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Evaluation of nitrogen loading in the last 80 years in an urbanized Asian coastal catchment through the reconstruction of severe contamination period

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Keywords: long-term, nitrogen loading, rapid urbanization, population growth, land use change, wastewater treatment plant

Abstract

Most semi-enclosed seas have experienced severe eutrophication owing to high nutrient loading from rivers during rapid population growth periods. In Japan, the coastal areas of some megacities (e.g. Tokyo and Osaka) experienced considerable economic growth during the 1960s–1970s. Therefore, determining the amount of nutrient loading during this period is essential to undertake measures for the conservation of coastal environments. However, determining the nutrient loading that occurred several decades ago is generally difficult owing to lacking water quality records. In this study, the nitrogen loading in the Yamato River catchment, an urbanized coastal catchment in Asia, for 80 years from the 1940s to the 2010s is reconstructed using the Soil and Water Assessment Tool. We considered factors such as population growth, wastewater treatment plant (WWTP) construction, and changes in land and fertilizer usage in different urbanization stages. Results show that the total nitrogen loading in the catchment peaked in the 1970s at 6616 tons yr^{-1} owing to untreated wastewater discharge and rapid increase in population growth. By reducing 57% of the nitrogen loading in the 2010s from the catchment, WWTPs have been instrumental in improving the water environment. The decrease in and integration of agricultural land has reduced nitrogen loading attributed to nonpoint sources; however, this reduction was not obvious because of the high fertilizer usage before the 2000s. Overall, the findings of this study provide a comprehensive understanding of the impact of rapid urbanization in an Asian coastal catchment on nitrogen loading during the high economic growth period in the past. This study will be useful for the long-term assessment of nutrient loading in other.

1. Introduction

Eutrophication in coastal areas is one of the earliest recognized problems related to the hydrosphere and biosphere [1]. This problem is still serious in several regions and threatens biodiversity [2, 3]. Severe eutrophication is mainly caused by anthropogenic activities such as domestic wastewater discharge, agricultural activities, and industrial development [4]. In particular, in semi-enclosed seas such as the Mediterranean Sea, the influence of nutrient loading from rivers is prolonged and severe eutrophication is caused in the coastal areas because of the circulation pattern and long residence time of the seawater [5, 6]. In the middle and late 20th century, nitrogen loading increased significantly owing to the rapid growth of population and economy [7, 8]. Fortunately, nitrogen loading can be curtailed by controlling anthropogenic activities, such as wastewater treatment [9]. Therefore, both the positive and negative impacts of human activities on nutrient loading must be considered in environmental research.

Table 1. Previous studies on long-term nutrient loading evaluation.

Research	Study area information	Period	Method	Source*
Van Meter *et al* [11]	Mississippi River (US) Susquehanna River (US) 1800–1960 only from literatures (10 year interval)	1800–2014	Modeling	PS & NPS
Savage *et al* [12]	Baltic Sea (SE) (10 year interval)	1880–2000	Observation	PS & NPS
Roberts and Marsh [13]	River Stour, River Tees, River Breat Ouse and River Thames (UK)	1930–1990	Observation	Total loading
José [14]	River Trent (UK)	1950–1990	Observation	Total loading
Pastuszak *et al* [15]	Vistula and Oder River (PL) 1955–1995 only from literatures (5 year interval)	1955–2015	Modeling	PS & NPS
Burt *et al* [16]	Slapton Wood Catchment (UK)	1970–1985	Observation	Total loading
Ouyang *et al* [21]	Upstream of Huanghe River (CHN)	1970–2010	Modeling (SWAT)	NPS
Yanagi and Ishii [22]	Seto Inland Sea (JP)	1979–1999	Observation	Total loading
Pulighe *et al* [23]	Sulcis catchment (ITA)	1982–2009	Modeling (SWAT)	NPS
Cozzi and Giani [17]	North of Adriatic Sea (EUR)	1990–2010	Observation	Total loading
Epelde *et al* [18]	Alegria River watershed (ES)	1990–2011	Modeling (SWAT)	NPS
Burkholder *et al* [19]	Neuse river (US)	1993–2004	Observation	Total loading
Räike *et al* [20]	Baltic Sea (EUR)	1995–2015	Observation	PS & NPS
Chotpantarat and Boonkaewwan [24]	Lower Yom River (TH)	2000–2013	Modeling (SWAT)	NPS
Jung and Kim [25]	Chungju dam watershe (KR)	2003–2010	Modeling (SWAT)	PS & NPS
Tong *et al* [26]	Yangtze River, Huanghe River, Liaohe River, etc (CHN)	2006–2012	Observation	Total loading
Tran *et al* [27]	Cau River (VN)	2007–2014	Modeling (SWAT)	PS & NPS

* PS: Point source, means this study considered point source loading; NPS: nonpoint source, means this study considered nonpoint source loading; Total loading means this study just considered total nutrient loading and not separate by point or nonpoint source.

Studying the long-term changes in the nutrients in river water is important to comprehensively understand complex environmental changes caused by anthropogenic activities, including both positive and negative effects [10]. For example, articles searched from the Web of Science with the keywords 'long-term' and 'nutrients' show considerable difference between the study period (table 1). Most studies in Europe, particularly in the UK, and North America began before the 1960s [11–15], and some studies were conducted in the 1970s–1990s [16–20]. Although some studies shown very long study periods, they do not provide results for continuous years in the early ages or cannot distinguish between point source and nonpoint source pollution.

In Asia, research in this regard mostly commenced later than 2000, except for some studies conducted in the 1970s, and these studies mainly focused on total river discharge or upstream nonpoint source pollution [21–27]. The Asian countries differ from the European and American countries in terms of both research time period and research objective owing to differences in urbanization. The UK underwent the earliest urbanization from the 1800s to 1900s [28]. The urbanization processes in the USA and Europe (excluding the UK) occurred during 1850–1950 and 1900–1980, respectively [29, 30]. Major urbanization processes with rapid economic growth in Japan and China occurred during 1960–1980 and 1985–2018, respectively [31, 32].

Hydrologic models are powerful tools for assessing the nitrogen loading on a catchment scale. The Soil Water Assessment Tool (SWAT) is widely used in long-term nutrient loading studies [33]. Few studies have combined both point and nonpoint sources in the SWAT model and accessed the comprehensive loading, particularly in the Asian countries before 2000. This is because water quality monitoring usually started after or during the urbanization stage, and because urbanization in Asia occurred later, water quality data were not recorded in the early ages. However, the population growth rates of Asian countries are considerably higher than those of the European and American countries, particularly in recent decades [34]. Rapid population growth and urbanization may induce extremely high nitrogen loading in aquatic systems. However, variations in nitrogen loading in such situations have not been fully studied yet and the severity of pollution is not sufficiently clear.

Osaka Bay is a part of the Seto Inland Sea and shows the worst water quality in this sea, particularly in the mid-1970s [35]. A study estimated long-term variations (1920–1995) in nitrogen loading in the regions from Osaka Prefecture to Osaka Bay as 'potential loading' based on a unit of output, suggesting that the nitrogen loading peaked around 1990 [36]. Another study estimated the nitrogen loading from all rivers inflowing to Osaka Bay as the 'net loading' based on observational data recorded by the government after the 1980s, reporting that the nitrogen loading peaked in the mid-1980s and then decreased after 1990s [37]. However, previous reports have shown that the rivers inflowing to Osaka Bay (e.g. Yamato River) were the most polluted in the 1970s, attributed to rapid economic growth [38]. Therefore, determining the 'net loading' of nutrients during the rapid economic growth period in Japan (the 1960s–1970s) when the monitoring of river water quality had not commenced is crucial.

The objective of this study is to evaluate the long-term changes in nitrogen loading in the last 80 years in the Yamato River catchment, an urbanized coastal catchment of Osaka Bay, based on the reconstruction of severe contamination periods. We used the SWAT to evaluate the influence of anthropogenic activities, in terms of both point and nonpoint sources, on the nitrogen loading.

2. Materials and methods

2.1. Study area

The Yamato River catchment covers an area of 1077 km^2, and the annual average precipitation in the catchment is approximately 1360 mm. Among the major rivers inflowing to Osaka Bay, the Yamato River shows the worst water quality [38]. The land use types in the catchment are mainly rice paddies, forest area, and residential area. The paddy fields accounted for 27.3% of the catchment in the 1970s, and by the 2010s, this area had shrunk to 15% because of urbanization and agricultural policies. The residential area in the catchment increased rapidly; as the total population increased from 650 000 to 2200 000 during 1940–2019, the residential area expanded from 17% to 29% during 1970s–2010s [39]. The forest area has remained approximately the same [40]. A vast residential area and high population are the reasons for the high proportion of wastewater discharge owing to anthropogenic activities in the catchment [38, 40].

Before the 1970s, no water quality management and monitoring measures were adopted in the Yamato River catchment [41]. To tackle the issue of pollution growth around Osaka Bay, the local government established several wastewater treatment plants (WWTPs) and a wastewater collection pipeline system in the catchment during 1970–1985 (figure 1). By the end of 2018, wastewater collection pipelines in Nara and Osaka Prefectures covered 80% and 96% of the areas, respectively [42, 43]. Moreover, some rural areas without WWTPs employ small-scale wastewater treatment equipment (SWWTE). The water quality of the SWWTE effluent is considered equivalent to that of the WWTP effluent [44]. However, some regions in the study catchment still directly discharge wastewater into the river without any treatment.

Although the observation of the total nitrogen (TN) concentration in the main channel of the Yamato River began in 1976, observations were conducted only four times per year, insufficient to analyze nitrogen discharge. The observation policy related to the monthly water quality was established in 1983 provided the minimum required data for modeling nitrogen loading.

2.2. Soil and water assessment tool

The SWAT is a catchment-scale hydrological model developed by the USDA Agricultural Research Service. It is designed to predict the impact of land use and management on water, sediment, and nutrient loading in ungauged watersheds [45, 46]. The SWAT is a semi-distributed model and is suitable for peak predictions and human impact and long-term estimations [39, 47]. In the SWAT, nitrogen transport describes the movement of nitrogen from lands to river and water bodies.

The SWAT model uses a digital elevation model (DEM) as the terrain data input, extracts the runoff direction and watershed boundary by calculating the aspect, and divides the watershed into several sub-catchments based on the topological relation of the river channels. Based on the land use, soil data, and slope information of the watershed, the SWAT can be used to build a detailed hydrological response unit (HRU). The HRU is the basic calculation unit of the SWAT and is the unit for simulating runoff, sediments, and nutrients. Detailed explanation can be found in the SWAT theoretical documentation [48].

Spatial datasets, including data related to the topography, land use, and soil, were converted to the SWAT 2012 input dataset using ArcSWAT, an external GIS interface [48]. Model calibration and validation work were performed in the SWAT Calibration Uncertainty Program. The Nash–Sutcliffe efficiency (NSE) and percentage of bias (PBIAS) were used to evaluate the simulation results. These parameters are calculated using equations (1) and (2) [49, 50]:

$$\mathrm{NSE} = 1 - \left[\frac{\sum_i (X-Y)^2}{\sum_i (X-X')^2}\right] \quad (1)$$

$$\mathrm{PBIAS} = \frac{\sum_i (X-Y)}{\sum_i X} \times 100 \quad (2)$$

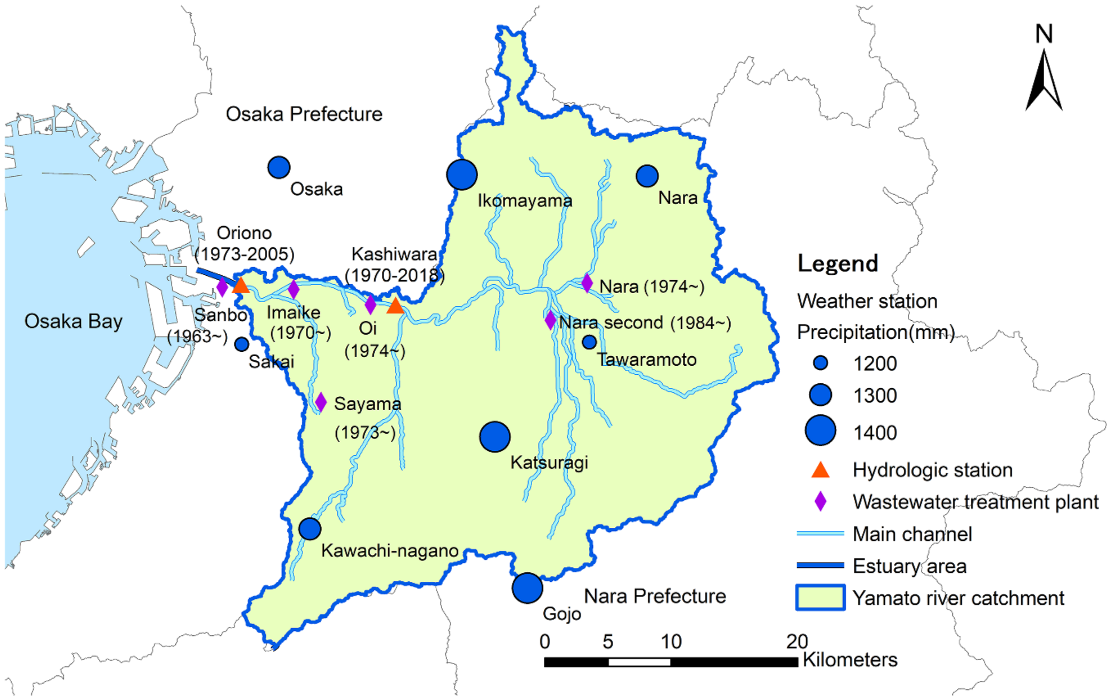

Figure 1. Overview of the Yamato River catchment.

where X, Y, X', and i represent the observed data, SWAT simulation result, means of the observed data, and number of observed or simulated data, respectively. A study provided the following criteria for nitrogen simulation: satisfactory when NSE > 0.35 and |PBIAS| < 30, good when NSE > 0.5 and |PBIAS| < 20, and very good when NSE > 0.65 and |PBIAS| < 15 [51].

2.3. Data collection and model construction

In this study, the land use data of 1976, 1997, and 2016 were used to estimate the effect of land use change on nonpoint source loading (figure 2). Meteorological data were collected from 1935 to 2019 from eight meteorological stations (figure 1). Streamflow and water quality observation data were collected from two hydrologic stations, Oriono and Kashiwara, for the available data period (figure 1).

The Yamato River catchment was divided into 39 sub-catchments and more than 2500 HRUs. Based on the construction of the WWTP and the change in the coverage of wastewater collection pipelines, the simulation was performed for three periods.

First period (1940–1984): The land use data of 1976 were adopted, and the commencement of operation of the wastewater treatment system.

Second period (1985–2008): The land use data of 1997 were adopted, and the popularization stage of wastewater equipment.

Third period (2009–2019): The land use data of 2016 were adopted, and the use of improved wastewater equipment.

We calibrated the streamflow, suspended sediment (SS) flux, and nitrate (NO_3–N) and TN flux. For the calibration run in the third period, daily data were used, and for the validation run in other periods, the monthly data were used. The parameters obtained from the daily calibration can better reflect the relation between precipitation and nutrient loading compared with those obtained from the monthly run [52]. However, for the first and second periods, the amount of data was insufficient for the daily simulation.

The agricultural schedule was set based on local agricultural practices and a previous study [53]. The total annual nitrogen fertilizer usage, including organic fertilizer obtained from local livestock, was 80, 90, 100, and 85 kg ha^{-1} for 1940–1962 (first half of the first period), 1963–1984 (second half of the first period), 1985–2008 (second period), and 2009–2019 (third period), respectively. Water for agriculture, domestic purposes, and industry was mainly provided from seven reservoirs in the catchment. Further, many small ponds in the catchment provide water for agriculture [54]. As a measure against the occurrence of frequent floods in the catchment, three reservoirs were constructed for flow control in the upstream area. The inflow and outflow of wastewater were provided by the local WWTPs. Because the Sanbo WWTP is not located in the catchment but in the estuary area (figure 1), its data were not input to the SWAT; however, its nitrogen loading was calculated separately and included in the total discharge from the Yamato River. All data sources and usage are listed in table 2.

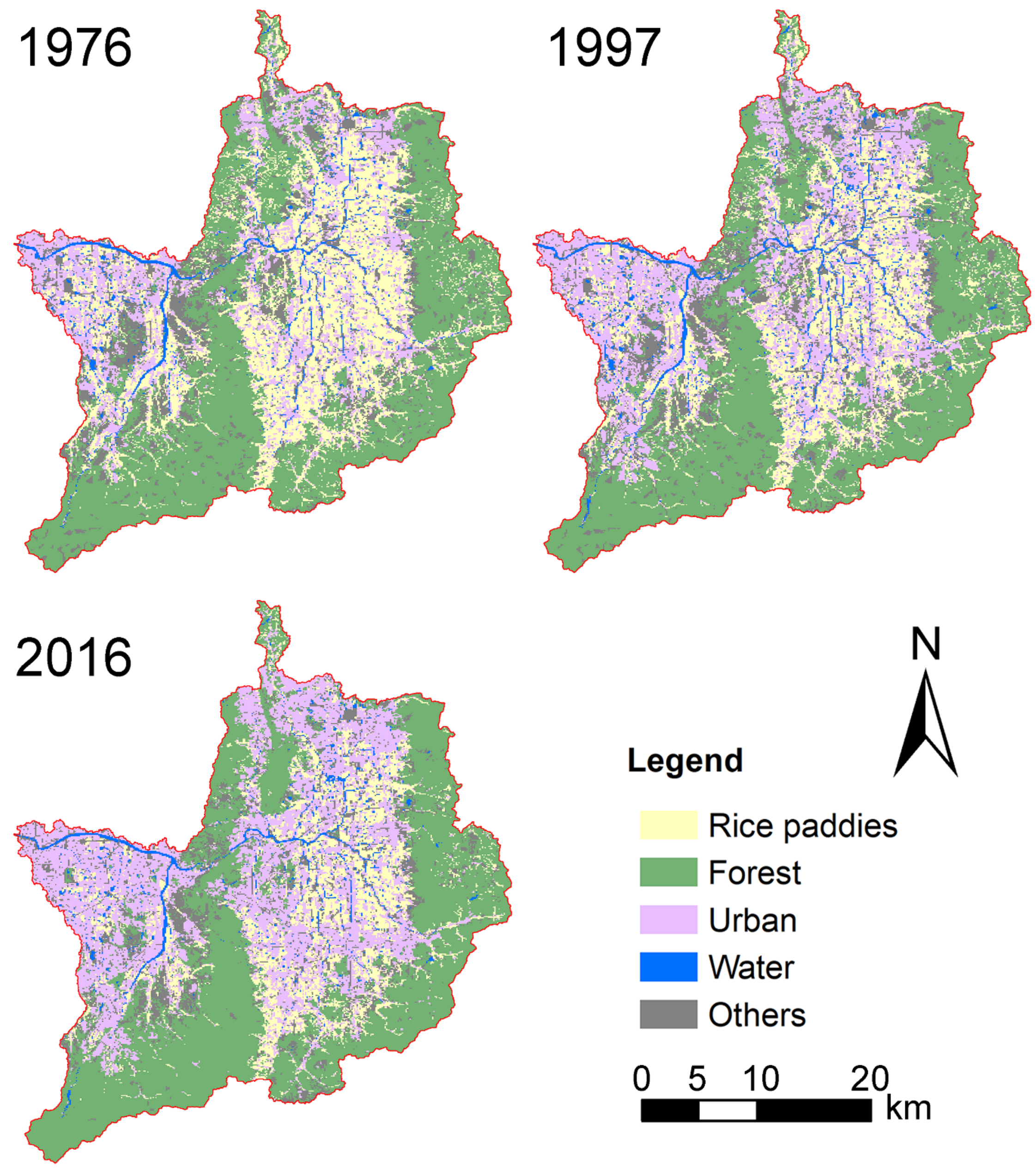

Figure 2. Land use maps of the three periods.

Nitrogen loading attributed to human wastewater in the catchment can be divided into three categories: discharge from WWTPs, discharge from SWWTEs (N_{SWWTE}), and untreated wastewater ($N_{\mathrm{untreated}}$). Discharge from WWTPs can be directly calculated from the outflow water quality data of each WWTP. The other two categories are estimated using equations (3) and (4):

$$N_{\mathrm{SWWTE}} = \frac{N_{\mathrm{out}}}{P_c} \times P_s \tag{3}$$

$$N_{\mathrm{untreated}} = \frac{N_{\mathrm{in}}}{P_c} \times P_u \tag{4}$$

where N_{out} is the nitrogen loading (kg N) in outflow water from a WWTP during a given time step; N_{in} is the nitrogen loading (kg N) in inflow water to the WWTP during a given time step; P_c is the population served by the WWTP; P_s is the population served by an SWWTE; and P_u is the population not served by any wastewater system. Each factor was calculated separately based on different wastewater treatment regions for each year. The calculation results of $\frac{N_{\mathrm{out}}}{P_c}$ and $\frac{N_{\mathrm{in}}}{P_c}$ can reflect the per capita load for treated domestic wastewater and per capita load for untreated domestic wastewater (CLU), respectively. The yearly estimated N_{SWWTE} and $N_{\mathrm{untreated}}$ can reflect the changes in wastewater quality owing to the anthropogenic factor corresponding to that year, such as improved wastewater treatment technology. For the early ages, when WWTPs were not built, $N_{\mathrm{untreated}}$ was estimated using the average CLU before the 1990s

Table 2. Data sources of this study.

Data description		Source and scale	Data usage
1	Topography	United States Geological Survey database (30 m)	Digital Elevation Model (DEM)
2	Land use data (1976, 1997, 2016)	Ministry of Land, Infrastructure, Transport and Tourism of Japan (MLIT; 100 m)	HRU division
3	Soil Data	MLIT (1000 m)	HRU division
4	Climate data	Japan Meteorological Agency	Weather input
5	Wastewater quality data	Local wastewater treatment plants, including inflow and outflow water quality	Point source estimation
6	Wastewater system coverage ratio	Osaka Prefecture office, Nara Prefecture office	Point source estimation
7	Hydrological observation data	MLIT	Model calibration and validation
8	Management practices	Nara Prefecture office	Planting types, harvesting, fertilizers application and the types, irrigation
9	Water Management	Japan Dam Association	Reservoir volume, domestic and industrial consumption, agricultural water withdrawal of weirs

as 11.26 g/capita/day. A previous study estimated that the CLU in early Japan was 11–13 g/capita/day [55], consistent with the range of CLU estimated in this study.

2.4. Simulating anthropogenic factor scenarios

We applied the actual situation as a baseline and designed two scenarios to simulate the impact of population growth on nitrogen loading and verified the effectiveness of the WWTP in improving environment.

Scenario 1: No WWTP was built in the catchment, and all wastewater was discharged directly without treatment. We calculated the nitrogen loading by subtracting N_{out} and adding N_{in} from the baseline results. This scenario reflected the change in potential loading owing to an increase in the population growth.

Scenario 2: Population growth stopped in 1970, and the total amount of domestic wastewater did not increase further. Wastewater was discharged with a gradual decrease in the new construction of wastewater treatment systems.

The nonpoint source loading in the two scenarios was the same as the baseline result.

3. Results

3.1. Model assessment and uncertainty analysis

Both monthly and daily TN flux simulations matched well with the observed values, including most of the peak values (figure 3). Based on the performance ratings [51], the streamflow, SS flux, and NO_3–N flux simulations were evaluated to be 'good,' 'satisfactory,' and 'satisfactory,' respectively (table 3). For the TN flux simulation during 1985–2018, NSE >0.7 and |PBIAS| <15, and the performance was evaluated to be 'very good.' For the TN flux simulation during 1970–1984, the NSE just reached the standard for 'good' but |PBIAS| reached the standard for 'very good.'

The uncertainty of this model mainly arises from the amount of untreated wastewater discharged because the nitrogen concentration in this part of wastewater is very high and strongly influenced by anthropogenic factors [55]. In the first and second periods, the quantity of untreated wastewater discharge was very high and this amount changed continuously. Contrarily, in the third period, the quantity of untreated wastewater was small and stable. Therefore, we selected the third period as the calibration period.

3.2. Estimation of long-term nitrogen transport

With the rapid population growth in 1960, the nitrogen loading in the catchment began to increase rapidly. It peaked in the 1970s and remained high in the first half of the 1980s. From the second half of the 1980s, the nitrogen loading began to decrease owing to a gradual improvement in wastewater treatment systems (figure 4). The average annual nitrogen discharge in the Yamato River catchment during each decade from the 1940s to the 2010s were 3980, 4677, 5750, 6616, 6480, 5363, 4024, and 3412 tons yr^{-1}, respectively. The untreated wastewater discharge in the second half of the 1960s and 1970s was 4722 tons yr^{-1}, considerably higher than those in the other periods and accounting for 72% of the TN loading in the catchment. With increasing WWTP discharge, the quantity of untreated wastewater effectively decreased. The WWTP-discharged nitrogen loading accounted for 12%, 24%, 37%, 49%, and 50% of the total catchment loading in each decade from the 1970s to 2010s, respectively. The TN loading during the 80 years was 403 024 tons and that

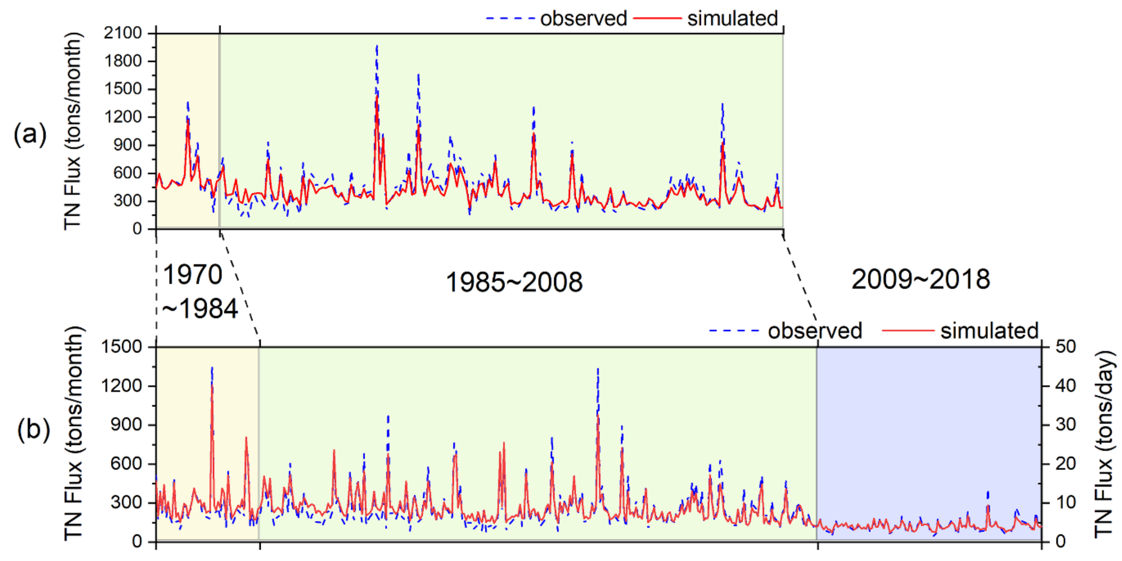

Figure 3. Observed and simulated TN fluxes at Oriono (a) and Kashiwara (b) hydrological stations in the period 1970–2018.

Table 3. Performance of the calibrated nitrogen flux in the SWAT model.

	Period	1970–1984		1985–2008		2009–2018
	Station name	Oriono	Kashiwara	Oriono	Kashiwara	Kashiwara
Streamflow	NSE	0.83	0.74	0.82	0.85	0.75
	PBIAS	7.0	4.5	4.4	4.3	7.3
SS flux	NSE	NA	NA	NA	0.47	0.55
	PBIAS	NA	NA	NA	18.9	16.7
NO_3-N flux	NSE	NA	NA	0.46	0.52	0.63
	PBIAS	NA	NA	24.6	−11.1	3.8
TN flux	NSE	0.56	0.55	0.71	0.72	0.76
	PBIAS	4.3	−9.8	4.2	−4.1	−0.6

during 15 years from 1970 to 1984 accounted for 25% of this value.

3.3. Impact of population growth and WWTP on nitrogen loading

The increase in the most important anthropogenic factors contributing to the nitrogen loading was the rapid population growth. In simulated scenario 1, the TN loading in the catchment was the maximum in the second half of the 1990s, reaching 10 611 tons yr^{-1}, twice the actual value for that period (figure 4). The TN loading in scenario 1 for each decade during the 1970s–2010s were 1.21, 1.47, 1.94, 2.49, and 2.81-folds that of the corresponding actual values, respectively. The population in 1970 was 1.33 million, 62.5% of the average population in the 2010s. Furthermore, the nitrogen loading in scenario 2 in the 2010s was 70% of the actual value in this decade. A comparison of the two scenarios with the actual situation indicates that although the population growth has a direct impact on the increase in the nitrogen loading, the WWTPs effectively suppress the increase in the nitrogen loading caused by population growth and improve the catchment environment. Moreover, based on the situation in 2019, if sufficient WWTPs can be established to service all the population in the catchment, the TN loading can be reduced by 22%.

3.4. Impact of agricultural policy and precipitation on nitrogen loading

Agricultural activities, particularly fertilizer application, are the main anthropogenic factors affecting nitrogen transport. The average loading over the studied 80 years was 1005 tons yr^{-1}. The average nonpoint source loading in the 2010s was 717 tons yr^{-1} (figures 4 and 5). The nonpoint source pollution decreased in 2010s because of two reasons: the reduction and integration of agricultural land and the reduction of fertilizer usage [53]. In the first period, the agricultural area was larger and its distribution was more scattered, leading to a high nonpoint source loading (figures 5 and 6). In the second period, although the area under agricultural land decreased, nonpoint source pollution remained high because of increased fertilizer usage. In the third period, both the area under agricultural land and fertilizer usage decreased, thus considerably reducing the nonpoint source loading. In addition to the agricultural factors, precipitation is another factor that directly affects nonpoint source pollution, showing a positive impact

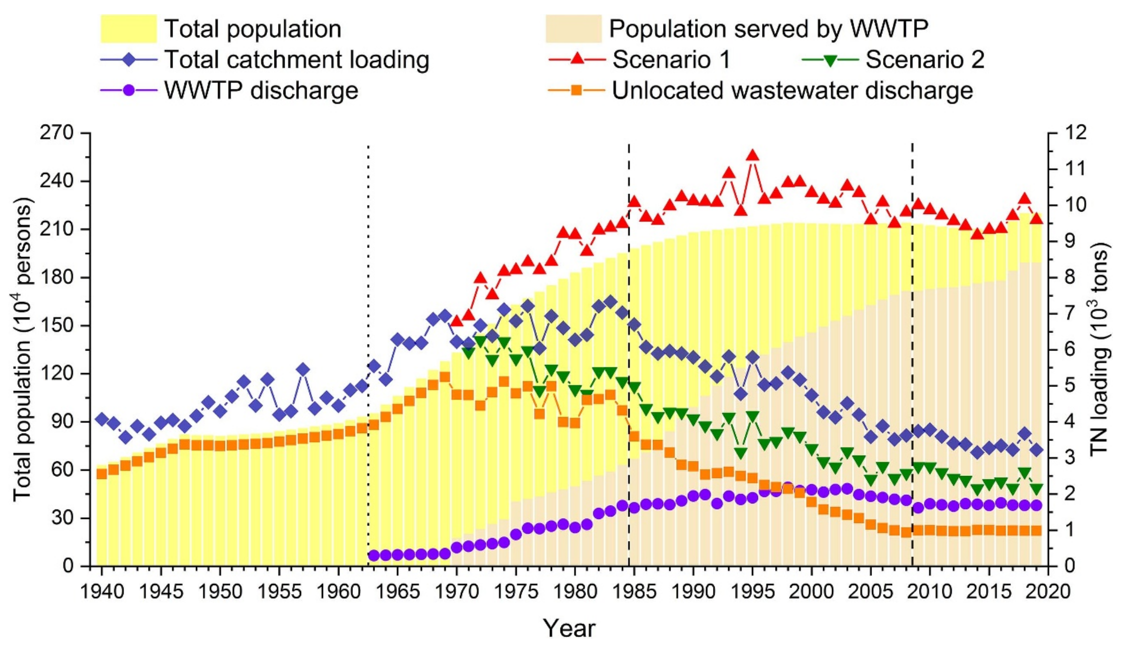

Figure 4. Yearly TN loading of the studied catchment and wastewater discharge.

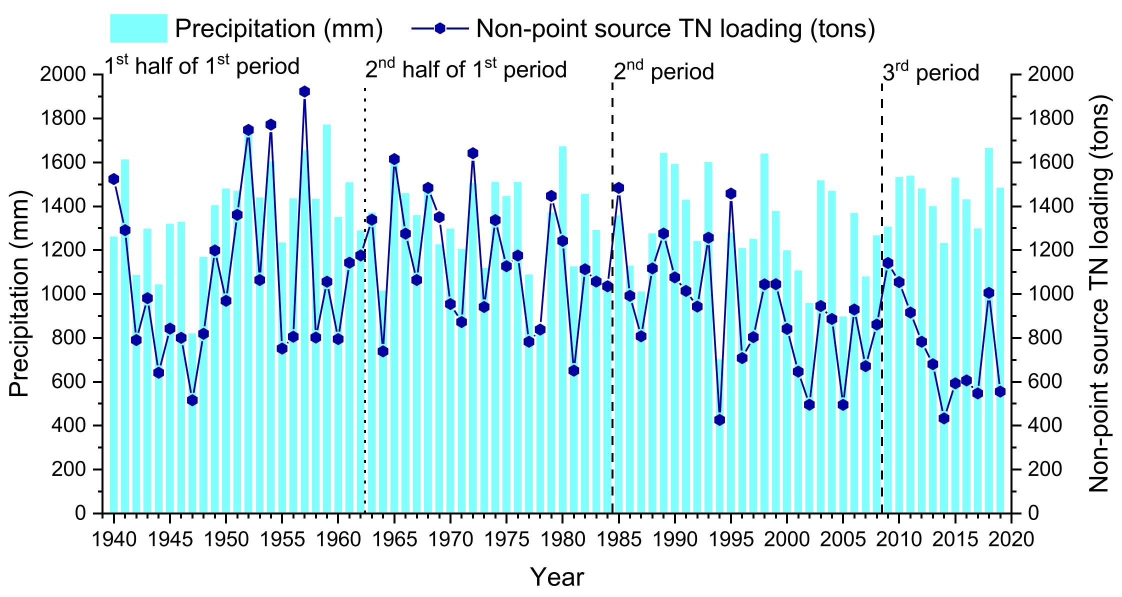

Figure 5. Yearly non-point source TN loading from study catchment.

on such pollution. During the study period, the maximum nonpoint source pollution of 1923 tons with 1652 mm precipitation occurred in 1957 and the minimum of 426 tons with 701 mm precipitation occurred in 1994. The maximum nonpoint source loading was almost five times the minimum one.

4. Discussion

The river water quality worsened rapidly since the 1960s, and the biochemical oxygen demand (BOD) was extremely high in the first half of the 1970s [38]. According to this study, this period corresponds to the period of peak untreated wastewater loading. A large amount of domestic wastewater was directly discharged into the river, thereby increasing the BOD concentration of the river water. The nitrogen loading in the Yamato River in the present study shows a trend similar to that in Osaka Bay in previous studies. Although the average water quality was the worst in the 1970s, the nitrogen loading of 6851 tons yr^{-1} in the first half of the 1980s was higher than that in the 1970s; this finding is similar that reported in a previous study [37]. However, after 1985, as the area served by WWTPs in the Yamato River catchment increased rapidly, the nitrogen loading began to

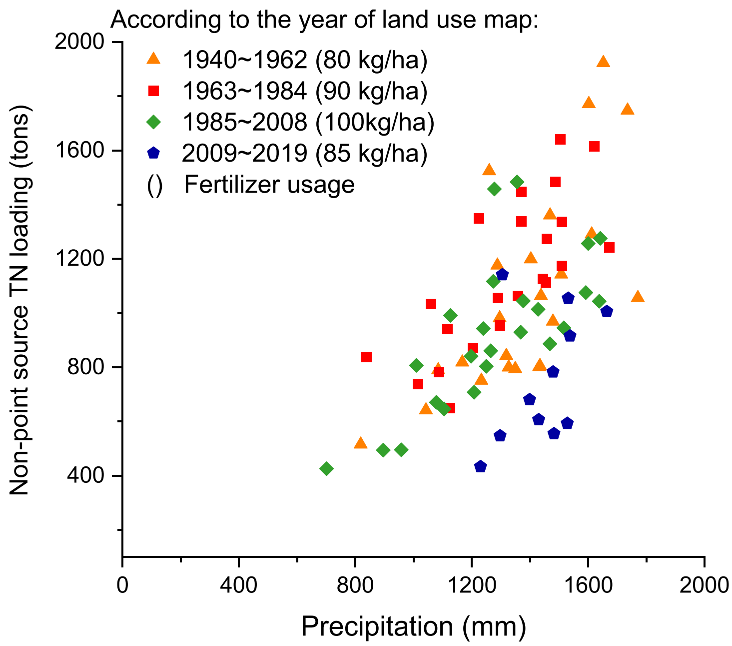

Figure 6. Relationship between non-point source TN loading and precipitation.

decrease remarkably. The nitrogen loading in scenario 1 has a similar meaning as 'potential loading' reported in a previous study [36], and both show peaks in the 1990s.

In several countries, during urbanization, open septic tanks were used, inducing groundwater pollution because of the leaching of nitrogen from the tanks to aquifers [56, 57]. However, in Japan and other countries where closed SWWTE systems were used in the urbanization stage for regions where it was difficult to install WWTPs, nitrogen leaching into aquifers was prevented [44, 55]. However, the problem of nitrogen leaching from agricultural lands in Japan and other Asian countries may be more serious than those in Europe or America. According to the SWAT results for the studied catchment, the nitrogen leaching was 15% of the applied fertilizer, considerably higher than that in a European upland field catchment [58]. This can be attributed to rice being the main crop in Asian countries, whereas wheat and corn are the main crops in Europe and America. Further, rice paddies required impoundment and must be kept flooded during the plowing season, causing greater nitrogen leaching from rice paddies [59]. This has also led to the denitrification in rice paddies-based catchments, which occurred at a higher rate than that in catchments where wheat is the main crop and the nitrogen fertilizer uptake rate is lower than in European and American countries [58]. Note that in some paddy soil under reducing conditions, nitrogen predominates in the form of ammonium. However, because ammonium easily undergoes oxidation, nitrogen contributed to the river is usually in the form of NO_3–N unless the river channel also under low dissolved oxygen [60].

As observed in previous studies, precipitation strongly affects the nonpoint source pollution of agricultural lands. The nitrogen loading in a high-precipitation year may be several times that in a low-precipitation year [24, 61–63] because non-point source pollution is driven by precipitation, particularly the amount of precipitation, following each fertilizer application during the plowing season [57, 64, 65]. Reducing and integrating agricultural lands are effective means of reducing nonpoint source pollution in the catchment [21, 66]. This is consistent with the results of this study. However, fertilizer usage is still an important factor [11].

Susquehanna River is one of most urbanized catchments in the USA. The population in this catchment has increased by 30% during 1900–2000. The peak nitrogen loading caused by population growth was 36 kg ha^{-1} in 1970s, considerably lower than that in the catchment studied in this work; however, the increased fertilizer usage after urbanization caused more severe environment issues in Susquehanna River [11]. The nitrogen loading in other urban rivers listed in table 4 was also considerably lower than the peak loading in the Yamato River catchment because of the urbanization speed here. In 1960–1990, the population housed in this catchment increased by 134%, considerably more rapid than those in other urban catchments. Rapid urbanization led to a very high nitrogen loading because the sewage treatment systems were constructed and installed at a much slower pace than the population growth rate. Catchments in more than 100 countries, such as China, Korea, and Thailand, that witnessed similar rapid urbanization or a population growth ratio higher than 100% in the past or recent 30 years

Table 4. TN loading caused by urbanization and population growth reported in previous studies worldwide.

Research	Study area information	High loading period	TN loading
Van Meter *et al* [11]	Susquehanna River (US)	1970s	36 kg ha^{-1}
Pastuszak *et al* [15]	Vistula River (PL)	1998	9 kg ha^{-1}
	Oder River (PL)	1985	11 kg ha^{-1}
Chotpantarat and Boonkaewwan [24]	Lower Yom River (TH)	After 2000	28 kg ha^{-1}
This study	Yamato River (JP)	1980–1984	64 kg ha^{-1}

may encounter similar high nitrogen loading situations [23, 25, 34, 67].

5. Conclusion

In this study, the nitrogen loading in the Yamato River catchment, an urbanized coastal catchment, was evaluated for a continuous period of 80 years. This study covers all stages of urbanization in an urban catchment in Asia. A large amount of wastewater data was used to accurately estimate the untreated wastewater loading in the catchment. Total 403 024-tons N was discharged from the Yamato River in the studied 80 years, and the peaking loading was 64 kg ha^{-1}. The peak nitrogen loading in the studied catchment was considerably higher than that caused by urbanization and population growth in European and American countries. This is because in the early stage of rapid urbanization, the increased untreated wastewater caused severe nitrogen pollution because the construction of wastewater treatment systems lagged behind the population growth rate.

Japan is a typical example of the effects of rapid urbanization and population growth. This study contributes to the understanding of the real N loading influenced by rapid urbanization and population growth. The method employed in this study can be used to study other catchments. It can also be employed by decision-makers for assessing long-term nutrient loading and some environmental issues from the past. For some countries that are going through the stage of urbanization and population growth, this research can help them predict possible future environmental problems and deploy measures in advance.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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