

Impact of N-Saturated Upland Forests on Downstream N Pollution in the Tataru River Basin, Japan

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ABSTRACT

This study evaluated whether nitrogen (N) saturated upland forests can degrade downstream water quality in the Tataru River Basin, northern Kyushu, western Japan. Our hypothesis is that elevated atmospheric N deposition degrades downstream water quality in a watershed containing N-saturated forests because a considerable amount of atmospherically deposited N passes into the streams without being retained. Synoptic stream water samplings were conducted at 23 sites across a wide range of land-use categories in the basin over 1 year. A long-term temporal analysis of downstream water quality over the last 30 years (1977–2007) was conducted and compared with long-term trends in related factors such as urban/agricultural activity, sewage wastewater treatment, atmospheric N deposition, and forest condition. The results showed that atmospherically deposited N to N-saturated forests can be a large enough non-point source of N leaving the watershed to impact

downstream water quality. This was highlighted by the reduction in pollutant exports derived from urban/agricultural activities, an increase in atmospheric N deposition, and the maturation of coniferous plantation forests in the past 30 years. These have led to reductions in total phosphorus and organic nitrogen concentrations in downstream water, whereas downstream nitrate (NO_3^-) concentrations increased over the last 30 years. The consequent increase in the downstream N:P ratio indicated P limitation. Reducing the NO_3^- exports from N-saturated upland forests is suggested as a strategy to improve regional downstream NO_3^- pollution, but involves intercontinental-scale action in reducing atmospheric N emissions.

Key words: N saturation; forest; atmospheric N deposition; non-point source; stream water quality; synoptic sampling; long-term trends.

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INTRODUCTION

Identifying and regulating the sources of instream nitrogen (N) and phosphorus (P) pollution responsible for coastal eutrophication is crucial to effectively reducing pollutant loads. Point sources such as municipal sewage treatment plants are relatively simple to measure and regulate, and can often be controlled by treatment at the source (Carpenter and others 1998). In contrast, non-point sources are

difficult to measure and regulate, because they often originate from large areas of land or from atmospheric deposition to receiving waters (Carpenter and others 1998). Agricultural and urban activities are major non-point sources of N and P pollution to aquatic ecosystems (Novotny and Olem 1994; Sharpley and others 1994; Carpenter and others 1998). Atmospheric N deposition is an additional source of N (Jaworski and others 1997; Whitall and Paerl 2001; Schaefer and others 2009) which can be an important part of the systems nitrogen budget. Determining the relative importance of different non-point sources of N and P pollution is a suitable strategy for the efficient reduction of non-point sources of N and P pollution.

Typically, forests are N limited and therefore have a great ability to retain atmospheric N deposition in vegetation and soils (Howarth and others 1996; Norton and Fisher 2000). Forests can act as a filter for atmospherically deposited N and can help maintain background concentration levels of N in streams (Miller and others 1997; Norton and Fisher 2000; Hayakawa and others 2006; Floyd and others 2009). Consequently, forests can mediate downstream water quality (Allan and others 1997; Wickham and others 2002). As forests generally occupy a large proportion of the total land area in many countries, including in Japan (64%), forests play a significant role in maintaining stream water quality (Wickham and others 2002).

Many studies, however, have indicated that elevated atmospheric N deposition may potentially lead to N saturation in forest ecosystems (Aber and others 1989; Vitousek and others 1997), resulting in a shift from net-N retention to high levels of net-N loss from forested watersheds (Aber and others 1989; Howarth and others 1996). Nitrogen oxide (NO_x) emissions, which are a source of N deposition, have remained high over the last two decades in Japan (Ohara and others 2007), and Japan has also received long-range transport of N compounds from Asia (Ohara and others 2007).

Nitrogen saturation occurs in forest ecosystems in Japan, North America and Europe. Okochi and Igawa (2001) found that N saturation is occurring in forested areas near the Tokyo metropolitan area where N deposition via bulk precipitation is estimated at approximately $10 \text{ kg N ha}^{-1} \text{ y}^{-1}$. Mitchell and others (1997) summarized the N biogeochemistry at 24 forested sites in Japan, and revealed that N saturation was occurring in high N load areas caused by a high degree of regional urbanization. Japan is also a recipient of long-range N transport from the Asian continent, where NO_x emissions

have increased 2.8 times during 1980–2003 (Ohara and others 2007). Therefore, the long-range transport of anthropogenic nitrogenous compounds from the Asian continent could also cause increased atmospheric N deposition and subsequently, N saturation in Japanese forest areas.

In addition to atmospheric N deposition, forest condition, including forest type and stand age, can have a significant affect on the retention or loss of N when inputs are similar (Ohrui and Mitchell 1997; Fukushima and others 2011). In Japan, a large proportion of the 10 million ha of plantation forests (typically Japanese cedar (*Cryptomeria japonica*) and Japanese cypress (*Chamaecyparis obtuse*)) were planted in the 1950s–1960s, and are now reaching maturation (Ministry of Agriculture, Forestry and Fisheries, Japan 2009). This could accelerate N saturation in forested ecosystems in Japan because N uptake is lower in older stands, leading to less N retention and high levels of NO_3^- leaching from forested areas (Ohrui and Mitchell 1997).

A few studies have suggested that N-saturated forests may considerably degrade downstream water quality (Stalnacke and others 1999; Howarth and others 2002; Tabayashi and Yamamuro 2009). However, to our knowledge, few studies have evaluated the impact of N-saturated forests on downstream quality. In the Tataru River Basin, the focus of this study (Figure 1), the upland forests exhibit N saturation, which may be the cause of the high nitrate (NO_3^-) concentration that has been observed in stream water in a small, forested sub-watershed in this basin. The atmospheric N deposition in the Ochozu experimental watershed (OEW), a forested watershed located in the Tataru River Basin, was moderately high ($11 \text{ kg N ha}^{-1} \text{ y}^{-1}$ via bulk deposition and $16 \text{ kg N ha}^{-1} \text{ y}^{-1}$ via throughfall + stemflow deposition), and significant levels of N leaching ($10.4 \text{ kg N ha}^{-1} \text{ y}^{-1}$) had occurred from the forested area (Chiwa and others 2010). The Tataru River flows into an enclosed coastal area, Hakata Bay, where a red tide (algal bloom) frequently occurs (Katayama and others 2009). Synoptic stream water sampling along a land-use gradient from forested upland areas to lowland agricultural and urban areas in this basin may provide insight into the effects of N-saturated upland forests on downstream water quality.

We hypothesize that the elevated atmospheric N deposition degrades downstream water quality in a watershed containing N-saturated upland forests by which considerable amounts of atmospheric N deposition passes into the streams without being retained. This is because the ability to retain N is

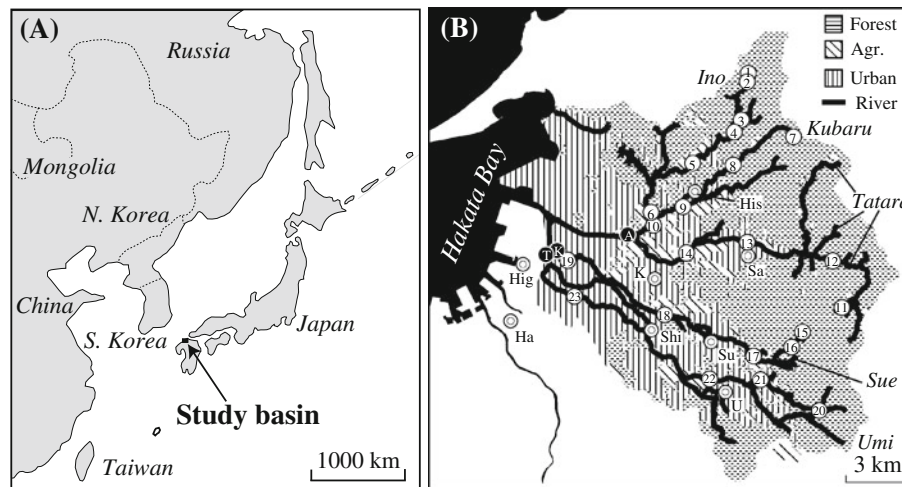


Figure 1. **A** Location of the Tataru River Basin in northern Kyushu, western Japan. **B** Map of the Tataru River Basin (199 km²) showing locations of synoptic sampling sites for this study (open circles) and government monitoring sites of downstream water quality (solid circles). Numbers and letters correspond to sampling sites shown in Table 1. Brevity codes indicate the government offices of Higashi ward (Hig), Hakata ward (Ha), and the towns of Umi (U), Sasaguri (Sa), Shime (Shi), Sue (Su), Hisayama (His), and Kasuya (K), respectively.

considerably low in N-saturated forests, resulting in high N loss from forested watersheds (Stoddard 1994). The maturation of coniferous plantation forests could also accelerate N saturation. As forests occupy large areas in this watershed, they can have a dominant influence on downstream water quality in a basin.

Any such effects of N-saturated forests on downstream water quality would depend on the extent of urban and agricultural activities as these are the main non-point sources of N and P pollution (Carpenter and others 1998). The relationship between long-term trends in downstream water quality and related factors such as the agricultural/urban area and use of an urban sewage treatment system, including advanced treatment of wastewater that can reduce N and P loads to stream water (Takeda 2000), can help to isolate the effects of urban and agricultural activities on downstream water quality from those related to lack of retention in N-saturated upland forests.

The objective of this study was to (1) to evaluate whether N saturation is occurring in the upland watersheds of the Tataru River Basin, (2) to determine the impact of N saturation in an upland forested watershed on downstream water quality, and (3) to determine the role of urban and agricultural activities on water quality.

Site Description

This study was conducted in the Tataru River Basin (199.3 km²), located in northern Kyushu

(Figure 1A), western Japan. The basin has an elevation ranging from 0 to 935 m above sea level (asl), and consists of five main rivers, the Ino, Kubaru, Tataru, Sue, and Umi rivers (Figure 1B). The proportions of forested, agricultural, and urban areas in this basin are 56, 8, and 36%, respectively. The underlying bedrock consists mainly of sedimentary rock in the plains areas, and metamorphic and igneous rock in mountainous areas. The annual mean precipitation and air temperature in the urban plains area (9 m asl) in this basin, from 2004–2007 were 1786 mm and 17.2°C, respectively, and in an upland forest area (260 m asl) were 1863 mm and 16.3°C, respectively. The urban areas in the basin consist of one city (the Higashi and Hakata-wards of Fukuoka City) and six towns (Umi, Sasaguri, Shime, Sue, Hisayama, and Kasuya). The population density in this basin was 2,667 people/km² in 2007.

METHODS

Sample Collection

Synoptic stream water sampling was conducted at 23 sites in this basin across a range of land uses, including forest, agricultural, and urban areas, from upstream to downstream on the five main rivers (Figure 1B, Table 1). The categorization of land uses into forestry, agriculture and urban was determined using Google Maps. The identification of the watershed at each sampling point was determined from a contour map. The proportion of

Table 1. Sampling Sites and Land-Use Percentage (%)

Site	River	Forest	Agriculture	Urban
1	Ino	100.0	0.0	0.0
2	Ino	99.3	0.0	0.7
3	Ino	99.6	0.3	0.1
4	Ino	99.3	0.5	0.3
5	Ino	92.7	3.0	4.3
6	Ino	85.5	6.7	7.7
7	Kubaru	100.0	0.0	0.0
8	Kubaru	95.8	3.0	1.2
9	Kubaru	81.8	9.5	8.7
10	Kubaru	73.3	10.2	16.5
11	Tatara	99.4	0.0	0.6
12	Tatara	95.6	3.3	1.1
13	Tatara	92.6	4.5	2.9
14	Tatara	88.1	4.3	7.6
15	Sue	100.0	0.0	0.0
16	Sue	100.0	0.0	0.0
17	Sue	72.3	8.8	19.0
18	Sue	91.2	5.8	3.0
19	Sue	58.6	11.5	29.9
20	Umi	100.0	0.0	0.0
21	Umi	86.4	4.9	8.8
22	Umi	79.1	4.4	16.5
23	Umi	54.7	6.9	38.5
A	Tatara	83.6	6.1	10.4
K	Sue	55.7	11.2	33.1
T	Umi	51.4	7.3	41.4

each land use (forest, agriculture, and urban) around each sampling point was calculated from the proportion of each land-use area to the watershed area at each sampling point.

At each sampling site, near surface water from the center of the channel was collected. Prior to sampling, the sample bottles (100 ml HDPE) were placed into a 1% Contaminon solution (Wako, Contaminon N) for at least 24 h and then washed with distilled water (Yamato, WG-25). The sampling was undertaken approximately every 2 months during non-precipitation periods from November 2008 to October 2009. Non-precipitation

periods were at least 3 days after a rainfall event (> 10 mm), with samples collected on 13 Nov 2008, 19 Jan 2009, 16 Mar 2009, 18 May 2009, 27 Aug 2009, and 5 Oct 2009. Each sampling across the entire Tatara River basin was conducted within 6 h.

Chemical Analysis

Stream water samples were placed on ice and were transported to the laboratory within 6 h. The samples were analyzed for total N (TN), total P (TP), NO_3^- , and NH_4^+ , with the analytical methods used, detection limits, and reproducibility shown in Table 2. Aliquots were filtered through a 0.45 μm membrane filter (GL Science, Chromatodisc, 25A) for the measurement of inorganic N (NO_3^- and NH_4^+). Organic N (ON) was calculated by subtracting the inorganic N concentration from the TN concentration. Unfiltered water samples (for TN and TP) and filtered water samples (for NO_3^- and NH_4^+) were stored in the dark at 4°C until chemical analysis, usually for a maximum of 2 months.

Analysis of Annual Trends

To evaluate the contributions of non-point N and P sources from urban and agricultural activities to downstream water, long-term trends in downstream water quality and related factors, such as agricultural area, the number of cattle, population size, and the usage rates for sewage treatment systems in the study basin, were analyzed. The number of farm animals in the study basin was not available, so the stocking rate for farm animals in Fukuoka Prefecture, where the study basin is located, was used, assuming that annual trends in cattle stocking rates in the study basin are equal to those in Fukuoka Prefecture. This assumption seems generally valid because changes in the number of farms between 1975 and 2005 are similar for the study basin and Fukuoka Prefecture (22–27% for dairy cows; 9–10% for beef cattle; 5–4% for pigs; and 7–5% for chickens). Long-term trends in downstream water

Table 2. Analytical Techniques, Detection Limit, and Reproducibility for TN, TP, NO_3^- , and NH_4^+ Determinations

	TN	TP	NO_3^-	NH_4^+
Analytical techniques	Ultraviolet absorptiometry after NaOH- $\text{K}_2\text{S}_2\text{O}_8$ digestion	Molybdenum blue absorptiometry after $\text{K}_2\text{S}_2\text{O}_8$ digestion	Ion chromatography	Ion chromatography
Detection limit ($\mu\text{g l}^{-1}$)	41	2.6	3.7	4.5
Reproducibility (%)	2.5	7.2	1.9	1.7

quality were analyzed using the government monitoring database reported in annual institutional reports (Fukuoka Prefecture 1978–2008). Concentrations of NO_3^- , ON, TN, and TP as measured at three monitoring sites (Site A (Amouzu), K (Kyuya), and T (Tounomoto); Figure 1A; Table 1) in this basin over the period of 1977–2007 were extracted. Sampling was conducted monthly for TN and TP and twice a year for NO_3^- . Long-term trends in agricultural area, the number of cattle, population size, and usage rates for sewage treatment systems in this basin over the last few decades were analyzed from the statistical database which is available from the Fukuoka Data Website (<http://www.pref.fukuoka.lg.jp/dataweb/>).

To determine whether atmospheric N deposition is high enough to induce N saturation and subsequently N leaching from forested areas in this basin, atmospheric N deposition was analyzed at two sites close to the basin (one site within a few kilometers and the other approximately 20 km southwest) and within a forested area of the basin. The atmospheric N deposition levels at the two sites close to the basin were taken from a monitoring database which is available from the Institute for Hygiene and the Environment; Fukuoka City, Fukuoka (<http://www.fch.chuo.fukuoka.jp/>), one site monitored bulk deposition from 1991 to 1998 followed by wet deposition from 1999 to 2007. The other Fukuoka site measured bulk deposition from 1991 to 1998. The data from the forested site were taken from the literature (Chiwa and others 2010) with bulk deposition of inorganic N collected during 2004–2007. Bulk (wet + dry) deposition was measured using a bulk sampler that was placed in the open, and wet deposition was measured using a wet-only sampler. The frequency of bulk and wet deposition sampling was weekly in Fukuoka City

and biweekly at the forest site. Because sampling was only weekly or biweekly, and biocide was not added into the samples as a preservative, the bio-consumption of nitrogenous compounds, especially NH_4^+ , may convert some of the initially deposited ions into an organic form (Ayers and others 1998) prior to chemical analysis. This could lead to the underestimation of atmospheric N deposition, but does not affect our conclusion.

To evaluate the changes in the forest condition in this basin over recent decades, long-term trends in the area of each type of forest and stand age in the Fukuoka forest management unit (where this basin is located) were investigated using the statistical handbook and annual institutional reports (Fukuoka Prefecture 1975–2005).

Statistical Analysis

The relationships between NO_3^- , ON, and TP concentrations in stream water and the percentage of urban/agricultural area were examined using Spearman's rank correlation coefficient. The Mann–Kendall test was performed to evaluate annual trends in the NO_3^- , ON, TN, TP concentrations, the fraction of NO_3^- in TN, and the molar N:P ratio in the downstream water. All statistical analyses were carried out using SPSS 11.0J (SPSS Japan Inc.).

RESULTS

Stream Water Quality Across the Basin

The NO_3^- concentrations in upland stream water from forested areas were high (0.6–1.2 mg l^{-1}) and similar to those in downstream water (Figures 2, 3), and there was no significant relationship between NO_3^- and the percentage of urban/agricultural area (Figure 3). In contrast, ON and TP concentrations

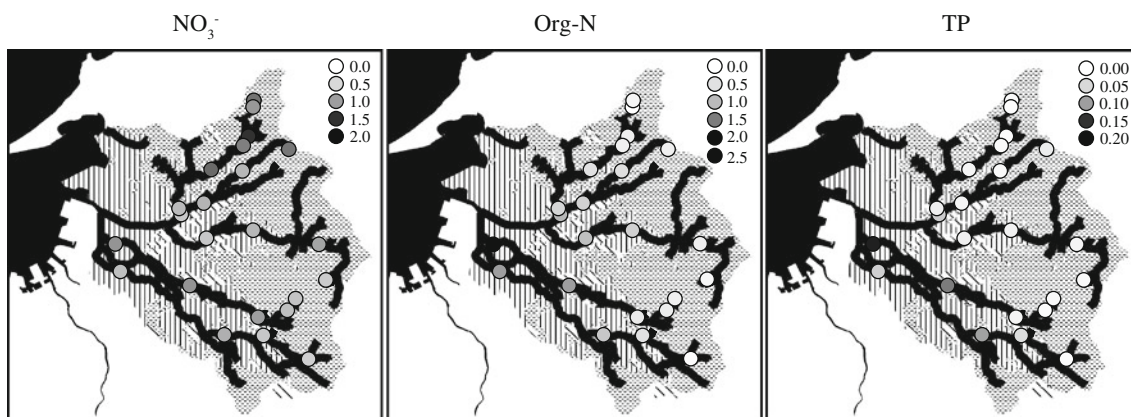


Figure 2. Spatial variations in NO_3^- , ON, and TP concentrations (mg l^{-1}) in stream water across the study basin

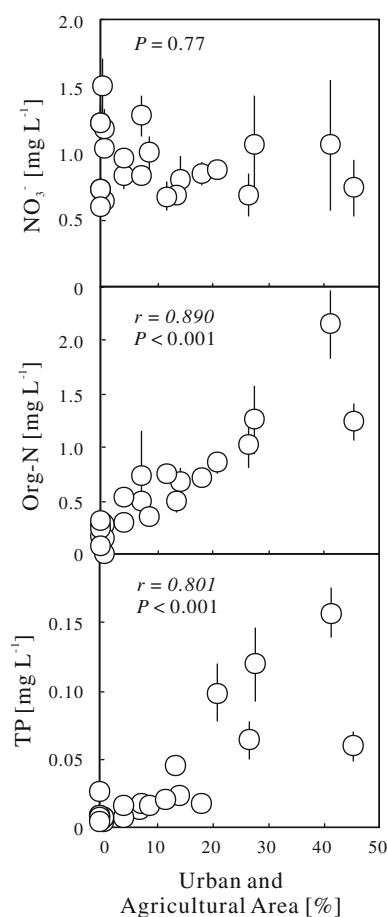


Figure 3. Relationships between annual mean concentrations of NO_3^- , ON, and TP in stream water and the percentage of urban/agricultural area in the study basin. Bars represent standard errors of annual mean concentrations.

were lower in the upland stream water from forested areas and higher in the downstream water (Figures 2, 3), and both increased significantly with the percentage of urban/agricultural area (Figure 3). The average ratio of NO_3^- to ON in the upland stream water was 5.4, with a range of 3.0–7.9 on five upland forested sites.

Annual Trends in Downstream Water Quality and Related Factors

Atmospheric N deposition at the sites near the study basin increased more than two-fold during 1991–1998, and exceeded $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ for most years after 2000 (Figure 4). In the Fukuoka forest management unit, which includes the Tataru River Basin, 56% of the forested area was planted in Japanese cedar and cypress in 2005. The percentage of mature (>46 years) Japanese cedar and

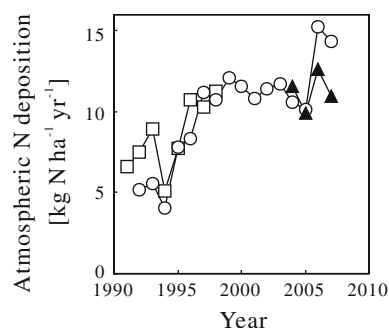


Figure 4. Annual trends in annual atmospheric N deposition at two sites close to the study basin (one site within a few kilometers, 1991–1998 (open square), the other approximately 20 km southwest of the basin, 1991–2007 (open circle), and a forested area of the study basin, 2004–2007 (filled triangle).

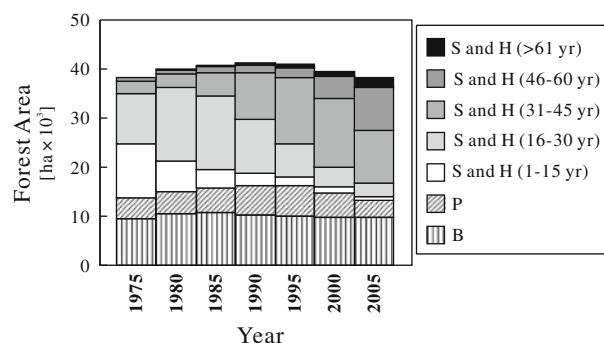


Figure 5. Long-term annual trends in the area of each forest type and classification of Japanese cedar and cypress plantations from 1975 to 2005 in the Fukuoka forest management unit. Brevity codes indicate Japanese cedar (Sugi; S), Japanese cypress (Hinoki; H), pine (P), and broadleaf forests (B), respectively.

cypress in the Fukuoka forest management unit was low (3%) in 1975, but increased to 43% of the total Japanese cedar and cypress plantation area in 2005 (Figure 5). The agricultural area in this basin decreased by 70% during 1975–2005 (Figure 6). The former agricultural areas (about 2,000 ha) were mainly converted to residential land [35% (665 ha)] and construction sites [39% (740 ha)] for commercial facilities (no figure). The number of dairy cows, pigs, and chickens in the basin decreased by 30–50% from 1990 but the number of beef cattle remained constant since 1980. The population in 2007 in Fukuoka City (C) and the five towns (T) increased by 70% compared with 1975 (Figure 6). Sewage treatment systems were used by almost 100% of Fukuoka City and approached 80% in the five towns in 2005 (Figure 6).

The TP and ON concentrations in downstream water decreased significantly during 1977–2007, to

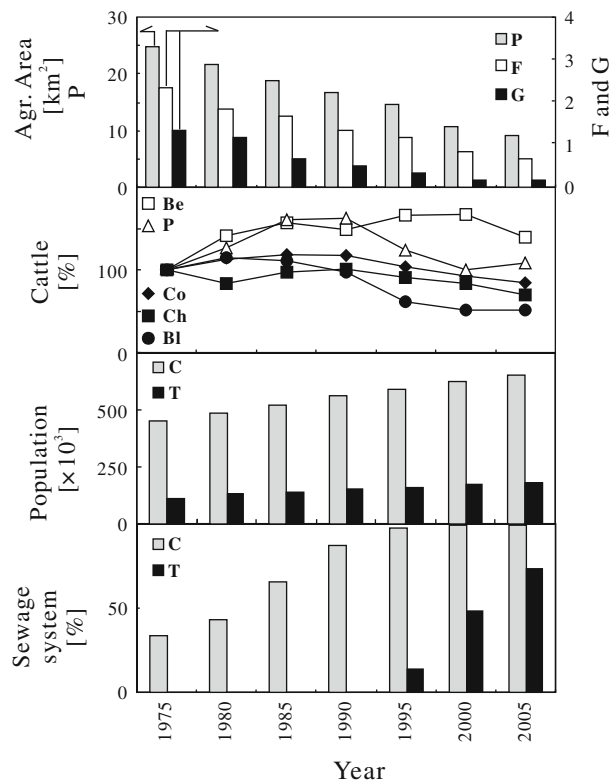


Figure 6. Annual trends in agricultural area, number of cattle farmers, population size, and usage rate for sewage treatment systems, 1975–2005. Brevity codes indicate paddy fields (P), fields (F), groves (G), cow (Co), beef (Be), pig (P), chicken (Ch), boiler chicken (Bl), the Higashi- and Hakata-wards in the city of Fukuoka (C), and the five towns (Umi, Sasaguri, Shime, Hisayama, and Kasuya Towns; T), respectively. The number of cattle in Fukuoka Prefecture was used (see “Methods” section) and was expressed as a percentage of the number in 1975.

approximately 30% of the 1977 levels, whereas the NO_3^- concentrations increased significantly over the same period (Figure 7). As NO_3^- concentrations increased, the reduction in TN concentration during 1977–2007 (approximately 50%) was less than the reduction in TP, and the NO_3^- fraction of TN increased to about 50% (Figure 7). Consequently, the N:P ratio in downstream water increased significantly during 1977–2007 (Figure 7).

DISCUSSION

N Saturation in Upland Forested Areas

High NO_3^- concentrations in stream water at the five sites in upland forests (Figures 2, 3) indicate that the upland forests in this basin exhibit symptoms of N saturation based on a study of the relationship between NO_3^- concentrations in stream

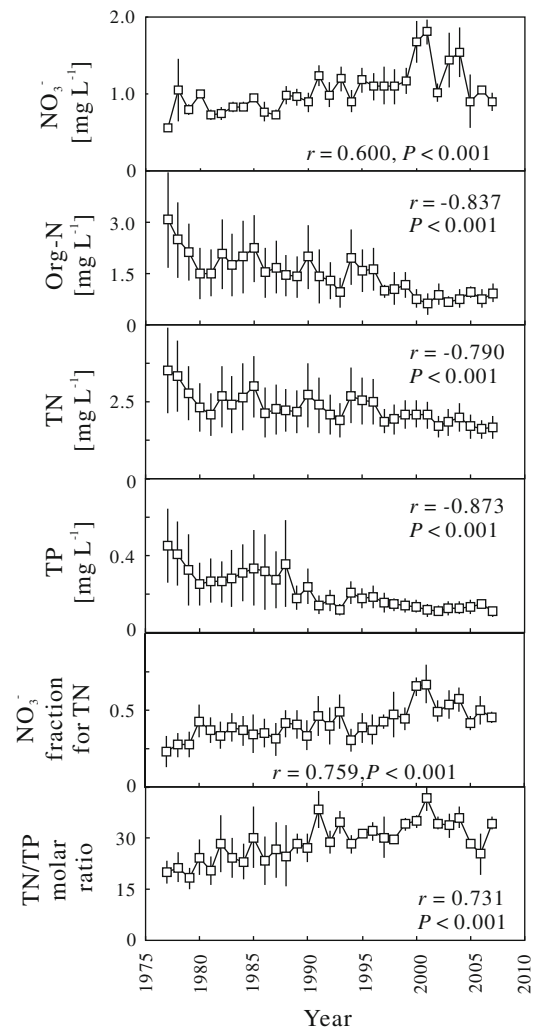


Figure 7. Long-term annual trends in NO_3^- , ON, TN, TP concentrations, the fraction of NO_3^- in TN, and the molar N:P ratio in downstream water, 1977–2007. Bars represent standard errors of three government monitoring sites.

water and N saturation status in forest ecosystems (Stoddard 1994).

The N saturation in forested areas of the Tataro River Basin is also associated with low N retention. The amount of annual inorganic N exports from the OEW, a forested watershed of this basin, via stream water was estimated at 10.4 (7.8–12.5) $\text{kg N ha}^{-1} \text{y}^{-1}$ [mean (range)] during 2004–2007 (Chiwa and others 2010) whereas the rate of N retention (N exports/atmospheric N deposition) was considerably lower at 33% (12–53%) [mean (range)] over the same time period (Chiwa and others 2010). The annual mean NO_3^- concentrations in stream water in the OEW [0.87 mg l^{-1} (0.76–0.97 mg l^{-1}) mean (range)] during 2004–2007 (Chiwa and others 2010) were comparable

with those in other forested sites in this basin (Figures 2, 3), and therefore the N exports from the OEW can be assumed to be representative of N exports from other forested areas in this basin.

In addition, atmospheric N deposition at the site closest to the forested area of this basin in recent years ($>10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$; Figure 4) was high enough to induce significant N leaching from forested areas. A sigmoid relationship between atmospheric N input and output was found by Ohrui and Mitchell (1997) (Figure 8), indicating that N inputs above $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ can induce high rates of N leaching. This phenomenon can also be applied to Japanese forests, including in OEW.

The forest condition, including forest type and stand age, can affect the retention or loss of N regardless of similar inputs (Ohrui and Mitchell 1997; Fukushima and others 2011). Nitrogen uptake by vegetation is an important factor regulating NO_3^- leaching (Vitousek and Reiners 1975). Ohrui and Mitchell (1997) demonstrated that lower N uptake in old stands can lead to lower N retention. They calculated that the difference in N uptake between 24- and 75-year-old stands of Japanese cedar and cypress plantations was approximately $30 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, resulting in higher levels of NO_3^- loss from older stands. Fukushima and others (2011) examined the N dynamics, including N uptake, of Japanese cedar plantation stands of different ages (5, 16, 31, 42, and 89 years), and found that N uptake was higher in younger stands (16-year-old) than in older stands (31, 42, and

89 years), and the difference in N uptake between the younger and older stands was approximately $20\text{--}30 \text{ kg N ha}^{-1} \text{ yr}^{-1}$. In the Fukuoka forest management unit, in the location of the Tataru River Basin, 56% of the forested area is Japanese cedar and cypress plantations, which have been approaching maturity (>46 year; Figure 5). Therefore, the maturing Japanese cedar and cypress plantations in recent decades may be an important factor leading to rapidly lowering N retention rates and significant N leaching from forested areas in the Tataru River Basin.

The evidence of N saturation caused by atmospheric N deposition in this basin is also supported by the dominance of NO_3^- over ON in stream water from forested areas. Although ON is the dominant form of N loss from unpolluted forests where atmospheric N deposition is naturally low (Hedin and others 1995; Williams and others 2001; Perakis and Hedin 2002), NO_3^- loss increases in response to anthropogenic N inputs and is dominant in polluted forests (Williams and others 2001; Stanley and Maxted 2008), due to increased soil ammonification and net-nitrification (Williams and others 2001; Aber and others 2003). Therefore, the ratio of inorganic N to ON in stream water can be used as an indicator of the N status of ecosystems (Williams and others 2001). The inorganic:organic N ratio in stream water from the five upland forested sites in this basin [5.4 (3.0–7.9); mean and range] was comparable with that in N-saturated forests [3.8 (Williams and others 2001)] and much higher than in unpolluted forests [<0.7 (Williams and others 1995; Kortelainen and others 1997; Perakis and Hedin 2002)].

N saturation across the forested areas in this basin could be caused partly by the long-range transport of anthropogenic nitrogenous compounds from East Asia causing the increase in atmospheric N deposition during the last few decades (Figure 4). Model calculations indicate that approximately 45% of the nitrogen deposition in Japan is received from East Asia (Lin and others 2008). In addition, the long-range transport of atmospheric N compounds from East Asia has been observed in an upland forested area of this basin (Chiwa 2010).

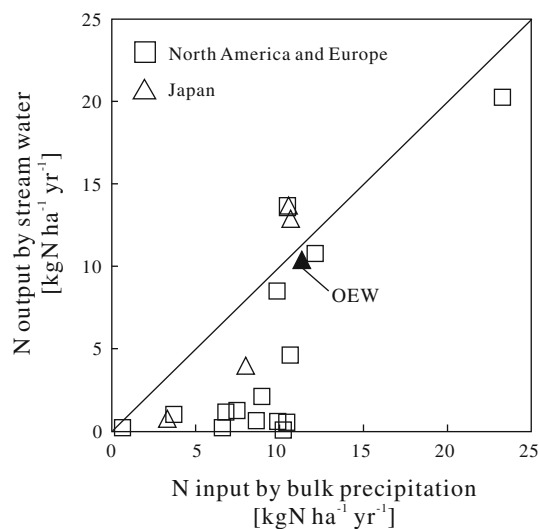


Figure 8. Relationship between the annual amounts of N from bulk deposition and in stream water for selected coniferous watersheds, modified from Ohrui and Mitchell (1997).

Impact of N-Saturated Forests on Stream Water Quality

Comparisons of the upstream and downstream water NO_3^- concentrations (Figures 2, 3) suggest that N leaching from N-saturated upland forests represents a large enough portion of the additional

N flux to have an impact on downstream water quality. This is important because there have been basin wide decreases in urban and agricultural sources of N, which will be discussed later. Previous studies have reported low NO_3^- concentrations in non-N-saturated forested areas and a negative relationship between NO_3^- concentrations and increasing non-N-saturated forest areas (Miller and others 1997; Norton and Fisher 2000; Salvia-Castellvi and others 2005; Hayakawa and others 2006; Floyd and others 2009). These previous studies suggest that upland non-N-saturated forests play a significant role in mitigating the degradation of downstream water quality. In this study, it is indicated that NO_3^- exports from N-saturated upland forests derived from atmospheric N deposition can be a non-point N source large enough to degrade downstream water quality in this basin, rather than mitigating the degradation of stream water quality as the ability to retain atmospheric N deposition is considerably reduced.

The instream processing of N, including biological uptake and denitrification in the stream, could lead to some changes in the nitrate concentration between forested areas and the coastal zone (Howarth and others 1996; Ohte and others 2010). Isotopic analyses including ^{15}N and ^{18}O (Ohte and others 2010) may be a potential way to evaluate the instream processing of N in future research.

The presence of a large non-point N source from forested areas in the Tataru River Basin is supported by the fact that the annual inorganic N export from OEW, a forested watershed in this basin, via stream water was higher than amounts reported from non-N-saturated forested watersheds, was comparable or lower than amounts reported from agricultural areas, and was comparable with reported exports from urban areas (Table 3). Thus, the N exports from N-saturated upland forest areas derived from atmospheric N deposition have become an important non-point source of downstream NO_3^- pollution in this basin.

Urban and Agricultural Activities

The impact of N-saturated upland forests on downstream NO_3^- pollution is highlighted by the reduction in N exports derived from urban and agricultural activities over the past few decades in this basin. The 70% decrease in agricultural areas during the 30 years from 1975 to 2005 (Figure 6) indicates a reduction in agricultural activity and the subsequent decrease in N and P exports derived from agricultural activities in this basin. This is supported by a 30–50% decrease in the number of dairy cows, pigs and chickens from 1990 to 2005 (Figure 6), which also indicates a decrease in N and P exports from these activities. The constant number of beef cattle since 1980 indicates that N and P exports from beef stock farming have not increased. The conversion of agricultural land to urban areas and the 70% increase in population over the same 30-year period (Figure 6) suggests that urban activity has been increasing and may subsequently cause increases in N and P exports from urban areas in this basin. However, the almost 100 and 80% usage rates for sewage treatment facilities in 2005 in Fukuoka City and the five towns in this basin, respectively (Figure 6) indicate that N and P pollutant exports to streams from urban areas have been constrained. Two sewage treatment facilities are also located in this basin and advanced treatment of wastewater occurs at both facilities (<http://gesui.city.fukuoka.lg.jp/>; <http://www.city.fukuoka.lg.jp/mizu/somu/index.html>), potentially leading to a reduction in N and P loads to stream water (Takeda 2000). In this study, the agricultural area, stock numbers, population, and sewage volumes were used as an index of non-point N and P sources from urban and agricultural areas. To further evaluate the N and P exports from urban and agricultural activities, a quantitative analysis is required. However, our qualitative analysis is suitable to evaluate annual trends in N and P exports from urban and agricultural activities.

Table 3. Outputs of Inorganic N from Forested, Agricultural, and Urban Areas

	Site	Output ($\text{kg N ha}^{-1} \text{y}^{-1}$)	Reference
Forest	Fukuoka, Japan	10.4	Chiwa and others (2010)
	Kyoto, Japan	1.4	Ohte and others (2001)
	Oregon, USA	<1	Vanderbilt and others (2003)
	Maryland, USA	0.1–0.2	Kaushal and others (2008)
	Ardennes region (Belgium–Luxembourg)	4	Salvia-Castellvi and others (2005)
Agriculture	Maryland, USA	7–30	Jordan and others (1997)
	Ardennes region (Belgium–Luxembourg)	27–33	Salvia-Castellvi and others (2005)
	Maryland, USA	3–39	Kaushal and others (2008)
Urban	Maryland, USA	2–15	Kaushal and others (2008)

The reduction in N and P exports to stream water from agricultural and urban activities is supported by the approximate 70% decreases in TP and ON concentrations and 50% decrease in TN concentration in downstream water over the 30-year period (Figure 7). Similar downward trends in P and N concentrations in stream water have also been reported by the pollutant reduction (Iital and others 2010). Furthermore, the significant positive relationships between TP and ON concentrations and the percentages of urban/agricultural area (Figure 3) support the idea that agricultural and urban activities are important sources of P and ON. These findings are consistent with previous studies that have identified agricultural and urban non-point sources of P (Miller and others 1997; Carpenter and others 1998) and ON (Kroeger and others 2006; Stanley and Maxted 2008).

Suggested Strategies for Improving Stream Water Quality

The NO_3^- concentration in downstream water in this basin increased significantly in spite of the reduction in agricultural activity and increases in sewage system wastewater treatment, unlike the TP, ON, and TN concentrations (Figures 6, 7). The increase in NO_3^- concentration may be caused by elevated atmospheric N deposition in recent decades (Figure 4) via three possible mechanisms. First, considerable amounts of atmospherically deposited N passes into streams without being retained by forests, leading to significant N leaching from N-saturated upland forests. The maturing of Japanese cedar and cypress plantations in recent

decades (Figure 5) may be an accelerating factor inducing further N leaching from forests, because maturation of these plantations could accelerate the reduction in N retention leading to significant N leaching from forested areas (Ohrui and Mitchell 1997). Second, elevated N deposition rates increase the mineralization/nitrification in forested areas, leading to significant N leaching from those forested areas (Aber and others 1998). Finally, elevated N deposition rates can increase the exports of atmospherically derived N from urban/agricultural areas because atmospheric N deposition is also a source of N from urban/agricultural areas (Jaworski and others 1997; Whittall and Paerl 2001; Schaefer and others 2009).

It should be noted that the discussion related to the increase in NO_3^- exports in recent decades in this basin is only based on stream water concentrations because flow measurements were not available for this watershed. However, there is no clear annual trend ($P > 0.05$; Mann-Kendall test) in the amount of annual precipitation in urban and forested sites in this basin (Figure 9), so changes in stream flow would have only limited impacts on the increase in NO_3^- exports in recent decades.

Approximately 50% of TN in downstream water comprises NO_3^- in recent years (Figure 7) indicating that NO_3^- is becoming a more important N pollutant species than ON. This means that the increased NO_3^- concentration has constrained the rate of reduction of TN, and the downstream N:P ratio increased during 1977 to 2007 (Figure 7). The N:P ratio is an important factor affecting whether primary production in estuaries and coastal waters is more likely to be limited by N or P, using the

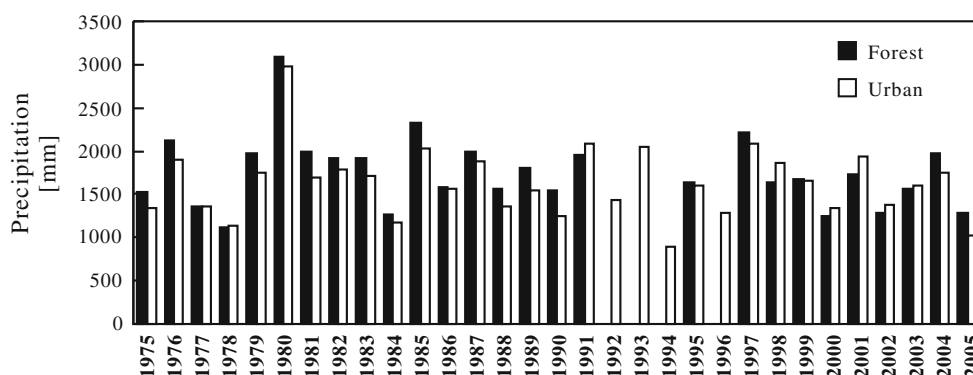


Figure 9. Long-term annual trends in annual precipitation in the urban (33°35'N, 130°23'E, 2.5 m a.s.l.) and forested site (33°38'N, 130°31'E, 70 m a.s.l.) in the Tataru River Basin from 1975 to 2005. The annual precipitation data were taken from a monitoring database provided by the Japan Meteorological Agency (<http://www.jma.go.jp/jma/index.html>) for the urban site, and from the Annual Report on Kyushu University Forests (http://leda.lib.kyushu-u.ac.jp/collection/co_l_id:236) for the forested site. The lack of bars between 1992 and 1994, and 1996 in the forested area indicates missing data.

Redfield ratio based on an approximate molar ratio of 16:1 (Howarth and others 1996). The N:P ratio in downstream water in this basin (Figure 7) indicates an apparent shift to P limitation over the past few decades, which may reduce phytoplankton diversity and impair food-web performance (Elser and others 2009). Thus, the sources of downstream NO_3^- should be reduced to improve stream water quality.

We propose a reduction in the NO_3^- export from forested areas as a strategy to reduce non-point source N pollution and improve downstream water quality. This strategy is proposed because the NO_3^- concentrations in upland stream water were comparable with those in downstream water and are an important non-point N source, as described earlier. In addition, as forests cover a large proportion of the total land area in this basin, forests can have a dominant influence on downstream water quality.

The effort to reduce NO_3^- exports from upland forest areas proposed here is a new strategy to reduce non-point source N pollution in a basin. To achieve this, it is necessary to improve N retention in upland forests and reduce atmospheric N deposition. To improve N retention in upland forest, regional-scale implementation, including reforestation, may be necessary. In addition to regional-scale implementation, our proposed strategy includes reducing atmospheric N emission at an intercontinental-scale to improve regional stream water quality. This is important as atmospheric N deposition in this basin has been affected by both long-range transport from East Asia and domestic transport, which have subsequently induced significant N leaching from the forested watershed.

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