{15}N in asymmetric NNO molecule, ^{15}N -site preference (SP) was added to bulk N and O isotope ratios as a new parameter to interpret N_2O production mechanisms and to estimate the global N_2O budget. SP, which is independent of the substrate's isotopic signature, has unique values reflecting on microbial production pathways. Regarding wastewater treatment systems, a few investigations described on production mechanisms and controlling factors of N_2O in a complex bacterial system associated with N_2O isotopomer ratios. However, more isotopic studies are needed to understand the relative contributions from each N_2O production pathway and the occurrence of N_2O reduction in wastewater treatment operated under different conditions. For instance, there are no studies that examined isotopic fractionation during N_2O reduction by activated sludge of WWTP.

Thus, the overall aim of this Ph.D research was undertaken (1) to elucidate the production and consumption pathways of N_2O in a real wastewater treatment by conventional activated sludge system under different oxygen conditions, (2) to investigate the dependence of the production and consumption mechanisms of N_2O during lab-scale wastewater treatment simulated with nitrification and denitrification on controlling factors that are known to affect N_2O production, including dissolved oxygen (DO), carbon to nitrogen ration (C/N), nitrogen (N) loading, and water temperature using a laboratory batch-scale reactor, (3) to reveal the effect of mixed liquor suspended solid (MLSS) and estimate the enrichment factors (ϵ) during N_2O reduction in N_2O decomposition experiment, and (4) to study the production mechanisms of N_2O in a PN aerobic granule reactor, respectively.

During real observational analysis in an aerobic reaction tank of conventional activated sludge treatment at WWTP located in Tokyo, aeration tank water was supersaturated with N_2O . The highest value, $3700 \text{ nmol kg}^{-1}$, was observed at the aeration tank end and in settled sludge under the lowest aeration condition. About 0.03% of the influent NH_4^+ was emitted as gaseous N_2O at the lowest aeration condition. The conversion rate was 0.14% under the highest aeration

condition. The SP values were significantly higher at the middle and end of aeration tanks under the highest aeration condition, but were nearly zero or slightly negative under lower aeration conditions. Under the highest aeration condition, NH_2OH oxidation (nitrification) was the main contributor to N_2O production of about 90% and 57%, respectively, at the aeration tank middle and end. At other sampling points, 55–63% of the N_2O was produced by bacterial NO_2^- reduction (nitrifier-denitrification) with a lower nitrification contribution. For all sampling points in lower aeration experiments, NO_2^- reduction was a major N_2O production pathway.

During lab-scale simulation experiments, in the course of anoxic N_2O decomposition experiment, N_2O reduction rate was affected by the high MLSS concentration. A positive correlation between $\delta^{18}\text{O}$ and $\delta^{15}\text{N}^{\text{bulk}}$ (slope = 2.21) and between SP and $^{15}\text{N}^{\text{bulk}}$ (slope = 0.94), that can be used as a characteristic for the occurrence of N_2O reduction process, was found. Moreover, isotopic enrichment factors, ϵ , which can be useful tracer to each reaction pathways, for N_2O reduction were estimated to be at -9.5‰ for the $\delta^{15}\text{N}^{\text{bulk}}$, -28.7‰ for the $\delta^{18}\text{O}$ and -10.0‰ for the SP of N_2O , respectively. In oxic (nitrification) condition, lower DO and higher N-loading cause greater N_2O accumulation. Observed low ^{15}N -site preference (SP, -2.6 –7.8‰ at 25°C and -7.2 – 9.2‰ at 18°C) of N_2O and relationship of nitrogen isotope ratios between N_2O and its substrate (NH_4^+) suggested that N_2O was produced mainly by NO_2^- reduction by autotrophic nitrifiers (nitrifier–denitrification). The N_2O production mechanism in this condition was not altered by the changes in DO and N-loading. In an anoxic condition (denitrification), NO_2^- reduction by denitrifying bacteria (heterotrophic denitrification) was dominant contributor to N_2O production, and N_2O reduction to N_2 occurred simultaneously, as implied by isotopomer analysis. The C/N ratio and N-loading had negligible effect on N_2O production mechanism. In an aerobic PN experiment, about 5.6% of the NH_4^+ load was converted and emitted as N_2O in the presence of ammonium and acetate. Analysis of dynamic concentration profiles during one cycle (4 h) of the sequence batch airlift reactor (SBAR) operation demonstrated that N_2O concentration in off-gas was increased up to around 400 ppm (v/v) at 1 hour after the beginning of aeration. Isotopomer analysis was identified the main N_2O production pathway in this reactor during one cycle. Results suggest that the majority (approximately 70-80%) of the N_2O produced was originated from NO_2^- reduction (heterotrophic denitrification) rather than NH_2OH oxidation.