Study Status and Prospect of N_2O Production and Reduction Control in Wastewater Biological Denitrification

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Abstract. Nitrous oxide （N_2O）is one kind of potent greenhouse gas which can destroy ozone layer and lead to acid rain. Furthermore, nitrous oxide is quite easy to be converted into the secondary particle pollutants which can aggravate haze in certain weather conditions. Effective control of N_2O emission has become imminent and the researches on N_2O reduction have been get more and more attention. Related researches indicate that the process of wastewater biological denitrification is one of main artificial sources of N_2O emission. And with the popularity of wastewater biological denitrification process, the emission quantity and harmness of N_2O increases gradually. In this work, the mechanism of N_2O generation in wastewater nitrification and denitrification process is revealed and the various influence factors to N_2O emission is summarized. And then, the present reduction control techniques of N_2O is analyzed in detail. On these basises, the future research directions of N_2O reduction control is prospected.

Introduction

With the rapid development of industrial and agricultural and mass consumption of fossil fuels, the greenhouse gases emissions increased year by year, which causing the global warming problem increasingly prominent. The current studies are generally believed that the three main greenhouse gases are carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O). N_2O can exist in the atmosphere stability in 114 years and its global warming potential (GWP) is approximately 298 ~ 320 times higher than CO_2 and 24 times higher than CH_4 although N_2O is trace gases. In addition, N_2O not only can destroy ozone layer and lead to acid rain,but also quite easy to be converted into the secondary particle pollutants to aggravate haze in certain weather conditions. The artificial cause is the immediate cause of its concentration increases according to the researches showed recently[1].

The studies have found that N_2O is a by-product of nitrification process and is intermediate product of denitrification[2]. The denitrification of denitrifying bacteria is the main process of N_2O production that is generate hydroxylamine (NH_2OH) and nitroxyl (NOH), which will promote to the production of N_2O under the action of active enzymes respectivly[3]. However, in terms of the denitrification process, the nitrous oxide reductase (Nos) activity of denitrifying bacteria is the decisive factor affecting N_2O, so keeping the higher activity of Nos is the key to achieve N_2O reduction of nitrogen(N_2), and it is likely that many factors have an impact on the activity of Nos. While some researches show that part of denitrifying bacteria have no Nos enzyme system.

In consideration of the serious environmental caused by N_2O emissions, in the process of sewage treatment of N_2O emissions control effectively has become a research hotspot in the field of air and water pollution control. Numerous factors can affact the production of N_2O like DO, C/N, pH, temperature, sludge retention time. Therefore, we should take many factors into consideration about the control theory research and the technology development on the emmision of N_2O. Recently our researches on control the emission of N_2O mostly concentrated in improving water quality, optimizing treatment process and operational states, regulating the microbial population structure in the sewage biological denitrification process. Nevertheless, in terms of engineering practice, it is not practical

operability to control the reduction strategies of N_2O by improving water quality and optimizing treatment process. Then the researches and practical of the reduction strategies of N_2O mainly focused on the optimization of operating conditions and regulation the microbial population structure in wastewater biological nitrogen removal process. We can reveal the kinetics regularity of N_2O producted and the microbial population succession ruls by researching the mechanism of N_2O producted and influence factors , and then we can come up emissions control strategies of N_2O.

Mechanism of N_2O production in wastewater biological nitrogen removal process.

It's have been seen that some intermediate products and by-products will be producted by different processes or crafts in wastewater removal process like NO, N_2O, N_2, N_4O_2.

The mechanism of N_2O production in wastewater nitrification process

Nitrification is mainly composed of nitrogen element in the following form:

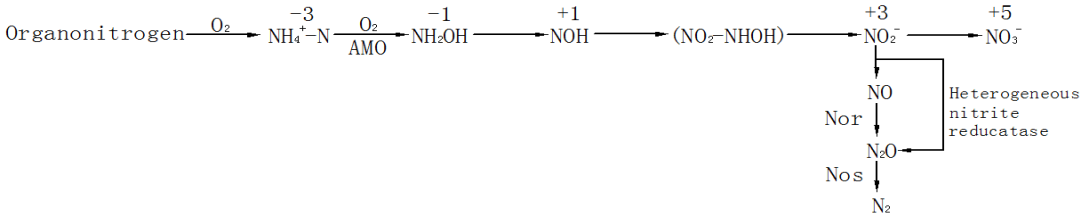

Fig.1 The mechanism of N_2O production in wastewater nitrification process

The researches have shown that N_2O is a by-product and mostly producted in denitrification of nitrification process. There are some product forms of N_2O in the nitrification process as follows:

a) The oxidation of NH_2OH and NOH. NH_2OH and NOH are the intermediate products.

Numberous researches have shown that a large number of NH_2OH could be oxidized under the hydroxylamine oxidoreductase if ammonia nitrogen concentration or pH is too high. However, the process cannot be particular biochemical explanted, which is need to be further demonstrated . NH_2OH could be transformed into NOH that be transformed into $NO_2{\cdot}NHOH$, and then $NO_2{\cdot}NHOH$ be hydrolyzed to N_2O by double molecular polymerization. However, this process cannot be happened because the NH_4^+ could not be oxidized under the condition of anaerobic[4].

b) The existence of aerobic denitrifying bacteria.

Poth and Focht[5] confirmed for the first time, when the lake of DO lead to the steady accumulation of NO_2^-, and then nitrifying bacteria come up alienation denitrification reaction to transform to N_2 by the accumulation of NO_2^-. Related Studies have also found that the production of N_2O is much more by revivifing NO_3^- of under the aerobic conditions than by revivifing NO_2^- under the condition of oxygen by denitrifying bacteria.

c) The heterotrophic nitrification

Stdied by Papen and others[6] found that discharge of N_2O in heterotrophic bacteria heterotrophic nitrification process is two orders of magnitude more than the oxygen release nitration process after discovering the existence of heterotrophic bacteria in the process of nitrification. Meanwhile, the concentration of DO have influenced on the N_2O emission by heterotrophic nitrification bacteria.

The mechanism of N_2O production in wastewater denitrification process

There are many intermediate in denitrification process, such as NH_2OH, N_2O, HNO. The denitrification process is showed as follows:

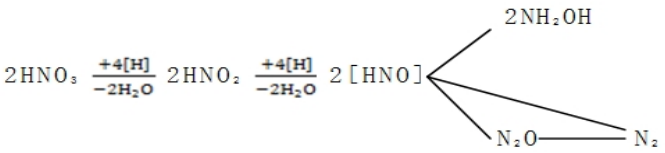

Fig. 2 The mechanism of N_2O production in wastewater denitrification process

The production of N_2O count on the activity of Nos. The overwhelming majority of Nos is a kind of soluble protein that active centre of its have cuprum.This is the reason why Nos have different on oxidation-reduction quality, spectral properties and enzyme activity[7]. On the one hand, there are

many factors that can affact the active of Nos like DO, NO_2^- and the concentration of metal ions. On the other hand, some denitrifying bacteria have no Nos system like *Fluorescent pseudomonads*, so N_2O have to release by final product[8].

The influence factors to N_2O emission in wastewater biological nitrogen removal process

We can see that N_2O is a reaction product caused by the incomplete of nitrification and denitrification according the above introductions, so the influence factors of N_2O is more complex. Combining previous research results of scholars both at home and abroad, what will affect the sewage biological denitrification process of N_2O factors summarized as the following aspects.

The concentration of dissolved oxygen

Dissolved oxygen(DO) plays an important role in the production and release of N_2O. Nowdays, most scholars believe that the release of N_2O can increase with the low concentration of DO [9]. There are studied that improve the system of DO also will increase the release quantity of N_2O, which could fell sewage denitrification efficiency. In terms of the denitrification process, the release of N_2O will increase with the increase of DO in the process of denitrification, which will lead to the decline of denitrification. It was found by Schulthess and others[10] from the reseach of the denitrifying activated sludge that the concentration of DO from 0 mg/L up to 4 mg/L, the N_2O accounts for the proportion of denitrifying gaseous products increased from 0% to 6%. On the one hand, the active of Nos could be damged by the DO. On the other hand, the aerobic respiration of denitrifying bacteria rather than the anaerobic respiration, at the same time in the molecular oxygen and N_2O to exist, molecular oxygen is priority in use and inhibit denitrification, and then the efficiency of denitrification is decline.

Temperature

Temperature is one of the important factors on the production of N_2O. The studied by Gejlsbjerg[11] have found that the temperature change will damage the balance between ammonia oxidation and nitrite oxidation in the process of nitrification. Higher reaction temperature can cause the NO_3^- accumulation, which further increases the nitration reaction of the release quantity of N_2O. In terms of denitrifying process, the temperature change nearly have no role in the release of N_2O, this is mainly due to the denitrifying enzyme activity has approximate correlation with temperature.

Carbon nitrogen ratio(C/N)

Carbon nitrogen ratio(C/N) is decide to degree of denitrifying process, and is one of the main influence factors. The higher the carbon nitrogen ratio, the better denitrifying denitrification effect. We can seen from the mechanism of denitrification that the denitrification reactions need to consume carbon source, so denitrifying bacteria using its endogenous carbon for denitrification when lack of exogenous carbon, and then the denitrification process is difficult to thoroughly, which result in the accumulation of NO_3^- and increase the release of N_2O. In the other words, the low carbon nitrogen ratio will lead to the increase emmision of N_2O. Currently, there are many explanations that low carbon nitrogen ratio lead to increase the emmision of N_2O, what is recognized as mentioned earsier that the lack of electron acceptor in denitrification reaction can not thoroughly, or electron acceptor serious lack of NO_2^- accumulation, and then make the N_2O gas produces[12].

Research progress on N_2O reduction control

Scholars at home and abroad also has carried out a lot of research works on the wastewater biological denitrification N_2O emissions control field. Nowadays, the researches and practices of N_2O reduction control of sewage biological denitrification process mainly focus on optimizing operation condition and regulation microbial population structure.

Hu and others[13] studied emission sources of N_2O by the chemical inhibition method, and the results showed that the main prodution source of N_2O is denitrification of nitrifying bacteria in the aerobic period. Lloyd and others[14] found that denitrifying bacteria in aerobic conditions reducing NO_3^- to generate the amount of N_2O than anoxic conditions have risen sharply, and then the production of nitrogen is decreased.

The studied by Hanaki and others[2] found that the N_2O production were higher at the DO of 0.5 mg/L, and the discharge of N_2O is least at the DO of 1.7 mg/L. Itokawa and others[15] studied the influence of carbon nitrogen ratio on the N_2O production, and the result showed that the discharge of N_2O is least at the carbon nitrogen ratio of 7.5, which took the removal efficiency of wastewater biological nitrogen into consideration.

Conclusions and Prospects

The release of N_2O is a very complex process, which involving the process by participation of various enzymes. There are two suggestions for researches of contronling the production and reduction of N_2O in the process of wastewater biological nitrogen removal.

Further defined the mechanism of N_2O, and then revealing its dynamic rules and analysising the relevant key dynamic factors of N_2O emission. Then we can guide and control the emissions of N_2O in the sewage biological denitrification process from the perspective of dynamic, and then the feasible control strategy will be formed to control the emmision of N_2O.

We can in-depth analysis the microbial population succession law that related to the produceof N_2O in the wastewater biological nitrogen removal process, and in the protection of wastewater biological nitrogen pollutants removal efficiency to screen dominant populations under the premise of relying on the means of molecular biology, which can realize N_2O reduction control. It is guidance the reduction of N_2O from the perspective of microbial ecology.

Acknowledgements

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