# Original Article Development of an Integrated Environmental Impact Assessment Model for Assessing Nitrogen Emissions from Wastewater Treatment Plants

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## ABSTRACT

Environmental impact assessments for wastewater treatment plants (WWTPs) have evaluated many endpoints including emissions of greenhouse gases, discharges of nutrients and discharges of toxic substances. The primary objective of this study was the development of an integrated environmental impact assessment model for wastewater treatment processes. The assessment model was based on an impact assessment methodology used in Japan for life cycle assessments. Specifically, eutrophication was taken into account in the model along with the impacts of free ammonia, because this chemical was known to have toxic effects on aquatic ecosystems. The model developed was then applied to an actual WWTP operating under two different conditions (case 1, without nitrification; case 2, with nitrification), and the best operating conditions were evaluated based on nitrogen emissions. The results showed that the main contributor to the environmental impacts of the WWTP in case 1 was ecotoxicity from discharges of NH<sub>4</sub>-N. In case 2, the main contributor was eutrophication from discharges of total nitrogen. These results demonstrated that the overall environmental impacts of WWTPs should decrease when nitrification is employed because this will reduce the impacts associated with the ecotoxicity of NH<sub>4</sub>-N.

Keywords: environmental impact, life cycle assessment, wastewater treatment plant

# **INTRODUCTION**

It is well known that wastewater treatment plants (WWTPs) consume a large amount of electricity in purifying wastewater. In addition, nitrous oxide (N<sub>2</sub>O) is emitted into the atmosphere during the process of nitrogen removal, and nitrogen nutrients are discharged as effluents into the water environment. It follows that environmental impact assessments of WWTPs should be considered from the points not only of CO<sub>2</sub> emissions as greenhouse gases from electricity use, but also of N<sub>2</sub>O emissions into the atmosphere and the discharge of nitrogen nutrients into the water environment.

Biological nitrogen removal is carried out in two steps: conversion of  $NH_4^+$  to  $NO_3^-$  by aerobic nitrification, followed by conversion of  $NO_3^-$  to  $N_2$  gas by anoxic denitrification. Nitrous oxide is known to be generated in these two steps [1]. The nitrification process is closely related to  $N_2O$  emissions and the discharge of nitrogen nutrients. In particular,  $N_2O$  has a greenhouse effect approximately 300 times greater than that of  $CO_2$  [2].

Most wastewater in Japan are treated by the conventional activated sludge process, which has two typical operating conditions: with or without nitrification. Our investigation [3] showed that a large amount of  $NH_4$ -N remained in the effluent, and that in the method without nitrification, the amount of aeration had decreased. Part of  $NH_4$ -N becomes free ammonia ( $NH_3$ -N) based on the chemical equilibrium, which is dependent on pH and water temperature [4]. The ratio of  $NH_3$ -N increased as pH or water temperature increased. Therefore, the ratio of  $NH_3$ -N discharged as  $NH_4$ -N

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Fig. 1 System boundaries for assessment of WWTP.

from WWTPs varies depending upon the pH and water temperature in the water environment. The high ecotoxicity of NH<sub>3</sub>-N to aquatic life, for example fish and crustaceans, was well reviewed by Camargo and Alonso [5], and the ecotoxicity of NH<sub>3</sub>-N for some species has been assessed in several laboratory experiments [6–8]. However, there has been no evaluation methodology to comprehensively consider all associated environmental impacts of WWTPs from the point of view of nitrogen emissions into the atmosphere and the water environment.

Life cycle assessment (LCA) is known as an effective method for evaluating various environmental impacts and has been widely applied to WWTPs [9-14]. Although a number of LCA methodologies have been developed recently [15–17], none have addressed the ecotoxicity of NH<sub>3</sub>-N. Corominas et al. [18] reported that decision-making in controlling wastewater nutrient removal systems was assessed using a combination of mechanistic process models using life cycle impact assessment models. An assessment tool, which employed mathematical models according to information from the literature, was developed in the report. However, there has been no report following the study of an actual WWTP in Japan, when inventories for various midpoints were actually measured as a case study. There had been no report as well in which the amount of electricity consumed for aeration, N<sub>2</sub>O emission to the atmosphere and nitrogen concentration in the effluent were measured and evaluated using a LCA model in an actual WWTP from the point of view of the nitrogen emissions.

This study aimed to develop an integrated environmental impact assessment model for wastewater treatment process-

es. We were especially concerned to evaluate the ecotoxicity impact of  $NH_4$ -N, estimate the damage factor of  $NH_4$ -N and develop a new ecotoxicity estimation model and introduce it into an existing LCA model. The model developed, which can assess integrated environmental impacts, was then applied to an actual WWTP operating under two different conditions and the best operating conditions were evaluated based on nitrogen emissions by nitrification.

# **METHODS**

#### Assessment of WWTP using an LCA model

The system boundaries for assessment of WWTP are shown in Fig. 1, where  $CO_2$  is emitted following electricity consumption for the aeration, N<sub>2</sub>O is emitted by the biological metabolism for nitrogen treatment in the aeration tank, and T-N and NH<sub>4</sub>-N are emitted in the effluent discharge from the WWTP. An environmental impact assessment of the WWTP using LIME2 (life cycle impact assessment method based on endpoint modeling ver. 2) was conducted [17,19]. This is the only life cycle impact assessment method developed for environmental conditions in Japan with human health (disability-adjusted life years; DALY), social assets (economic value) and biodiversity (expected increase in the number of extinct species; EINES) as endpoint indicators [17,19]. The environmental impacts for global warming, eutrophication and ecotoxicity as midpoints affecting human health, social assets and biodiversity as endpoints can be assessed using LIME2. This LCA model is achieved by multiplying economic value conversion factors and damage factors, which indicate impacts on endpoints needing protection



Fig. 2 Calculation flow in the ecotoxicity estimation model.

as endpoints per unit environmental load, with evaluation of environmental loads.

It is possible to evaluate these impacts by converting the environmental impact into an economic value in this model. The actors related to the inventory of  $CO_2$ ,  $N_2O$  and T-N have already been developed in LIME2; however, the damage factor of NH<sub>4</sub>-N has not yet been estimated. Therefore, the NH<sub>4</sub>-N damage factor was developed and introduced into LIME2 (see the following section).

#### Development of ecotoxicity impact evaluation

Ecotoxicity impact on biodiversity using the damage factor of  $NH_4$ -N was evaluated by the ecotoxicity estimation model according to the procedure and the equations shown in **Fig. 2** and **Table 1**,  $r_{NH3-N}$  (–) is the ratio of  $NH_3$ -N in

NH<sub>4</sub>-N calculated from pH (–) and temperature (°C), [NH<sub>3</sub>-N] (mgN/L) is the NH<sub>3</sub>-N concentration in environmental water, DF<sub>NH3-N</sub>(EINES·L/mgN) is the damage factor of increasing NH<sub>3</sub>-N concentration, EINES is the unit of expected increase in the number of species becoming extinct, D<sub>j</sub> (L/mgN) is the marginal increase in expected percentage of extinction in rank j species following increase in NH<sub>3</sub>-N concentration, LC50<sub>i</sub> is LC50 (an index of ecotoxicity) of species in group i, N(i,j) is the number of species in group i and rank j, i is a species group (fish and crustaceans), j is a rank at threat of extinction, DF<sub>NH4-N</sub> (EINES/kgN) is the damage factor of NH<sub>4</sub>-N emissions, wr (m<sup>3</sup>) is the renewable water resource in Japan, p is the NH<sub>4</sub>-N decrease factor in environmental water following nitrification, EI (yen/y) is the impact of ecotoxicity on biodiversity from NH<sub>4</sub>-N discharge,

Process in Fig. 2	Equation
(1)	$r_{\rm NH3-N} = f (pH, temp)$
(2)	$DF_{NH3-N} = \Sigma_i \Sigma_j D_j (LC50_i, [NH_3-N]) \cdot N(i,j)$
(3)	$DF_{NH4-N} = DF_{NH3-N} \cdot r_{NH3-N} \cdot wr^{-1} \cdot p \cdot 10^{3}$
(4)	$EI = DF_{NH4-N} \cdot e_{NH4-N} \cdot WF$

 Table 1 Equations in the ecotoxicity estimation model.

 $e_{\rm NH4-N}$  (kgN/d) is the amount of NH<sub>4</sub>-N emissions, WF (yen/ EINES) is the weighting factor (economic value conversion factor).

The data of NH<sub>4</sub>-N concentration, pH, water temperature, and water resources obtained were input into the ecotoxicity estimation model to obtain the environmental impact of ecotoxicity at definitive economic value. The value of  $r_{NH3-N}$  was calculated from pH and temperature using equation (1) according to Emerson *et al.* [4]. The value of  $r_{NH3-N}$  can be used for the initial NH<sub>3</sub>-N concentration in the water environment to calculate the increase factor of extinction rate with LC50, rate of natural increase, environmental capacity, etc. The LC50 of fish and crustaceans was included in the parameter of the ecotoxicity estimation model in LIME2. For other parameters in this model (e.g. rate of natural increase and environmental capacity), the values in LIME2 were used.

The impact of ecotoxicity by  $NH_3$  was assessed by the expected number of EINES over a defined number of years. The number of extinct species was calculated using the increased concentration in the water environment determined by the model based on LIME2 procedures. Increase in EINES by the unit of  $NH_3$  concentration, which is the damage factor of increasing  $NH_3$  concentration, was calculated as  $DF_{NH3-N}$  by multiplying the inclination of function and number of species by species groups and rank at threat of extinction using equation (2).

The damage factor of NH<sub>4</sub>-N emissions was calculated as  $DF_{NH4-N}$  by multiplying  $DF_{NH3-N}$ , the increase factor of NH<sub>3</sub>-N and NH<sub>4</sub>-N decrease factor, using equation (3). The impact of ecotoxicity by NH<sub>4</sub>-N emissions decreased because of nitrification in the water environment, and so an NH<sub>4</sub>-N decrease factor was introduced into equation (3). The 40% of NH<sub>4</sub>-N discharged from WWTP was thought to be oxidized to NO<sub>2</sub>-N or NO<sub>3</sub>-N in the water environment by nitrification according to water pollution control law in Japan. The 60% of NH<sub>4</sub>-N was assumed to remain as NH<sub>4</sub>-N in the water environment, and so a value of 0.6 as the NH<sub>4</sub>-N decrease factor was used in this model. The environmental impact of ecotoxicity as an economic value was calculated by multiplying the  $DF_{NH4-N}$ , the weighting factor and the amount of  $NH_4$ -N emissions using equation (4).

It was necessary to examine the sensitivity of  $DF_{NH4-N}$ , because the development of  $DF_{NH4-N}$  was new. Therefore, a sensitivity analysis was conducted to evaluate the variability of the effect of  $DF_{NH4-N}$ . The change in damage factor due to the change of -50 to +50% in pH, water temperature,  $NH_4$ -N concentrations, and water resource was evaluated. The range of possible values in each parameter was also evaluated from the obtained values.

### WWTP case study

One treatment line of an actual WWTP in Saitama Prefecture, Japan, was used as the case study to apply the developed LCA model including the ecotoxicity estimation model. The plant was operated by the conventional activated sludge process with continuous aeration under two operating conditions (case 1 and case 2) [3]. The wastewater was treated with less aeration in case 1, to save electricity consumption, without nitrification. In case 2, nitrification was accelerated with a large rate of aeration.

The effluent to be discharged into the water environment was sampled and the NH<sub>4</sub>-N, T-N and dissolved N<sub>2</sub>O were measured. Nitrous oxide in gases emitted from the aeration tank was collected before entering the deodorizing tower and was measured using an N<sub>2</sub>O meter (Model 46i, Thermo Fisher Scientific, USA) or ECD gas chromatography (GC-14B, Shimadzu, Japan). Two kinds of N<sub>2</sub>O discharge from the wastewater treatment process, gaseous discharge into the atmosphere and dissolved discharge into the water environment, have been reported [20]. Therefore, both gaseous and dissolved N<sub>2</sub>O were measured, and totaled as the N<sub>2</sub>O emissions from the WWTP in this study. The emissions of CO<sub>2</sub> from the aeration process were estimated based on electricity consumption for the aeration according to the operating data

Parameter	Units	Data Case1 Case2		Midpoints	Data source
Water environment					
pH		7.6		Ecotoxicity	[21]
Water temperature	°C	17		Ecotoxicity	[21]
NH <sub>4</sub> -N concentration ([NH <sub>4</sub> -N])	mgN/L	0.31		Ecotoxicity	[21]
Water resource (wr)	m <sup>3</sup>	4.1×10 <sup>11</sup>		Ecotoxicity	[23]
Wastewater treatment plant					
N <sub>2</sub> O emission from WWTP	kgN2O/d	1.2	2.9	Global warming	This study
NH <sub>4</sub> -N concentration in effluent	mgN/L	9.9	1.1	Ecotoxicity	This study
T-N concentration in effluent	mgN/L	17	11	Eutrophication	This study
Electricity consumption for aeration	kWh/d	2400	3000	Global warming	Operating data from WWTP monitoring reports
Flow rate of wastewater	m <sup>3</sup> /d	24000		Eutrophication, Ecotoxicity	Operating data from WWTP monitoring reports

Table 2 Data obtained in the water environment and WWTP.

from WWTP monitoring reports.

# **RESULTS AND DISCUSSION**

#### **Estimation of damage factor**

The data of pH and water temperature in the water environment were collected from the water quality measurement result at public water bodies in Saitama Prefecture, Japan, in 2011 [21]. The medians of the data (shown in **Table 2**) from 94 sampling points were obtained and used to estimate the damage factor. Data in **Table 2** allowed a calculation of 0.0119 for  $r_{NH3-N}$ .

The ECOTOX database [22] was used for this study, with geometrically averaged LC50 values for each species group according to LIME2 procedures. As there were insufficient data on other species groups, our impact estimate was only targeted on fish and crustaceans, in contrast to LIME2, which targets six species groups. In the model, the increase of species extinction implements a function between NH<sub>3</sub>-N concentration and extinction probability according to species group and rank (by threat of extinction) was estimated with the same procedure as LIME2. The geometrically averaged value of LC50 for fish (n = 22) and crustaceans (n = 24) was 11.9 mgN/L and 11.9 mgN/L, respectively.

The data obtained from the water environment shown in **Table 2** were input into the ecotoxicity estimation model. The calculated value of  $DF_{NH3-N}$  was 1.12 EINES·L/mgN. Then,  $DF_{NH3-N}$  was multiplied by the increase factor of NH<sub>3</sub>-N ( $r_{NH3-N}$ /wr), and  $DF_{NH4-N}$  was definitively estimated to 1.93 × 10<sup>-11</sup> EINES/kgN. This value is the 644th highest value in the materials of 907 listed in LIME2 estimated by the same procedure. This  $DF_{NH4-N}$  value also meant that NH<sub>4</sub>-N had almost the same ecotoxicity as toluene (2.11 × 10<sup>-11</sup>) or pyrocatechol (1.95 × 10<sup>-11</sup>) in the water environment.

#### Sensitivity analysis of damage factor

A sensitivity analysis was conducted to evaluate the variability of the effect of DF<sub>NH4-N</sub> as the damage factor by the change in pH, water temperature, NH<sub>4</sub>-N concentrations, and water resource. The results are shown in Fig. 3, where the dotted line shows the calculated value and the solid line shows the range of possible values. The range of possible values of pH (7.4 - 7.8), water temperature ( $15 - 18^{\circ}$ C) and  $NH_4$ -N concentrations (0.1 - 1.1 mgN/L) were used as the statistical range of 25 - 75% from the universal data obtained. The water resource data obtained from Japanese Ministry of Land, Infrastructure, Transport and Tourism [23] were limited to the average and the case of the risk of shortage (third quartile of yearly data). For this reason, the value of  $3.3 \times 10^{11}$  (shortage)  $-4.1 \times 10^{11}$  (average) m<sup>3</sup> was used as the available range of water resource in the analysis. Increases in pH and water temperature increased the magnitude of the damage factor, although increase in water resource resulted in a decrease of it. Because the gradient of pH increase was higher, it was thought that the influence of pH on the damage factor was larger compared with the other parameters. The value of 75% of pH was 7.8; however, pH sometimes exceeded 8.5, which was the allowable maximum value of the environmental standard, especially in the summer season. It was therefore thought that pH was the most important and sensitive component of the damage factor. In contrast, NH<sub>4</sub>-N concentrations had little effect on the damage factor. In addition to these sensitivity analyses, further research should also be done to clarify NH<sub>4</sub>-N decrease factor (p) in detail because it is a very important factor to evaluate NH<sub>4</sub>-N concentration in the water environment.



Fig. 3 Sensitivity analysis of damage factor.



Fig. 4 Estimated results of the integrated environmental impacts.

# Integrated environmental impact assessment from WWTP

Assessment of the integrated environmental impacts using LIME2, including the ecotoxicity estimation model, was carried out using the data shown in **Table 2**. The electricity emission factor of 0.000463 tCO<sub>2</sub>/kWh in 2011 published by the Japanese Ministry of the Environment [24] was used to evaluate the impact on global warming from electricity consumption. The estimated results are shown in **Fig. 4**. The results show that the main contributor to the environmental impacts from WWTP in case 1 was ecotoxicity due to discharges of NH<sub>4</sub>-N. The impacts of ecotoxicity in cases 1 and 2 were estimated to be 2.7 and 0.3 yen/m<sup>3</sup>, respectively. Because  $NH_4$ -N concentration was decreased by nitrification in case 2, the impacts of ecotoxicity also decreased. The impact for global warming due to the discharge of  $CO_2$  and  $N_2O$  was lower than the other impacts in both cases.

Saitama Sewage Systems Agency reported that it costs 42 yen to treat 1 m<sup>3</sup> of wastewater in WWTP at Saitama Prefecture, Japan, in 2014 [25]. It was understood that the cost of  $NH_4$ -N ecotoxicity was about 6.4% of the total wastewater treatment cost. On the other hand, N<sub>2</sub>O emission from WWTP was estimated to be 1.2 and 2.9 kgN<sub>2</sub>O/d (shown in **Table 2**) which meant 50 and 120 mgN<sub>2</sub>O/m<sup>3</sup>, respectively.

These values were lower than the value of  $160 \text{ mgN}_2\text{O/m}^3$  reported by Japanese Ministry of Land, Infrastructure, Transport and Tourism [26] and larger than the value reported as N<sub>2</sub>O emission factors in the conventional activated process by Tsushima *et al.* [27].

Godin et al. [28], Niero et al. [12] and Tsurumaki and Noike [29] estimated life cycle greenhouse gas emissions from wastewater treatment, including sludge treatment, to be 0.192, 0.195 and 0.300 kgCO<sub>2</sub>/m<sup>3</sup>, respectively. Niero et al. [12] also reported the main source of life cycle global warming potential. The estimated global warming potential in the present study was  $0.062 \text{ kgCO}_2/\text{m}^3$  in case 1 and 0.098 $kgCO_2/m^3$  in case 2. These values are lower than the two previous studies because this study focuses on the biological treatment process without sludge treatment process. Hospido et al. [30] reported input data of wastewater treatment by processes. Electricity use for preliminary biological treatment was 0.103 kWh/m<sup>3</sup>, nearly the same as in case 1 of this study (0.101 kWh/m<sup>3</sup>). Godin et al. [28] and Niero et al. [12] also estimated ecotoxicity, considering impacts both from effluent and other processes, but not including the impact of NH<sub>3</sub>. Estimated values were 1.7 and 2.6 g1,4DBeq/ m<sup>3</sup>(converted to the equivalent of 1,4-paradichlorobenzene) in the report of Godin et al. [28] and Niero et al. [12], respectively. Ecotoxicity in this study was equivalent to 0.27  $g_{1,4}DBeq/m^{3}$ . The difference in the ratio compared with the two previous studies suggests that the impact of NH<sub>3</sub> should be taken into account when estimating life cycle ecotoxicity impact, to allow a more precise calculation.

The total impacts for global warming, eutrophication and ecotoxicity in cases 1 and 2 were estimated to be  $3.6 \times 10^7$  and  $1.2 \times 10^7$  yen/y, respectively. These results demonstrated that the overall environmental impacts from the WWTP should decrease when nitrification is employed because this would reduce the impacts associated with the ecotoxicity of NH<sub>4</sub>-N. In case 2, the main contributor was eutrophication due to discharges of T-N. Trade-offs in relationships, mainly between the impact of eutrophication caused by T-N and the impact of ecotoxicity of NH<sub>4</sub>-N, were identified in this study. It was revealed that complete nitrification of NH<sub>4</sub>-N to NO<sub>3</sub>-N would be the best operating method, with low negative environmental impacts from the point of view of nitrogen emissions from WWTPs.

# CONCLUSIONS

This study aimed to develop an integrated environmental impact assessment model for wastewater treatment processes.

To evaluate the influence of  $NH_4$ -N ecotoxicity in the context of LCA, the damage factor of  $NH_4$ -N was estimated. The ecotoxicity estimation model developed was then introduced into LIME2 to evaluate the integrated environmental impact for global warming, eutrophication and ecotoxicity, and then a case study on an actual WWTP was carried out. The results show that the main contributor to environmental impacts of the WWTP in case 1 was ecotoxicity due to discharges of  $NH_4$ -N. In case 2, the main contributor was eutrophication due to discharges of T-N. These results demonstrate that the overall environmental impacts of a WWTP should decrease when nitrification was conducted because this will reduce the impacts associated with the ecotoxicity of  $NH_4$ -N.

The estimated damage factor and the ecotoxicity estimation model developed in this study can be used as core modules and models to evaluate various wastewater treatments and aquatic environments in the future.

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