

Temperature effect on aerobic denitrification and nitrification

XIE Shu-guang^{*}, ZHANG Xiao-jian, WANG Zhan-sheng

(Department of Environmental Science and Technology, Tsinghua University, Beijing 100084, China. E-mail: xsg00@mails.tsinghua.edu.cn)

Abstract: Nitrogen loss without organic removal in biofilter was observed and its possible reason was explained. A lower hydraulic loading could improve aerobic denitrification rate. Aerobic denitrification was seriously affected by low temperature (below 10°C). However, nitrification rate remained high when the temperature dropped from 15°C to 5°C. It seemed the autotrophic biofilm in BAF could alleviate the adverse effect of low temperature.

Keywords: aerobic denitrification; nitrification; biofilter; temperature effect

Introduction

Nitrogen removal has become an important aspect of water and wastewater treatment. Two different populations are involved in nitrification, ammonia and nitrite oxidizers. It is commonly believed that ammonium and nitrite oxidation always happen together. Under normal conditions, nitrate is generally the only end product of nitrification. However, some environmental conditions can favor high nitrite accumulation, such as low temperature (Randall, 1984a), elevated pH (Suthersan, 1986) and low dissolved oxygen (Alleman, 1984). DO concentration is an important factor for nitrification. Jayamohan *et al.* (Jayamohan, 1988) reported that continuous nitrification under low DO leads to a high nitrite accumulation. Nicolas *et al.* (Nicolas, 2001) suggested nitrite oxidizer's growth rate decreases more than ammonia oxidizers under low DO concentrations and nitrite accumulation could occur. Recently aerobic deammonification or aerobic denitrification with low demand of oxygen has been of great interest for the treatment of nitrogen rich wastewater (e.g. industrial water or landfill leachate) (Helmer, 1999). The operative microorganisms are assumed to be autotrophic populations, which could denitrify under low DO conditions. This kind of oxygen-limited autotrophic nitrification-denitrification (OLAND) system can directly remove ammonium without converting it into nitrate.

However, OLAND processes are always used to treated high-strength ammonia wastewater. Reports concerning aerobic denitrification in low-strength ammonia surface or river water treatment processes are sparse. Moreover, surface or river water temperatures vary seasonally. Therefore, the temperature effect on biological treatment must be seriously considered, especially in cold climate. However, temperature on aerobic denitrification has never been reported up to date. It was the aim of this study to investigate the temperature effect on aerobic denitrification. The temperature effect on nitrification was also discussed.

1 Materials and methods

1.1 Experimental setup

A novel process (Fig.1) was designed and used to investigate the feasibility of biological pretreatment of river water entering into the Guanting Reservoir (China). All reactors consisted of Plexiglas column with a 20 cm inside diameter and of 300 cm length. The filters were filled with ceramic particles up to 1.2 m depth. Ceramic particles had an average diameter of 3—5 mm, a porosity of 0.09, a density of 1560 kg/m³, and a specific surface area of 2.5 m²/cm³. Biofilter I and Biofilter II were nonaerated, while Biofilter III was aerated. All reactors were provided with liquid sampling ports at different height.

The aim of this process was: Biofilter II was to achieve aerobic denitrification at low oxygen concentrations (about 1 mg/L); Biofilter I was to removal organic carbon and reduce the DO concentration entering into Biofilter II; Biofilter III was to remove residual organic carbon and convert residual ammonium into nitrate.

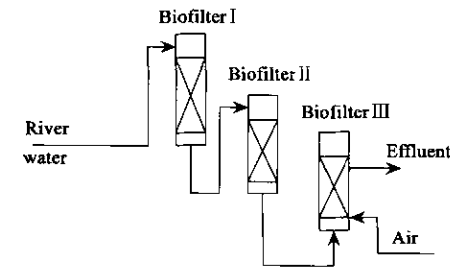


Fig.1 Experimental setup of the combined biofilters system

The study was performed near an ammonia-rich river and the filters were directly fed with river water. The study was conducted after an initial start-up period of more than 50 days to assure the biomass establishment (during this period, the water temperatures were about 15–30 °C). The river water temperature gradually dropped from 15 to 5 °C.

1.2 Analysis

For all liquid samples ammonia, nitrite and nitrate concentrations were measured according to standard methods (APHA, 1995). Organic carbon concentration was indirectly expressed by KMnO₄ index(WWMA, 1988). Temperature and

dissolved oxygen(DO) values were determined using selective electrodes. All liquid samples were taken and measured under steady conditions at different temperatures.

2 Results and discussion

2.1 Aerobic denitrification and nitrification

Some nitrogen loss without organic carbon removal under low oxygen concentrations was observed as shown in Table 1. When Biofilter I effluent entered into Biofilter II, its dissolved oxygen (DO) concentration was slightly raised as the result of natural regeneration of oxygen. In this study the numbers in bracket all indicated the real DO concentration of water entering into Biofilter II. Table 2 demonstrates fluctuation of nitrite concentration in Biofilter II, indicating that nitrite was an intermediate during nitrogen loss.

Table 1 Operating result of the whole process under hydraulic loading of 5 m³/(m²·h) at 15 °C

	River water	Biofilter I effluent	Biofilter II effluent	Biofilter III effluent
COD _{Mn} , mg/L	9.2	7.6	7.6	6.2
DO, mg/L	2.3	0.6(1.3)	0.7	5.0
NH ₄ -N, mg/L	7.6	7.6	5.0	0.1
NO ₂ -N, mg/L	0.4	0.4	0.2	0.0
NO ₃ -N, mg/L	0.0	0.0	0.0	5.1
Total inorganic nitrogen, mg/L	8.0	8.0	5.2	5.2

This kind of nitrogen loss could not be explained by conventional heterotrophic denitrifying activity within anoxic biofilm microniches. The reasons were as follows: There was no consumption of organic carbon which was needed by conventional denitrification; therefore these nitrogen losses were the activity of aerobic denitrification, which could be regarded as a kind of autotrophic denitrification. One possible hypothesis for nitrogen loss without organic carbon at low DO concentration was presented by Siegrist *et al.*

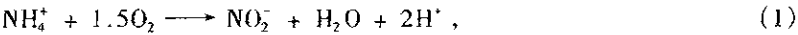
Table 2 Nitrogen conversion profile in Biofilter II under hydraulic loading of 5 m³/(m²·h) at 15 °C

	L ₀ , inlet	L _{0.2}	L _{0.6}	L _{1.0}	L _{1.2} , outlet
DO, mg/L	1.4	1.3	1.0	0.7	0.6
NH ₄ -N, mg/l.	8.0	7.6	6.7	5.9	5.5
NO ₂ -N, mg/L	0.5	0.6	0.4	0.5	0.2
NO ₃ -N, mg/l.	0.2	0.2	0.2	0.2	0.2

Notes: L₀, L_{0.2}, L_{0.6}, L_{1.0} and L_{1.2} represented the sequential liquid sampling ports, 0, 0.2, 0.6, 1.0 and 1.2 m, from the top of filter bed respectively

(Siegrist, 1998).

Nitrite, produced in the aerobic biofilm layer by *Nitrosomonas* close to the surface, diffuses into the anoxic layer below and is reduced in parallel with ammonium oxidation.



In a biofilm system, the fast-growing bacteria tend to occupy the surface of the biofilm(Wanner, 1986). This feature favor ammonia oxidizers. Under low DO conditions, oxygen will be the limiting substrate, only available at the surface of the biofilm due to the mass transfer limitation(Dennac, 1983). The ratio of ammonia oxidizers to nitrite oxidizers should increase with the growth of the biofilm until ammonia oxidizers colonize the whole surface of the biofilm. The evolution of the ecology in biofilm should be the disappearance of nitrite oxidizers and growth of ammonia oxidizers, at least on upper layers(Nicolas, 2001). DO concentration was a key factor for aerobic denitrification. If it were too high, nitrite produced by ammonia would be converted into nitrate. However, if it were too low conventional denitrification would happen instead of aerobic denitrification or ammonia oxidizers could not be enriched. A proper maintenance of DO concentration was essential for aerobic denitrification.

Biofilter Ⅲ was used to further remove residual organic carbon and ammonium(nitrification). As to biological aerated filter(BAF) autotrophic bacteria(nitrifying bacteria) and heterotrophic bacteria compete for space and oxygen. When influent organic carbon concentration is low, the growth of heterotrophic bacteria will be limited with the gradual reduction of organic carbon. Therefore, autotrophic bacteria (nitrifying bacteria) could also become predominant organism. Simultaneous organic carbon and ammonium removal were observed as shown in Table 1 and Table 3.

2.2 Temperature effect

Operating results of the whole process under hydraulic loading of 3 m³/(m²·h) at different temperature are shown in Table 4—8. In comparison with the results shown in Table 1, Table 4 indicates a much higher nitrogen loss at low hydraulic loading. Aerobic denitrifiers in Biofilter Ⅱ could only remove ammonium and nitrite. Therefore, we took the difference of total amount of ammonium and nitrite between influent and effluent as the amount as nitrogen loss. Temperature effect on aerobic denitrification is shown in Fig.2. When the temperature was above 10℃, its effect on aerobic denitrification was not very significant. However, aerobic denitrification was seriously affected when the temperature was below 10℃. Therefore, temperature is also an important factor for aerobic denitrification.

Table 3 Simultaneous organic carbon and ammonium removal profile in Biofilter Ⅲ under hydraulic loading of 5 m³/(m²·h) at 15 ℃

	<i>L</i> ₀ , inlet	<i>L</i> _{0.2}	<i>L</i> _{0.6}	<i>L</i> _{1.0}	<i>L</i> _{1.2} , outlet
COD _{Mn} , mg/L	7.8	6.8	6.2	6.2	6.2
NH ₄ -N, mg/L	5.8	4.3	0.2	0.0	0.0

Notes: *L*₀, *L*_{0.2}, *L*_{0.6}, *L*_{1.0} and *L*_{1.2} represented the sequential liquid sampling ports, 0,0.2, 0.6, 1.0 and 1.2 m, from the bottom of filter bed respectively

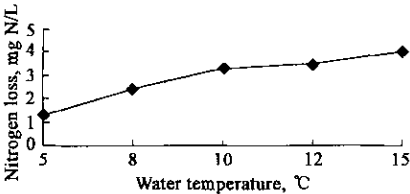


Fig.2 Nitrogen losses in Biofilter Ⅱ at different temperature

Table 4 Operating result of the whole process under hydraulic loading of 3 m³/(m²·h) at 15℃

	River water	Biofilter Ⅰ effluent	Biofilter Ⅱ effluent	Biofilter Ⅲ effluent
COD _{Mn} , mg/l.	9.0	7.2	7.2	6.0
DO, mg/L	2.5	0.7(1.3)	0.7	5.2
NH ₄ -N, mg/L	7.8	7.7	3.9	0.0
NO ₂ -N, mg/L	0.1	0.2	0.0	0.0
NO ₃ -N, mg/L	0.1	0.1	0.1	4.0
Total inorganic nitrogen, mg/L	8.0	8.0	4.0	4.0

Table 5 Operating result of the whole process under hydraulic loading of 3 m³/(m²·h) at 12℃

	River water	Biofilter I effluent	Biofilter II effluent	Biofilter III effluent
COD _{Mn} , mg/L	16.8	13.0	13.0	10.0
DO, mg/L	2.8	0.4(1.4)	0.8	5.4
NH ₄ -N, mg/L	10.0	10.0	6.6	0.1
NO ₂ -N, mg/L	0.3	0.3	0.2	0.0
NO ₃ -N, mg/L	0.1	0.1	0.1	6.8
Total inorganic nitrogen, mg/L	10.4	10.4	6.9	6.9

Table 6 Operating result of the whole process under hydraulic loading of 3 m³/(m²·h) at 10℃

	River water	Biofilter I effluent	Biofilter II effluent	Biofilter III effluent
COD _{Mn} , mg/L	9.4	7.8	7.8	6.0
DO, mg/L	3.0	1.0(1.5)	0.8	5.0
NH ₄ -N, mg/L	7.3	7.2	4.0	0.4
NO ₂ -N, mg/L	0.3	0.4	0.3	0.0
NO ₃ -N, mg/L	0.1	0.1	0.1	4.0
Total inorganic nitrogen, mg/L	7.7	7.7	4.4	4.4

Table 7 Operating result of the whole process under hydraulic loading of 3 m³/(m²·h) at 8℃

	River water	Biofilter I effluent	Biofilter II effluent	Biofilter III effluent
COD _{Mn} , mg/L	9.1	8.2	8.2	5.5
DO, mg/L	2.9	0.7(1.2)	0.6	5.2
NH ₄ -N, mg/L	8.2	8.0	5.7	0.6
NO ₂ -N, mg/L	0.4	0.6	0.5	0.0
NO ₃ -N, mg/L	0.4	0.4	0.4	6.0
Total inorganic nitrogen, mg/L	9.0	9.0	6.6	6.6

Table 8 Operating result of the whole process under hydraulic loading of 3 m³/(m²·h) at 5℃

	River water	Biofilter I effluent	Biofilter II effluent	Biofilter III effluent
COD _{Mn} , mg/L	12.2	9.4	9.4	8.0
DO, mg/L	3.0	0.9(1.3)	0.8	5.5
NH ₄ -N, mg/L	7.2	6.9	5.1	0.9
NO ₂ -N, mg/L	0.2	0.5	1.0	0.0
NO ₃ -N, mg/L	0.9	0.9	0.9	6.1
Total inorganic nitrogen, mg/L	8.3	8.3	7.0	7.0

It is commonly known that nitrification rate significantly decreases at low temperature. It was reported an 85% nitrification rate was achieved at 5℃ in BAF(Rogalla, 1991). Nitrification rate remained high when the temperature dropped from 15℃ to 5℃ as shown in Table 4—8. It seemed the autotrophic biofilm in BAF could alleviate the temperature effect.

Moreover, it seemed aerobic denitrifiers in Biofilter II were more sensitive to temperature change than nitrifiers in Biofilter III. One possible hypothesis is that the ammonia-oxidizing bacteria in Biofilter II and Biofilter III were different. The former was more easily affected by low temperature. Helmer *et al.* (Helmer, 1999) had revealed the presence of large microcolonies of at least three different types of ammonia-oxidizing bacteria in those biofilm regions where extremely high nitrogen losses occurred using in situ hybridization with 16s rRNA-Targeted probes.

3 Conclusions

A novel process was designed and used to investigate the feasibility of biological pretreatment of river water entering into the Guanting Reservoir. Nitrogen loss without organic removal under low oxygen concentrations in Biofilter II was observed and its possible reason was explained. A lower hydraulic loading could stimulate aerobic denitrification rate. When the temperature was above 10℃, its effect on

aerobic denitrification was not very significant. However, aerobic denitrification was seriously affected below temperature (below 10°C). Nitrification rate remained high when the temperature dropped from 15°C to 5°C. It seemed the autotrophic biofilm in BAF alleviated the adverse effect of low temperature. Moreover, it seemed aerobic denitrifiers in Biofilter II were more sensitive to temperature change than nitrifiers in Biofilter III.

References:

- Alleman J E, 1984. Elevated nitrite occurrence in biological wastewater treatment systems[J]. *Wat Sci Tech*, 17(2/3): 409—419.
- APHA, 1995. Standard methods for the examination of water and wastewater[M]. 19th ed. Washington, DC, USA: American Public Health Association.
- Dennac M, Uzman S, Tanaka H *et al.*, 1983. Modeling of experiments on biofilm penetration effects in a fluidized bed nitrification reactor[J]. *Biotechnol Bioengrg*, 25:1841—1861.
- Helmer C, Kunst S, Juretschko S *et al.*, 1999. Nitrogen loss in a nitrifying biofilm system[J]. *Wat Sci Tech*, 39(7): 13—21.
- Jayamohan S, Ohgaki S, Hanaki K, 1988. Effect of DO on kinetics of nitrification[J]. *Water Supply*, 6:141—150.
- Nicolas B, Peng D C, Jean-Philippe D *et al.*, 2001. Nitrification at low oxygen concentration in biofilm reactor[J]. *Journal of Environmental Engineering*, 127(3): 266—271.
- Randall C W, Buth D, 1984. Nitrite build-up in activated sludge resulting from temperature effects[J]. *J Water Pollut Control Fed*, 56(9): 1039—1044.
- Rogalla F, Payraudeau M, Sauvergrain P *et al.*, 1991. Reduced hydraulic detention time for complete nutrient removal with innovative biological reactors[J]. *Wat Sci Tech*, 24(10): 217—229.
- Siegrist H, Reithaar S, Lais P, 1998. Nitrogen loss in a nitrifying rotating contactor treating ammonium rich leachate without organic carbon [J]. *Wat Sci Tech*, 37(4—5): 589—591.
- Suthersan S, Ganczarczyk J J, 1986. Inhibition of nitrite oxidation during nitrification: some observations[J]. *Water Pollut Res J Can*, 21(2): 257—265.
- Wanner O, Gujer W, 1986. A multispecies biofilm model[J]. *Biotechnol Bioengrg*, 28:314—328.
- WWMA, 1988. Methods for the examination of water and wastewater[M]. 3th ed. Beijing: China Environmental Science Press.

(Received for review June 28, 2002. Accepted August 6, 2002)