

The Increased Contribution of Atmospheric Nitrogen Deposition to Nitrogen Cycling in a Rural Forested Area of Kyushu, Japan

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Abstract Japan has been receiving increasing levels of atmospheric nitrogenous pollutants from the East Asian continent over the last few decades, so it is critical to evaluate the impact of this increased atmospheric nitrogen (N) deposition on N cycling even in rural forests. This study evaluated the contribution of the current level of atmospheric N deposition to N cycling in a rural forested area. Bulk precipitation and stream water were collected during 2007–2011 at the Shiiba Research Forest (SRF) located in the central Kyushu mountain range of southern Japan. Litterfall was also collected to investigate the contribution of atmospheric N deposition to total N input (litterfall N+atmospheric N deposition). The results showed that atmospheric depositions of both nitrate (NO_3^-) and ammonium (NH_4^+) were a few times higher during 2009–2011 than in 1991. This could be the result of additional N deposition from the increased long-range transport of nitrogenous pollutants from the East Asian continent. The current level of annual N deposition ($9.7 \text{ kg N ha}^{-1} \text{ year}^{-1}$) at the SRF was comparable with that at many urban sites and was close to the

reported threshold values causing N saturation in forest ecosystems. Although current atmospheric N deposition was an important component (23 %) of total N input ($43 \text{ kg N ha}^{-1} \text{ year}^{-1}$) at the SRF, the concentrations of NO_3^- in stream water were consistently low ($<10 \mu\text{mol L}^{-1}$). These results indicate that atmospheric N deposition is currently largely incorporated into forest ecosystems without excess N export from forested watersheds.

Keywords Atmospheric N deposition · Rural forest · N cycling · Long-range transport · East Asia

1 Introduction

Increased atmospheric nitrogen (N) deposition can have a large impact on N cycling in forest ecosystems (Aber et al. 1989; Vitousek et al. 1997; Fujimaki et al. 2009). When atmospheric N deposition is naturally low, temperate forests are limited by the supply of biologically available N (Vitousek et al. 1997), and N input from atmospheric N deposition is a small contribution to N cycling compared with the large input of litterfall N (Likens and Bormann 1979). However, as atmospheric N deposition increases, it can become an important component of N input that changes N cycling, resulting in enhanced growth and carbon sequestration of trees in the initial stage (Magnani et al. 2007; Solberg et al. 2009; Thomas et al. 2010). Further chronic atmospheric N deposition can lead to N saturation of forests (Aber

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et al. 1989; Vitousek et al. 1997), which is an excess of inorganic N (NO_3^- and NH_4^+) over biotic demand (Aber et al. 1989) and can cause excess N export from forested watersheds (Chiwa et al. 2012).

Despite this, the impacts of increased atmospheric N deposition on N cycling have not been intensively evaluated in rural forested areas as much as in urban forests, defined as urbanized and industrial areas, and those immediately outside the area, according to the Acid Deposition Monitoring Network in East Asia (EANET). While atmospheric N deposition is high in urban forests (Mitchell et al. 1997; Okochi and Igawa 2001; Chiwa et al. 2010; Tokuchi et al. 2011), rural ecosystems, defined as areas that are more than 20 km distant from large pollution sources like cities, power plants, and highways according to EANET, have been thought of as unpolluted areas less affected by anthropogenic perturbations including air pollutants because there are no large local anthropogenic sources. However, Japan, including its rural areas, has been receiving air pollutants from the East Asian continent through long-range transport (Wakamatsu et al. 1996; Nagafuchi et al. 2001; Shimohara et al. 2001; Akimoto 2003; Takiguchi et al. 2008; Chiwa 2010). This is because N oxide (NO_x) emissions in Asia increased 2.5 times between 1980 and 1997 (Akimoto 2003), a further 2.8 times between 1980 and 2003 (Ohara et al. 2007). In addition, atmospheric N deposition has been increasing across Japan during the last two decades even in rural areas (Seto et al. 2004; Morino et al. 2011). The effects of increased atmospheric N deposition caused by long-range transport of pollutants on forests can be widespread because rural forests commonly occupy a larger proportion of the total forest area than urban/suburban forests.

Rural forested areas are more important than remote areas, defined as areas that are more than 50 km distant from large pollution sources like cities, power plants, and highways and more than 500 m apart from main roads, which are farther from regional pollutant sources in terms of the impact of increased atmospheric N deposition on N cycling. This is because regionally derived pollutants enhance atmospheric N deposition in rural areas in addition to long-range transport of pollutants. Regionally derived pollutants around remote areas are very few. Therefore, atmospheric N deposition in rural forests can be greater than in remote areas, and the effect of increased atmospheric N deposition derived from long-range transport on N cycling can be larger in rural areas than in remote areas.

The objective of this study was to evaluate the contribution of the current level of atmospheric N deposition to N cycling in rural forests. To achieve this objective, atmospheric deposition and stream water N concentration were quantified at the Shiiba Research Forest (SRF) from 2007 to 2011. Litterfall was also collected to investigate the contribution of atmospheric N deposition to total N input (litterfall N+ atmospheric N deposition).

2 Materials and Methods

2.1 Study Area

This study was conducted at the SRF of Kyushu University ($32^\circ 22' \text{N}$, $131^\circ 09' \text{E}$; elevation 660–1,000 m), situated in the central Kyushu mountain range of southern Japan (Fig. 1). The SRF, a rural forested area according to the EANET definition, is situated in the village of Shiiba. The population in the village has declined over the last few decades, from 4,611 in 1990 to 3,092 in 2010 (Shiiba Village Website, <http://www.vill.shiiba.miyazaki.jp>), and the population density in the village was 5.6 people/ km^2 in 2012. In the village, forest accounts for 96 % of the total area (536 km^2). Land use in Shiiba has changed little over recent decades; agricultural area fell from 378 ha in 1990 to 343 ha in 2005 (Ministry of Agriculture, Forestry and Fisheries, <http://www.maff>).



Fig. 1 Location of the Shiiba Research Forest study site. *Short dashed lines* indicate national boundaries. Codes represent sampling sites for bulk/wet deposition: Tokyo (T), Banryu (B), Fukuoka (F), Yoshino (Y), Sagamihara (S), Ijira (I), Oku-Nikko (Oku), Rishiri (R), Ogasawara (Og), Oki (Oki), and Hedo (He)

go.jp/j/tokei/index.html). The SRF is a cool temperate mixed forest, consisting of coniferous trees such as *Abies firma* Sieb. et Zucc. and *Tsuga sieboldii* Carr. and deciduous broad-leaved trees such as *Quercus crispula* Blume, *Fagus crenata* Blume, and *Acer sieboldianum* Miq (Murata et al. 2009). The annual temperature and annual precipitation at the SRF office (32°22'N, 131°09'E; altitude 600 m) was 13.5 °C during 2010–2011 and 357.7 cm during 2009–2011, respectively.

2.2 Water Sample Collection and Chemical Analysis

Bulk precipitation was collected for 3 years from January 2009 to December 2011. A polyethylene funnel with a diameter of 240 mm was used to collect bulk precipitation and was installed on the roof of the SRF office (~10 m above the ground). The method for collecting bulk precipitation followed that of Inokura et al. (1994) where a polyethylene container was placed and precipitation samples were collected biweekly or monthly. The biweekly sampling of stream water was conducted in the Oyabu basin (5.30 km², 32°22'N, 131°10'E; altitude 990–1,480 m) during 2009–2011 and in three adjoining small sub-basins of the Oyabu basin (A 0.10 km², B 0.03, and C 0.03 km², 32°22'N, 131°09'E; altitude 1,050–1,210 m) during 2007–2011. Before sampling, bottles and funnels were placed into 1 % Contaminon solution (Contaminon N, Wako, Osaka, Japan) for at least 24 h and then washed with deionized water.

The samples were transported to the laboratory within ~1 h of collection. The samples were filtered with a 0.45- μ m pre-washed glass fiber filter (GC50, Advantec, Tokyo, Japan). Filtered and unfiltered water samples were frozen in the dark until chemical analysis. The concentrations of anions (chloride (Cl⁻), nitrate (NO₃⁻), and sulfate (SO₄²⁻) and cations (sodium (Na⁺), ammonium (NH₄⁺), potassium (K⁺), magnesium (Mg²⁺), and calcium (Ca²⁺)) in the filtered sample were determined using an ion chromatograph (DX-120, Dionex, Osaka, Japan). For total phosphorus (TP) analysis, unfiltered water samples were digested using potassium peroxodisulfate (K₂S₂O₈) and then measured by molybdenum blue (ascorbic acid) absorptiometry.

2.3 Litterfall Collection and Nitrogen Analysis

A study plot of 100×100 m was established in the SRF (32°22'N, 131°10'E; altitude 1,092–1,133 m). Dominant

tree species of the plot were *T. sieboldii*, *A. firma*, *Q. crispula*, *Castanea crenata*, and *Pinus densiflora*. The basal area of the plot was 52.4 m²/ha. The plot was separated into twenty-five 20×20 m quadrats. Twenty-five 0.5-m² litter traps were established at the center of each quadrat. Litterfall was collected monthly from April 2008 to April 2009. Litter samples were separated into pine needles, leaves of other conifer species, leaves of other species, and other components. Separated samples were dried at 70 °C for 48 h, weighed, pooled, and ground to a fine powder for chemical analysis. Total N in the samples was measured with a CN analyzer (CN corder MT-700, Yanaco Co., Ltd., Tokyo, Japan).

2.4 Data Handling and Statistical Analysis

Annual deposition (flux) of each chemical species via bulk precipitation (kg ha⁻¹ year⁻¹) was calculated by multiplying volume-weighted mean concentrations (mg L⁻¹×10⁻²) by the annual precipitation depth (cm). Precipitation depth (cm) was calculated by dividing collected water volume (mL) by the funnel area (cm²). The concentrations of non-sea-salt (nss)-SO₄²⁻ were calculated from the equivalent ratios of SO₄²⁻/Na⁺ (0.0603) in seawater. Annual trends in agricultural areas and the number of cattle in Japan during 1990–2010 were analyzed from the statistical database of the Census of Agriculture Forestry, which is available from the Ministry of Agriculture, Forestry and Fisheries (<http://www.maff.go.jp/j/tokei/census/afc/index.html>). The N amount in litterfall (kg N ha⁻¹ year⁻¹) was calculated by multiplying litterfall amount (kg dw ha⁻¹ year⁻¹) by the litter N content (kg N kg⁻¹ dw).

3 Results and Discussion

3.1 Increase in Atmospheric N Deposition at the SRF

Atmospheric NO₃⁻ and NH₄⁺ deposition during 2009–2011 was a few times larger than in 1991 (Fig. 2). Annual atmospheric deposition for each ion except for K⁺ tends to increase with annual precipitation (Fig. 3). These trends can be applied for Cl⁻, nss-SO₄²⁻, Na⁺, and Mg²⁺ during both periods (1991 and 2009–2011). However, atmospheric deposition for NO₃⁻ and NH₄⁺ in 1991 is much lower than that from 2009–2011, despite the high annual precipitation in 1991. To explain the increase in atmospheric N deposition in this study

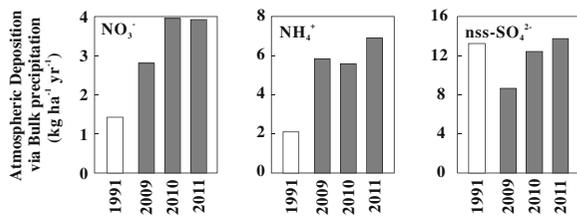


Fig. 2 Atmospheric depositions of NO_3^- , NH_4^+ , and nss-SO_4^{2-} via bulk precipitation ($\text{kg ha}^{-1} \text{ year}^{-1}$) in 1991 and 2009–2011 at the Shiiba Research Forest. Deposition data in 1991 were taken from Inokura et al. (1994)

site during the past few decades, factors other than precipitation are needed.

The increase in atmospheric NO_3^- deposition during the last two decades at the SRF is likely to be the result of increased long-range transported air pollutants from East Asia. Analogous increases in atmospheric NO_3^- deposition during the last two decades have been reported (Seto et al. 2004; Morino et al. 2011; Chiwa et al. 2012). Morino et al. (2011) found that wet deposition rates of NO_3^- across Japan during 1989–2008 increased

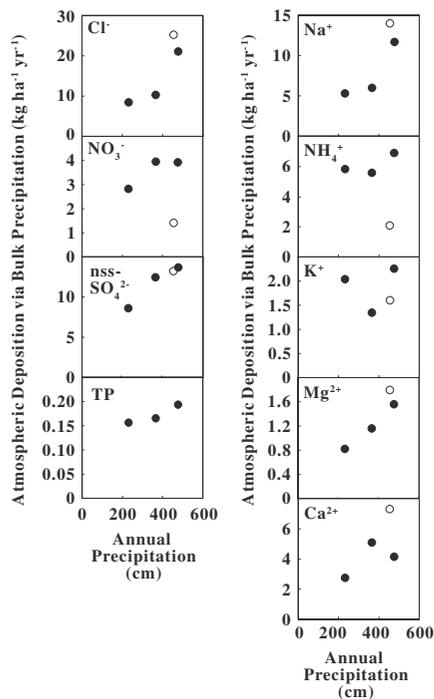


Fig. 3 Relationship between annual precipitation depth (cm) and atmospheric depositions of Cl^- , Na^+ , Mg^{2+} , Ca^{2+} , K^+ , and total phosphorus (TP) via bulk precipitation ($\text{kg ha}^{-1} \text{ year}^{-1}$) in 1991 (white circle) and 2009–2011 (black circle) at the Shiiba Research Forest. Deposition data from 1991 were taken from Inokura et al. (1994). TP deposition in 1991 was not analyzed

by 2–5 %/year, and the authors attributed this mostly to the increased emissions of atmospheric pollutants from China. NO_x emission in China was 6.55 Mt in 1990 but had increased to 14.4 Mt in 2003 (Ohara et al. 2007), while in Japan, NO_x emissions slightly decreased during 1990–2003 (Ohara et al. 2007) and decreased from 2000 to 2008 by 17 % (Regional Emission inventory in Asia, <http://www.nies.go.jp/REAS/>). Air pollutants including NO_x emitted from the East Asian continent are transported to Japan by the prevailing westerly winds (Nagafuchi et al. 2001; Shimohara et al. 2001; Akimoto 2003; Takiguchi et al. 2008; Chiwa 2010).

The increase in atmospheric NH_4^+ deposition during the last two decades at the SRF could also be the result of the increased long-range nitrogenous pollutant transport. NH_3 emission is reportedly derived mostly (80 %) from agricultural activities including livestock and fertilizer (Streets et al. 2003; Huang et al. 2012). Agricultural activity in local and domestic areas could not be responsible for the increase in atmospheric NH_4^+ deposition at the SRF. This might be because the agricultural area (378 and 343 ha in 1990 and 2005, respectively) and the number of cattle (1,350 and 1,370 in 1993 and 2005, respectively) have not changed much during the last few decades in the village of Shiiba (Ministry of Agriculture, Forestry and Fisheries, <http://www.maff.go.jp/j/tokei/index.html>). In addition, the decrease in agricultural area (Fig. 4a) and the number of cattle (Fig. 4b) in Japan during 1990–2010 implies a reduction in agricultural activity and subsequently a decrease in NH_3 emission derived from agricultural activities. In support of this view, NH_3 emission in Japan decreased from 2000 to 2008 by 10 % (Regional Emission

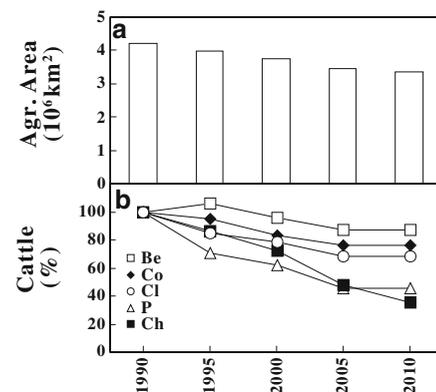


Fig. 4 Annual trends in **a** agricultural areas and **b** the number of cattle in Japan during 1990–2005. Codes indicate beef (Be), cow (Co), boiler chicken (Bl), pig (P), and chicken (Ch). The number of cattle was expressed as a percentage of the number in 1990

inventory in Asia, <http://www.nies.go.jp/REAS/>). On the other hand, NH₃ emission on the East Asian continent increased from 10.6 Tg in 1990 to 14.2 Tg in 2000 (Streets et al. 2003), and NH₃ emissions in Asia increased from 2000 to 2008 by 17 % (Kurokawa et al. 2013), which could be responsible for the increase in atmospheric NH₄⁺ deposition at the SRF.

A temporal change in nss-SO₄²⁻ deposition in this study site during the last few decades was elusive (Fig. 2). SO₂ emissions in Japan decreased slightly during 1990–2003 (Ohara et al. 2007) and decreased from 2000 to 2008 by 12 % (Kurokawa et al. 2013). In contrast, SO₂ emission increased by 1.5 times during 1990–2003 in Asia (Ohara et al. 2007) and increased from 2000 to 2008 by 34 % (Kurokawa et al. 2013). Both of these factors could have affected the changes in nss-SO₄²⁻ deposition in this study site during the last two decades. However, a clear explanation could not be achieved in this study. The deposition of nss-SO₄²⁻ at the SRF in 1991 (13.2 kg S ha⁻¹ year⁻¹) was close to the values (14.0 kg S ha⁻¹ year⁻¹) recorded for precipitation collected at 29 sites in Japan including urban, rural, and seaside sites during 1986–1988, while NO₃⁻ (1.4 kg N ha⁻¹ year⁻¹) and NH₄⁺ (2.1 kg N ha⁻¹ year⁻¹) deposition in 1991 was lower than the national average at 29 sites (3.8 and 6.0 kg N ha⁻¹ year⁻¹, respectively; Tamaki et al. 1991). In addition, Na⁺ and Cl⁻ depositions whose sources are mainly marine aerosol are also lower at the SRF (25.3 and 14.0 kg ha⁻¹ year⁻¹, respectively) than the national average at 29 sites (66.6 and 34.4 kg ha⁻¹ year⁻¹, respectively; Tamaki et al. 1991). These results imply that nss-SO₄²⁻ deposition was already high in 1991. Seto et al. (2004) reported the decreasing trend of annual nss-SO₄²⁻ deposition throughout Japan during 1989–1998 due to the decrease in domestic SO₂ emissions. During 2000–2005, by contrast, SO₄²⁻ deposition in Japan increased because of the rapid increase of SO₂ emission from China (Kuribayashi et al. 2012). The latest trends on SO₂ emission in Asia and SO₄²⁻ deposition in Japan are needed. Further investigation will be required to understand the annual trend in nss-SO₄²⁻ deposition and its relationship with domestic and East Asian emissions of SO_x.

Table 1 shows the current level of annual atmospheric N deposition in urban and rural sites in Japan. It was higher in urban and rural sites including the SRF than those in remote sites. This can be explained by regionally derived pollutants around these areas in addition to long-range transport of pollutants from East Asia. In addition,

the current level of annual N deposition at the SRF was surprisingly comparable with that at the urban sites. Comparable deposition at the SRF with urban sites suggests that long-range nitrogenous pollutant transport from the East Asian continent is also an important component of current atmospheric N deposition.

The value of annual N deposition at the SRF (9.7 kg N ha⁻¹ year⁻¹) was close to the reported threshold values causing N saturation in other forested ecosystems. The threshold value was estimated to be 9–13 kg N ha⁻¹ year⁻¹ for a whole watershed in the northeastern United States (Aber et al. 2003). In addition, Dise and Wright (1995) found that below an N deposition threshold of 10 kg N ha⁻¹ year⁻¹, no significant N leaching occurred from European forests, and at intermediate levels of 10–25 kg N ha⁻¹ year⁻¹, N leaching occurred at some forested sites. In Japan, Ohruai and Mitchell (1997) demonstrated that significant nitrate leaching occurred from the forests that received more than 10 kg N ha⁻¹ year⁻¹. However, it should be noted that other factors such as climate, vegetation type, land use history, and hydrologic flow path influence ecosystem responses to N deposition (Campbell et al. 2004; Tokuchi et al. 2011).

3.2 Impact of Increased Atmospheric N Deposition on N Cycle in Rural Forested Ecosystems

Nitrogen input via litterfall at the SRF was 33 kg N ha⁻¹ (Table 2), and atmospheric N deposition accounted for 23 % of total N input (43 kg N ha⁻¹ year⁻¹), indicating that atmospheric N deposition is an important component of total N input at the SRF. Litterfall N input at the SRF was comparable with that in a Japanese black pine stand (44 kg N ha⁻¹ year⁻¹; Enoki et al. 1997); Japanese cedar stands (26–34 kg N ha⁻¹ year⁻¹; Fukushima et al. 2011); six Japanese stands of Hinoki cypress and Japanese red pine, beech, and oak (15–49 kg N ha⁻¹ year⁻¹; Kawahara 1971); and five European stands of Norway spruce, Sitka spruce, fir, beech, and oak (29–45 kg N ha⁻¹ year⁻¹; Hansen et al. 2009).

The NO₃⁻ concentration level of stream water at the SRF (<10 μmol L⁻¹; Table 3) was much smaller than in the N-saturated forested watersheds in urban/suburban areas of Japan (~100 μmol L⁻¹ (Ohruai and Mitchell 1997); 76 μmol L⁻¹ (Okochi and Igawa 2001); 80–90 μmol L⁻¹ (Shibata et al. 2001); 59–66 μmol L⁻¹ (Zhang et al. 2008); 54–69 μmol L⁻¹ (Chiwa et al. 2010)). Low NO₃⁻ concentrations (typically less than

Table 1 Annual N and S deposition ($\text{kg ha}^{-1} \text{ year}^{-1}$) via bulk or wet precipitation in urban/suburban, rural, and remote sites in Japan in recent years

	Location	Periods	NO_3^-	NH_4^+	DIN ($\text{NO}_3^- + \text{NH}_4^+$)	nss- SO_4^{2-}	Bulk or wet	Reference	
Urban	Tokyo	Tokyo	2007–2008	4.7	6.0	10.7	8.1	Wet	Network center of EANET ^b
	Shimane	Banryu	2001–2008	4.4	3.5	7.9	8.3	Wet	Network center of EANET
	Fukuoka	Fukuoka	2000–2007	5.3	6.7	12.0	12.4	Wet	Fukuoka Data Website ^c
	Fukuoka	Kasuya	2009–2011	5.1	8.4	13.5	13.9	Bulk	Chiwa, unpublished data
Rural	Nara	Yoshino	2002–2004	3.3	3.7	7.1	14.2	Bulk	Fukushima and Tokuchi (2009)
	Kanagawa	Sagamihara	2004–2005	2.0	5.0	7.0	NA	Bulk	Ham et al. (2010)
	Gifu	Ijira	2001–2008	8.0	7.1	15.1	14.9	Wet	Network center of EANET
	Tochigi	Oku-Nikko	2000–2001	2.4	5.1	7.5	19.6 ^a	Bulk	Oura (2010)
	Miyazaki	Shiiba (SRF)	2009–2011	3.6	6.1	9.7	11.6	Bulk	This study
Remote	Hokkaido	Rishiri	2001–2008	1.7	2.6	4.3	4.6	Wet	Network center of EANET
	Tokyo	Ogasawara	2001–2008	0.9	1.0	1.9	2.4	Wet	Network center of EANET
	Shimane	Okii	2001–2008	3.3	2.5	5.8	6.4	Wet	Network center of EANET
	Okinawa	Hedo	2001–2008	2.4	2.9	5.3	6.3	Wet	Network center of EANET

The site classifications of urban, rural, and remote sites are followed by their EANET definitions

^a As SO_4^{2-}

^b <http://www.eanet.cc/jpn/>

^c <http://www.city.fukuoka.lg.jp/kankyo/hokanken/>

$10 \mu\text{mol L}^{-1}$) in baseflow stream water have been observed in non-N-saturated forested areas (Stoddard 1994; Zhang et al. 2008). Therefore, we conclude from this that N saturation is not occurring in the SRF forest ecosystem. The lack of N saturation symptoms in the SRF forest ecosystem, despite a high deposition level comparable with the N saturation threshold values, indicates that atmospheric N deposition at the SRF is currently largely incorporated into the forest ecosystem without excess N export. Leaching of N from forested watersheds as a result of low N retention may occur much later than internal N cycle changes (Emmett 2007;

Phoenix et al. 2012). Therefore, atmospheric N deposition should be taken into consideration regarding N cycling in forest ecosystems even in rural areas.

4 Conclusions

This study showed that there has been increased atmospheric N deposition at the SRF as a result of the

Table 2 Litterfall N input at the Shiiba Research Forest

	Litterfall amount ($\text{g m}^{-2} \text{ year}^{-1}$)	Litter N content ($\text{g kg}^{-1} \text{ dw}$)	Litterfall N amount ($\text{kg N ha}^{-1} \text{ year}^{-1}$)
Average	489	6.9	33
Median	505	6.4	33
Min	242	5.7	16
Max	700	9.2	46
SD	91	1.1	7.5
<i>n</i>	25	25	25

SD standard deviations

Table 3 Stream water NO_3^- concentrations ($\mu\text{mol L}^{-1}$) at the Shiiba Research Forest

	Oyabu	Sub-A ^a	Sub-B ^a	Sub-C ^a
Average	7.0	6.0	9.4	7.5
Median	7.3	5.7	9.1	7.1
Min	0.7	1.8	1.0	3.0
Max	14.3	12.6	22.3	14.8
SD	2.9	2.5	4.3	2.7
<i>n</i>	63	80	80	91
Periods	2009–2011	2007–2011 ^b	2007–2011 ^b	2007–2011

SD standard deviations

^a Small sub-basins of Oyabu basin

^b Except for the periods from October 2008 to February 2009

increased emissions in East Asia during the last few decades. Currently, increased atmospheric N deposition caused by the long-range transport of pollutants from the East Asian continent is an important component of N input even in rural forested areas. Consequently, this study highlights the importance of considering atmospheric N deposition with regard to N cycling in rural forested areas. Long-term ecological monitoring including atmospheric N deposition is needed to investigate the current and future impact of the atmospheric N deposition on rural forest ecosystems.

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