II-446 Production of Nitrous Oxide from Nitrite in Denitrification

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INTRODUCTION

Nitrification -denitrification process has been used in various kind of wastewater treatment. However, there is possibility that N2O (greenhouse gas) is released to the atmosphere in nitrification as a by-product and in defitrification as an intermediate.

The laboratory-scale experiments were conducted to examine the N2O production during denitrification. 1) Low COD/NO3-N, short SRT and low pH were favorite conditions for N2O production. High content of N2O in nitrogenous gas was often found together with accumulation of NO2 or with left over of NO3 concurrently in both continuous and batch experiments. It seems that high concentration of NO2 brings release of N2O. However, as both NO2 and N2O are intermediates in denitrification, these two can appear together during an incomplete denitrification even if there is no direct relationship between them. therefore, the batch experiments were conducted to investigate the effect of substrate concentration and different type of sludge on N2O production.

MATERIALS AND METHODS

68ml serum vials were used. 40ml of mixed liquor from the continuous reactor in steady state condition was transferred to a vial and mixed with 10ml of various substrate solution. An anoxic incubation was conducted at 25°C. N2O gas in head space was determined by a gas chromatography equipped with a thermal conductivity detector using helium as carrier gas. NO2-N and NO3-N were determined by a liquid chromatography.

For study of the substrate concentration on N2O production, the mixed liquor was taken from the continuous reactor fed with COD/NO3-N of 2.5 at an SRT of 3 days. To investigate the effect of different type of sludge, the sludge taken from the continuous reactor fed with COD/NO3-N of 3.5 at an SRT of 5days was used for a comparison. The substrate contained acetate and yeast exact (CODcr ratio fixed at 10:1) as carbon source. Nitrite was used as nitrogen source. NO2-N was added so that COD/NO3-O was 0.53, which is equivalent to the COD/NO3-N ratio of 1.5. The composition of substrate is shown in Table 1.

Table 1 compsition of substrate

Case	COD (mg/l)	NO2-N (mg/l)
1	1000	1111
2	400	444
3	200	222
4	50	56
KH_2PO_4		200mg/l
$CaCL_2.2H_2O$		30mg/l

RESULTS AND DISCUSSION

Effect of substrate concentration on N2O production

N2O production per volume of liquid with substrate having the highest concentration at initial TOC of 393mg/l and NO2-N of 1028mg/l is shown in Fig.1-1. Removal of NO2-N and NO3-N (concentration of NO3-N which remained in the sludge was about 100mg/l), removal of TOC which represents the removal of organic matter obtained in the substrate are shown in Fig.1-2 and Fig.1-3. During the first 26 hours, only 6% of NO2-N, 11% of NO3-N and 30% of TOC were removed. In this period, just 10% of N2O as compared with the final amount was observed. The various analyzed parameters showed that denitrification was not vary active in this step. It might be due to the inhibitory effect of high concentration of NO2 on organisms. After this stagnate step, the active degradation of organic matter and consumption of nitrogen took place. N2O-N dramatically increased in this period up to the highest value of 187mg/l which accounts for 37% of consumed nitrogen.

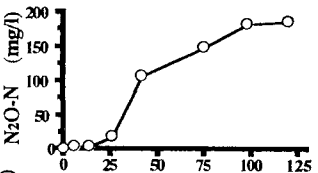

Fig.1-1 N_2O accumulation

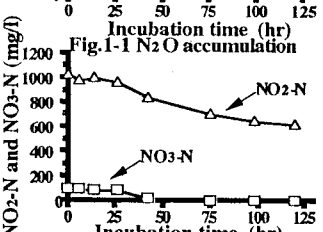

Fig.1-2 Remained NO_2 and NO_3

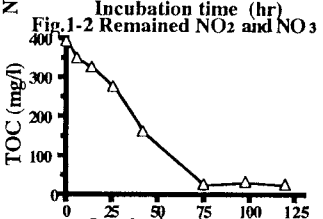

Fig.1-3 TOC degradation

Fig. 2-1 to Fig.2-3 show the denitrification during the batch incubation with reduced nitrogen and organic matter concentration, which was 40% of the first case. At the 26th hour, the concentration of N2O reached over half of the final value. 82% of TOC, 62% of NO3-N, 21% of NO2-N were removed. After most of organic matter had been consumed, with gradual reduction of NO2, concentration of N2O increased until it reached relatively steady state. The final conversion of N2O from the consumed nitrogen was 23%, removal of NO2-N and NO3-N were 33% and 64%, respectively. The concentration of organic matter and nitrogen were further reduced to 20% and 5% of the first case, the behavior of N2O production and degradation pattern of nitrogen plus organic matter were also observed. The concentration of N2O did not show the difference from these two cases, however, the conversion of N2O in case 4 was found higher than case 3. It seems that with low concentration of organic matter, NO2 is more easily used as electron donor by organisms than NO3. In other word, high concentration of organic matter gives high removal of NO3.

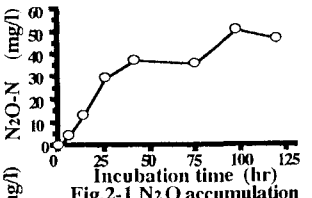

Fig.2-1 N2O accumulation

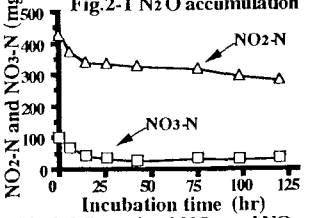

Fig.2-2 Remained NO2 and NO3

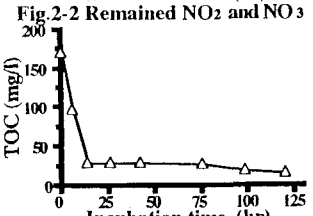

Fig.2-3 TOC degradation

The concentration of N2O-N and N2O conversion as consumed nitrogen with various substrate concentration is shown in Fig.3. N2O production seems to be divided into two steps. In the first step, N2O is released together with active denitrification and high N2O producing rate. The second step takes place after organic matter has been consumed, with using remained nitrogen, N2O released gradually proceeds till it reaches a relatively steady state condition. With high substrate concentration, the degradation rate of organic matter was much smaller than the other 3 cases. The active denitrification step could be kept longer, at the same time, the highest N2O producing rate and conversion were found in this case.

Effect of type of sludge on N2O production

Fig.4 show the batch experimental results in which sludge having no ability to produce N2O during the continuous experiment was used with same substrate as case 1. 10mg/l of N2O was detected in head space, and only about 2% of removed nitrogen was converted as N2O. The degradation rate of nitrogen and organic matter were also bigger than case 1. These differences are suggested comes from the sludge source. The sludge fed with just satisfactory carbon source in the substrate might have small amount of N2O producing species compared with the sludge fed with insufficient carbon. N2O-producing species were perhaps developed and accumulated with favorite condition (low COD/NO3-N, or short SRT). The bacteria which produce N2O in continuous operation can continue producing N2O in batch experiments. N2O production in batch incubation depending on species of bacteria was also observed in previous studies.

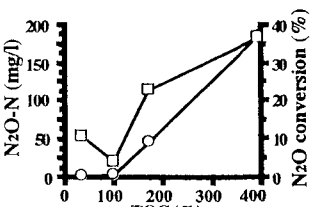

Fig. 3 N2O-N concentration and conversion

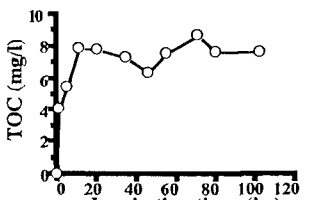

Fig.4 TOC degradation

SUMMARY

High concentration of substrate can cause high amount of N2O production, up to 37% of consumed nitrogen converted. production of N2O depends on species of bacteria, with the organisms which can produce N2O during the continuous operation, high N2O producing rate was found together with active denitrification in batch experiments. With limited NO3, high concentration of substrate brings high removal of NO3-N, with low concentration of organic matter, NO2 is easily used by organisms and further it might cause high conversion of N2O. Excess nitrogen (when carbon source is completely consumed) is favorite condition for N2O remaining in gas phase.

REFERENCE: K.HANAKI, Z.HONG and T.MATSUO (1992). Production of nitrous oxide gas during denitrification of wastewater. IAWPRC, 16th Biennial conference.