Current Status of Nitrogen Saturation in the Upper Reaches of the Kanna River, Japan

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***Abstract*—**Nitrogen saturation has become one of the serious issues in the field of forest environment. The watershed protection forests located in the downwind hinterland of Tokyo Metropolitan Area are believed to be facing nitrogen saturation. In this study, we carefully focus on the balance of nitrogen between load and runoff. Annual nitrogen load via atmospheric deposition was estimated to 461.1 t-N/year in the upper reaches of the Kanna River. Annual nitrogen runoff to the forested headwater stream of the Kanna River was determined to 184.9 t-N/year, corresponding to 40.1% of the total nitrogen load. Clear seasonal change in NO_3-N concentration was still observed. Therefore, watershed protection forest of the Kanna River is most likely to be in Stage-1 on the status of nitrogen saturation.

***Keywords*—**Atmospheric deposition, Nitrogen accumulation, Denitrification, Forest ecosystems.

I. Introduction

SEWAGE effluent and/or drainage from agricultural fields is/are generally believed to be possible sources of the nitrogen contamination of stream water. In the last decade, however, much literature has pointed out remarkable nitrogen contamination in forested headwater streams surrounded by nature [1]-[3]. Despite the absence of anthropogenic sources, high concentrations of nitrogen in excess of the national average of 0.4 mg-N have been observed [4]. In our previous study, we demonstrated that the upper reaches of the Kanna River, which is one of the headwater streams flowing into the Tone River, is also contaminated with high concentration of NO_3-N [5]. We mainly discussed the possibility of nitrogen saturation due to the decline of forestry in the watershed protection forests of the Kanna River. Briefly, nitrogen saturation means the situation in which the nitrogen load exceeds the capacity of nitrogen uptake and denitrification in the forest ecosystems. As a result, non-negligible amounts of nitrogen runoff may be observed. As we discussed in the previous study, several policies such as carbon credit and carbon offset platforms may trigger the revitalization of forestry in Japan. However, for a better understanding of the current status of nitrogen saturation, we need to pay more attention to not only the integrity of forest ecosystems, but also the nitrogen deposition via air pollution, because the hinterland of megacities is generally affected by polluted air. The upper reaches of the Kanna River are located in the hinterland situated in the northwest mountainous areas approximately 100 km from the center of Tokyo. Therefore, the atmospheric deposition is likely to be one of the significant causes of nitrogen saturation. The current study carefully focuses on atmospheric nitrogen deposition including dry and wet depositional processes.

In Japan, the Air Pollution Control Act was established in 1968 by Ministry of the Environment, and the emissions from stationary and mobile sources have been controlled. Although the current NO_2 concentration has met its environmental standards to date, depositional fluxes of nitrogen compounds tend to be increasing in the downwind hinterland areas of Metropolitan Tokyo [6]. Many articles have been concerned with the relationship between the nitrogen saturation of forest ecosystems and the overload of nitrogen compounds by an atmospheric deposition [7]-[9]. On a global scale, the atmospheric emissions of nitrogen pollutants tend to increase continuously [10], [11]. Hence, the nitrogen balance in the ecosystem probably changes incrementally [12].

This study is intended to estimate the balance between the nitrogen load and its runoff to the headwater stream of the Kanna River. First, we roughly estimated dry and wet depositional fluxes of nitrogen compounds by using the database from Acid Deposition Survey in Japan [13], [14]. Next, we determined the amount of nitrogen runoff to the Kanna River based on the field monitoring data. Finally, by focusing on the balance between nitrogen load and runoff, we discuss the current status of nitrogen saturation from the viewpoint of functions such as nitrogen uptake and denitrification in forest ecosystems.

II. Experimental

A. Observation Sites

The observation sites in the current study are the same as in our previous work [5]. Briefly, the Kanna River is one of the headstreams of the Tone River (gross length: 322km; gross basin area: 16,840km^2, the largest river in Japan). The upper reaches of the Kanna River are located in the northwest mountainous areas situated in the downwind hinterland of Tokyo Metropolitan Area during the warm season. In this study, three monitoring stations (St.1, St.2, and St.3 in Fig. 1) were placed in the upper reaches of the Kanna River (gross length: 87.4km; gross basin area: 407km^2). Most of the water catchment area (approx. 94%) is covered with forest. Forestry was once active but now it is facing serious decline due to the falling price of ligneous sources and a staff shortage.

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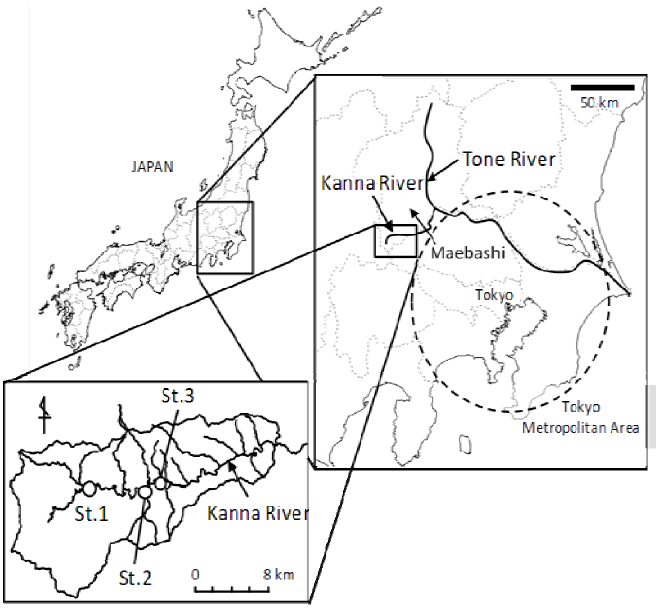

Fig. 1 Location of the upper reaches of the Kanna River (The map is cited from our previous paper [5])

B. Estimation of Nitrogen Loading via Atmospheric Deposition

The datasets of dry and wet deposition were cited from Acid Deposition Survey in Japan, Phase 4. This national survey was set up to investigate the trend of acid rain, and was conducted from 2003 to 2005 at more than 60 monitoring stations [13], [14].

Particulate (p) and gaseous (g) compounds (as a dry deposition) were collected weekly by using the filter-pack method [15]. A NILU holder packed with four different types of filters (1st stage: PTFE filter for aerosol sampling, 2nd stage: polyamide filter for HNO_3 sampling, 3rd stage: cellulose filter for SO_2 and HCl sampling, 4th stage: phosphoric acid impregnated cellulose filter for NH_3 sampling) was used. Sample loaded filters were put into plastic bottles with pure water, and extracted by using an ultrasonic bath. The extracts were then filtered by using a membrane filter with a pore size of 0.45 µm, and ionic species (SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, Na^+, K^+, Ca^{2+} and Mg^{2+}) were analyzed by using an ion chromatography [13].

Rain water samples (as a wet deposition) were collected weekly by using an automatic wet-only rain sampler [16]. The samples were filtered by using a membrane filter with a pore size of 0.45µm, and then ionic species were analyzed by using an ion chromatography [13].

The depositional fluxes of respective species were estimated based on an inferential method [15], [17], [18]. In this study, we cite the datasets of nitrogen deposition (dry deposition: NH_4^+(p), NO_3^- (p), NH_3 (g), and HNO_3 (g), wet deposition: NH_4^+ and NO_3^-) obtained from 33 monitoring stations in which the datasets of both dry and wet depositions were simultaneously determined.

C. Estimation of Nitrogen Runoff to the Kanna River

River water samples were collected monthly at each monitoring station (St. 1, 2 and 3 in Fig. 1) from April 2012 to March 2013. Approximately 3 L of surface water was collected from the center of the river. Samples were filtered by using a membrane filter with a pore size of 0.45µm. Dissolved inorganic nitrogen (DIN) species, namely ammonium ion (NH_4^+), nitrite ion (NO_2^-), and nitrate ion (NO_3^-), were analyzed by using an ion chromatography. The amount of nitrogen runoff was determined by the same method applied in our previous work [5]. Briefly, the annual water runoff to the Kanna River was calculated by subtracting the possible evapotranspiration from annual precipitation. Annual nitrogen runoff was determined by multiplying the annual water runoff by the average DIN concentrations.

III. Results and Discussion

A. Distribution of Nitrogen Deposition

Fig. 2 illustrates the distribution of annual depositional fluxes of nitrogen. The dry depositional fluxes ranged from 0.14 to 1.46 t-N/km^2/year, and the average value was 0.51 t-N/km^2/year (Fig. 2 (a)). Higher values were typically observed at the suburban sites located in the hinterland of large cities. Here we focus on the data of Maebashi that is the nearest city to our research site. In Maebashi, a relatively high value (0.82 t-N/km^2/year) was observed. Gaseous compounds accounted for approximately 90% of the total dry depositional flux. In particular, NH_3 (g) was the significant contributor. Shimoda et al. have pointed out that NH_3 concentration in Maebashi is extraordinarily high as compared to other neighboring sites [6]. The simple reason for this is that Maebashi is located in an area of concentrated livestock industries.NH_3 concentration in Maebashi is generally four times as high as that in other neighboring sites unaffected by livestock industries. On the other hand, depositional fluxes of HNO_3 (g), NH_4^+(p), and NO_3^- (p) in Maebashi are comparable with the neighboring sites. By considering the overload of NH_3 in Maebashi, dry depositional flux of nitrogen in the upper reaches of the Kanna River can be roughly estimated as 0.47 t-N/km^2/year.

The wet depositional fluxes ranged from 0.34 to 1.87t-N/km^2/year, and the average value was 0.97 t-N/km^2/year (Fig. 2 (b)). The data in Maebashi was 1.87 t-N/km^2/year, which was the highest value of all the stations. According to the national survey database, there were no significant differences in the wet depositional fluxes among Maebashi and its neighboring sites. Therefore, we estimated wet depositional flux of nitrogen in the upper reaches of the Kanna River as 1.87 t-N/km^2/year which is the same value as in Maebashi.

The total depositional fluxes ranged from 0.48 to 2.69t-N/km^2/year, and the average value was 1.47 t-N/km^2/year (Fig. 2 (c)). The data in Maebashi was 2.69 t-N/km^2/year, which was the highest value of all the stations. From the above estimation, total depositional flux of nitrogen in the upper reaches of the Kanna River can be roughly estimated as 2.34 t-N/km^2/year. Hence, it is confirmed that our research sites are severely affected by the polluted air transported from the Tokyo Metropolitan Area.

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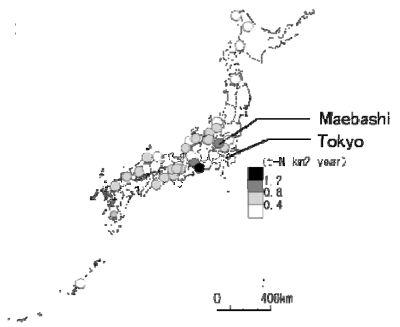
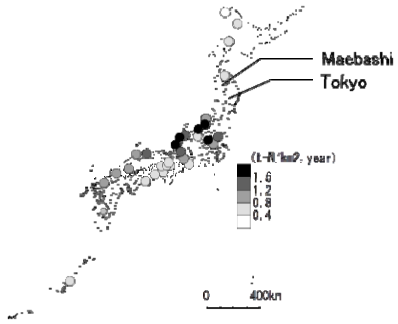
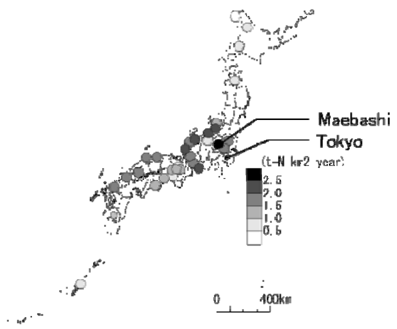
By multiplying the total depositional flux by the water catchment area (197.03km^2), annual nitrogen load via atmospheric deposition can be estimated as 461.1 t-N/year.



(a) Dry deposition



(b) Wet deposition



(c) Total deposition

Fig. 2 Distribution of nitrogen deposition

B. Estimations of Nitrogen Runoff from the Forest Ecosystems

Table I summarizes the average DIN concentrations from April 2012 to March 2013. The limits of determination (LOD) for respective DIN species have improved as compared to our previous data owing to the highly sensitive analysis from using an ion chromatography. Considerably high concentrations of NO_3-N (0.84–0.92 mg-N/L) were determined. These values were more than twice the national average of 0.4 mg-N. This suggests that the watershed protection forest of the Kanna River has been facing a decline in functions of forest ecosystems.

According to meteorological observations, annual precipitation in the upper reaches of the Kanna River was estimated to be 1,443 mm/year. By using Thornthwaite's method, annual evapotranspiration was calculated to be 433.8 mm/year. Consequently, the amount of annual water runoff can be determined as 1009.17mm/year. This corresponds to 198.8 × 10^{6}m^3/year of annual water flow at St. 3 in which the water catchment area is 197.03km^2.Therefore, the amount of nitrogen runoff at St. 3 can be estimated as 184.9 t-N/year by multiplying the annual water flow by the total DIN concentration of 0.93 mg-N/L (here, the concentrations of NH_4-N and NO_2-N were assumed to be half the value of their LODs).

TABLE I
CONCENTRATIONS OF DISSOLVED INORGANIC NITROGEN SPECIES

Species	Concentration (mg-N/L)		
	St.1	St. 2	St. 3
NH_4-N	0.01	0.02	<0.01
NO_2-N	<0.01	<0.01	<0.01
NO_3-N	0.84	0.91	0.92

C. Comparison between the Nitrogen Load and Runoff

Fig. 3 summarizes the relationship between the nitrogen load and its runoff to the headwater stream of the Kanna River. Estimated nitrogen runoff (184.9 t-N/year) accounted for 40.1% of the nitrogen load (461.1 t-N/year).

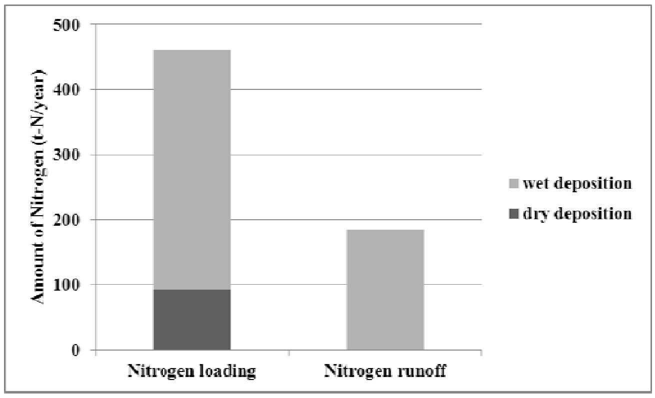


Fig. 3 Balance of nitrogen load and runoff at the upper reaches of the Kanna River

Because of its bioavailability, nitrate is generally taken up by plants as a nutrient, or denitrified by soil microbes. Therefore, nitrogen compounds hardly leach from the forest ecosystems (see Fig. 4) [1], [11]. In fact, a temperate forest is generally situated in nitrogen limitation [8], [10]. When the nitrogen loading exceeds the capacity of nitrogen uptake and denitrification in the forest ecosystems, however, overloaded nitrogen starts leaching into the headwater streams (see Fig. 4) [1]. We refer to this situation as nitrogen saturation. In this case, forest ecosystems will be free from nitrogen limitation.

According to the research by Stoddard, the status of nitrogen saturation can be classified into four stages depending on the seasonal characteristics of NO_3-N concentration [19]. InStage-0, there is no significant NO_3-N runoff in the streams. In Stage-1, the forest grows rapidly by increasing the nitrogen deposition. We can observe a certain amount of NO_3-N in the stream water with clear seasonal change. In Stage-2, the forest will be free from nitrogen limitation due to the overloading of nitrogen. A considerable amount of NO_3-N will be found, and clear seasonal change will not be observed. In Stage-3, a large amount of NO_3-N runoff exceeding the total load will be observed.

Many articles have shown concern the great majority of forests located in the hinterland of Tokyo Metropolitan Area are high in nitrogen saturation [20]. In some cases, considerable amounts of NO_3-N corresponding to the full load were observed in the headwater streams connecting to the Tone River [7]. This case is generally believed to be in Stage-2. As estimated above, nitrogen runoff in the upper reaches of the Kanna River will correspond to 40.1% of the total load. In addition to this, clear seasonal change in NO_3-N concentration was still observed. Consequently, watershed protection forest of the Kanna River is most likely to be in Stage-1. In other words, the functions of forest are facing a gradual decline, and causing the nitrogen runoff. Overload of nitrogen compounds via atmospheric deposition will worsen the status of nitrogen saturation.

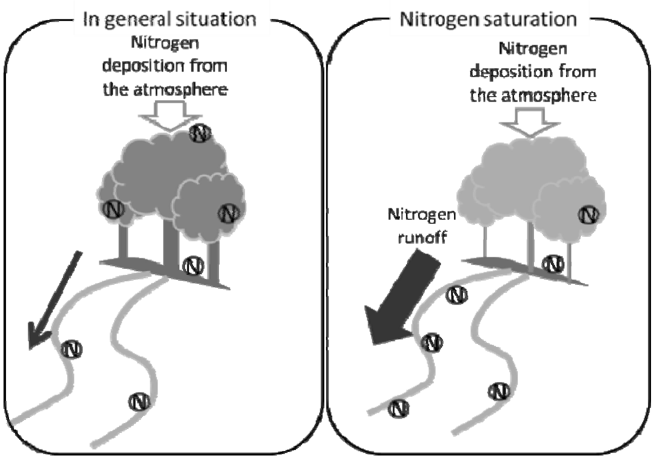


Fig. 4 Schematic view of the nitrogen load and runoff

From the difference between total nitrogen load and runoff, accumulation flux in our research sites is estimated to be 1.4 t-N/km^2/year. This seems to be slightly lower than that of the other sites (*e.g.*, 2.2 t-N/km^2/year in Tsukuba [21]), suggesting that the capacity of nitrogen uptake or denitrification has dropped. We cannot conclude whether the forest ecosystems have already lost their functions, however it is important they regain the functions that we expected.

D.Relationship between the Status of Nitrogen Saturation and Forestry Industry Trends in Japan

Nitrogen runoff is typically found in areas receiving high nitrogen load (>2.5 t-N/km^2/year). In moderately loaded areas (0.5–2.5 t-N/km^2/year), nitrogen concentration in the stream water varies depending on the surroundings. Ohrui and Mitchell have demonstrated that cedar forests situated in moderately loaded areas (1.05t-N/km^2/year) have been facing nitrogen saturation [22]. This indicates that there are some important factors other than the increase in nitrogen load via atmospheric deposition.

Gundersen et al. investigated the relationship among several parameters and nitrogen runoff. Parameters that correlated with the nitrogen runoff were defined as N status. For instance, tree species and age, C/N ratio in soil, course of disturbance due to meteorological event or vermination, and history of the land use were identified as the N status. It is difficult to elucidate the functions of forest ecosystems which are complicatedly related to above mentioned parameters [23]. However, nitrogen saturation will be dominated by not only the increase in nitrogen load via atmospheric deposition but also the combination of N status parameters.

From viewpoints of the history of land use and tree age, the worsening of nitrogen saturation might be associated with the increase in abandoned artificial forests. As mentioned in our previous study, the falling price of ligneous sources and a staff shortage have caused a rapid collapse of Japan's forestry. As a result, tree age in artificial forests is getting higher [5]. In addition, forestry collapse has caused serious damage to growing forests by leaving them without management. In the abandoned forest, the accumulation rate of nitrogen will drop while the nitrification rate will increase [24], [25]. Overload of nitrogen via atmospheric deposition may accelerate this process, and finally cause the nitrogen runoff [26]. The upper reaches of the Kanna River are surrounded by great swathes of artificial forest. However, most of them are far from adequately managed. In addition to the increase in atmospheric deposition, the decline of forestry is also an important N status parameter in our research sites.

E. Key Issue and Future Prospect of the Policy on Atmospheric Environment

Several policies contributing to forest preservation have been on the right track as reviewed in our previous study. On the other hand, there is still room for improvement on the policies for conserving the atmospheric environment.

When we focus on the atmospheric nitrogen pollutants, NO is a primary pollutant emitted from combustion sources. During advection and dispersion, NO is oxidized to NO_2.Furthermore, NO_2 reacts with ・OH to form HNO_3.This causes the formation of secondary particulate matter, acid rain, and acid fog. On the other hand, NO_2 can photolyze to NO and O. This process contributes to the generation of photochemical oxidant. Thus, NOx is an important precursor of secondary pollutants. Today, two major regulations, the Air Pollution Control Act and the Law Concerning Special Measures for Total Emission Reduction of Nitrogen Oxides and Particulate Matter, have been enforced in Japan. Owing to these regulations, the achievement rate of Environmental Quality Standard for NO_2 has been kept in favorable condition so far (100% in residential sites and 99.5% in roadside sites in 2011) [27]. However, air pollution by the above mentioned secondary pollutants remains a matter of grave concern for hinterlands located in the downwind area of megacities. In the northwest suburban area of Tokyo, for instance, high concentration of nitrate is generally found in fine particulate matter [28]. Moreover, acid fog with pH less than 3.5 has been occasionally observed in mountainous areas [29]. Furthermore, very high concentration (more than 120 ppm) of photochemical oxidant is frequently observed in the warm season [27]. Overload of nitrogen via atmospheric deposition will not be unrelated to these phenomena. No wonder many researchers point out the effect of air pollution as a dominant contributor of nitrogen contamination in stream water. Consequently, further reduction of air pollutants is required for improving the nitrogen balance in the forest ecosystems.

IV. Conclusion

Our findings reveal that 40.1% of total nitrogen load has been leached to the headwater stream, suggesting that the watershed protection forest of the Kanna River has been facing nitrogen saturation. In addition to the overload of nitrogen via atmospheric deposition, the increase of abandoned artificial forest due to the decline of forestry may also have caused the disruption of biogeochemical cycling of nitrogen in forest ecosystems. A multidirectional approach based on both the conservation of the atmospheric environment and the revitalization of forestry is required to regain the functions of forest ecosystems.

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