Organic and Inorganic Nitrogen Deposition in Two Contrasting Forest Types in Gifu Prefecture, Central Japan

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(岐阜県の対照的な二つの森林生態系における有機態および無機態の窒素沈着)

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The United Graduate School of Agricultural Science, Gifu University

Science of Biological Environment

(Gifu University)

CAO RUOMING

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Chapter 1

INTRODUCTION

1.1. The N cycle of forest ecosystems

A third of the Earth's land surface is covered by forests, and these ecosystems provide multiple services (e.g., fiber production, biodiversity, purification of air and water, and conservation of equable climate; Jones et al., 2014). Nitrogen (N), as a major essential nutrient for plants, its cycle of forest ecosystems is an important part of the global biogeochemical cycle. Various forest types with different tree species, soil types, topography, meteorology, and adjacent land use, cause a challenge to gain integrated insights into the N cycle of forest ecosystems. Therefore, studies about the N cycle of forest ecosystems are still urgently needed.

N cycle (Fig. 1.1) includes three parts: N input, internal N cycle (N fluxes and N pools), and N output (Gundersen, 1991). N inputs into forest ecosystems through N_2 fixation or N deposition. N_2 fixation amounts to 1–2 kg N ha^{-1} year^{-1} (Cole, 1995), while the amount of N deposition depends on anthropogenic N emission. The main processes in the internal N cycle consist of mineralization, nitrification, immobilization, litterfall, and uptake by

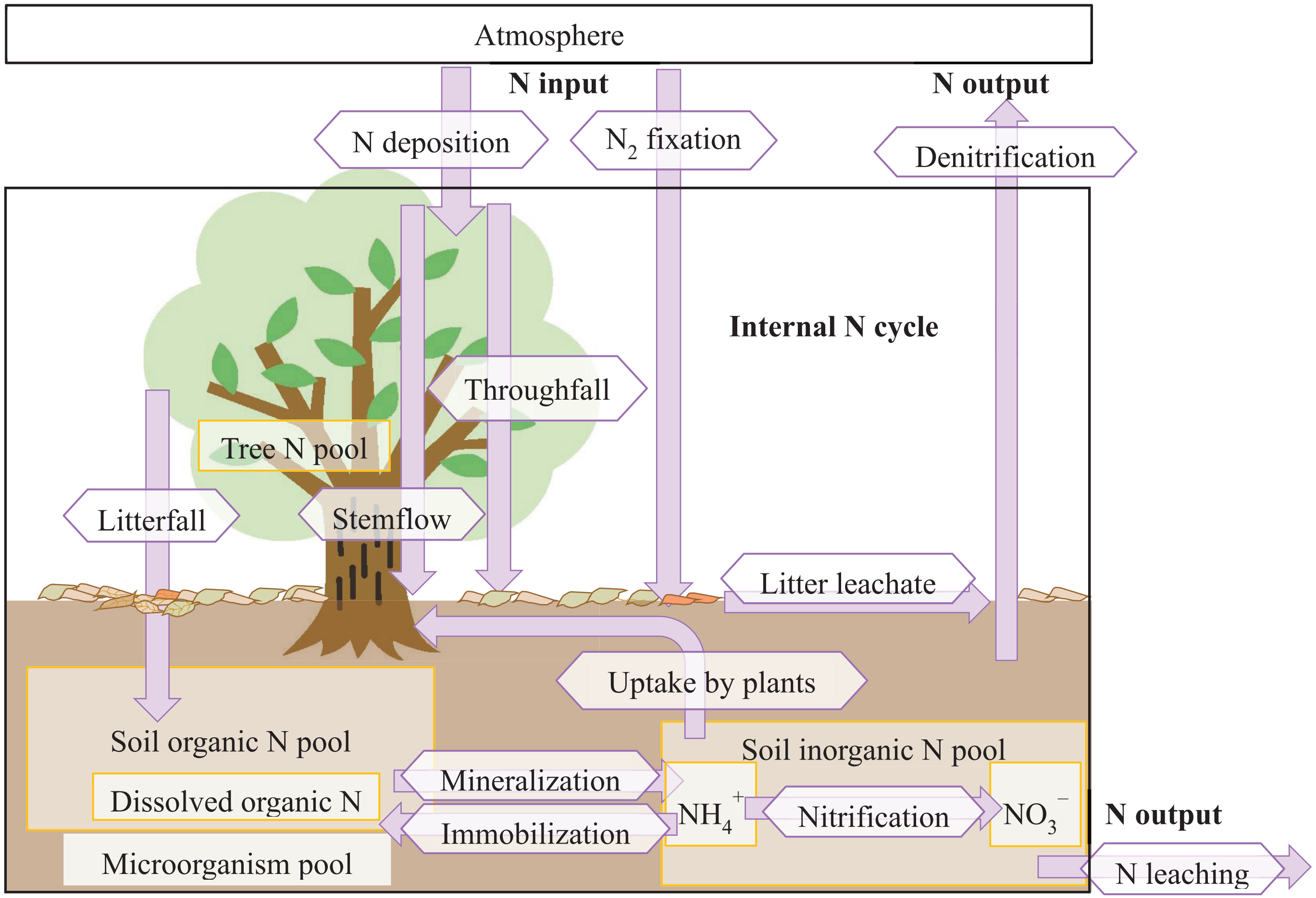

Fig. 1.1. N cycle of forest ecosystems.

plants. N output from forest ecosystems includes denitrification and N leaching. N deposition, as a primary process between forest ecosystems and atmosphere, is supposed to be a significant process in the N cycle of forest ecosystems.

1.2. N deposition in N cycle of forest ecosystems

N deposition (Fig. 1.2) includes wet deposition (WD) via precipitation, and dry deposition (DD) as gases and particles. Wet or bulk deposition (BD) can be directly measured and quantified using wet-only or bulk collectors, while it is difficult to estimate total N deposition (wet and dry) in forest ecosystems. Due to the relative simplicity, the most known and widely used method to measure total N deposition in forest ecosystems is the "throughfall method" (Butler and Likens, 1995). Total N deposition onto the forest floor can be calculated by measuring the quantity and quality of the precipitation passing through the forest canopy, which collects the dry deposition from the canopy's surface. These canopy-derived flows are referred to as throughfall (TF) and stemflow (SF). N deposition is comprised of dissolved inorganic N (DIN: NH_4–N and NO_3 + NO_2–N) and dissolved organic N (DON) in water fluxes. The difference (or enrichment) between TF (plus SF) and WD (or BD) is called net TF, which was first proposed by Bredemeier (1988). It reflects the DD and possible canopy exchange (uptake or leaching) as incident precipitation occurs.

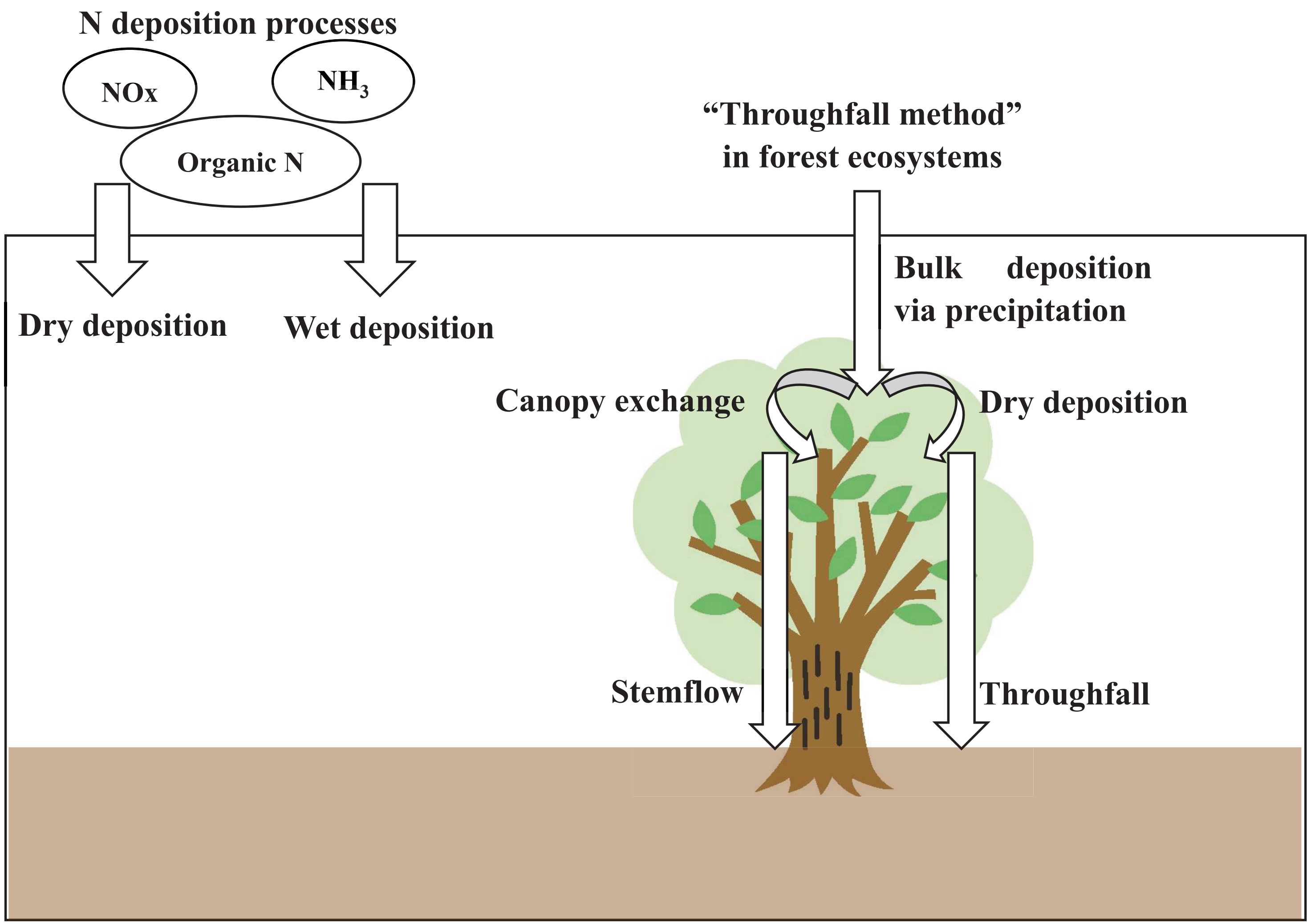

Fig. 1.2. Pathways of nitrogen deposition into the forest ecosystems.

Studies about N deposition (BD, TF, and SF) in Japanese forest ecosystems have started mainly in artificial evergreen forests (Japanese cypress, Japanese cedar, and Japanese red pine) since the 1960s (Maruyama et al., 1965; Iwatsubo and Tsutsumi, 1967; Iwatsubo and Tsutsumi, 1968; Nishimura, 1973). Bulk NH_4–N deposition range was 2.1–4.3 kg N ha^{-1} $year^{-1}$, and bulk NO_3–N deposition range was 1.4–2.6 kg N ha^{-1} $year^{-1}$. N fluxes in TF varied with forest types, which were higher or lower than those in BD. N fluxes in SF were lower compared with BD and TF, with 6%–23% contribution to total N deposition (TF + SF). N budget (N input – N output) in the Japanese forest ecosystems was positive estimated by the difference between BD and stream discharge, indicated that excess N leaching didn't occur. The N cycle of the Japanese forest ecosystems was thought to be a closed system with limited N at this stage. It was reported that BD contributed 8% to annual N requirement for tree growth (Maruyama et al., 1965). Therefore, N deposition was recognized as an important nutrient supply for N input in N cycle at this stage, which is not able to be overlooked.

1.3. Double-sided roles of chronic N deposition under human activities in N cycle of forest ecosystems

With increasing N pollution of the atmosphere under frequent human activities (e.g., fossil fuel combustion), the adverse effects of enhanced N deposition on forest ecosystems have been highlighted as significant concerns. At the same time, acid rain, as one of the global environmental issues, attracted researchers' attention since the 1970s. It was reported that NO_3–N concentration in rainfall has increased approximately three times in forest sites in Kyoto and Shiga Prefecture since the second half of the 1970s (Naoko and Goro, 1992). Since the 1980s, the forest dieback of Japanese cedar subsequently occurred in the Kanto district of Japan. Under these conditions, the research topic focused on the negative effects of acid deposition on forest ecosystems. The negative impacts of acid precipitation on plants were clarified, such as injury to foliage and flower (Nouchi, 1990). Soil pH (pH < 4 in 0–10 cm soil depth) was lower in decaying Japanese cedar sites than not decaying sites in the Kanto district, possibly due to acid deposition (Matsuura et al., 1991). N cycle of Japanese forest ecosystems tended to be N saturated (Mitchell et al., 1997); excessive N leaching from forests was found (e.g., Ogura et al., 1986; Ohrui and Mitchell, 1997; Ham et al., 2007). On the other hand, it was still a closed system in some N limited forests; output in stream water was low although annual N input amounted to 3–17 kg N ha^{-1} $year^{-1}$ (Naoko and Goro, 1992; Feng et al., 1999), which was utilized and retained in the internal N cycle of forests. Therefore, double-sided roles of N deposition on forests was realized with positive effects (e.g., increased tree carbon storage, Thomas et al., 2010; accumulation of litter decomposition, Zhu et al.,

2015) or negative effects (e.g., acceleration of soil acidification, Lu et al., 2014; the threats to ecosystem biodiversity, Bobbink et al., 2010) at this stage.

Nowadays, developed countries (the United States and Central and Western Europe) have experienced a decrease in N deposition since the 1980s because of decreased emissions under associated abatement policies (Gilliam et al., 2019; Schmitz et al., 2019). Inorganic N emissions in Japan have also decreased because of strict emission abatement policies (e.g., air NO_2 emission has decreased five-fold compared with that in the 1970s, Japan's Ministry of the Environment). However, it is unclear about current status of N deposition in Japanese forest ecosystems, which is needed to clarify.

1.4. Uncertainties and urgent future research needs

Although monitoring efforts have substantially improved our understanding of N deposition and its effects in Japanese forest ecosystems, uncertainties still remain. Five unclear points reveal as the followings.

Most of the previous studies were conducted on artificial forest sites, which were artificially planted to meet the demand for wood production since the 1950s. However, only two previous studies were conducted in natural forest sites, which involved Veitch's fir (*Abies Veitchii*), Birch (*Betula platyphylla* Sukatchev var. *japonica* Hara), and Konara oak (*Quercus serrata*) tree species (Oura et al., 2006; Seki et al., 2010). Half of the forest area is covered by natural forests in Japan, so that it is necessary to fill the gap of N deposition on the natural forests with various forest types to comprehensively understand the national distribution and effects of N deposition on Japanese forest ecosystems.

N deposition in rainfall has well been estimated, while previous studies have rarely reported the N deposition in snowfall. Merely Oura et al., (2006) estimated the contribution of snowfall to N deposition in a cool-temperate forest, which amounted to 45% of N deposition. Snowfall, as a non-negligible component of precipitation in cool-temperate forest ecosystems, may have deep implications for N deposition processes in cool-temperate forests.

Comparative studies on N deposition levels and dynamics have been conducted based on rural and urban site classifications (Aikawa et al., 2006; Chiwa et al., 2003). These studies indicated that N deposition were higher in urban sites than in rural sites. Jia et al. (2014) reported a negative correlation between N deposition and the distance between the forest sites and the nearest large city in China. However, there is still a lack of data on N deposition to urban forests in Japan, which makes it difficult to understand the general patterns of N deposition processes along urban-rural gradients in Japanese forest ecosystems.

DIN deposition has long been routinely assessed and quantitatively evaluated in

Japanese forest ecosystems; however, data on DON deposition are still rare due to the complexity of analysis and past neglect. DON is considered an important part of the dissolved organic matter (DOM) pool that plays a vital role in many biochemical processes in soil (McDowell, 2003), especially in microbial activities (Jones et al., 2004). As a component of N deposition and a source of organic N in the soil, the importance of DON deposition to forest floor should be paid attention to evaluate the effects of total N deposition on forest ecosystems. Thus, further work needs to be conducted on the N deposition fluxes (DIN and DON) of forests, in order to comprehensively improve the existing understanding of interactions between forest ecosystems and atmosphere in biogeochemical cycles.

DD is one of the knowledge gaps in Japanese forest ecosystems due to measurement difficulties. Mitchell et al. (1997) found that there was a significant positive relationship between BD and N drainage in stream water ($r^2 = 0.34$, $p < 0.01$). However, large N drainage unexpectedly occurred in some forest sites with low BD input (e.g., 7.6 kg N ha^{-1} $year^{-1}$ of input < 28 kg N ha^{-1} $year^{-1}$ of output, Mitchell et al., 1997). DD and DON deposition (also mentioned before) may cause the large N output, which was not considered in previous studies. Thus, further work needs to be conducted on the N deposition fluxes (DIN and DON), including DD and BD, in order to comprehensively improve the existing understanding of total N deposition. Moreover, the forest type affects the DD into the forest ecosystems, which may capture more DD and subsequently cause large N drainage from forest ecosystems. De Schrijver et al. (2007) reported that the forest type significantly affected net TF deposition, because a higher DD was captured in evergreen forests due to their greater leaf area index (LAI). It is necessary to estimate N deposition in various forest types to enrich the knowledge of N deposition mechanisms at Japanese forest fields.

1.5. Purposes of the study

For this study, we assessed N deposition in the BD, TF, and SF of two natural forest sites (chapter 2 and chapter 3) in Gifu Prefecture with different forest types (a deciduous broad-leaved forest and an evergreen broad-leaved forest), different climate types (cool-temperate and subtropical), and different site types (rural and urban), to fill the current knowledge gaps. The main objectives of this study were to evaluate (1) Snowfall contribution in BD (2) BD characteristics concerning DIN and DON under two site types (3) TF and SF characteristics concerning DIN and DON under two forest types (4) DIN and DON enrichment characteristics in net TF.

Chapter 2

N DEPOSITION IN A COOL-TEMPERATE DECIDUOUS BROAD-LEAVED FOREST

2.1. Introduction

N deposition into the cool-temperate forest ecosystems with abundant annual precipitation on Asia have been neglected compared with those on North America and Europe having limited precipitation (Izquieta-Rojano et al., 2016; Lovett and Lindberg, 1993; Pelster et al., 2009; Verstraeten et al., 2016). Therefore, it is critical to identify DIN and DON deposition levels for supplying a premise to get a more complete understanding of N deposition in the cool-temperate forest ecosystems.

The primary point of contact between a forest ecosystem and N deposition is the canopy. From the perspective of the hydrological cycle, BD can be divided into two pathways by which precipitation reaches the soil: TF and SF. Soil N cycle may be impacted by water flows (TF and SF). TF is known to be an important hydrological pathway that reflects canopy interactions and DD in the internal N cycle of forest ecosystems. In contrast, we are yet to fully understand N dynamics in SF, which can concentrate a greater water flux into the root zone (Bellot and Escarre, 1998). Furthermore, the TF of understory plants has often been overlooked. In forest ecosystems with dense understory plants, there may be a major influence of the understory canopy on TF passing through the tree canopy to the soil (Chen and Mulder, 2007). Therefore, TF and SF are considered important conduits between the atmospheric N deposition input and the soil N cycle of forest ecosystems; as such, it is necessary to better understand the fate of N deposition based on the hydrological cycle and N nutrient cycling in forest ecosystems.

The present study in this chapter was designed to estimate the concentrations and fluxes of dissolved N in BD, TF above a bamboo canopy (TF_a), TF below a bamboo canopy (TF_b), and SF over three years in the Takayama forest site, Central Japan, which is a cool-temperate forest with a dense bamboo understory. The site was established as one of the AsiaFlux network sites for measuring CO_2 flux in 1993 (Saigusa et al., 2002). Not only eddy covariance-based net ecosystem production (NEP) but also biometric-based carbon (C) flux measurement has been conducted intensively at this site; therefore, where and how the forest stores C is well known (Ohtsuka et al., 2007). Moreover, Ohtsuka et al. (2009) revealed that net primary production (NPP) of annual woody tissue varied markedly at the Takayama forest site and was positively correlated with eddy covariance-based NEP. They suggested that the interannual variability in the ecosystem C exchange was directly responsible for much of the interannual variation of C accumulation in the woody components. Thus, the direct effects of N deposition on woody growth greatly affect C sequestration. Moreover, Chen et al. (2017) measured the concentrations and fluxes of dissolved organic C (DOC) in the Takayama forest site and quantified the contribution of DOC from various forest water fluxes such as TF and SF. The present study aims to further comprehend the links between C and N cycling in forest ecosystems.

To understand N cycling in the Takayama forest over three years, N fluxes (BD, TF, and SF) were used; these included snowfall, which is a non-negligible component of precipitation in cool-temperate forest ecosystems. The effects of the understory canopy on N fluxes derived from N deposition were also quantified at the study site. We hypothesized that (i) N deposition to the Takayama forest site would be characterized by a low DIN contribution but high DON contribution (including from snowfall deposition) and (ii) the dense canopy layers of trees and understory dwarf bamboo would have substantial effects on N deposition. We attempted to address the following questions to test these hypotheses: (1) What is the contribution of dissolved N in rainfall and snowfall to N deposition? (2) What are the dynamics and characteristics of the concentrations and fluxes of DON and DIN in SF, TF_a, and TF_b? (3) What are the responses of the tree canopy and bamboo canopy to N deposition?

2.2. Materials and Methods

2.2.1. Study site

The study was conducted in a cool-temperate deciduous broad-leaved forest (Takayama forest), located on mid slope of Mt. Norikura in the Takayama Forest Research Station belonged to the River Basin Research Center, Gifu University, central Japan (36°08'N, 137°25'E, 1420 m a.s.l.). A permanent plot of 1 ha (100 m × 100 m) was set on a west-facing slope for field measurements since 1998 (Fig. 2.1).

Takayama forest site is an approximately 50-year-old forest study site dominated by *Quercus crispula* (26.9% in total basal area), *Betula ermanii* (24.6%) and *B. platyphylla* var. *japonica* (14.6%), only 2.8% of evergreen species are present (Ohtsuka et al., 2005). The forest floor is covered (ca. 40 stems m^{-2}) by dense dwarf bamboo grass *Sasa senanensis* at 1–1.5 m height (Nishimura et al., 2004). The soil at the study site was classified as an andisol along with Japanese volcanic ash soils (Chen et al., 2017). The climate is seasonal cool-temperate, with a mean annual air temperature of 7.2°C and a mean annual precipitation of 2,215 mm (average 40% contribution from snowfall) during the period from 2010 to 2018. Fig. 2.2 shows the precipitation and air temperature at the study site during the sampling time. The snow depth is usually 1–2 m in the snow season (December–April).

2.2.2. Sample collection

In consideration of the presence of snow cover during the snow season, we set up samplers to collect BD, TF, and SF during the growing season and collected snow samples during the snow season. During the growing season of years 2015–2017, BD was collected in bottles (20L) equipped with funnels (collection area: 0.0346 m^2). The bottles (3 replicates from July 2016) were set up in areas near the permanent plot but without a tree canopy. The TF collectors were the same as those for BD, except for the volume of the bottles (12 L) and the fact that they were evenly distributed within the permanent plot (9 replicates). Owing to the dense cover of dwarf bamboo on the forest floor, a pair of TF collectors was set up to collect TF_a and TF_b. An SF collector consisted of a polyethylene film surrounding a tree trunk (like a collar), a tube connecting the film to a rain gage (HOBO RG3), and a reservoir tank (24 L). SF collectors were set up on three major tree species (3 replicates each). After measuring total volume using a measuring cylinder or rain gage, subsamples from each type of collector were placed into polyethylene bottles (100 mL) for chemical analysis in the same manner once per month. After each sampling time, 10 mL of 0.1 mg L^{-1} $Cu(Br)_2$ solution was added to the collectors to prevent microbial alteration during collector storage and transit.

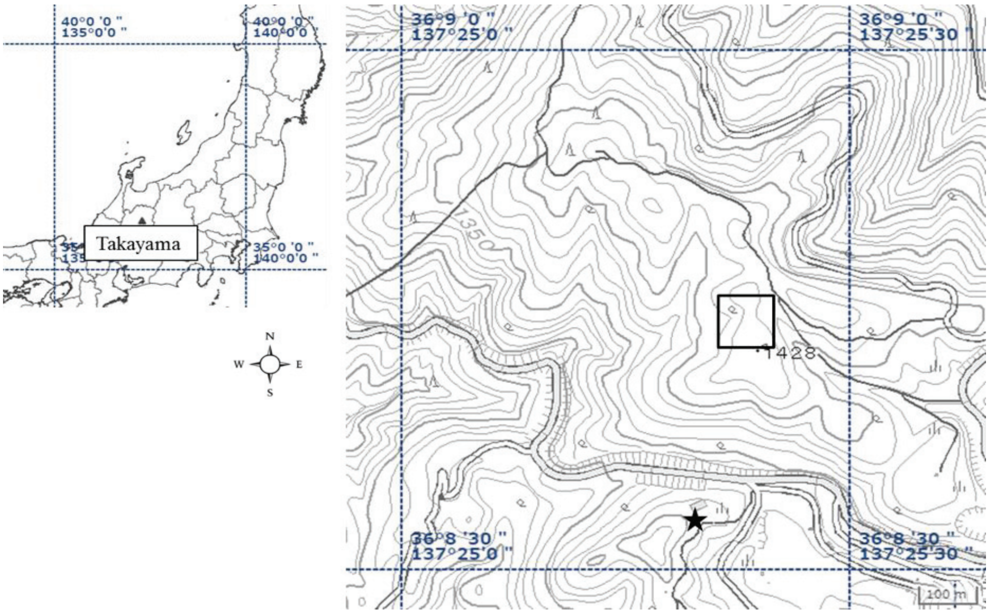

Fig. 2.1. Location of the study site at Takayama Forest Research Station (★). Square indicates the permanent plot (100 m × 100 m). Map is from the Geospatial Information Authority of Japan.

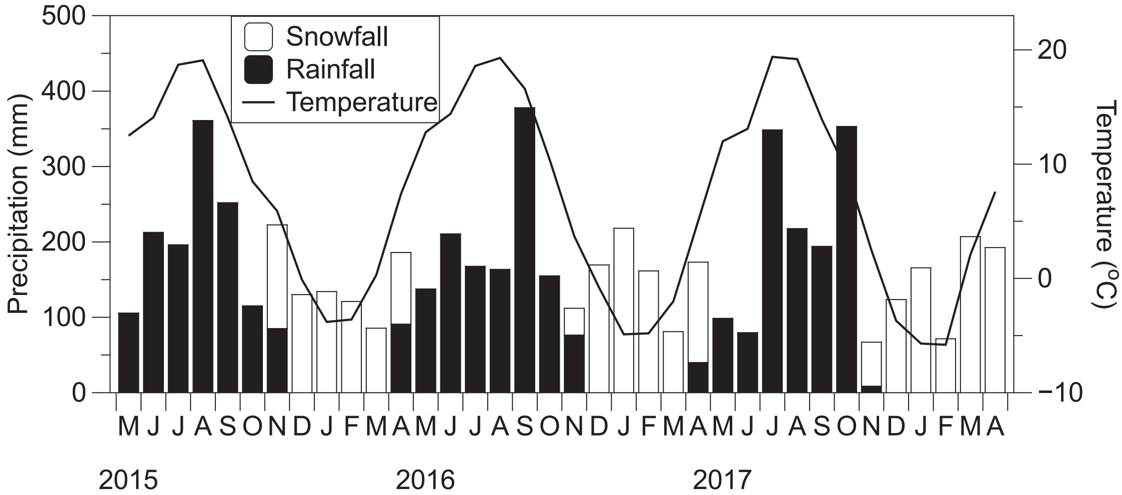

Fig. 2.2. Precipitation (mm) and air temperature (°C) at the study site from May 2015 to April 2018. Data were obtained from the meteorological station at the Takayama Field Station.

During the snow season, snowpack samples were collected in January 2016, January 2017, and March 2018, each on three sampling occasions. Because there was no canopy cover (understory dwarf bamboo was overwhelmed by thick snowpack during the snow season), we assumed that TF was the same as BP during this period. Moreover, no SF was considered to have occurred. Three random locations were selected as sampling points within the permanent plot. Snow samples were collected using a 100 mL soil corer from the snow surface to the soil surface in 5-cm depth layers (35–30 cm, 25–20 cm, 15–10 cm, and 5–0 cm for sampling in 2016; 105–100 cm, 85–80 cm, 65–60 cm, 45–40 cm, 25–20 cm, and 15–10 cm for sampling in 2017; and 105–100 cm, 80–75 cm, 60–55 cm, 40–35 cm, 20–15 cm, and 10–5 cm for sampling in 2018). Snow sampling was conducted when the snowpack had accumulated to a maximum. The samples were placed into sealed plastic bags. Later, the melted-snow samples were transferred into 100 mL bottles for storage prior to chemical analysis.

2.2.3. Chemical analysis

Water samples were immediately filtered through a 0.45 μm nitrocellulose membrane after transporting to the laboratory within 24 h, and then were frozen in the dark until chemical analysis within a month. Concentrations of NH_4–N, NO_3 + NO_2–N, and total dissolved N (TDN) were measured colorimetrically by a nutrient auto-analyzer (QuAAtro 2-HR, BL TEC Co., Ltd., Tokyo, Japan) using a continuous flow analysis. TDN was ultimately oxidized to NO_3^- by an alkaline persulfate via combustion at high temperature in the analyzer for colorimetry. NH_4–N was measured with the indophenol blue method at a wavelength of 630 nm. NO_2–N was measured after reaction with the sulfanilamide and N-(1-naphthyl)-ethylenediamine to form a red azo dye at a wavelength of 550 nm. NO_3–N was measured after cadmium reduction to NO_2–N. Concentration of DON was then calculated as difference between TDN and DIN. The detection limit was 0.01 mg L^{-1} for NH_4–N, $NO_3 + NO_2$–N concentration, defined as the sum of mean value and three times of standard deviation of blank samples.

2.2.4. Flux calculation

N fluxes (kg N ha^{-1} $episode^{-1}$) were calculated based on average concentration (mg N L^{-1}) and water depth (mm) in each sampling time (Chen et al., 2017).

N fluxes in SF and BD were calculated as follows:

$$F = hC/100, \tag{1}$$

where F (kg N ha^{-1} $episode^{-1}$) is N flux in each sampling time, h (mm) is water depth in each sampling time and C (mg N L^{-1}) is average concentration of N in each sampling time. Data of water depth in BD (rainfall and snowfall) were obtained from the meteorological station at the Takayama Field Station.

And water depth in SF was calculated based on basal area of trees in the permanent plot (DVWK, 1992).

$$h_{SF} = (V_{SF}/b)\ (B/S), \tag{2}$$

where h_{SF} (mm) is water depth in SF, V_{SF} (L) is volume collected by SF, b (m^2) is basal area of the sample tree, B (m^2) is total basal area of all trees in the permanent plot and S (m^2) is the permanent plot area.

N flux in TF was calculated as follows:

$$F = V_{TF}C/100S_{TF}, \tag{3}$$

where V_{TF} (L) is volume collected by TF in each sampling time and S_{TF} (m^2) is collection area of funnel.

Annual N flux (kg N ha^{-1} $year^{-1}$) was calculated as follows:

$$F_a = \sum F, \tag{4}$$

where F_a (kg N ha^{-1} $year^{-1}$) is annual N flux. During the snow season, N flux in TF was assumed to be equal to N flux in BD and N flux in SF was assumed equal to zero.

Assessment of the relative importance of DD and WD in BD was made according to the assumptions of (Kopaček et al., 1997): (i) DD from local sources dominates total deposition if a strong negative correlation exists between rainfall and average N concentration while N deposition is independent of rainfall and (ii) WD from long-range transport is prominent if average N concentration is independent of rainfall while rainfall and N deposition show a positive correlation.

We used net TF to identify the N fluxes that influenced by canopy. Net TF was calculated using flux data:

$$\text{Net } TF_a = TF_a + SF - BD, \tag{5}$$

$$\text{Net } TF_b = TF_b - TF_a, \tag{6}$$

in this study, the SF of the understory dwarf bamboo was neglected because of (i) possible low contribution from the SF of bamboo forests to N fluxes in internal forest cycles (Tu et al., 2013) and (ii) measurement difficulties owing to the crowded (ca. 40 stems m^{-2}) and thin culms (diameter at breast height < 1 cm) of the understory dwarf bamboo.

Correlations between rainfall in each rain event or water fluxes in TF_b and N fluxes in each net TF were estimated to understand the canopy processes. This approach was first proposed by Lovett and Lindberg (1984) and then improved by Aguillaume et al. (2017). It has been stated that a positive correlation between each rain event or water fluxes in TF_b and N fluxes in each net TF indicates leaching from the canopy, whereas a negative correlation indicates consumption by the canopy and no significant correlation may be a result of DD.

2.2.5. Statistical analysis

Significant differences among average concentration of dissolved N in each snow depth were assessed by one-way analysis of variance (ANOVA) with post hoc Tukey's Honestly Significant Difference (HSD) tests. Significant difference of dissolved N concentrations between TF_a and TF_b was analyzed by paired two-sample t-tests. Spearman′s rank correlation coefficient for BD and net TF was applied to test significant correlations between water flux and average dissolved N concentration or fluxes at each sampling time in the growing seasons. All statistical analyses were performed by R version 3.4.4 (R Core Team, 2018). Statistically significant differences were set at $p < 0.05$.

2.3. Results

2.3.1. N deposition characteristics in bulk deposition

The average monthly concentration of DON in BD followed a clear seasonal pattern in each year, with highest concentrations in spring (May to June) followed by a tendency to decrease over time (Fig. 2.3a). The dynamics of DIN concentrations also revealed decreasing trends from spring to winter in 2015 and 2017; although there was no clear seasonal trend in 2016 (Fig. 2.3a). For the average dissolved N flux pattern, DON fluxes were dominant in total N deposition (Fig. 2.3b). A higher dissolved N flux was deposited during summer, except for in 2017 when the flux during autumn was also high (Fig. 2.3b). The depth of the snowpack was 35 cm, 105 cm, and 105 cm during the three years, respectively (Fig. 2.4). Significant differences were observed between the concentration of NO_3 + NO_2–N at the top of the snow layer and that at the remaining snow depths and between the concentration of NH_4–N at the top of the snow layer and at a depth of 60–55 cm in March 2018 ($p < 0.05$) (Fig. 2.4c). During the three years, snowfall flux fluctuated (2.1 ± 0.3 kg N ha^{-1} year^{-1}, 5.2 ± 0.9 kg N ha^{-1} year^{-1}, and 5.5 ± 0.6 kg N ha^{-1} year^{-1}, respectively) owing to variations in the snowfall amount across the three years (659 mm, 799 mm, and 871 mm, respectively) (Fig. 2.3).

As BD water fluxes, Takayama forest received 1,344 ± 25.6 mm of rainfall and 776 ± 107 mm of snowfall during the three years (Table 2.1). The average concentrations of DON in rainfall and snowfall were 0.41 ± 0.03 mg N L^{-1} and 0.32 ± 0.16 mg N L^{-1}, respectively, whereas the average concentrations of DIN were 0.13 ± 0.04 mg N L^{-1} and 0.23 ± 0.09 mg N L^{-1}, respectively (Table 2.1). The average fluxes of DON in rainfall and snowfall amounted to 5.3 ± 0.4 kg N ha^{-1} period^{-1} and 2.6 ± 1.6 kg N ha^{-1} period^{-1} in each year, respectively, whereas the average fluxes of DIN were 1.5 ± 0.1 kg N ha^{-1} period^{-1} and 1.7 ± 0.9 kg N ha^{-1} period^{-1}, respectively (Table 2.1). Overall, 11.1 ± 1.7 kg N ha^{-1} year^{-1} of dissolved N was deposited onto the study site by BD, with a 78% DON flux contribution and 22% DIN flux contribution (Table 2.1). In terms of snowfall, average snowfall flux contributed 37% of N deposition (23%, 43%, and 46%, respectively, for the three sampling years) (Fig. 2.3 and Table 2.1).

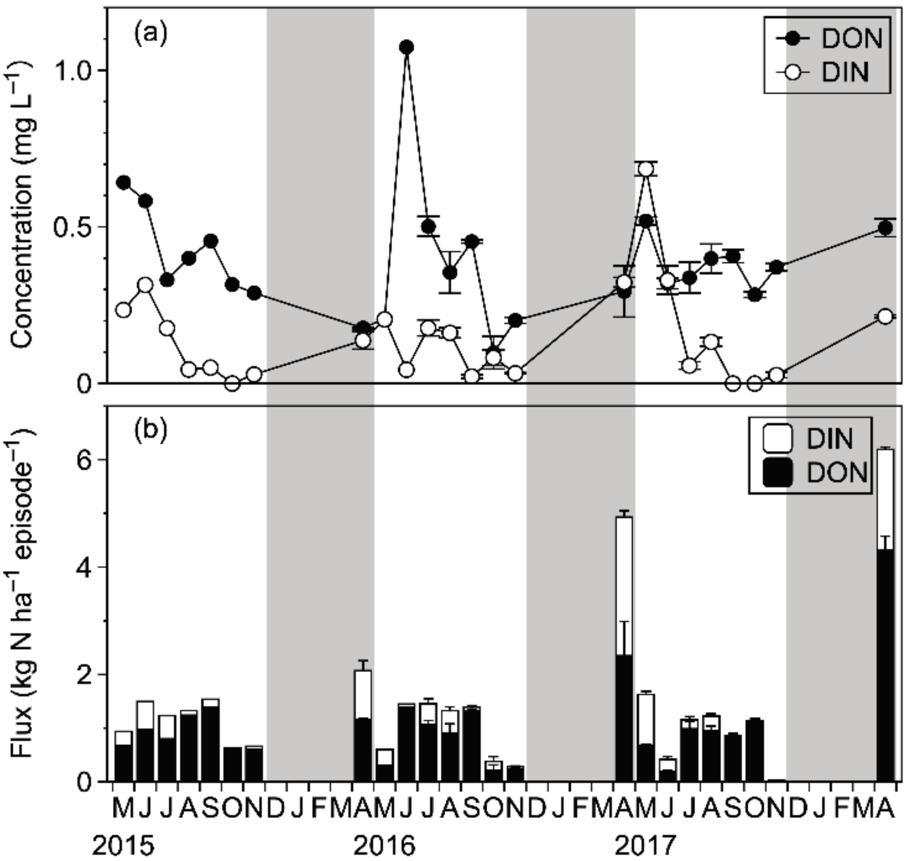

Fig. 2.3. Average monthly concentration (mg N L^{-1}) (a) and average flux (kg N ha^{-1} $episode^{-1}$) (b) of dissolved nitrogen in bulk precipitation from May 2015 to April 2018. Shaded areas show snow seasons. Sampling in the snow season was once per season. Error bars show standard error (n = 3). DON: Dissolved organic nitrogen, DIN: Dissolved inorganic nitrogen.

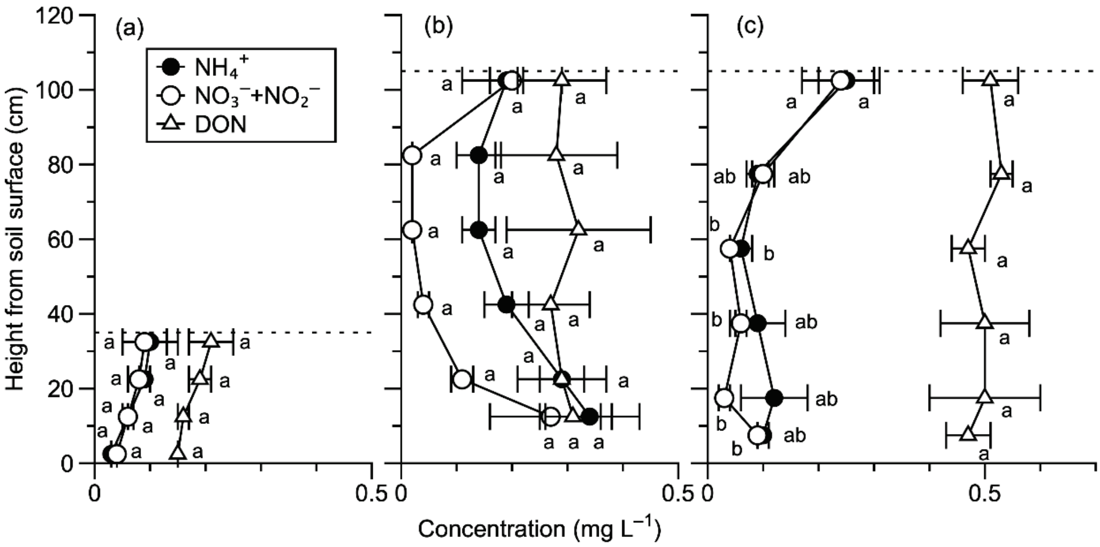

Fig. 2.4. Average concentration (mg N L^{-1}) of dissolved nitrogen at each snowpack depth in January 2016 (a), January 2017 (b), and March 2018 (c). Error bars show standard error (n = 3). DON: Dissolved organic nitrogen. Dashed lines show snow depth. Different letters indicate significant differences of dissolved nitrogen concentration among snow depths at $p < 0.05$.

Table 2.1. Precipitation (mm), average nitrogen concentration (mg L^{-1}), and average flux (kg N ha^{-1} $period^{-1}$) in bulk deposition during three years (May 2015–April 2018). Values in parentheses are standard deviation (n = 3). DON：Dissolved organic nitrogen, DIN: Dissolved inorganic nitrogen, TDN: Total dissolved nitrogen.

	Rainfall	**Snowfall**	**Total**	**Contribution of snowfall**
Precipitation (mm)	1,344 (25.6)	776 (107)	2,120 (117)	0.37 (0.03)
Concentration (mg L^{-1})				
DON	0.41 (0.03)	0.32 (0.16)	–	–
DIN	0.13 (0.04)	0.23 (0.09)	–	–
TDN	0.55 (0.04)	0.53 (0.19)	–	–
Flux (kg N ha^{-1} $period^{-1}$)				
DON	5.29 (0.39)	2.61 (1.60)	7.90 (1.22)	0.32 (0.15)
DIN	1.52 (0.09)	1.67 (0.85)	3.19 (0.80)	0.50 (0.13)
TDN	6.81 (0.32)	4.28 (1.97)	11.1 (1.71)	0.37 (0.13)
Contribution of DON	0.78 (0.02)	0.59 (0.13)	0.78 (0.02)	–

For DIN, there were no significant relationships between average monthly concentrations or average fluxes and rainfall (Table 2.2). For DON, average monthly concentrations remained almost constant with increasing rainfall, whereas average fluxes dramatically increased with increasing rainfall. This indicates that wet deposition dominated DON fluxes in BD (Table 2.2).

2.3.2. N deposition characteristics in throughfall and stemflow

In SF, there were no clear seasonal trends in DON concentrations, whereas DIN concentrations tended to peak around summer in each of the three years (DON concentration range: 0.35–1.36 mg N L^{-1}; DIN concentration range: 0.02–0.40 mg N L^{-1}) (Fig. 2.5a). The dissolved N flux in SF was extremely low in each sampling period (DON flux range: 0.01–0.04 kg N ha^{-1} $year^{-1}$; DIN flux range: 0.00–0.02 kg N ha^{-1} $year^{-1}$) (Fig. 2.5b).

In TF, variations in the concentration of DON in TF_a and TF_b showed similar tendencies, with two peaks being observed around June and August (Fig. 2.6). There was no clear seasonal variation in DIN concentration (Fig. 2.6). Significant differences between DIN concentration in TF_a and TF_b were found for some sampling times in 2016 and 2017 (Fig. 2.6). DON and DIN fluxes in TF showed highest contributions during summer, except in 2017 when the DON flux during autumn was also high (Fig. 2.7).

Table 2.2. Spearman correlations between rainfall (mm) and average monthly concentrations of dissolved nitrogen (mg N L^{-1}) or average fluxes of dissolved nitrogen (kg N ha^{-1} $episode^{-1}$) in bulk deposition (n = 21). DIN: Dissolved inorganic nitrogen, DON: Dissolved organic nitrogen.

	NH_4–N	**$NO_3 + NO_2$–N**	**DIN**	**DON**
Concentrations	–0.15	–0.32	–0.24	–0.04
Fluxes	0.08	–0.06	0.02	0.67*

* indicates significant at $p < 0.05$.

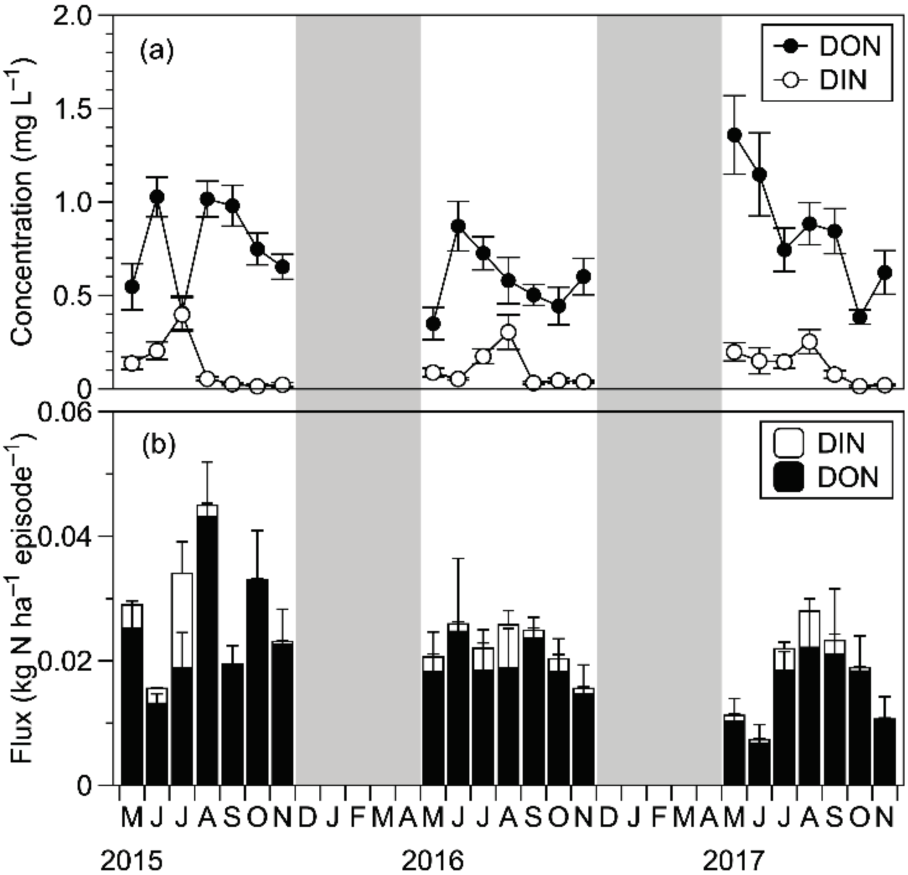

Fig. 2.5. Average monthly concentration (mg N L^{-1}) (a) and average flux (kg N ha^{-1} episode^{-1}) (b) of dissolved nitrogen in stemflow from May 2015 to November 2017. Shaded areas show the snow seasons. Error bars show the standard error (n = 9). DON: Dissolved organic nitrogen, DIN: Dissolved inorganic nitrogen.

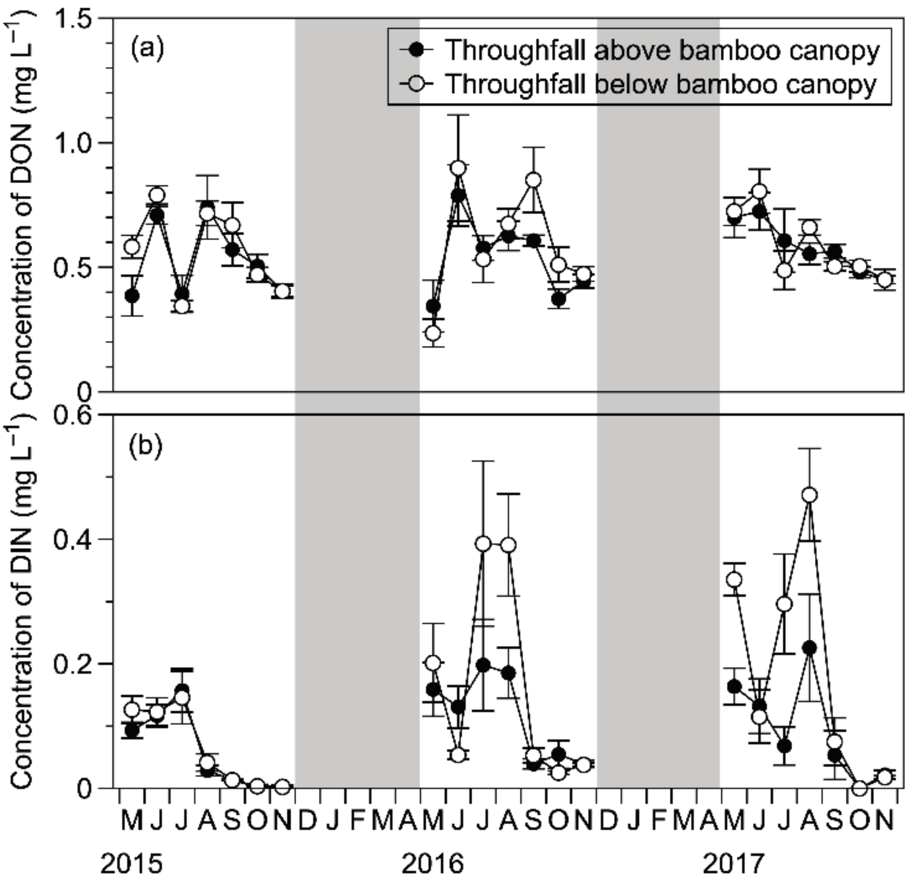

Fig. 2.6. Average monthly concentration (mg N L^{-1}) of dissolved organic nitrogen (DON) (a) and dissolved inorganic nitrogen (DIN) (b) in throughfall above and below the bamboo canopy from May 2015 to November 2017. Shaded areas show the snow seasons. Error bars show the standard error (n = 9).

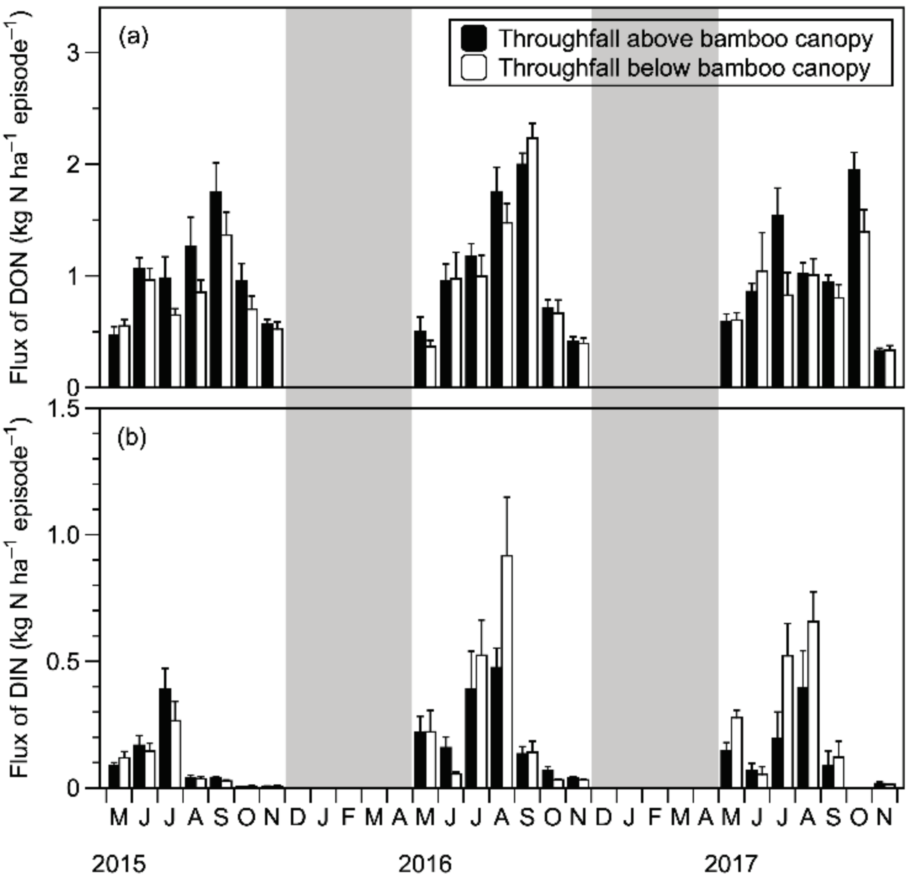

Fig. 2.7. Average flux (kg N ha^{-1} episode^{-1}) of dissolved organic nitrogen (DON) (a) and dissolved inorganic nitrogen (DIN) (b) in throughfall above and below the bamboo canopy from May 2015 to November 2017. Shaded areas show the snow seasons. Error bars show the standard error (n = 9).

The amount of water flux decreased from BD, TF_a, SF to TF_b. TF_a and SF intercepted 50 mm of precipitation, with TF_b subsequently intercepting 283 mm of precipitation (Table 2.3). For DIN, the average annual flux of $NO_3 + NO_2$–N in net TF_a was negative, indicating that the tree canopy consumed 0.7 ± 0.2 kg N ha^{-1} $year^{-1}$ as $NO_3 + NO_2$–N. However, a positive DIN value (0.4 ± 0.4 kg N ha^{-1} $year^{-1}$) was found for net TF_b (Table 2.3). For DON, leaching showed a positive value (2.1 ± 0.4 kg N ha^{-1} $year^{-1}$) for net TF_a, whereas a negative value (–1.0 ± 0.6 kg N ha^{-1} $year^{-1}$) was found for net TF_b (Table 2.3). The results of Spearman correlations between rainfall and DON fluxes in net TF_a at each sampling time showed a positive relationship between rainfall and DON fluxes in net TF_a but a negative relationship between water fluxes in net TF_a and DON fluxes in net TF_b ($p < 0.05$) (Table 2.4). For DIN flux, there was no significant correlation between rainfall and DIN fluxes in net TF_a, or between water fluxes in TF_a and DIN fluxes in net TF_b (Table 2.4).

Table 2.3. Water flux (mm), dissolved nitrogen fluxes (kg N ha^{-1} year^{-1}) in bulk deposition (BD), stemflow (SF), throughfall above bamboo canopy (TF_a), throughfall below bamboo canopy (TF_b), net TF_a, and net TF_b during the three study years. Dissolved nitrogen fluxes in TF_a and TF_b during the snow seasons were assumed the same as that in BD during the snow seasons; SF was assumed equal to zero during the snow seasons. Values in parentheses are standard deviation (n = 3). DIN: Dissolved inorganic nitrogen, DON: Dissolved organic nitrogen.

	Water flux (mm)	**NH_4–N (kg N ha^{-1} year^{-1})**	**NO_3 + NO_2–N (kg N ha^{-1} year^{-1})**	**DIN (kg N ha^{-1} year^{-1})**	**DON (kg N ha^{-1} year^{-1})**
BD	2120 (117)	1.66 (0.68)	1.53 (0.12)	3.19 (0.80)	7.90 (1.22)
SF	21.4 (3.53)	0.02 (0.01)	0.00 (0.00)	0.02 (0.01)	0.14 (0.03)
TF_a	2048 (114)	2.05 (0.91)	0.80 (0.30)	2.82 (1.21)	9.87 (1.67)
TF_b	1764 (104)	2.22 (1.12)	0.98 (0.39)	3.17 (1.52)	8.85 (1.86)
Net TF_a	–50.0 (11.7)	0.41 (0.24)	–0.73 (0.19)	–0.35 (0.41)	2.11 (0.42)
Net TF_b	–283 (57.0)	0.17 (0.28)	0.18 (0.18)	0.35 (0.44)	–1.02 (0.55)

Table 2.4. Spearman correlations between rainfall amount (mm) and fluxes of dissolved nitrogen in net throughfall above the bamboo canopy (Net TF_a) (kg N ha^{-1} $episode^{-1}$), or between water fluxes in net TF_a (mm) and fluxes of dissolved nitrogen in net throughfall below the bamboo canopy (Net TF_b) (kg N ha^{-1} $episode^{-1}$) (n = 21). DIN = Dissolved inorganic nitrogen, DON = Dissolved organic nitrogen.

	NH_4–N	**NO_3 + NO_2–N**	**DIN**	**DON**
Net TF_a	0.11	–0.01	0.36	0.46*
Net TF_b	0.04	0.08	0.20	–0.63*

* indicates significant relationships at $p < 0.05$.

2.4. Discussion

2.4.1. Bulk N deposition into the Takayama forest site

The average monthly DIN concentration (0.13 ± 0.04 mg N L^{-1}) in rainfall was lower than that reported for China (0.15–5.10 mg N L^{-1} at 38 forest stands) (Du et al., 2014), Europe (0.18–1.33 mg N L^{-1}) (Cape et al., 2012), and Japan (0.49–1.92 mg N L^{-1}) (Mitchell et al., 1997). The present study site was in a rural region with active agricultural activities, far from urban areas. The lower DIN concentration coincided with urban hotspots of DIN input (Cape et al., 2012), indicating that DIN decreased with increasing distance to the nearest large city with heavy traffic or fossil fuel combustion (Jia et al., 2014). Although the major global sources of NH_3 were emissions from livestock and fertilization use (Asman et al., 1998), residence time of NH_3 in the atmosphere was relatively short, with NH_3 tending to be deposited nearly to the emission sources (Ferm, 1998; Krupa, 2003).

In contrast, the average monthly concentration of DON (0.41 ± 0.03 mg N L^{-1}) in rainfall was within the range of that reported for China (0.05–0.86 mg N L^{-1} at 3 sites) (Song et al., 2017) but higher than that reported for Europe (0.02–0.18 mg N L^{-1} at 18 sites) (Cape et al., 2012). Some studies reported that agricultural activities were possible sources of DON because of higher DON concentration found at the agricultural site compared with other types of sites (Izquieta-Rojano et al., 2016; Song et al., 2017). At this study site, temporal variations in DON concentration showed highest concentrations in spring at the study site. This is in accordance with agricultural activities, such as the use of compost, which is mainly applied in spring, implying that fertilizer application was a possible source of DON to the study site.

For snowfall, different annual variation patterns were observed with respect to the DIN concentrations at snowpack depths during three years. DIN movements, such as sublimation from the top of the snow layer, downward movement at the subsurface layers, and N mineralization along the ground, would occur upon the accumulation of the snowpack. There is little previous research on the annual variation of the dissolved N concentration at various snow depths; however, the seasonal variability of DIN concentration at snowpack depths in the alpine regions would be explained by the air temperature contrasts (Hiltbrunner et al., 2005), which would cause atmospheric mixing and air mass movements as well as the percolation of DIN into the subsurface layers (Bowman, 1992). These factors would cause annual variations of the DIN concentrations, which will require further research. The significantly high concentration of $NO_3 + NO_2$−N observed at the top of the snow layer in March 2018 implied that the NO_x from NO_3^- photolysis regenerated NO_3^- in the temporary tops of the snow layer and was ultimately deposited at the top of the snow layer when the snowpack reached its maximum level (Fibiger et al., 2016). In total, N deposition from snowfall was expected to become almost half of the TDN flux in the Takayama forest site, despite the extremely low contribution of snowfall to N fluxes (23%) in the snow season between 2015 and 2016.

The average annual DIN flux (3.2 ± 0.8 kg N ha^{-1} year^{-1}) was extremely low when compared with that at other study sites across the world (Ban et al., 2016; Ham et al., 2007; Jia et al., 2014; Li et al., 2012; Zhang et al., 2008) and slightly low when compared with that at 24 Japanese forested sites (3.5–10.5 kg N ha^{-1} year^{-1}) (Mitchell et al., 1997). The average annual DON flux (7.90 ± 1.22 kg N ha^{-1} year^{-1}) was high compared with that in Europe (0.15–1.74 kg N ha^{-1} year^{-1} at 18 sites) (Cape et al., 2012) and in China (6.84 kg N ha^{-1} year^{-1} on average at 32 sites) (Zhang et al., 2012). For the study site, it can be deduced that the low DIN flux was due to low DIN concentrations (despite a high precipitation contribution), whereas the high DON flux was due to both high DON concentrations and a high precipitation contribution.

In all, 11.1 ± 1.7 kg N ha^{-1} $year^{-1}$ TDN was input by BD to the Takayama forest; this level is lower than that found in other studies (Balestrini and Tagliaferri, 2001; Ham et al., 2007; Li et al., 2012; Zhang et al., 2008) even though it is slightly higher than that found at European sites (1.4–10 kg N ha^{-1} $year^{-1}$) (Cape et al., 2012) because of the low precipitation contribution (an annual average of 802 mm) at the European sites. The contribution of DON to TDN (up to 78%) is higher than that found in most regions (Balestrini and Tagliaferri, 2001; Cape et al., 2012; Ham et al., 2007; Li et al., 2012; Zhang et al., 2012) but agrees well with studies reported by Zhang et al. (2008) for forests in the Sichuan province (79%) and Tibet (72%) in China, both of which are located in remote areas. Clearly, DON was dominant at the present study site. Our data verified previous research that denoted that DON was the main component of the dissolved N input into the remote forest regions (Pacheco et al., 2004; Zhang et al., 2008), achieving a complete picture and comprehension of N deposition in the forest ecosystems.

2.4.2. Responses of forest structure to N deposition in internal N cycle

The Spearman correlation between rainfall amount and DIN fluxes in net TF_a indicated that dry deposition played a vital role in DIN fluxes for tree canopy interactions. Consumption of DIN by the tree canopy has been reported in many studies (Aguillaume et al., 2017; Balestrini and Tagliaferri, 2001; Fenn et al., 2013; Izquieta-Rojano et al., 2016). Direct foliage or bark uptake, epiphyte uptake, and microbial action can cause the consumption of DIN fluxes. Enrichment of DIN by the tree canopy also was reported by some studies maybe due to dry deposition (Aguillaume et al., 2017; Izquieta-Rojano et al., 2016). Buffering dry deposition and biological nitrification in tree canopies processing with DIN deposition input were demonstrated by isotopic tracers (Guerrieri et al., 2015). Net annual negative value of $NO_3 + NO_2$–N in net TF_a implied that annual consumption of $NO_3 + NO_2$–N by trees or microbes was greater than any dry deposition or canopy nitrification that might occur. Net consumption of $NO_3 + NO_2$–N in the tree canopy amounted to 0.7 ± 0.2 kg N ha^{-1} $year^{-1}$, a low level compared with that reported by Lovett and Lindberg (Lovett and Lindberg, 1993). In consideration of the dry deposition and canopy nitrification effects on net TF_a, total DIN consumption by the tree canopy should have been more than 0.7 ± 0.2 kg N ha^{-1} $year^{-1}$.

For the DON flux, 2.1 ± 0.4 kg N ha^{-1} $year^{-1}$ of DON was leached by the tree canopy. The leaching of DON from a canopy is a common phenomenon (Lovett and Lindberg, 1993), which was originated from pollen, insect excretions, and microbial activities (Gaige et al., 2007; Le Mellec et al., 2010).

The Spearman correlation between water fluxes in TF_a and DIN fluxes in net TF_b indicated that dry deposition also played a vital role in DIN fluxes in dwarf bamboo canopy interactions. This dry deposition may have derived from the washing out of dissolved N gathered on the surfaces of bamboo leaves. Completely opposite responses of the overstory tree canopy and understory dwarf bamboo canopy to DIN deposition were found in the study site. Leaf wettability was determined to be a crucial factor for the ability of foliar DIN uptake at leaf level of evergreen and deciduous tree species (Adriaenssens et al., 2011; Wuyts et al., 2015). The evergreen dwarf bamboo canopy probably hardly absorbed the DIN, as compared with the deciduous tree canopy, because the fibrous foliage was slowly wet as water flowed through the dwarf bamboo canopy.

A significant negative relationship between water fluxes in TF_a and DON fluxes in net TF_b showed that consumption of DON by the dwarf bamboo canopy may have occurred. There are two possible reasons for this contradiction to the common leaching process of DON from canopies. Firstly, it was reported that plants can consume some organic N directly from solution without microbial mineralization (Hinko-Najera Umana and Wanek, 2010; Izquieta-Rojano et al., 2016; Vonk et al., 2008), which was called "short circuit" in the N cycle of ecosystems by Neff et al. (2007). Agricultural activities were a possible source of DON at the present study site and these may have produced bioavailable DON, such as amino acids and urea (Cornell, 2011), which were readily available to plants or microbes. Secondly, from the perspective of the complete hydrological cycle, TF_a should be partitioned into two pathways: TF_b and SF of bamboo. It seems that the contribution of dissolved N in SF of the understory bamboo should not be neglected because of the high stem density of the dwarf bamboo at the study site; this was not the same as the distribution of tree stems, which would have provided a negligible contribution of dissolved N in SF in the complete hydrological cycle (Rodrigo et al., 2003). Completely opposite responses of the overstory tree canopy and understory dwarf bamboo canopy to DON deposition were found in the study site. Studies on controlling factors for DON exchange at leaf level were rarely reported. However, the dwarf bamboo canopy seemed to be inclined to consume DON, given the abundant DON supply in the water flows at the study site, and assuming that DON uptake by the understory canopy may be a supplementary pathway when facing the fierce competition with overstory trees in the root system.

2.4.3. Ecological implications of N deposition in the deciduous broad-leaved forest

In total, 3.2 kg N ha^{-1} year^{-1} of DIN occurred from TF_b and SF. Total DIN deposition was extremely lower than the minimal value of critical loads of inorganic N deposition in forest ecosystems, which mostly ranged from 10 to 20 kg N ha^{-1} year^{-1} (Bobbink et al., 2010). It indicates that we can consider DIN deposition as a nutrient supply for forest growth in the N-limited forest site without negative effects. In contrast to DIN, studies concerning the effects of DON deposition (9.0 kg N ha^{-1} year^{-1} from TF_b and SF in the present study site) on forest ecosystems are scarce, which needs to study further.

2.5. Summary

Our results in this chapter showed that N deposition in the Takayama forest site amounted to 11.1 ± 1.7 kg N ha^{-1} year^{-1}, with a high contribution of DON (78%) and low contribution of DIN (22%). The snowfall contribution was 37% of the total N input to the study site. The large DON contribution indicates that DON estimation is essential for critical load determination of N deposition, especially in rural areas. The substantial snow contribution shows the importance of N deposition estimation during the snow season; this finding may have deep implications for estimation of the melted-snow flux at the soil surface in cool-temperate forests.

The canopy layers of the trees and dwarf bamboo played a vital role in N fluxes derived from BP in the internal N cycle of the Takayama forest site. The tree canopy leached DON but consumed DIN, which was in accordance with other studies. DIN was released from the surface of the dwarf bamboo canopy, but DON was consumed by the dwarf bamboo canopy, indicating that the dwarf bamboo canopy has an impact on dissolved N fluxes in the internal N cycle. Field observations and estimations of N fluxes passing through understory plants should be conducted to more accurately assess the N deposition input to soil layers.

Chapter 3

N DEPOSITION IN A SUBTROPICAL EVERGREEN BROAD-LEAVED FOREST

3.1. Introduction

Urban forest ecosystems could provide more diverse benefits for human society; for example, environmental benefits (improvement of urban air quality, mitigation of noise pollution, modification of temperature and microclimate, water regulation, etc.), economic benefits (tourism, development of local economy, etc.), and social benefits (improving human health, promoting education, etc.) (Dwyer et al., 1992; Nowak and Dwyer, 2007). However, little is known about the N deposition processes of urban forest ecosystems, which are potential hotspots for N deposition and suffer from more extensive human disturbance compared to natural areas. Several studies have shown that positive N flux in net TF is a common characteristic of polluted sites compared to relatively protected sites, due to the dominant washout process of dry deposition (Aikawa et al., 2006; Juknys et al., 2007; Rodrigo et al., 2003). Comparative studies on N deposition levels and dynamics have been successively conducted based on different site classifications, ranging from urban to peri-urban, as well as montane, lake, and agricultural sites (Aikawa et al., 2006; Chiwa et al., 2003; De Souza et al., 2015; Rodrigo et al., 2003). These studies indicated that N deposition tended to increase as the distance from the pollution sources decreased (urban hotspots of N deposition, Du et al., 2014). In terms of forest ecosystems, Jia et al. (2014) reported a negative correlation between N deposition and the distance between the forest sites and the nearest large city.

The present study in this chapter was designed to estimate the concentrations and fluxes of dissolved N in BD, TF, and SF over three years in Mt. Kinka forest site, located in Central Japan, in the center of Gifu City, which has a population of more than 400,000. Mature subtropical/warm-temperate evergreen broad-leaved forest (mainly *Castanopsis cuspidata*) predominates on the entire mountain (Chen et al., 2017). Due to the historically harvesting for the requirement of charcoal and firewood production, remnant evergreen broad-leaved forests in Japan are very rare, and except for artificial evergreen coniferous forests (Aikawa et al., 2006), few studies of N deposition have been conducted in natural evergreen forests in urban areas. We hypothesized that 1) N deposition in BD is higher compared to that in the forests in rural areas; and 2) a high level of dry N deposition contributes to total N deposition on the forest floor, with positive net TF values in the urban forest. We tested these hypotheses using the "throughfall method," with special emphasis on the characteristics of N species such as NH_4–N, NO_3 + NO_2–N, and

DON.

3.2. Materials and Methods

3.2.1. Study site

The study was conducted on Mt. Kinka (Fig. 3.1), which is located in Gifu City, Gifu Prefecture, central Japan (35°26′N, 136°47′E, 329 m a.s.l.). A study plot of 0.7 ha (70 m × 100 m) has been established on the lower slope of Mt. Kinka (ca. 60 m a.s.l.) since 1989 (Chen et al., 2017). Japanese red pine (*Pinus densiflora*) previously predominated in Mt. Kinka, which was utilized for charcoal and firewood production. Since 1889, the harvesting of the forest was prohibited, and natural evergreen broad-leaved trees gradually invaded after abandonment. The forest site is dominated by evergreen broad-leaved tree species *Castanopsis cuspidata* (Thunb.) Schottky (87.8% in total basal area) and only 5.5% of the forest consisted of deciduous trees as of 2017 (Chen et al., 2017). *C. cuspidata* is one of the typical dominant species in the subtropical/warm-temperate regions from the coastal area of central Japan to southwestern Japan. A heavy traffic highway (Japan National Route 256) with 34,954 vehicles day^{-1} (data collected from Japan's Ministry of Land, Infrastructure, Transport and Tourism) is 300 meters from the study site, which is located at the northern periphery behind the forest area in the center

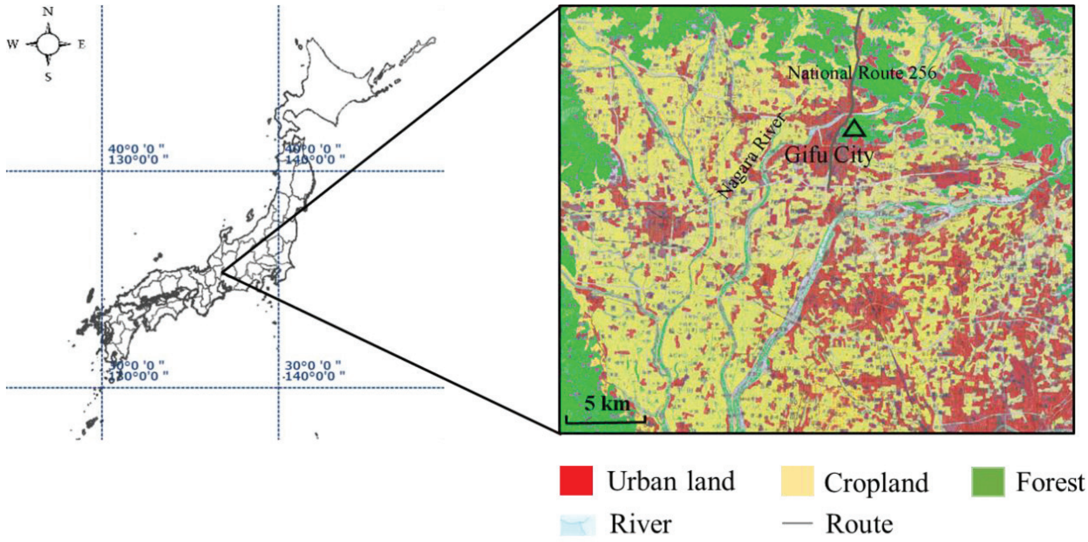

Fig. 3.1. Location of the study site at Mt. Kinka (△) and land use patterns around the site.

Map source was obtained from the Geospatial Information Authority of Japan.

of Gifu City (population of 400,118 in 2020), and there is an agricultural area used with N fertilizers in the direction of the prevailing northwest wind (Fig. 3.1).It has a subtropical temperate climate with a mean annual air temperature of 15.8℃ and a mean annual precipitation of 1,827.5 mm during the period from 1981 to 2010 (data collected from the Japan Meteorological Agency). Fig. 3.2 depicts the air temperature and precipitation at the study site during the study period. The meteorological data was obtained from the Japan Meteorological Agency, which was collected at a weather station situated approximately 4 km from the study site.

3.2.2. Sample collection

Water samples of BD, TF and SF were collected fortnightly from July 2016 to June 2019. Sampling frequency was performed on a monthly basis during dry periods (rainfall less than 10 mm). BD was collected in an open area near the study plot. Three collectors consisting of a polyethylene (PE) funnel (collection area: 0.0346 m^2) connected to a 20 L PE bottle (Cao et al., 2019). Glass wool was placed in the neck of the funnels as a plug, and a draining mesh bag was placed on the top of the funnels to prevent contamination from litterfall, insects or others. The TF collectors were the same as those for BD, except for the volume of the bottles (12 L). They were evenly distributed within the study plot (9 replicates). SF was collected by system consisting of a PE film surrounding a tree trunk

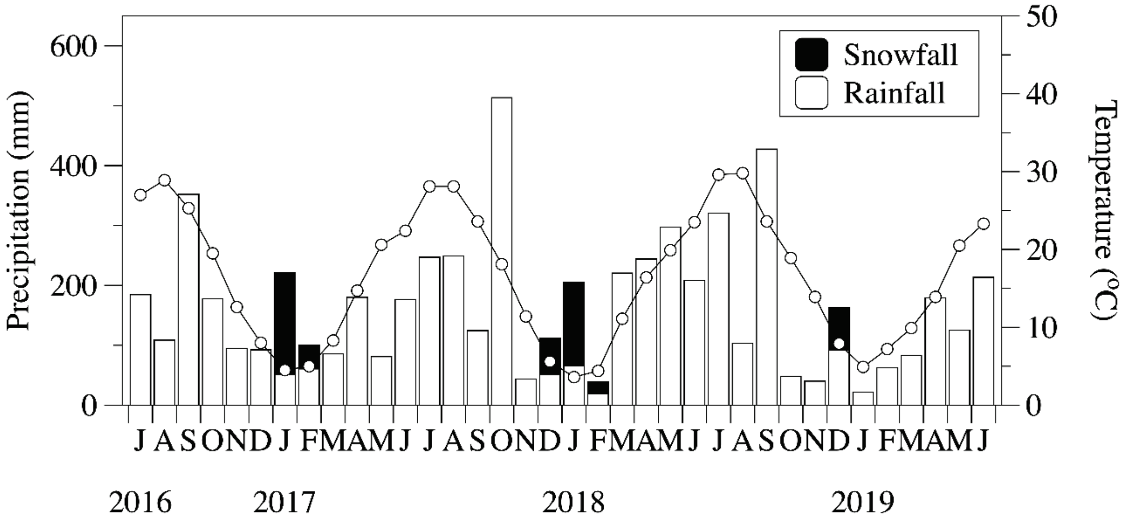

Fig. 3.2. Monthly sum precipitation (mm) and monthly average air temperature (℃) (○) at the study site from July 2016 to June 2019. The data was obtained from the Japan Meteorological Agency.

(like a collar), a tube connecting the "collar" to a rain gauge, and a reservoir tank (24 L) under the rain gauge as described in Chen et al. (2019). SF collectors (15 replicates in total) were set up on *C. cuspidata* (different diameter at breast height classes: 50 cm, 40 cm, 30 cm and 20 cm, three replicates each) and three deciduous trees (*Eleutherococcus sciadophylloides*, *Magnolia obovata*, and *Quercus serrata* in 40 cm diameter at breast height class). Water samples of BD, TF and SF were stored cold until further treatment in the laboratory. After each collection, the collectors were cleared from litterfall or other debris, and 10 mL of 0.1 mg L^{-1} $CuBr_2$ solution was added into the collectors to prevent microbial alteration during collector storage (Thimonier et al., 2019). The collection bottles were cleaned for reuse, by soaking overnight in HCl solution (3%) and then being rinsed with deionized water 3 times.

3.2.3. Chemical analysis

Water samples were immediately filtered through a 0.45 μm nitrocellulose membrane after transporting to the laboratory within 24 h, and then were frozen in the dark until chemical analysis within a month. Concentrations of NH_4–N, NO_3 + NO_2–N, and total dissolved N (TDN) were measured colorimetrically by a nutrient auto-analyzer (QuAAtro 2-HR, BL TEC Co., Ltd., Tokyo, Japan) using a continuous flow analysis. TDN was ultimately oxidized to NO_3^- by an alkaline persulfate via combustion at high temperature in the analyzer for colorimetry. NH_4–N was measured with the indophenol blue method at a wavelength of 630 nm. NO_2–N was measured after reaction with the sulfanilamide and N-(1-naphthyl)-ethylenediamine to form a red azo dye at a wavelength of 550 nm. NO_3–N was measured after cadmium reduction to NO_2–N. Concentration of DON was then calculated as difference between TDN and DIN. The detection limit was 0.01 mg L^{-1} for NH_4–N, NO_3 + NO_2–N concentration, defined as the sum of mean value and three times of standard deviation of blank samples.

3.2.4. Flux calculation

Water fluxes of BD and TF were calculated by dividing the volume (L) by the collection area (m^2) of the funnel, and water fluxes of SF were related to the basal area of the trees (DVWK, 1992), which is mentioned in 2.2.4.

The mean monthly dissolved N fluxes (kg N ha^{-1} $month^{-1}$) in different water fluxes were calculated by the sum of dissolved N fluxes corresponding to each sampling time in one month. Likewise, annual fluxes of dissolved N deposition (kg N ha^{-1} $year^{-1}$) in different water fluxes were calculated by the sum of the mean monthly dissolved N fluxes for one year.

3.2.5. Statistical analysis

Because the data did not fit a normal distribution according to the Shapiro-Wilk test, Spearman's rank correlation coefficient was applied to test for significant correlations between monthly precipitation (mm) and N fluxes (kg N ha^{-1} $month^{-1}$) in BD, TF, SF, and net TF. All statistical analyses were performed by R version 3.4.4 (R Core Team, 2018). Statistically significant differences were set at $p < 0.05$ or $p < 0.01$.

3.3. Results

3.3.1. N deposition characteristics in bulk deposition

Annual precipitation at the site was 1,916 ± 316 mm (Table 3.1). The canopy intercepted 585 ± 131 mm of the water flux. The annual VWM concentrations of the DON and DIN were 0.39 ± 0.05 mg N L^{-1} and 0.25 ± 0.09 mg N L^{-1}, respectively. The annual TDN flux in BD was 10.6 ± 3.5 kg N ha^{-1} $year^{-1}$ (NH_4–N:1.5 kg N ha^{-1} $year^{-1}$; NO_3 + NO_2–N kg N ha^{-1} $year^{-1}$: 2.2; DON: 6.5 kg N ha^{-1} $year^{-1}$), with a 66% average contribution from DON. The annual DON flux was approximately two times higher than the annual DIN flux in BD. The VWM concentration of DON in BD peaked separately in May, the end of September, and August during each year of sampling (Fig. 3.3a). On the other hand, the VWM concentration of DIN in BD peaked separately in July, February, and August during each year of sampling (Fig. 3.3a). The monthly N fluxes in BD (Fig. 3.4a) followed a clear seasonal pattern, with the highest N deposition (DON fluxes: 1.2, 2.0, and 1.2 kg N ha^{-1} $month^{-1}$, respectively; DIN fluxes: 1.4, 0.8, and 0.3 kg N ha^{-1} $month^{-1}$, respectively) occurring during the summer and early autumn (rainy season) in the three sampled years (Fig. 3.2). There was a significantly positive relationship between precipitation and the monthly bulk N deposition (DIN: $p < 0.05$; DON: $p < 0.01$) (Table 3.2).

3.3.2. N deposition characteristics in throughfall and stemflow

The annual VWM concentrations of all N species were higher in TF and SF than in BD (Table 3.1). The annual VWM concentration of the TDN in TF was 2.00 ± 0.16 mg N L^{-1}, which consisted of 1.31 ± 0.15 mg N L^{-1} of DIN and 0.72 ± 0.03 mg N L^{-1} of DON. The annual VWM concentration of DIN (both NH_4–N and NO_3 + NO_2–N) in TF was more than five times higher than that of the BD. The annual VWM concentration of DON in TF was approximately two times higher than that in the BD. The annual VWM concentration of the TDN in SF was 1.47 ± 0.34 mg N L^{-1}, which consisted of 0.63 ± 0.18 mg N L^{-1} of DIN and 0.77 ± 0.09 mg N L^{-1} of DON. The annual VWM concentration of DIN (both NH_4–N and NO_3 + NO_2–N) and DON in SF was also approximately two times higher than that in the BD. The annual VWM concentration of DIN in TF was approximately two times higher than that in the SF, whereas the annual VWM concentrations of DON were similar between TF and SF. The VWM concentration of

DON in TF peaked separately in June, the end of September, and May during each year of sampling (Fig. 3.3b). The highest VWM concentration of DIN appeared at the end of August over the three years of sampling. For SF, there were also different peaks for the VWM concentration of DON, namely June, the end of September, and October, during the three sampled years (Fig. 3.3c). The highest VWM concentration of DIN occurred in August.

The annual N fluxes varied similarly, with a higher N deposition in TF than in BD, except for SF with minor fluxes compared with TF or BD (Table 3.1). In total, over the three-year study period, there was 20.4 kg N ha^{-1} $year^{-1}$ of TDN from the input of TF and SF to the forest floor at the study site, with 12.5 kg N ha^{-1} $year^{-1}$ of DIN (61% contribution) and 8.1 kg N ha^{-1} $year^{-1}$ of DON (39% contribution). The monthly N fluxes in TF (Fig. 3.4b) and SF (Fig. 3.4c) also showed a clear seasonal pattern with peaks in the summer, which had a significantly positive correlation with precipitation ($p < 0.01$).

The net TF showed positive values for DIN, with 3.7 ± 1.1 kg N ha^{-1} $year^{-1}$ of NH_4–N and 5.1 ± 0.5 kg N ha^{-1} $year^{-1}$ of $NO_3 + NO_2$–N (Table 3.1). In total, DIN was enriched by 8.8 ± 1.5 kg N ha^{-1} $year^{-1}$ from the tree canopy in the evergreen broad-leaved forest, with a 71% contribution to the total DIN deposition onto the forest floor. The DON deposition in net TF also showed positive values (1.5 ± 0.6 kg N ha^{-1} $year^{-1}$), with a 19%

contribution to total DON deposition onto the forest floor. There were no significant relationships between precipitation and the monthly N fluxes in the net TF (Table 3.2).

However, the net TF of DON and DIN varied greatly with season (Fig. 3.5). The DON fluxes of net TF were lower during winter and early spring (even in negative values) compared to summer (Fig. 3.5a). Additionally, the DIN fluxes of net TF also showed a clear seasonal change, with peaks in summer, and a negative value occurring during May (Fig. 3.5b).

Table 3.1. Water flux (mm), volume-weighted mean (VWM) concentration (mg N L^{-1}), and annual nitrogen (N) fluxes (kg N ha^{-1} $year^{-1}$) in bulk deposition (BD), throughfall (TF), stemflow (SF), and net TF during the three study years (July 2016–June 2019). Values in parentheses are standard deviation (n = 3). DIN: Dissolved inorganic N; DON: Dissolved organic N; TDN: Total dissolved N.

	BD	TF	SF	Net TF
Water flux (mm)	1,916 (316)	1,254 (188)	76.7 (3.86)	–585 (131)
VWM concentration (mg N L^{-1})				
NH_4–N	0.10 (0.03)	0.50 (0.09)	0.26 (0.07)	–
$NO_3 + NO_2$–N	0.15 (0.05)	0.81 (0.08)	0.39 (0.11)	–
DIN	0.25 (0.09)	1.31 (0.15)	0.63 (0.18)	–
DON	0.39 (0.05)	0.72 (0.03)	0.77 (0.09)	–
TDN	0.65 (0.17)	2.00 (0.16)	1.47 (0.34)	–
N flux (kg N ha^{-1} $year^{-1}$)				
NH_4–N	1.49 (0.71)	5.07 (0.33)	0.15 (0.06)	3.72 (1.07)
$NO_3 + NO_2$–N	2.19 (1.21)	7.10 (1.03)	0.18 (0.04)	5.09 (0.53)
DIN	3.68 (1.88)	12.2 (0.86)	0.32 (0.10)	8.86 (1.46)
DON	6.52 (1.64)	7.51 (0.99)	0.54 (0.07)	1.53 (0.64)
TDN	10.6 (3.48)	19.5 (1.75)	0.86 (0.15)	9.82 (2.12)
DON contribution	0.66 (0.11)	0.38 (0.02)	0.63 (0.06)	0.16 (0.06)

Table 3.2. Spearman correlation between monthly precipitation (mm) and nitrogen fluxes (kg N ha^{-1} $month^{-1}$) in bulk deposition (BD) or net throughfall (TF) (n = 36).

	$NO_3 + NO_2$–N flux		**NH_4–N flux**		**DON flux**	
	BD	Net TF	BD	Net TF	BD	Net TF
Monthly precipitation	0.30*	0.30	0.37*	0.24	0.78**	0.10

* indicates significant relationships at $p < 0.05$, ** indicates significant relationships at $p < 0.01$. DON: Dissolved organic nitrogen

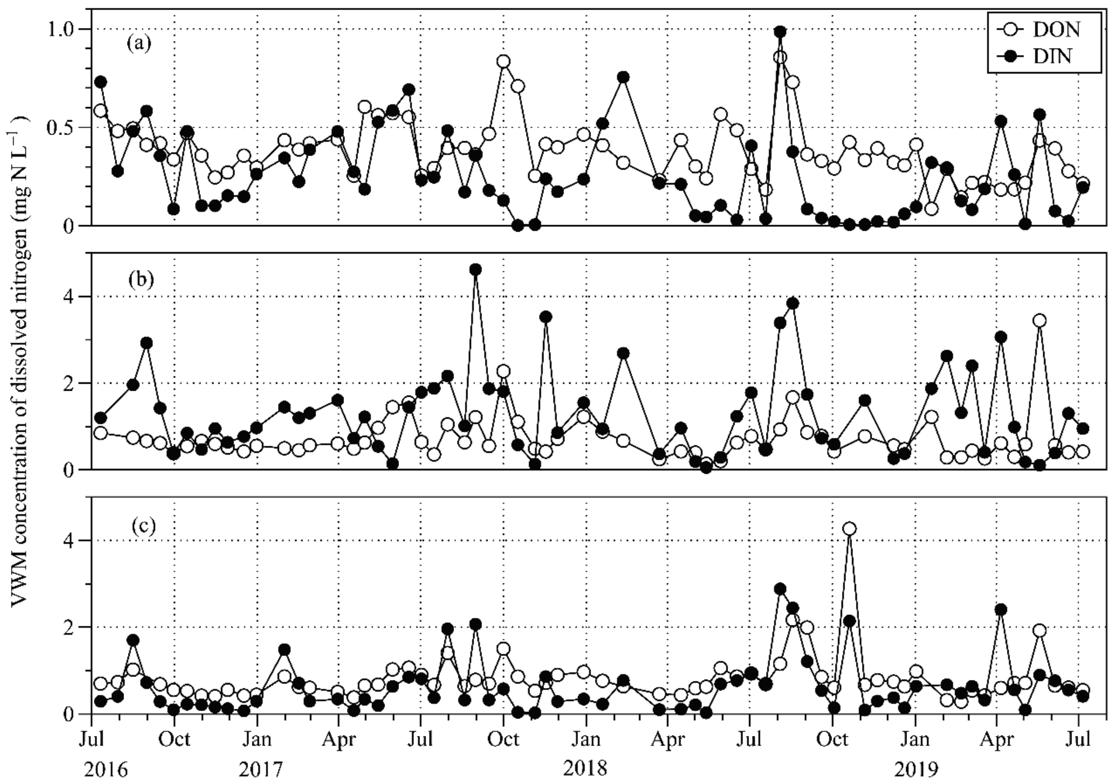

Fig. 3.3. Volume-weighted mean (VWM) concentration (mg N L^{-1}) of dissolved nitrogen in (a) bulk precipitation, (b) throughfall, and (c) stemflow during each period of sampling from July 2016 to June 2019. DON: Dissolved organic nitrogen; DIN: Dissolved inorganic nitrogen.

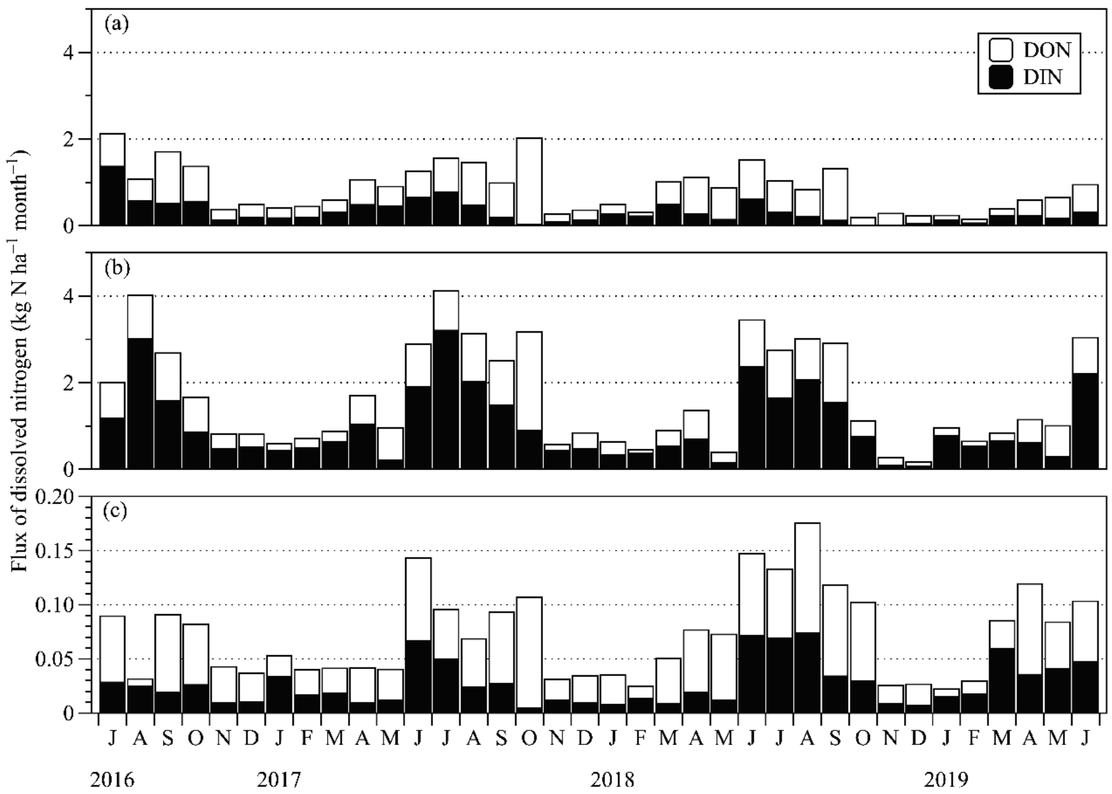

Fig. 3.4. Average monthly fluxes (kg N ha^{-1} month^{-1}) of dissolved nitrogen in (a) bulk deposition, (b) throughfall, and (c) stemflow from July 2016 to June 2019. DON: Dissolved organic nitrogen; DIN: Dissolved inorganic nitrogen.

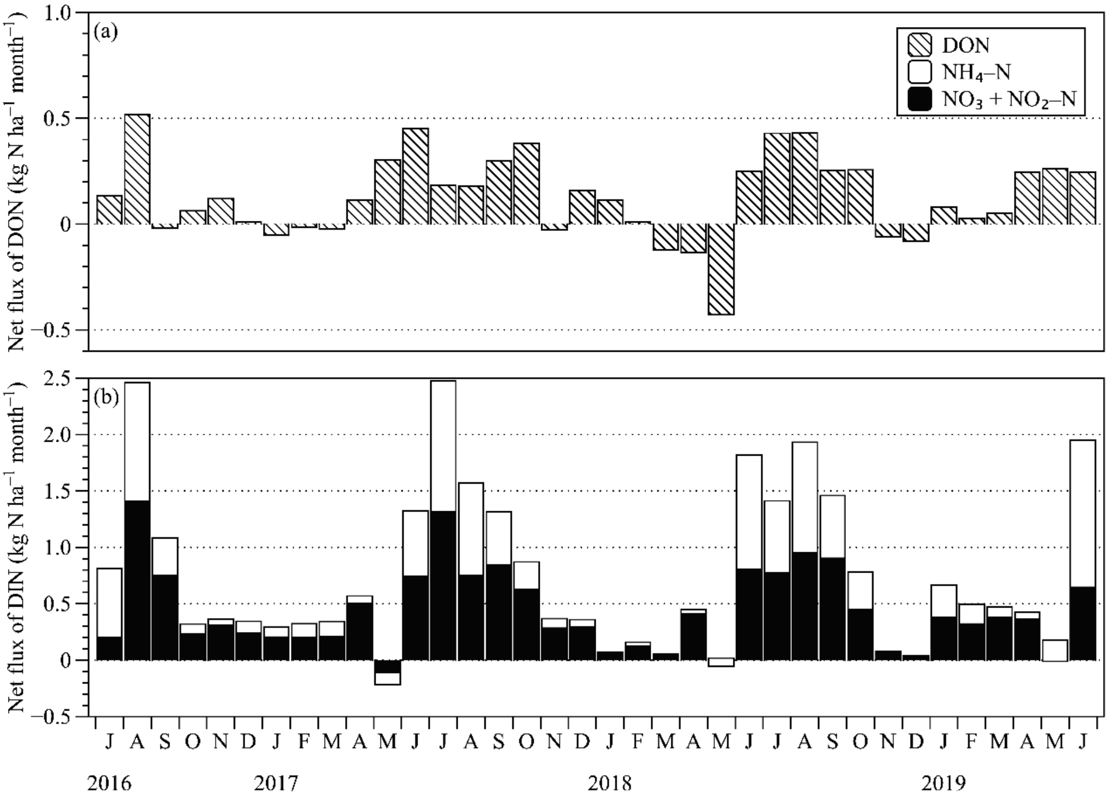

Fig. 3.5. Monthly net throughfall fluxes (kg N ha^{-1} $month^{-1}$) of (a) dissolved organic nitrogen (DON) and (b) dissolved inorganic nitrogen (DIN) from July 2016 to June 2019.

3.4. Discussion

3.4.1. Bulk N deposition into the Mt. Kinka forest site

The bulk $NO_3 + NO_2$–N (2.2 kg N ha^{-1} $year^{-1}$) and NH_4–N (1.5 kg N ha^{-1} $year^{-1}$) deposition levels were within the ranges of what had been reported for Japanese forest sites (1.4–5.6 kg N ha^{-1} $year^{-1}$ for NO_3–N, and 0.5–5.3 kg N ha^{-1} $year^{-1}$ for NH_4–N) (Mitchell et al., 1997). But bulk DIN deposition tended to be low compared with that at other study sites worldwide (e.g., 7.4–11.0 kg N ha^{-1} $year^{-1}$ in southeastern Brazil, de Souza et al., 2015; 44.3 and 83.8 kg N ha^{-1} $year^{-1}$ in Indonesia, Gillett et al., 2000), especially in Chinese forest sites (2.7–57.5 kg N ha^{-1} $year^{-1}$ and 14.0 kg N ha^{-1} $year^{-1}$, on average) (Du et al., 2014), which is approximately four times higher than that in the present study. Du et al. (2014) proposed that N deposition increased with increasing proximity to large cities in China because of local emission sources (e.g., waste disposal, energy consumption, and traffic) (urban hotspots of N deposition). Unexpectedly, our findings of a low level of bulk DIN deposition were contrary to the urban hotspots of N deposition. The DON deposition (6.5 kg N ha^{-1} $year^{-1}$) at the present study's site is significantly higher than the global range (25%–40%), as reviewed by Cornell (2011), which suggested that DON may be a vital portion of bulk N deposition in Japanese forest sites. Studies about the sources of DON are rare and have not been completely clarified.

Some studies have suggested that agricultural activities are possible sources of DON deposition, due to a higher proportion of DON deposition being found at agricultural sites (Izquieta-Rojano et al., 2016; Song et al., 2017).

N deposition fluxes showed distinct seasonal variations, with peaks in summer and early autumn, which is similar to other studies across the Asian monsoon area (Fu et al., 2019; Song et al., 2017; Wang et al., 2018; Wang et al., 2019). A different seasonal variation was found in northwestern Spain that experiences greater N deposition during its rainier winter season due to its Eurosiberian climate (Calvo-Fernández et al., 2017). Monthly N deposition was significantly related to monthly precipitation in the present study site (DIN: $p < 0.05$; DON: $p < 0.01$) (Table 3.2). These data show that the seasonal amount of precipitation is the main factor determining the bulk N deposition. Most studies concerning the effects of N deposition on forest ecosystems involve annual application of N addition (e.g., Lovett and Goodale, 2011). This general finding highlights the importance of the effects of monthly N deposition on forest ecosystems in various climates.

3.4.2. N deposition characteristics under the forest canopy

DIN was enriched by 8.8 kg N ha^{-1} $year^{-1}$, due to the tree canopy in the evergreen broad-leaved forest. These enrichments reflect the joint influence that dry deposition and canopy exchange have on N fluxes. Correlations between precipitation amount and N fluxes in net TF were applied in order to understand the canopy processes proposed by Lovett and Lindberg (1984). A positive correlation between precipitation amount and N fluxes in each net TF indicates canopy leaching; a negative correlation indicates canopy uptake; and no significant correlation suggests dry deposition. For the site of this present study, no significant correlation between precipitation amount and DIN fluxes in net TF (Table 3.2) indicates that dry deposition dominated in the net TF process.

Of note is that 71% of DIN was enriched from canopy for total DIN deposition to the forest floor (Table 3.1). Bettez and Groffman (2013) estimated that dry inorganic N deposition is relatively higher in urban sites compared to nonurban areas, due to the considerable emissions from human activities. The NH_4–N fluxes (3.7 kg N ha^{-1} $year^{-1}$) in net TF at the present study's site were higher than in the other Japanese forest sites (Aikawa et al., 2006; Matsumoto et al., 2020). The major sources of NH_3 are emissions from agricultural activities (livestock and fertilizer use) (Asman et al., 1998), and the major sources of NO_x are emissions from combustion of fossil fuel (Galloway et al., 2013). At the present study's site, there was an urbanized area (annual concentration of NO_x in the air: 8.6 ppb, Gifu City Official Website) as well as an agricultural area in the direction of the prevailing northwest wind (Fig. 3.1). The times in which fertilizers were added in rice cultivation during June, July, and August in Gifu City were consistent with the peaks of NH_4–N fluxes in net TF, which may explain the increased dry deposition of NH_4–N.

In contrast to DIN, DON was also enriched by 1.5 kg N ha^{-1} $year^{-1}$, and only 19% of DON was enriched from canopy for total DON deposition to the forest floor (Table 3.1). Le Mellec et al. (2010) suggested that the sources of DON in net TF are pollen deposition, insect excretions, and microbial activities from the forest canopy.

3.4.3. Ecological implications of N deposition in the evergreen broad-leaved forest

In total, 12.5 kg N ha^{-1} $year^{-1}$ of DIN occurred from TF and SF. Total DIN deposition was slightly higher than the minimal value of critical loads of inorganic N deposition in forest ecosystems, which mostly ranged from 10 to 20 kg N ha^{-1} $year^{-1}$ (Bobbink et al., 2010). We began monitoring forest dynamics and net primary production (NPP) at the study site of the present study at the foot of Mt. Kinka in 1989 (Chen et al., 2017). The area of Mt. Kinka was previously managed as productive forest, and harvesting was prohibited. The area has been protected as a natural secondary forest since 1947 because of previous over-use. During the 28-year study period starting in 1989, self-thinning of *C. cuspidata* governed the stand development process. The stem density of *C. cuspidata* decreased considerably from 666 to 404 stems ha^{-1}, while the aboveground biomass increased from 131.7 to 180.6 Mg ha^{-1}. No age-related decline of NPP occurred during the study period, and the growth of individual trees still increased in the nearly 70-year-old forest. Thus, deleterious effects of DIN deposition on the forest ecosystem, such as threats to ecosystem functioning and aggravation of eutrophication, are unlikely.

In contrast to DIN, studies concerning the effects of DON deposition on forest ecosystems are scarce. Annual total DON deposition (8.1 kg N ha^{-1} $year^{-1}$) from TF and SF contributed only 0.6% of soil organic N pool (1.4 ton N ha^{-1} at 5 cm soil depth, unpublished data) in the present study site. However, DON plays a substantial role in microbial activities in soil (e.g., acceleration of decomposition and subsequent ammonification and nitrification rate in soil, Jones et al., 2004). To our current knowledge, the effects of DON deposition on forest ecosystems have not been systematically studied. Merely the roles and processes of DON present in soil have been clarified. Therefore, we appeal to clarify the DON deposition and its effects on forest ecosystems to improve the existing understanding of interactions between forest ecosystems and atmosphere in biogeochemical cycles, especially for Japanese forest ecosystems. High DON contribution originates not only from ecosystem-external wet deposition (e.g., agricultural sources), but also occurs as canopy enrichment originating from biological sources in Japanese forest ecosystems.

3.5. Summary

Our results in this chapter showed that the bulk DIN deposition in the Mt. Kinka forest site totaled 3.7 kg N ha^{-1} $year^{-1}$. Bulk DIN deposition of the urban forest was within the ranges of previous studies of Japanese forests, and equal to that in rural areas. The bulk DON deposition was 6.5 kg N ha^{-1} $year^{-1}$, and contributed 66% of bulk TDN deposition, which suggests the importance of bulk DON deposition in Japanese forest ecosystems. When precipitation passed through the forest canopy, the total N deposition to the forest floor was 20.4 kg N ha^{-1} $year^{-1}$ (61% DIN and 39% DON). For each net TF (NH_4–N, NO_3 + NO_2–N, and DON), there was no significant correlation between precipitation amount and N fluxes; thus, the study site had a considerable amount of dry deposition, and the evergreen forest's ability to capture the dry deposition was greater. Our results also suggest that DIN in net TF originated as dry N deposition from N emissions outside the urban forest, while DON in net TF originated from recirculating N within the urban forest.

Chapter 4

DISCUSSION

4.1. Characteristics of bulk deposition in Japanese forest ecosystems

Our results showed that the snowfall contribution amounted to 37% of total N input to the Takayama forest site. There has been little previous research on the contribution of snowfall to N deposition; merely Fahey et al. (1985) studied the N cycle (snowfall contribution = 32%) in lodgepole pine forests of the Medicine Bow Mountains in western North America, and Oura et al. (2006) estimated the contribution of snowfall (45% of total N deposition) in a cool-temperate forest in Japan. Snowfall contribution in the Takayama forest site was similar with these two previous studies. Moreover, large differences in the dissolved N flux (23%, 43%, and 46%, respectively, for the three sampling years) were observed during the snow season, indicating the necessity of the long-term estimation of snowfall because of unpredictable year-to-year variations in snowfall. The relatively high contribution of the snow flux indicates that snow flux should be considered with respect to N deposition in the cool-temperate forests.

It seems that bulk DIN deposition in Japanese forest sites was irrespective of them being rural or urban (Table 4.1). Two possible reasons exist for the generally low level of bulk DIN deposition in Japanese forest ecosystems. First, developed countries (the United

States and Central and Western Europe) have experienced a decrease in N deposition since the 1980s because of decreased emissions under associated abatement policies (Gilliam et al., 2019; Schmitz et al., 2019). Inorganic N emissions in Japan have also decreased because of strict emission abatement policies (e.g., air NO_2 emission has decreased five-fold since the 1970s, Japan's Ministry of the Environment). Second, bulk DIN deposition may not be governed simply by local urban emission sources. De Souza et al. (2015) found that N deposition decreased by approximately 2.4 kg N ha^{-1} $year^{-1}$ from a coastal to an inland site in southeastern Brazil, influenced by oceanic emissions of N at the coastal site. Because Japan is a country surrounded by sea, the dominant sources of bulk DIN deposition likely come from oceanic emissions.

DON deposition in Japanese forest sites has rarely been studied (Table 4.1), with the only example being Mt. Fuji (4.1 kg N ha^{-1} $year^{-1}$), reported by Matsumoto et al. (2020). The DON deposition (7.9 kg N ha^{-1} $year^{-1}$ in Takayama forest site; 6.5 kg N ha^{-1} $year^{-1}$ in Mt. Kinka forest site) at the present study's sites is comparable to the Mt. Fuji, without urban-rural gradients (Table 4.1). DON contributed more than half of bulk N deposition (78% at Takayama forest site; 66% at Mt. Kinka forest) in both the present study's site, as well as the rural areas in the previous study mentioned (51% at Mt. Fuji). These values are significantly higher than the global range (25%–40%), as reviewed by Cornell (2011).

Studies about the sources of DON are rare and have not been completely clarified. Some studies have suggested that agricultural activities are possible sources of DON deposition, due to a higher proportion of DON deposition being found at agricultural sites (Izquieta-Rojano et al., 2016; Song et al., 2017). One characteristic of organic N compounds is their ability to be transported over long distances and deposited over a wide area (Cornell, 2011). Considering the possible ecological roles of DON (e.g., bioavailable or toxic to biota, as reviewed by Cornell, 2011; an important source of N for microorganisms in soil, Jones et al., 2004), DON deposition and the sources of DON could be an emerging research focus concerning Japanese forest ecosystems.

4.2. Characteristics of throughfall and stemflow in Japanese forest ecosystems

When precipitation passed through the forest canopy in growing season (May–November), the DIN flux in TF was approximately eight times higher in the Mt. Kinka forest site than in the Takayama forest site (Table 4.2). In contrast to DIN, there was similar DON flux in TF between two forest sites. One possible reason is that there are more human activities (e.g., traffic) near Mt. Kinka than Takayama, which could supply more DD in Mt. Kinka. Another possible reason is that the evergreen tree could catch more DD through canopy surface from atmosphere than deciduous tree canopy, and then more DD could be washed off into TF. De Schrijver et al. (2007) reported that the forest type significantly affected net TF deposition, because a higher DD was captured in evergreen forests due to their greater LAI. In Japanese forests, the mean LAI of evergreen forests (7.3 ± 2.0 for broad-leaved forests and 6.2 ± 2.0 for coniferous forests) is usually higher than that of deciduous broad-leaved forests (5.1 ± 1.7) (Scurlock et al., 2001). Thus, for the capturing of DD, the Mt. Kinka forest site may have a greater capacity to do this than the deciduous Takayama forest site would.

SF contributed a minor component of N fluxes (1% and 4% of total N deposition to the forest floor in Takayama Forest site and Mt. Kinka Forest site, respectively). However, the DIN flux was approximately ten times higher in the Mt. Kinka forest site than in the Takayama forest site. N fluxes in TF and SF showed a clear seasonal pattern with peaks in the summer and early summer, which had a significantly positive correlation with precipitation ($p < 0.01$), except for N fluxes in SF in the Takayama forest site. It indicated that the effects of N exchange with stems in SF should not be neglected in the Takayama forest site, which implies that the N flux in SF is more affected by different bark morphologies than water flux. The effects of different bark morphologies on SF needs to study further.

4.3. Characteristics of net throughfall in Japanese forest ecosystems

DIN enrichment in net TF tended to increase along rural-urban gradients in Japanese forest ecosystems (Table 4.3). DIN enrichment in Takayama forest site (NH_4–N: 0.4 kg N ha^{-1} year^{-1}; NO_3 + NO_2–N: –0.7 kg N ha^{-1} year^{-1}) was extremely lower than those in Mt. Kinka forest site (NH_4–N: 3.7 kg N ha^{-1} year^{-1}; NO_3 + NO_2–N 5.1 kg N ha^{-1} year^{-1}). However, the mechanisms of DD processes remain unclear, as do the reasons for the only increased DD (not including BD) in urban areas.

The major limitation of the "throughfall method" is the effect of canopy exchange on the estimation of dry inorganic N deposition, which has been reported in many deciduous and coniferous forest ecosystems (Adriaenssens et al., 2012; Draaijers et al., 1997; Fenn et al., 2013; Talkner et al., 2010). Negative correlations between precipitation and DIN deposition in net TF, and the annual negative value of DIN fluxes in net TF were found in the Takayama forest site. It suggested that net TF of DIN (especially for NO_3 + NO_2–N) in the tree canopy totaled 0.7 kg N ha^{-1} year^{-1}. However, the estimated value was much lower than the N uptake capability of deciduous forests (2.8–3.0 kg N ha^{-1} year^{-1}) (Talkner et al., 2010), mainly due to the DD effect.

Therefore, we need to separate the canopy exchange and DD on the forest canopy surface to clarify the N deposition processes under the forest canopy. A canopy budget model was proposed and applied in recent studies (Adriaenssens et al., 2012; Adriaenssens et al., 2013; Matsumoto et al., 2020) in order to clarify the DD and canopy exchange in net TF by using the equation "TF + SF = BD + DD + canopy exchange" and a tracer method (Na^+) to identify the deposition factor (dry/wet deposition). For example, Matsumoto et al. (2020) estimated that NH_4–N and NO_3 + NO_2–N in net TF amount to −0.5 kg N ha^{-1} year^{-1} and 0.7 kg N ha^{-1} year^{-1}, respectively. However, using the canopy budget model, the actual DD of NH_4–N and NO_2 + NO_3–N amounted to 4.1 kg N ha^{-1} $year^{-1}$ and 2.5 kg N ha^{-1} $year^{-1}$, respectively. Therefore, the DD of DIN in the present study sites would be expected to be more. Further research to identify the canopy exchange processes using the canopy budget model would facilitate a more precise quantification of N deposition using the "throughfall method" in forest ecosystems.

In contrast to DIN, DON was also enriched by 2.1 kg N ha^{-1} $year^{-1}$ in Takayama forest site, and 1.5 kg N ha^{-1} $year^{-1}$ in Mt. Kinka forest site, respectively. Moreover, DON deposition in net TF was nearly identical, for either urban or rural forest types (Table 4.3). This indicates that the DON might come from recirculating N within the forest, and that the anthropogenic effects for DON flux in net TF is negligible.

Table 4.1. Bulk deposition (BD) (kg N ha^{-1} year^{-1}) in Japanese forest ecosystems.

Study site	Population density (People/km^2)	Forest type	Dominant species	BD					References
				NH_4–N	NO_3 + (NO_2)–N	DIN	DON	TDN	
Kamigamo Experimental Forest Station, Kyoto	1,760 (Urban)	Evergreen conifer	*Chamaecyparis obtusa*	4.1	2.3	6.4	–	–	Maruyama et al., 1965
Kiryu, Ohtsu City	738 (Rural)	Evergreen conifer	*Pinus densiflora, C. obtusa*	4.0	1.4	5.4	–	–	Nishimura, 1973
Experiment forests, Tokyo University of Agriculture and Technology, Gunma Prefecture	6,368 (Urban)	Evergreen conifer	*Cryptomeria japonica*	4.1	4.4	8.5	–	–	Wu et al., 1996

Table 4.1. Continued.

Study site	Population density (People/km^2)	Forest type	Dominant species	BD					References
				NH_4–N	NO_3 + (NO_2)–N	DIN	DON	TDN	
Ryuoh Town, Shiga Prefecture	270 (Rural)	Evergreen conifer	*C. japonica*	0.6	2.7	3.3	–	–	Tokuchi and Iwatsubo, 1992
Tomakomai City, Hokkaido	310 (Rural)	Evergreen conifer	*P. strobus, P. koraienses*	2.2	2.5	4.7	–	–	Mitchell et al.,1997
Tsukuba City, Ibaragi Prefecture	860 (Rural)	Evergreen conifer	*C. obtusa, C. japonica*	5.3	5.2	10.5	–	–	
Mt.Fuji, Fujiyoshida City	384 (Rural)	Evergreen coniferous	*P. densiflora*	2.7	1.3	4.0	4.1	8.1	Matsumoto et al., 2020
Mt.Rokko, Kobe City	2,723 (Urban)	Evergreen coniferous	*C. japonica*	2.4	2.0	4.4	–	–	Aikawa et al., 2006

Table 4.1. Continued.

Study site	Population density (People/km^2)	Forest type	Dominant species	BD					References
				NH_4–N	NO_3 + (NO_2)–N	DIN	DON	TDN	
Mt.Awaga, Tanba City	124 (Rural)	Evergreen coniferous	*C. japonica*	3.4	3.4	6.9	–	–	Aikawa et al., 2006
Takayama forest, Takayama City	39 (Rural)	Deciduous broad-leaved	*Quercus crispula, Betula ermanii,* B. *platyphylla* var. *japonica*	1.7	1.5	3.2	7.9	11.1	Present study
Mt.Kinka, Gifu City	1,965 (Urban)	Evergreen broad-leaved	*Castanopsis cuspidata*	1.5	2.2	3.7	6.5	10.2	Present study

Table 4.2. Average bulk deposition (BD), throughfall (TF), and stemflow (SF) in Takayama forest site and Mt. Kinka forest site during growing season (May–November) (kg N ha^{-1} 7 $months^{-1}$).

	Takayama forest site				Mt. Kinka forest site			
	NH_4–N	NO_3 + NO_2–N	DIN	DON	NH_4–N	NO_3 + NO_2–N	DIN	DON
BD	0.6	0.9	1.5	5.3	0.9	1.1	2.0	5.2
TF	1.0	0.2	1.2	7.3	4.2	5.2	9.4	6.2
SF	0.02	0.0	0.02	0.1	0.1	0.1	0.2	0.4

Table 4.3. Nitrogen fluxes (kg N ha^{-1} $year^{-1}$) in net TF for Japanese forests. Net TF = TF (+ stemflow) – BD. DON: Dissolved organic nitrogen.

Study site	Population density (People/km^2)	Forest type	Dominant species	Net TF			References
				NH_4–N	NO_3 + (NO_2)–N	DON	
Mt.Fuji, Fujiyoshida City	384 (Rural)	Evergreen coniferous	*Pinus densiflora*	–0.5	0.7	1.6	Matsumoto et al., 2020
Mt.Rokko, Kobe City	2,723 (Urban)	Evergreen coniferous	*Cryptomeria japonica*	1.4	11.4	–	Aikawa et al., 2006
Mt.Awaga, Tanba City	124 (Rural)	Evergreen coniferous	*Cryptomeria japonica*	0.4	4.6	–	
Takayama forest, Takayama City	39 (Rural)	Deciduous broad-leaved	*Quercus crispula, Betula ermanii,* B. *platyphylla* var. *japonica*	0.4	–0.7	2.1	Present study
Mt.Kinka, Gifu City	1,965 (Urban)	Evergreen broad-leaved	*Castanopsis cuspidata*	3.7	5.1	1.5	Present study

Chapter 5

CONCLUSIONS

The snowfall contribution was 37% of the total N input to the Takayama Forest site, with large annual variations (23%, 43%, and 46%, respectively, for the three sampling years). The substantial snow contribution highlights the importance of snowfall N deposition estimation in heavy snow zones along the Sea of Japan.

The bulk DIN deposition in Japanese forest ecosystems is generally low, irrespective of rural-urban gradients (only 3.2 kg N ha^{-1} $year^{-1}$ and 3.7 kg N ha^{-1} $year^{-1}$ in Takayama Forest site and Mt. Kinka Forest site, respectively), which is in contrast to the urban hotspots of N deposition in China (Du et al., 2014). This is attributable mainly to decreasing N emissions in developed countries. The bulk DON deposition contributed 78% and 66% of TDN deposition (7.9 kg N ha^{-1} $year^{-1}$ and 6.5 kg N ha^{-1} $year^{-1}$ in Takayama Forest site and Mt. Kinka Forest site, respectively) in the present two study sites, respectively. It suggests the bulk DON deposition is an indispensable component of N deposition in Japanese forest ecosystems.

When precipitation passed through the forest canopy in growing season (May–November), the DIN flux in TF and SF was significantly higher in the Mt. Kinka forest site (TF: 9.4 kg N ha^{-1} $year^{-1}$; SF: 0.2 kg N ha^{-1} $year^{-1}$) than in the Takayama forest site

(TF: 1.2 kg N ha^{-1} $year^{-1}$; SF: 0.02 kg N ha^{-1} $year^{-1}$). This may due to the non-negligible N emissions near Mt. Kinka and possibly because of the evergreen broad-leaved forest's greater ability to capture N. In contrast to DIN, there was similar DON flux in TF and SF. SF contributed a minor component of N fluxes (1% and 4% of total N deposition to the forest floor in Takayama Forest site and Mt. Kinka Forest site, respectively).

The contribution of dry DIN deposition estimated in net TF tended to increase along rural-urban gradients in Japanese forest ecosystems (NH_4–N: 0.4 kg N ha^{-1} $year^{-1}$ in Takayama Forest site, 3.7 kg N ha^{-1} $year^{-1}$ in Mt. Kinka Forest site; NO_3 + NO_2–N: –0.7 kg N ha^{-1} $year^{-1}$ and 5.1 kg N ha^{-1} $year^{-1}$, respectively). The mechanism behind only DD increase (not BD in urban forests) is presently unclear, although the net TF method is critical for forest types because it enables differentiating between capturing DD and canopy uptake abilities. In other words, evergreen broad-leaved forests may have a high capacity for capturing DD. On the other hand, total DON deposition in net TF (2.1 kg N ha^{-1} $year^{-1}$ and 1.5 kg N ha^{-1} $year^{-1}$, respectively) was nearly identical for either urban or rural forest types and indicated that the anthropogenic effects for DON deposition in net TF are negligible. Higher contribution of bulk DON deposition and DON enrichment under the forest canopy suggests the importance of DON deposition in Japanese forest ecosystems.

Future studies should focus on clarifying the regulations of BD and DD processes and use data from more different types of Japanese forest ecosystem, which may not be governed simply by local N emissions along the rural and urban gradients, but detailed land-use pattern near the sites.

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