Estimation of Nitrogen Loading to Surface Water from Agriculture Based Area and Its Application for Water Pollution Mitigation

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ABSTRACT

Eutrophication of surface water is a globally widespread environmental problem. Similarly, in Thailand, the Nakhon Nayok River (NNR), located in an agriculture-based area, has been markedly affected by eutrophication problems. However, there are limited studies on significant nitrogen (N) sources during agricultural activities in the area. Therefore, this study examined the major sources and key flows of N loading to the surface water by applying material flow analysis (MFA) to the relevant seven subsystems in 2018. The results showed that aquaculture and rice cultivation were the main sources of nitrogen inputs and outputs. Both considerably contributed to the nitrogen loading to the surface water, yet nitrogen released from the aquaculture was five times higher than the rice cultivation. The nitrogen flux found in the study area was 0.11 kg/ha. Accordingly, creating wetlands for aquaculture wastewater treatment that could potentially remove nitrogen by 12% was recommended.

1. INTRODUCTION

Eutrophication is a global problem (WHO and EC, 2002; Smith and Schindler, 2009). Agriculture was reported to have significantly accelerated eutrophication (FAO and IWMI, 2017). For example, fertilizer application doubled the level of global nitrogen fixation (Vitousek et al., 1997) and the excess N from land flowing to surface water can promote algal blooms that affect surface water quality due to hypoxia (oxygen depletion) (Chislock et al., 2013). Drinking water produced from the water contaminated with toxins released from some algae is a risk to human health (WHO and EC, 2002).

There were 34 occurrences of eutrophication reported in the Gulf of Thailand in 2008-2009 (Marine Knowledge Hub, 2020). In 2016, eutrophication killed plenty of fish (MCRC, 2020) and impacted the economy especially for tourism and aquaculture. The NNR basin is in the agriculture-based area and empties into the Bang Pakong River Basin connected to the Gulf of Thailand. The use of N as the eutrophication indicator in the river, suggested by Yang et al. (2008), disclosed that the NNR had a high risk of eutrophication because total N concentration exceeded 300 µg/L (Kammuang, 2010; REO7, 2020).

Additionally, the ammonia and nitrite concentrations in groundwater in Nakhon Nayok were greater than the standard (DGR, 2006). Although the nitrate concentration (34 mg/L) was allowable by the drinking water standard, this value higher than 27.4 mg/L in Wisconsin where the blue baby syndrome occurred (Knobeloch and Anderson, 2000).

In Thailand, point source pollutants such as industrial wastewater are controlled by regulations (e.g., MNRE, 2016); however, the wastewater standard does not focus on total nitrogen pollution. On the other hand, non-point sources possibly caused by the leaching and runoff of nutrients from soil in agricultural area are more difficult to control. Several complaints indicated that water pollution in the NNR basin is from agricultural wastewater such as pig farming (TPBS, 2017), rice field (DOF, 2019), and duck farming (NPLO, 2020). Hence, the NNR water quality has been classified as deteriorated especially in the agricultural and municipal area (REO7, 2017; REO7, 2020).

Using MFA could help provide comprehensive information about sources, flows, and sinks of N (Brunner and Rechberger, 2004). Several studies agreed that MFA is a powerful tool for nutrient

management in the watershed (Schaffner et al., 2011; Kupkanchanakul et al., 2015; Alvarez et al., 2018; Ta et al., 2018). MFA results can be used to support environmental protection (Wang et al., 2015), to recognize environmental problems early (Elshkaki and Graedel, 2013), and to establish priority for environmental measures (Kwonpongsagoon et al., 2007; Schaffner et al., 2009; Wongsoonthornchai et al., 2016). Thus, MFA was chosen to investigate the N flows in the NNR basin. The objectives of this study were to indicate the significant sources of N; to estimate the N loading to surface water; and to suggest the mitigation for N management.

2. METHODOLOGY

Material flow analysis (MFA), a systematic model based on the law of mass conservation, was applied in this study to estimate the flows and stocks of nitrogen through products and processes in the NNR basin. This study focuses on nitrogen emissions especially N loading to the surface water and recommendations to decrease the excess nitrogen loading.

2.1 System analysis
2.1.1 Study area

The NNR Basin, located in Nakhon Nayok Province, Central Thailand, covers 212,200 ha with hills in the north and flat toward the south. The land is mainly used for agriculture extending over 54% of the total area, with rice fields occupying 72% of the total agricultural area. The basin is also taken up by roughly 30% forest, 9% community and industrial area, and 7% other areas (water body, grass land, shrub land, and soil pit) (LDD, 2018) (Figure 1).
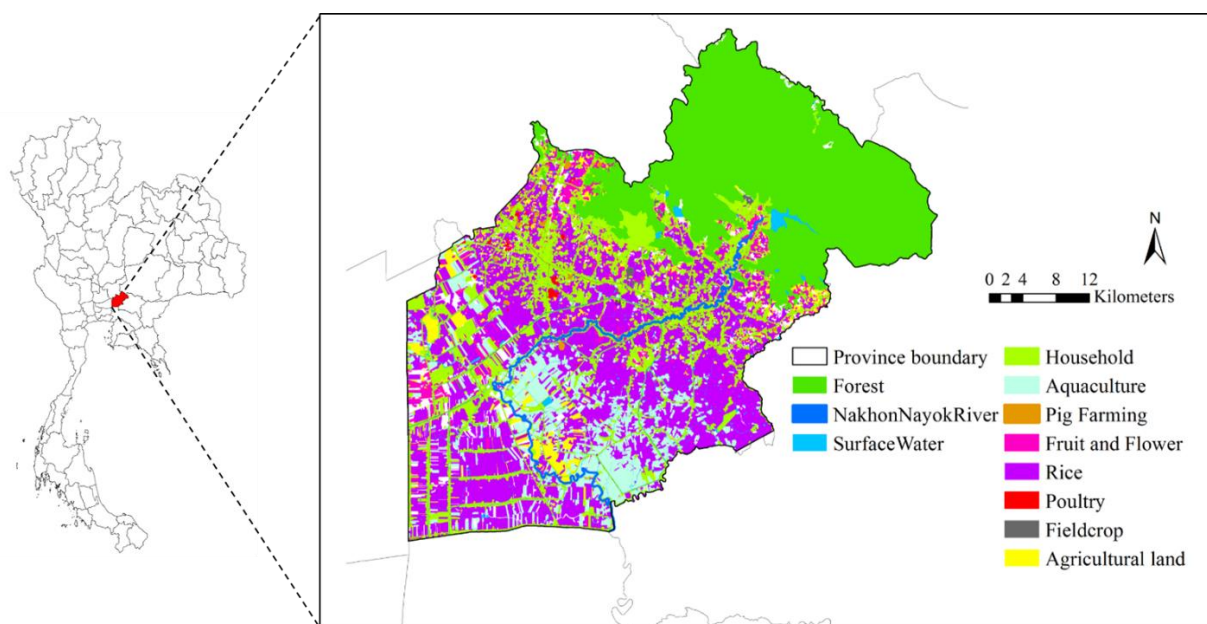

Figure 1. System boundary and land use of the Nakhon Nayok River Basin

The Nakhon Nayok River, the main river in this sub basin, is known as a tourist attraction. The NNR is also an important water source of local people for consumption and recreation. Nevertheless, it has been reported to have the high concentration of N compounds which can lead to eutrophication (Figure 2).

2.1.2 System boundary

The geographic area of this study is the NNR Basin and the data used for estimating N flows were collected in 2018. The seven subsystems were studied based on the MFA methods. The subsystems included (1) rice cultivation; (2) pig farming; (3) aquaculture; (4) poultry farming; (5) field crop cultivation; (6) fruit and flower farming; and (7) households (Figure 2). The N inputs flowing into the rice system included fertilizer ($I_{fert}^{(N)}$), rain ($I_{rain}^{(N)}$), fixation ($I_{fix}^{(N)}$), and water irrigation ($I_{irri}^{(N)}$). The outflows were emission ($O_{emiss}^{(N)}$), rice yield ($O_{yield}^{(N)}$), and drainage ($O_{drain}^{(N)}$) as shown in Figure 3.
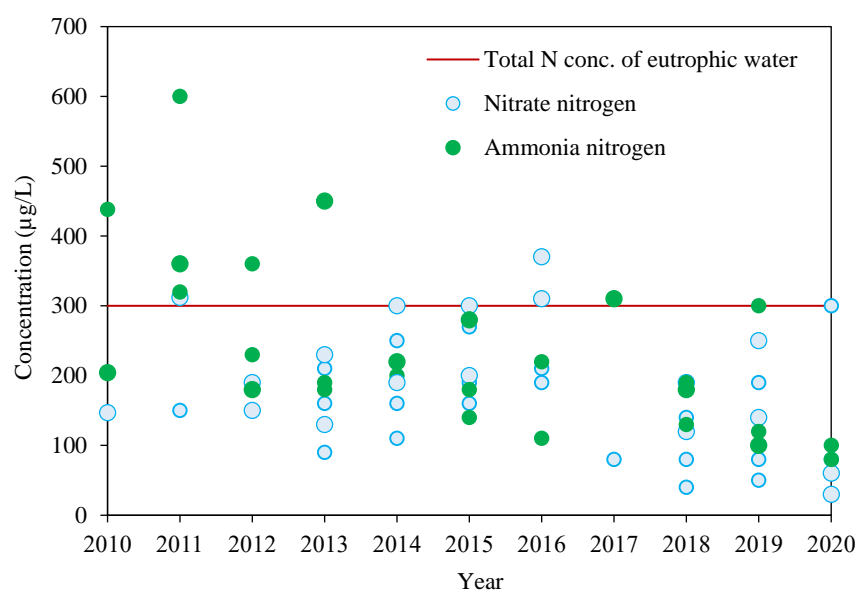
Figure 2. The temporal nitrate nitrogen and ammonia nitrogen in Nakhon Nayok River Basin during 2010-2020 (REO7, 2020).
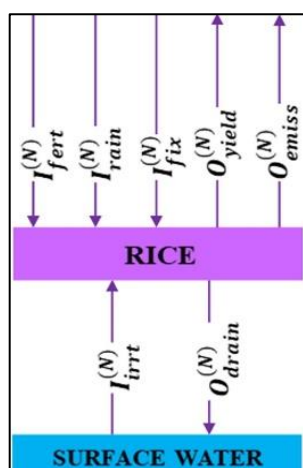

Figure 3. The system analysis of nitrogen flows through the rice system in the Nakhon Nayok River Basin

2.2 Model Approach

The Model approach of this study consists of a general model and specific models based on the mass balance principle. The general model of mass balance is in Equation (1):

$$\sum_{i=1}^{n} m_{input} = \sum_{i=1}^{n} m_{output} + m_{storage} \qquad (1)$$

Where; $\sum_{i=1}^{n} m_{input}$ indicates the sum of all inputs for process i, and $\sum_{i=1}^{n} m_{output}$ is the sum of all outputs for process i. Parameter "m" is the mass flow in a system, and n is the number of processes.

Most specific models of N flows for each subsystem were adopted from Schaffner (2007). To calculate the inflows and outflows of N, the equations and the values of rice cultivation are shown as an example in Table 1 and Table 2, respectively. Scenario analysis was the final step to establish the recommendations based on the key flows of N loading and possibilities in the area.

Table 1. Equations for calculation of nitrogen input through rice cultivation

Variables	Definition	Value	Unit	Reference
Input	$\sum input_{rice}^{(N)} = I_{fert}^{(N)} + I_{rain}^{(N)} + I_{irrg}^{(N)} + I_{fix}^{(N)}$			
Fertilizer	$I_{fert}^{(N)} = P_{freq} \cdot P_{area} \cdot P_{i\_fert} \cdot C_{i\_fert}^{(N)}$			
P_{freq}	Frequency of rice cultivation	2	crop/year	This study
P_{area}	Rice area	81,854	ha	LDD (2018)
P_{i_fert} - 16-20-0 - 46-0-0	 NPK fertilizer Urea fertilizer	 187.29 102.11	 kg/ha-crop kg/ha-crop	 This study This study

Table 1. Equations for calculation of nitrogen input through rice cultivation (cont.)

Variables	Definition	Value	Unit	Reference
$C_{i\_fert}^{(N)}$				Schaffner (2007)
- 16-20-0	N concentration in NPK	160	mg/kg	
- 46-0-0	N concentration in urea	460	mg/kg	
Rainfall	$I_{rain}^{(N)} = P_{rain} \cdot P_{area} \cdot C_{rain}^{(N)}$			
P_{rain}	Rainfall	1,692.9	mm/year	RIHC (2020)
$C_{rain}^{(N)}$	N concentration in rain	3	mg/L	Schaffner (2007)
Irrigation	$I_{irrg}^{(N)} = [M_{evp} + M_{drain} + M_{per} - M_{efr}] \cdot C_{NR}^{(N)}$			
M_{evp}	$M_{evp} = P_{freq} \cdot P_{area} \cdot P_{evap} \cdot 10000$			
M_{dra}	$M_{drain} = (P_{Land} + P_{depth}) \cdot P_{freq} \cdot P_{area} \cdot k_{drain}$			
M_{per}	$M_{per} = P_{perc} \cdot P_{area} \cdot 365$			
M_{efr}	$M_{efr} = (1 - k_{roff}) \cdot P_{area} \cdot P_{rain}$			
P_{evap}	Evapotranspiration	575	mm/crop	DOED (1992)
P_{Land}	Water for land preparation	275	mm/crop	RID (2016)
P_{depth}	Water depth rice	150	mm/crop	Schaffner (2007); DOED (1992)
P_{perc}	Percolation	1	mm/day	Schaffner (2007)
P_{water}	Total water requirement	1,000	mm	DOR (2020)
k_{drain}	TF drain with field water	0.5	-	Schaffner (2007)
k_{roff}	TF rainfall to runoff	0.43	-	
$C_{NR}^{(N)}$	N concentration in water	0.32	mg/L	This study
Fixation	$I_{fix}^{(N)} = P_{area} \cdot P_{freq} \cdot C_{fix}^{(N)}$			
$C_{fix}^{(N)}$	N fixation by rice	5	kg/ha-crop	Schaffner (2007)

Table 2. Equations for calculation of nitrogen output from rice cultivation

Variables	Definition	Value	Unit	Reference
Output	$\sum output = O_{yield}^{(N)} + O_{NR}^{(N)} + O_{emis}^{(N)}$			
Yield	$O_{yield}^{(N)} = P_{freq} \cdot P_{area} \cdot P_{yield} \cdot C_{yield}^{(N)}$			
P_{yield}	Grain yield	5,000	kg/ha-crop	This study
$C_{yield}^{(N)}$	N concentration in grain yield	12	g/kg	Schaffner (2007)
To River	$O_{NR}^{(N)} = O_{roff}^{(N)} + O_{drain}^{(N)}$			
$O_{roff}^{(N)}$	$O_{roff}^{(N)} = k_{roff}^{(N)} \cdot I_{fert}^{(N)}$			
$O_{drain}^{(N)}$	$O_{drain}^{(N)} = k_{drain} \cdot k_{drain}^{(N)} \cdot O_{surplus}^{(N)}$			
$O_{surplus}^{(N)}$	$O_{serplus}^{(N)} = \sum input_{rice}^{(N)} - O_{yield}^{(N)} - M_{residual}^{(N)} + M_{ash}^{(N)}$			
$M_{residual}^{(N)}$	$M_{residual}^{(N)} = M_{rice} \times P_{res} \times P_{res}^{(N)}$			
$M_{ash}^{(N)}$	$M_{ash}^{(N)} = \left(1 - k_{burn}^{(N)}\right) \times k_{burn} \times M_{residual}^{(N)}$			
k_{drain}	TF drain with field water	0.5	-	Schaffner (2007)
$k_{drain}^{(N)}$	TF N surplus drain	0.65	-	
P_{res}	Crop residual rice	4,062.5	kg/ha-crop	Lertkrai (n.d.)
$P_{res}^{(N)}$	N concentration in crop residual rice	5.38	g/kg	
$k_{runoff}^{(N)}$	TF N runoff	0.1	-	Schaffner (2007)
Emission	$O_{Emis}^{(N)} = I_{input}^{(N)} \cdot k_{emis}^{(N)}$			
$k_{emis}^{(N)}$	TF of N emission	0.0117	t N_2O-N/t N	USEPA (1995)

2.3 Data acquisition and calibration

There are two types of data in this study: primary and secondary. For the primary data, systematic random sampling was applied to collect water samples from 15 points with different land uses along the NNR. The water samples were obtained from mid-river, stored at 4°C, and returned to the laboratory immediately. Then, TKN, nitrite nitrogen, and nitrate nitrogen were analyzed using APHA section 4500-Ammonia, 4500-NO$_2$-B, and 4500-NO$_3$-B, respectively. Furthermore, the primary data also included interviews with 40 farmers on the patterns and frequency of cultivation, types, frequency, and fertilizer application. The data were analyzed by arithmetic mean ($\overline{X}$) as shown in Equation (2):

$$\overline{X} = \frac{\sum_{i=1}^{n} x_i}{n} \qquad (2)$$

Where; n is the total number of observations in the data set and x_i represents the observation value.

The secondary data were divided into three groups: (1) the general data such as rainfall and land use were derived from national statistics; (2) the specific data for the subsystems (e.g., rice area, and rice yield) were collected from the government sector and literature; and (3) transfer coefficients (TF) were obtained from Schaffner (2007). The specific data collected in the study area by the local government sector were prioritized in this study.

For calibration, the quantitative data were crosschecked with other sources. For example, the rice area data from the Land Development Department (LDD, 2018) were identical to the data from the Nakhon Nayok Statistical Office (NNSO, 2019). In addition, the uncertainty intervals of the information sources were determined (Danius, 2002) for data quality. The uncertainty range of the data in this study was between 10% and 40% which presented moderately because the data can be variated up to 80% when determining by expert judgement (Zoboli et al., 2016).

3. RESULTS AND DISCUSSION
3.1 Nitrogen flow

The flows of nitrogen through the NNR basin in 2018 were illustrated in Figure 4. The total input and output of nitrogen were approximately 54,542 tons/year and 41,232 tons/year, respectively. The system storage calculated from the difference between the total input and output was 13,310 tons/year. Aquaculture (36%) and rice cultivation (33%) were the major sources of N inputs, followed by poultry

farming (21%). The main sources of N outputs were also aquaculture (44%) and rice cultivation (32%).

Aquaculture was the predominant source of N flows despite not being the main type of land use in the NNR basin. About 99% of its N inputs were from the feeds containing N needed for fish and shrimp growth. The catfish feed was the largest N input with around 550 tons/ha, about 1.5 times higher than the snakehead fish feed. For the shrimp, the main N input was the water due to the large farming area. Moreover, catfish farming was also the main source of N outputs (68%), with 90% of its N inputs discharged to the river. The low level of N recovered in catfish (approximately 14%) could be the reason (Worsham, 1975).

Rice cultivation was the second-largest source of N inputs and outputs in the basin. Although the fertilizer application was lower than the average at 209-350 kgN/ha/crop in China (Sui et al., 2013; Chen et al., 2014; Ding et al., 2020), the fertilizer was still the largest N input responsible for nearly 70% of the total N input of the subsystem. Additionally, The N input from the rain was 12 times higher than the irrigation ascribable to the higher nitrogen concentration in the rain than in the river; both sources contributed around 23% of the total N input. Regarding N outputs, about 74% of N was discharged from the system with yield while roughly 24% to the NNR.

Pig and poultry farming were the third and the fourth largest sources of N inputs in the basin. Almost 100% of N flowed into both systems via the feeds. Pig farming is a minor contributor of N loading to the surface water due to Thailand's regulation for wastewater from pig farming. However, pig farming played an important role in N emission as N$_2$O was present during the composition, aerobic and anaerobic treatments of pig slurry and dung.

The NNR basin is also famous for flower planting. For the fruit and flower subsystems, fertilizer was the main N input, contributing to 98% of the total N input. While 68% of N flowed out the subsystems with yield, about 5% went to the river through runoff and leaching processes.

Nitrogen flowing into household subsystem was 98.7% via food and 1.3% via water. In terms of on-site wastewater treatment plants, most households in the NNR basin had either a septic tank or cesspool. Nevertheless, some households directly discharged the greywater into the river; consequently, nitrogen flowed out with the wastewater and solid waste was around 727 tons/year and 237 tons/year, respectively.
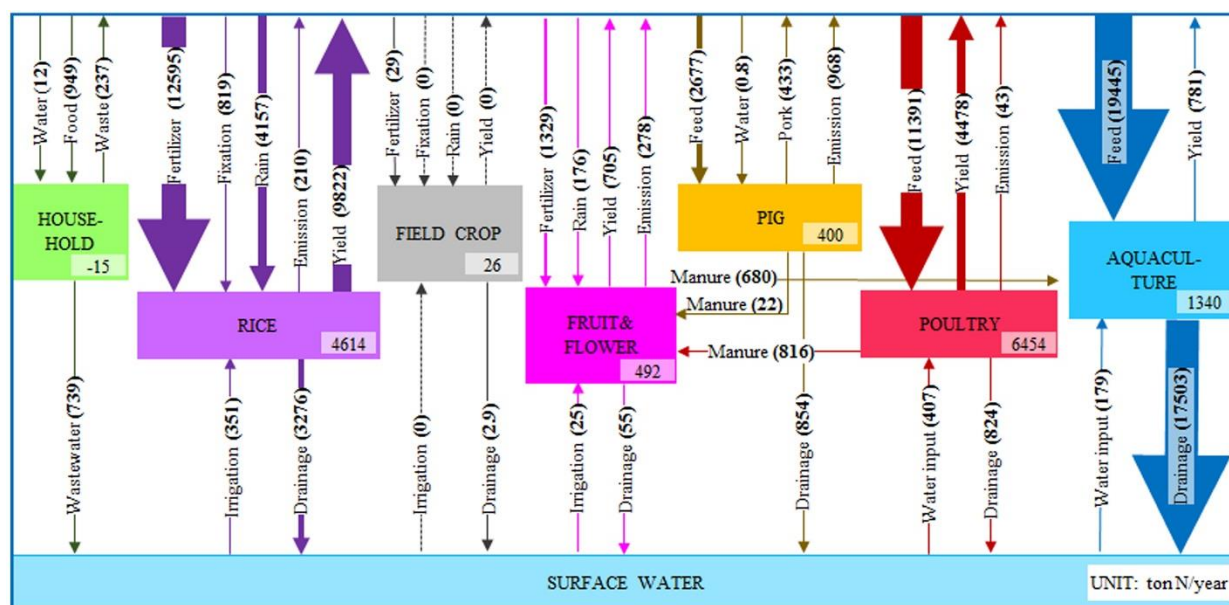
Figure 4. Nitrogen flows through the rice system in Nakhon Nayok River Basin, 2018

3.2 Nitrogen loading to the surface water

Around 23,254 tons/year of N or 42% of N inputs reached the surface water. Aquaculture was responsible for 17,503 tons/year of N discharge, which was five times higher than that of the rice system. The ratio of N loading to the NNR to the N input of aquaculture was about 0.89 being in the same range of 0.73 to 0.86 reported by Lazzari and Baldisserotto (2008). The poultry subsystem had the highest storage (48%); consequently, its ratio of N loading to N input (0.07) was lower than that of the rice system (0.18) (Figure 5).
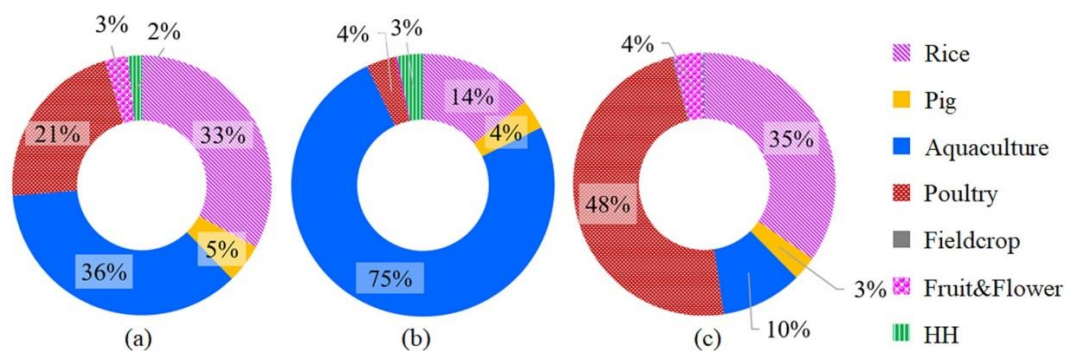

Figure 5. Proportion of nitrogen flows from each subsystem in Nakhon Nayok River Basin (a) nitrogen input; (b) nitrogen loading to NNR; and (c) nitrogen storage

As presented in Table 3, aquaculture and rice cultivation were the key flows of N discharged to the NNR, Thachin, and Lower Bang Pakong, except for the Maeklong River Basin (Schaffner, 2007; Tipsaeng, 2014; Pharino et al., 2016). The nitrogen flux of the NNR basin was roughly 2-3 times higher than other areas; however, it was comparable to the non-forest area of the Meklong River Basin. The possibility was that despite being the smallest area among the others, as discussed in section 3.1, the NNR Basin received N mostly from three times more catfish farms existing in the area than in the Thachin River Basin.

3.3 Scenario analysis

There were five recommendations for reducing N loading from aquaculture, rice cultivation, and households. The details for each scenario were described below.

Scenario 1: Implementation of wastewater treatment for aquaculture

The wetland system was recommended for aquaculture due to the high removal efficiency of nitrogen. As pond systems are the most common aquacultural systems in Nakhon Nayok Province,

accounting for approximately 99.89% (NNPFO, 2018), the wetland is an alternative system that can be applied for wastewater management. Lin et al. (2002) reported the average nitrogen removal efficiencies of 86% to 98% for ammonia, >99% for nitrite nitrogen, 82% to 99% for nitrate nitrogen, and 95% to 98% for the total inorganic nitrogen. Considering aquaculture wastewater law (MNRE, 2008), big farms (larger than 1.6 ha) are obliged to control N concentration. Although aquacultural farms in Nakhon Nayok are small with the average area of 1.34 ha (NNPFO, 2018), wastewater treatment should be implemented for at least 5% in each district to control N pollution. This scenario also suggested that 20% of the recent aquacultural farming area should have been wetland for aquaculture wastewater treatment.

Table 3. The key flows of nitrogen input through the river basins in Thailand

River Basin	Nitrogen loading to surface water (ton/year)	Nitrogen flux		Key flow of N discharge	Reference
		(ton/ha)	(ton/cap)		
Nakhon Nayok	23,254	0.11	0.09	Aquaculture Rice	This study
Thachin	45,036	0.05	0.02	Aquaculture Rice	Schaffner et al. (2007)
Lower Bangpakong	15,001	0.03	0.02	Aquaculture Rice	Tipsaeng (2014)
Meklong	25,911	0.04	0.03	Household Industry	Pharino et al. (2016)

Scenario 2: Reduction of 10% of catfish culture

Schaffner (2007) suggested that reduction of catfish culture could decrease the largest amount of N discharge; however, it can be difficult to implement since catfish is the economic fish in the area. Therefore, reducing only 10% of the catfish farming was instead recommended.

Scenario 3: Reduction of fertilizer use in rice cultivation during the rainy season

The results from MFA showed that fertilizer was the main cause of the excessive N discharged from rice cultivation. The survey results also revealed that the fertilizer application in the rice system were higher than the recommended rates by the Thai Department of Rice at 125-156 kg/ha/crop for NPK fertilizer and 31-63 kg/ha/crop for urea fertilizer (DOR, 2020). Therefore, reducing fertilizer application in the rainy season was encouraged.

Scenario 4: Reduction of fertilizer application in rice cultivation during both rainy and dry seasons

Even though the NPK fertilizer application in dry season was in between the recommended range at 156-219 kg/ha/crop, the urea fertilizer application was higher than recommended at 63-94 kg/ha/crop (DOR, 2020). Thus, this scenario suggested reducing the fertilizer application in rice system during both rainy and dry seasons.

Scenario 5: Construction of a centralized municipal wastewater treatment plant

The construction of a centralized municipal wastewater treatment plant was recommended for Nakhon Nayok Province. The activated sludge (AS) wastewater treatment plant could remove about 67% of the nitrogen from wastewater (Hsu, 1998).

Table 4 displays the scenario analysis from the assumptions in MFA. The five scenarios showed that construction of wetland on 20% of the recent aquacultural area could successfully remove 2,800 tons/year of the N loading. The reduction of only 10% of catfish farming can also lower the amount of N released to the river by 1,754 tons/year. The reduction of fertilizer application during rainy and dry seasons also cuts N loading to the surface water by approximately 753-2,285 tons/year. Lastly, the construction of a centralized wastewater treatment plant in the area helps decrease the N discharge around 500 tons/year. To further reduce N loading, Nakhon Nayok province should promote the wetland for aquaculture and suggest the farmers to reduce their fertilizer application in rainy and dry seasons.

Table 4. The scenario analysis and the potential reduction

No.	Scenario	N (ton/year)	Potential reduction
0	Current situation	23,254	-
1	Implementation of wastewater treatment from aquaculture	20,454	12.0%
2	Reduction of 10% of catfish farming	21,500	7.5%
3	Reduction of fertilizer use in rainy season	22,578-21,606	3.0-7.0%
4	Reduction of fertilizer use in rainy season and dry season	20,969-22,501	3.2-10%
5	Construction of the central municipal wastewater treatment plant	22,754	2.2%

4. CONCLUSION

This research estimated the overall N flows and indicated the key flows of N loading causing the risk of euthrophication in the NNR Basin. Despite the limited data on the study area, MFA was able to provide the comprehensive flows of N loading to the NNR and to establish priorities for environmental measures. The total input and output of nitrogen in 2018 were approximately 54,542 tons/year and 41,232 tons/year, respectively. The NNR received around 23,254 tons/year of N from anthropogenic activities and aquaculture was identified as the main source and key flows of N pollution. Thus, the reduction of 10% of catfish farming was prioritized to decrease the risk of eutrophication in the NNR Basin.

ACKNOWLEDGEMENTS

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