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The control of point and non-point source nitrogen to prevent eutrophication of the Nakdong River basin, Korea

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Abstract. Eutrophication of surface waters is commonly caused by excessive inputs of nutrients such as nitrogen and phosphorus. Nakdong River basin was chosen as the study area to investigate the effect of point and non-point source pollution of nitrogen on eutrophication in water body. Non-point source inputs of nitrogen accounted for approximately 84% in the total nitrogen input of the upper Nakdong river watershed, which mainly consists of agricultural land and forests. However, point source inputs of nitrogen accounted for 58~85% in the total nitrogen input of the middle and lower watersheds, including urban area. Therefore, for watershed near urban area, control of point source inputs of nitrogen may be an optimal method to control eutrophication. In this respect, the enforcing reduction of nitrogen in the final effluent of wastewater treatment facilities is needed. On the other hand, to enact more stringent nitrogen regulations, the LOT (limit of technology) and environmental impact should be considered. In this study nitrogen data were analyzed to propose new nitrogen regulations.

Keywords: dissolved organic nitrogen; eutrophication; limit of technology; nitrogen budget; nitrogen regulation

1. Introduction

Excessive nitrogen and phosphorus inputs from urban and agricultural areas have accelerated eutrophication of water bodies (Bergstrom and Jansson 2006, Duce *et al*. 2008). The ratio of nitrogen to other nutrient was reported to be one of the main factors influencing the composition of phytoplankton communities (Domingues *et al*. 2011). The high N: Si ratio can stimulate diatoms growth (Sommer 1994, Roberts *et al*. 2003), and the N:P ratio affects the growth of cyanobacteria (Orr and Jones 1998, Watanabe and Oishi 1985). The biochemical processes of nitrogen cycle in the environment include nitrogen fixation, nitrogen assimilation, ammonification, nitrification, and denitrification (Tchobanoglous *et al*. 2003). The several different chemical forms of nitrogen converted by the biogeochemical cycle enter the water body. During the cycle, nitrogen (N_2) in the atmosphere is synthesized as ammonia (NH_3/NH_4^+) through nitrogen-fixing bacteria. Ammonia introduced into the basin undergoes nitrification/denitrification and is converted into nitrite (NO_2^-) and nitrate (NO_3^-), and is present in the water. These nitrogen sources can be diffused into the water system through a point or non-point sources. Nitrogen remaining in water promotes the growth of microorganisms as well as aquatic plants with phosphorus and is known to be a major source of water pollution such as eutrophication (Pedersen and Borum 1996, Schindler 2006). Algal blooms due to eutrophication have been more frequent than in the past, expanded globally (Peperzak 2003, Heisler *et al.* 2008). In particular, the increase in artificially generated nutrients, such as inorganic chemical fertilizers, induces an algal bloom, increasing the difficulty of algae management. In Korea, the phosphorus concentration from municipal wastewater treatment plants (MWWTPs) effluent is strongly regulated at 0.2~0.5 mg/L, albeit the nitrogen is still regulated at a relatively high concentration above 20 mg/L.

Rockstrom *et al*. (2009) reported that nitrogen accumulation in aquatic environment was around three times higher than phosphorus accumulation due to excessive use of inorganic fertilizers. On the other hand, beside dissolved inorganic nitrogen (DIN), dissolved organic nitrogen (DON) has been considered as a contributor influencing algal blooming (Carlsson *et al*. 1993, Seitzinger and Sanders 1997, John and Flynn 1999).

For USA, stringent regulations for N and P in effluent discharges from public sewage treatment facilities have set according to national pollutant discharge elimination system (NPDES). For European countries, nutrients discharge has been regulated by water quality standard of water framework directive (WFD). In case of nitrogen accumulation, although Republic of Korea has ranked among the top countries in OECD countries (Howarth 2008), there is little understanding of the effects of different forms of nitrogen on water environment.

Especially, Korea has strongly regulated the phosphorus concentration from municipal wastewater treatment plants since 2012, there is growing concerned about high DON inputs because public sewage treatment works are

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combined with various industrial wastewater treatments (Randall 2006). In this study, nitrogen discharge load was estimated for the Nakdong River basin, the representative river of Republic of Korea. Also, based on the characteristics of nitrogen pollution sources and wastewater treatment plants near the river basin, new nitrogen regulations were proposed.

2. Characterization of the Nakdong river basin and its nitrogen budget

Nakdong river (525 km of total flow length and 32,280 km^2 of watershed area) is the longest river in Republic of Korea (Fig. 1). There are 5 multipurpose dams and 8 dams (less than 15 m in height), where contaminants can accumulate in the water because of stagnant zones created by dams. Also, since many urban areas such as Gumi, Daegu and Busan cities are located and industrial complexes are clustered near the river, water may be susceptible to contamination. In addition, a certain amount of organic nitrogen and nitrate can enter the river because of agricultural and forest lands distributed over the whole surface. These characteristics of the Nakdong river system have caused high fluctuation of nitrogen concentration, resulting in challenges of nitrogen management (Jung *et al.* 2019).

To characterize total nitrogen (TN) input to the Nakdong river basin, its fluxes were classified into two categories: point and non-point sources. In addition, the annual trend of TN, total phosphorus (TP), and Chl-a concentrations was estimated by using the information gathered from 10 points of telemonitoring system (TMS) in the Nakdong River basin, national water quality automatic system of ministry of environment (MOE), Republic of Korea (Fig. 1). Estimation of TN inputs from point source was made by the sum of the daily average discharges from MWWTPs and industrial wastewater treatment plants (IWWTPs) surveyed in 2017 by the Ministry of Environment. It has been found that the contribution of IWWTPs and small-scale MWWTP spread over the basin was less than 5% of the total discharge, so MWWTPs with a capacity of more than 500 m^3/d located in the study area were selected. In addition, all IWWTPs in the study area selected.

On the other hand, TN inputs from non-point source are transported by storm water and other runoff (Choi *et al.* 2019). The TN non-point sources include fertilizer application, soil erosion, and agricultural operations, and runoff from roads. It is difficult and complicated to determine non-point source pollutant loads, so non-point TN and TP loads in this study were estimated using the load method based on the land use types such as agricultural, forest, and urban areas (Table 1).

3. Nitrogen and phosphorus inputs to the Nakdong river basin

The point source (PS) and non-point source (NPS) flowing into the Nakdong river basin could be classified as the municipal and industrial wastewater treatment plant and

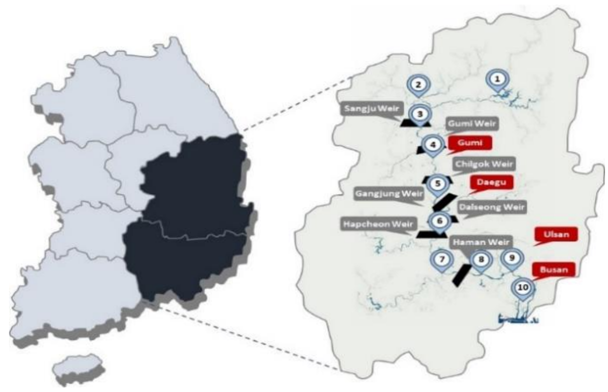

Fig. 1 Water TMS locations of the study area (Nakdong River basin)

Table 1 The standard TN and TP loading rate of each areas according to the load method (https://wamis.go.kr)

	Agriculture ($g/m^3/d$)	Forest ($g/m^3/d$)	Urban ($g/m^3/d$)	Etc. ($g/m^3/d$)
TN	8.0	2.20	0.06	13.69
TP	0.43	0.14	0.03	2.10

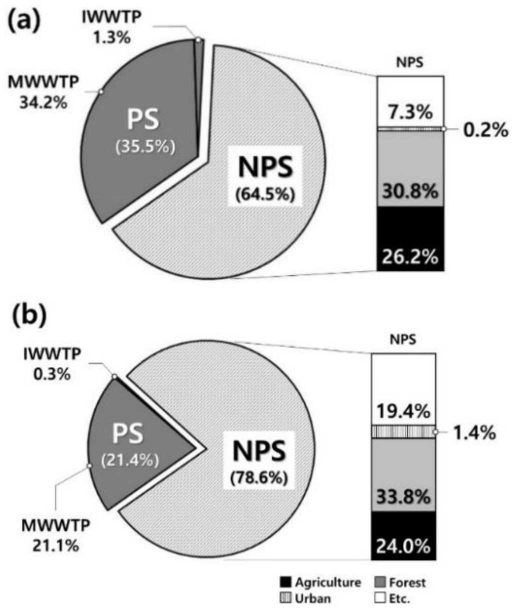

Fig. 2 Source composition of TN (a) and TP (b) in the Nakdong River basin

agriculture, forest, urban area, respectively. The minor distribution contains roads, railroads, wildland, as well as livestock and agricultural wastewater from small facilities (Choi *et al.* 2019). For non-point source pollution, TN and TP inputs were estimated to be 64.5% and 78.6% of total nitrogen and phosphorous loads, respectively. The TN inputs to the river basin seemed to be primarily attributed to nutrient runoff from agriculture and forest lands (Fig. 2). This result is in good agreement with the literature (Wu and Chen, 2013). TP inputs from point sources have been reduced by approximately 21.4% of the total TP load before the new regulations effective. Similarly, TP are mainly attributed to nutrient runoff from agriculture and forest lands.

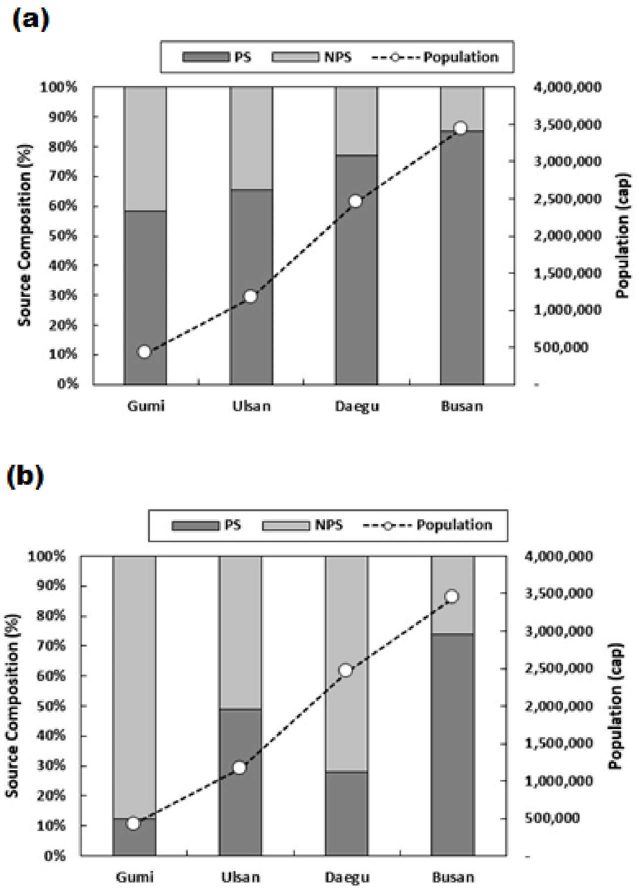

Fig. 3 Source composition of TN (a) and TP (b) in urban area

Nutrient (TN or TP) source composition in different urban areas located the Nakdong river basin are shown in Fig. 3. Contrary to the tendency of the entire the Nakdong river basin, TN inputs from point sources in urban areas were estimated to be 58~85%, which were higher than those from non-point sources. Also, the trend of higher TN inputs from point sources compared to non-point sources were clear as population size increases. This agrees with the previous literature where the ammonia and nitrate discharge trends from urban non-point sources are significantly different from non-urban areas such as agricultural area (Lee *et al.* 2019). In case of TP inputs from point sources, as population size increases, the trend of higher TP inputs from point sources compared to non-point sources was shown, but less significant compared to TN. These results imply that effective control of TN discharges from MWWTPs as well as nutrients from non-point sources is required for the water quality improvement of the river basin.

4. Algal blooms in the Nakdong river basin

Based on the average monthly TN data gathered from TMS in 2015, the average annual TN concentrations ranged between 1.3 and 5.1 mg/L. These values are maintained at a relatively high concentration in the winter season (November to February) (Fig. 4).

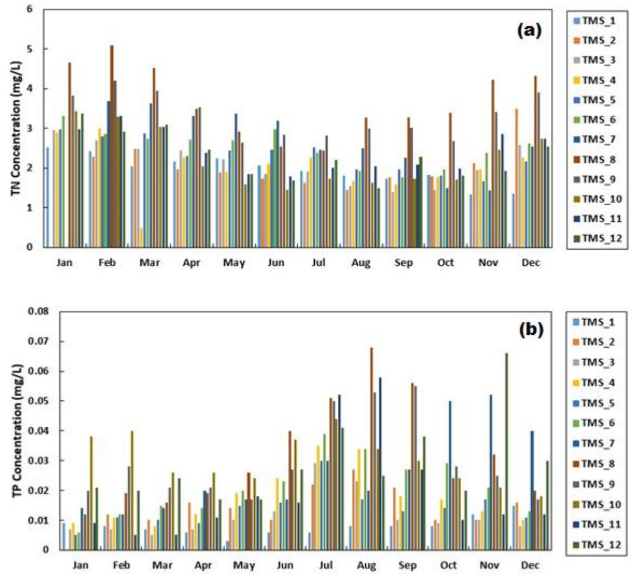

Fig. 4 Average monthly concentrations of TN (a) and TP (b) in Nakdong River basin

However, TN concentrations were lower than the average monthly TN concentration during the rainy season from June to August. For urban areas such as Gumi and Daegu, high TN concentrations were observed at TMS locations 8 and 9, where tributary streams flow into the mainstream. After strengthening the effluent standard in sewage treatment plants in 2012, average annual TP concentrations for all TMS locations ranged from 0.01 to 0.06 mg/L, which were 10 times less than those for the past few years (Lee *et al*. 2002). Unlike TN, the variation of TP concentrations was relatively small (Fig. 4). However, it shows that strong rainfall during the rainy season (July to August) can increase the phosphorus discharge from the NPS, increasing the TP loads up to 0.08 mg/L. Based on the annual trend of TN and TP concentrations gathered from each TMS location, Nakdong River basin can be classified as eutrophication level (Dodds *et al*. 1998).

Most average monthly Chl-a concentrations in the area of the most dams located in the river exceeded 15 mg/m^3 of algal bloom alert standard. Fig. 5 shows average monthly Chl-a concentrations for Sangju, Dalsung, and Haman dams.

For Sangju dam (TMS 3) which is located in the upper course of the river, average monthly Chl-a concentrations were lower than 15 mg/m^3 except for April, June, and September. This low level Chl-a concentration may be attributed to low TN inputs from urban areas. Also, this result is likely due to DON which is the predominant species in the forms of nitrogen from forest area. However, for Dalsung (TMS 6) and Haman dams (TMS 8) in the middle and lowest courses of the river, respectively, most average monthly Chl-a concentrations exceeded 15 mg/m^3 of except for November and December. This result may be attributed to high TN inputs from urban areas such as Gumi and Daegu, leading to nitrogen accumulation followed by eutrophication. Regarding seasonal variation of Chl-a concentration in 2015, Chl-a concentrations for Sangju and Dalsung dams increased during the period of February to

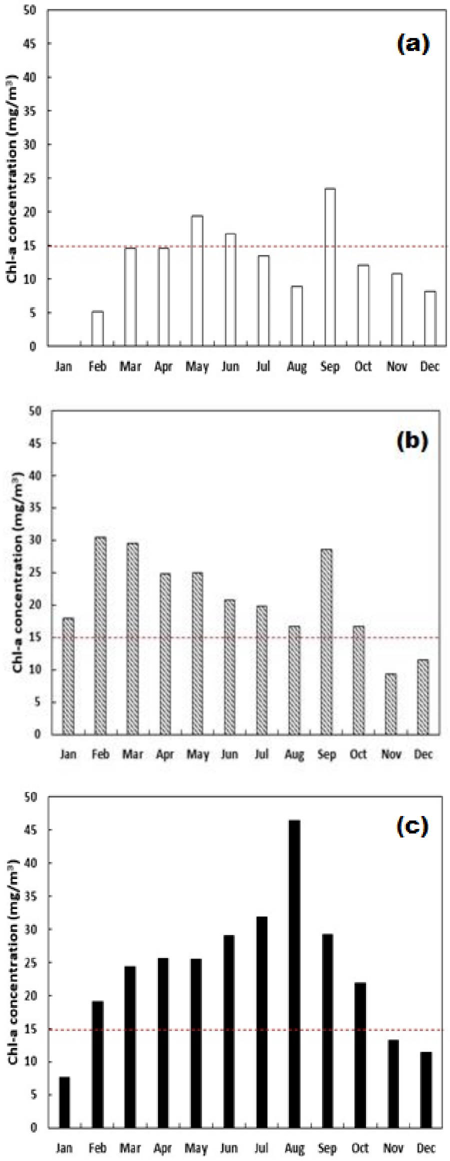

Fig. 5 Phytoplankton concentration in the Nakdong River basin: (a) Sangju, (b) Dalsung, and (c) Haman dam

May, but they decreased during the rainy season from June to August. For Haman dam in the lowest course of the river, Chl-a concentration gradually increased from February, and reached the highest level in August. This result implies that the degree of nutrient accumulation has a greater influence on Chl-a concentration than rainfall amounts in the lower course of a river. Also, the decrease in flow rate in the lower course may have influence on Chl-a concentration.

Fig. 6 shows average monthly concentration of cyanobacteria in the river basin. From the upper to lower course of the river, average monthly concentration of cyanobacteria increased at the all dams located in the river. Cyanobacteria concentration at Haman dam in the lowest course was higher than other dams.

On the other hand, the relation between Chl-a concentration and N/P ratio is shown in Fig. 7. Since strengthening the MWWTP effluent TP standard from 2.0 to 0.2 mg/L in 2012, the N/P ratio has increased. Nevertheless,

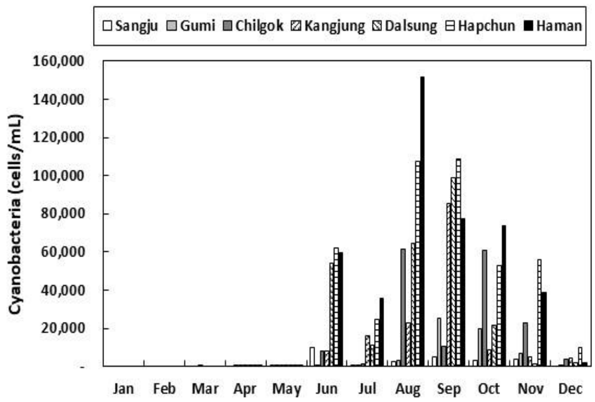

Fig. 6 Cyanobacteria concentration in the Nakdong river (Water Environment Information System, MOE, https://water.neir.go.kr)

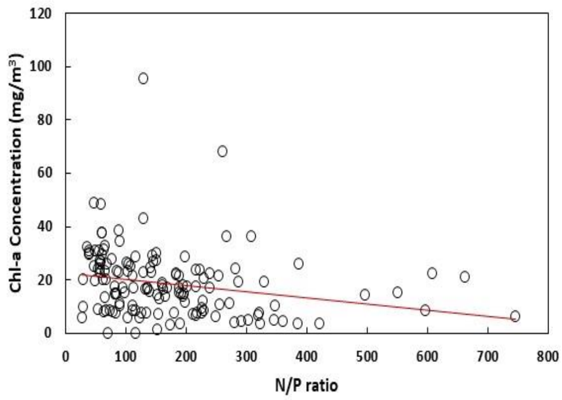

Fig. 7 Relation between Chl-a concentration and N/P ratio

algae tended to occur at high concentrations at N/P ratios of less than 200. In other words, to prevent unbalanced N/P ratios and to control algal blooms, it can be indicated that the appropriate nitrogen management to maintains an N/P ratio between 200 and 300 may help.

5. Present status of public sewage treatment plants in the Nakdong river basin

Effluent water quality standards of wastewater treatment plants (WWTPs) in the world are summarized in Table 2. In case of USA, site-specific criteria incorporating watershed features and maximum load have been proposed for TN and TP according to NPDES (Sedlak, 1991). For Japan, basically the government set uniform national standards applicable to all public waters. In addition, TN and TP water quality criteria of wastewater treatment plants are established for various levels according to their treatment processes. For example, TN and TP water quality standards of wastewater treatment plants near Biwa lake ranged from 6.1 to 10.0 mg/L and 0.05 to 0.5 mg/L, respectively. In case of Germany, effluent water quality standards of WWTPs were set based on the capacities of wastewater treatment plants. However, uniform national water quality standards for discharge of all WWTPs were set in Republic of Korea.

The water quality standards for effluent TN of public sewage treatment plants need to be developed according to LOT (limit of technology). Also, the standards need to be set depending on the current state of each water body. To achieve very low TN and TP levels, technology based nutrient limitations need to be considered in many advanced wastewater treatment processes, which are summarized in Table 3 (Pargilla 2009). Among the processes, Johannesburg shows the highest TN removal efficiency.

Table 2 Effluent water quality standards of wastewater treatment plants (WWTPs)

Countries	City	WWTP	TN (mg/L)	TP (mg/L)
Korea			20	0.2
Japan[1]	Shiga (Lake Biwa)	Konan Jubu	6.7	0.1
		Kosei	6.1	0.1
		Tohoku	6.7	0.1
		Takashima	8.0	0.1
USA[2]	San Diego, California	Pad Dam	1.0	0.1
	Arkansas	Fayetteville	2.0	1.0
			5.0	1.0
	Madison, Wisconsin	Nine Springs	1.8	-
			4.1	-
	Connecticut	Danbury	1.9	1.0
			4.0	1.5
	New Hampshire	Keene	2.1	0.2
			12.0	10.0
	Florida	Hookers Point	3.0	7.5
		River Oaks	1.2	0.4
		Reno-Sparks	5.0	0.4
	Maryland	Chesapeak Bay Tributaries	8.0	2.0
	New Jersey	Landies Sewerage Authority	0.5	-
Germany[3]			15.0	2.0
			10.0	1.0

[1]Department of sewer system in Tokyo, 2012 (www.shiganogesui.jp)
[2]Sedlak R.I., 1991
[3]German Federal Environment Agency, 2013

Table 3 Limit of technologies for advanced treatment processes for removal of TN (Pargilla 2009)

Process	Influent conc.(mg/L)	Effluent conc.(mg/L)	Efficiency (%)
Johannesburg	7.86	2.03	74.2
A2O[1]	10	7.3	27.0
MBBR[2]	10	5.8	42.0
MLE[3]	4.35	2.2	49.4
4-stage Bardenpho	5	3.5	30.0
SBR[4]	4.59	1.6	65.1
Step-feed AS[5]	5.25	3.7	29.5
Biological aerated filters	3.61	1.4	61.2
5-stage Bardenpho	2.32	1.24	46.6

[1]A2O = anaerobic/anoxic/oxic
[2]MBBR = moving-bed biofilm reactor
[3]MLE = modified Ludzak-Ettinger
[4]SBR = sequencing batch reactor
[5]AS = activated sludge

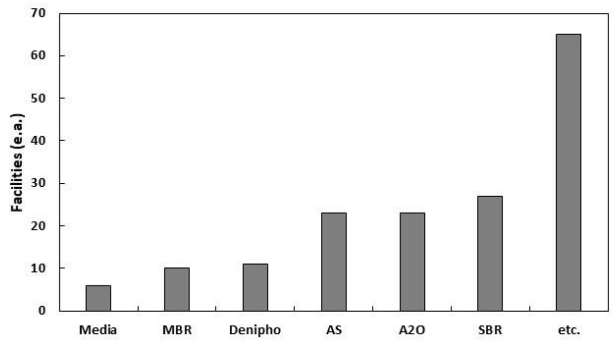

Fig. 8 Number of WWTP process in the Nakdong river basin (MBR: membrane bioreactor, AS: activated sludge, A2O: anaerobic/anoxic/oxic, SBR: sequencing batch reactor)

On the other hand, Fig. 8 shows 165 MWWTPs with a capacity of more than 500 m^3/d in the Nakdong river basin were classified according to treatment processes. Among various treatment processes, commonly used treatment options included AS (activated sludge), A2O (anaerobic/anoxic/oxic), and SBR (sequencing batch reactor). SBR was reported to be able to meet nitrogen level less than 5 mg/L (Grady *et al.* 1999). Either A2O or AC was reported to meet nitrogen level less than 10 mg/L. However, either A2O or AC combined with disc filtration was able to meet nitrogen level less than 3 mg/L.

In order to strengthen the maximum contaminant level of TN in effluent of sewage treatment was set at 5 mg/L in Nitrogen river basin, a combination of disk filtration and current processes such as A2O and AC can be an alternative. Further, novel technologies need to be developed for meeting future stringent water quality standards. According to WERF report, to meet as low as 3 mg/L TN in effluent of sewage treatment plants, greenhouse gas emission increased upto 1,000 ton/year (Falk *et al.* 2011).

Total nitrogen is comprised of TDN (total dissolved nitrogen) and TPN (total particulate nitrogen). TDN is comprised of DIN (dissolved inorganic nitrogen) and DON (dissolved organic nitrogen). In general, DIN includes NH_4^+, NO_2^-, and NO_3^- (Tchobanoglous *et al.* 2003). TPN can be removed by physical wastewater treatment units such as filtration and sedimentation, whereas, DIN can be removed by biological wastewater treatment processes. It is difficult to remove DON attributed to agricultural and industrial activities by conventional wastewater treatment systems. Therefore, DON represents most of the dissolved nitrogen in the effluent of wastewater treatment plants. Range of DON concentrations for various water systems are summarized in Table 4. For main sewage treatment plants in USA, DON accounted for more than 60% of TN, ranging from 0.5 to 2.8 mg/L. In case of Japan and Netherlands, DON concentrations in lakes ranged from 0.05 to 1.8 mg/L. The content of DON in lake and river basin may be affected by vegetation type. For Republic of Korea, there is no reference regarding DON concentration in river, but some references on DON in sewage treatment plants. DON in the influent of sewage treatment plants ranged from 1.6 to 3.2 mg/L, most of which was discharged to water environment

Table 4 Range of DON concentrations in selected water bodies

Countries	DON(mg/L)	Reference
USA (Effluent)	0.5 ~ 2.8	Jimenez *et al*. 2007
USA (Rivers)	0.3 ~ 1.9	Seitzinger and Sanders 1997
Japan (Lake Biwa)	0.05 ~ 0.1	Mitamura and Matsumoto 1981
Netherlands (Lake Ysel)	0.6 ~ 1.8	Wetzel 2001
Korea (sewage)	1.6 ~ 3.2	Im and Gil, 2017

because it was not effectively removed. Therefore, DON control is also important to maintaining the eutrophication in Nitrogen river basin. As part of efforts to treat DON, studies on characteristics and distribution of DON in the river basin need to be preceded.

6. Conclusions

For the control of eutrophication in Nitrogen river basin, depending on LOT and environmental characteristics, the effluent water quality standards for TN need to be set. Especially, effective removal treatment of DON is of great concern for public sewage treatment practices since it cannot be effectively removed by wastewater treatment processes. DON removal efficiency of only 30% was reported to be achieved in biological nutrient treatment processes. Therefore, technology innovation in wastewater treatment processes is needed for effective treatment of DON. In addition, for the control of eutrophication at this time NH_4^+-N management is still important because NH_4^+-N accounts for 60~70% of TN in the effluent of sewage treatment plants. The removal efficiency of NH_4^+-N is reported to be 25~50% in biological nutrient removal process. In view of NH_4^+-N removal efficiency, effluent TN concentration of 20 mg/L can be at an appropriate level because little nitrification occurs during cold winter season, however, effluent TN concentration may be maintained at 8 mg/L during summer season.

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