Comparison of Nitrogen Dry Deposition on Cedar and Oak Leaves in the Tama Hills Using Foliar Rinsing Method

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Abstract Amount of nitrogen (N) dry deposition on the leaf surface of oak and cedar was compared using throughfall method and foliar rinsing method. The study was conducted at FM Tamakyuryo, located at the northeast edge of the Tama Rolling Hills, Tokyo, Japan. The throughfall method showed that the total N deposition were 19.7, 19.1, and 50.6 kg N ha^{-1} year−¹ for open bulk deposition and total deposition in oak forest and cedar forest, respectively. The deposition of ammonium nitrogen on leaf surface were 0.29 ± 0.38 (average \pm standard deviation) and $0.75\pm$ 1.06 mg N m^{-2} for oak and cedar, respectively, and that of nitrate nitrogen were 0.15 ± 0.18 and $0.35 \pm$ 0.28 mg N m⁻², respectively. Dry deposition on leaf

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surface was influenced by the cumulative amount of rainfall before sampling and was found to be easily washed off with high amount of precipitation within a few days.

Keywords Dry deposition \cdot Foliar rinsing method \cdot Leaf surface . Forest . Nitrogen

1 Introduction

Dry deposition is the transfer of chemical species in the atmosphere onto the earth surfaces in the form of gases and particles whereas wet deposition is the transfer of the chemicals by rain and snow in the form of aqueous solution. Dry deposition, as well as wet deposition, is a major mechanism of atmospheric nitrogen (N) input to the forest canopy (Lindberg et al. [1986](#page-8-0)). Due to its three-dimensional structure, forest canopies serve as a very effective receptor for airborne substances. Surface properties greatly influence the rates of dry deposition of particles and gases, both directly through chemical reactions with the surface, and indirectly through perturbation of the quasi-laminar adjacent to the surface (Seinfeld and Pandis [1998](#page-8-0)).

For the evaluation of dry deposition on leaf surfaces, foliar rinsing technique is prevailing (e.g., Hanson and Lindberg [1991](#page-8-0); Duyzer and Fowler [1994](#page-8-0)). It directly removes accumulated dry deposits on the foliage (Hanson and Lindberg [1991](#page-8-0)). Various

methods of foliar rinsing have been used so far for both laboratory and field experiments. Usually, leaves are washed with deionized water before exposure to open air (Dasch [1987](#page-8-0)) or enriched air with aerosols in closed chambers (Marshall and Cadle [1989](#page-8-0)) to calculate the deposition velocity. Most studies have been dealing with seedlings since higher positioned leaves are difficult to access and difficult to expose to aerosols. Washing the surface by deionized water could recover more than 95% of the N deposition on leaf surfaces exposed to environment enriched with HNO3 for 2–4 h (Dolske [1988](#page-8-0); Marshall and Cadle [1989](#page-8-0); Cadle et al. [1991](#page-8-0)). Whereas leaves are just rinsed with deionized water in most cases, sample leaves were put into deionized water continuously agitated by a magnetic stirrer by the study of Dolske [\(1988](#page-8-0)). In that case, the extracted nitrate nitrogen $(NO₃⁻-N)$ concentration reached a plateau after 30 min stirring. Leaching from leaves could be excluded in the case of $NO₃⁻-N$ (Dolske [1988](#page-8-0)); however, uptake of $NO₃⁻-N$ from leaf surface was found by $15N$ tracing as a rate of 35% to 68% of the applied NO_3^- -N to pine and spruce (Cadle et al. [1991](#page-8-0)). The amount of uptake from the surface increased as the amount of application increased for spruce and pine. The duration of exposure also increases the amount of cumulated deposition and thus to increase the amount of $NO₃⁻-N$ uptake. Even though detail foliar rinsing studies have been conducted under controlled condition, there are only few reports on in situ seasonal change of deposition (Chiwa et al. [2003](#page-8-0)). For artificial rinsing, monitoring stands (or towers) have to be established in the forest, which in turn limits the sampling site, since deposition may vary with the height of the leaf, distances from the forest edge, or slope direction (Weathers et al. [2006](#page-8-0)). Surrogate surfaces does not fully represent the structure and chemical composition of the leaf surface, nor is the measured amount of dry deposition the result of leaching and uptake processes of leaf, even though it can be placed at any places, and is a good indicator of dry deposition (Raymond et al. [2004](#page-8-0)). A more flexible and simple method is needed to monitor the actual dry deposition onto forest ecosystem.

The objective of this study is to examine a simple foliar rinsing method to estimate N dry deposition on forest ecosystem with special attention on different tree species—oak (Quercus serrata Thunb.) and cedar (Cryptomeria japonica D. Don)—and further to explore the influence of rainfalls on the N dry deposition. In order to clarify the amount of N dry deposition accumulating during the dry period, the influence of rain events on the surface compounds was analyzed. A modified foliar rinsing method was used and compared to the conventional throughfall method.

2 Study Site

The study area was the Field Museum Tamakyuryo, a field experimental station of Tokyo University of Agriculture and Technology. The station is located at the northeast edge of the Tama Rolling Hills, which spread to the southeast of the Kanto plane, which includes Tokyo Metropolitan and neighboring prefectures and has the highest population density in Japan. Due to this location, a large amount of N deposition ranging from 15.3 to 31.8 kg N ha⁻¹ year⁻¹ has been observed here (Baba et al. [1995](#page-8-0)) and this area is reported as a forest of "N saturation". The FM Tamakyuryo is nominated as one of the Japan Long-Term Ecological Research Network (JaLTER) associated sites, whose mission is to provide appropriate scientific information and predictive understanding of ecological processes for scientific community, policy makers, and general societies through long-term ecological research in Japan (JaLTER Information Management Committee [2007](#page-8-0)). The most dominant tree types oak (Q. serrata Thunb.) and cedar (C. japonica D. Don) were chosen as representative tree types in this region.

3 Materials and Methods

Daily precipitation and air temperature were monitored at the field station from May 25, 2005 to December 7, 2006. Leaves from oak and cedar were sampled from several trees at 3 and 5 m height of each tree type with a long-branch cutter. For cedar, only current-year foliage was used. Each foliage sample was placed in a plastic bag by using plastic gloves. Two replicates were taken at each height. Two grams of leaves were put in 50 ml ion exchanged water and then washed in an ultrasonic bath for 1 h. A preliminary test has shown that the amount of N found by leaf surface washing by ultrasonic bath was

equal to that from repeated washing (five times wash by 20 ml water each) of leaf surface. The slope of regression lines for the amount of N from ultrasonic extraction against that from repeated washing were 0.91 (p <0.05) and 1.02 (p <0.01) for the amount of NH_4^+ -N and NO_3^- -N, respectively.

The throughfall precipitation and stemflow was monitored at both forest sites. An open bulk deposition sampler was set outside the forest, around 200 m from the study site. The throughfall and bulk precipitation were collected using 200 cm^2 polyethylene funnels attached to 10 l polyethylene bottle. The top of the funnel had a margin of 10 cm height, which was surrounded by wooden sticks to prevent birds from sitting down. The top of the funnel was 1 m from the soil surface. Five-milliliter chloroform was added to the polyethylene bottle after each sampling to prevent biological activity in the sample.

All water samples were filtrated with 0.45-μm micropore filter and analyzed for NH_4^+ -N and NO_3^- -N. Ammonium ion was measured by the Indophenol blue method (BioSpec-1600, SHIMADZU) and the NO_3^- -N concentration was quantified by the capillary electrophoresis method (Capi3300, Otsukadennshi). Leaf area was analyzed by scanning the spread leaves. The image was analyzed by Adobe Photoshop 6.0. Since the leaf profile of the cedar has a trapezoid shape and are arranged to the stem in a three-dimensional shape, the error of the scanning was revised using a factor of 1.33. This factor was created by multiplying 1.25, which is the ratio of the surface length of the trapezoid to the diameter of the leaf using profile slices of the leaf $(n=5)$, and 1.06, which is the ratio of the area of leaflets scanned separately displayed on the scanner to the original leaf area $(n=5)$.

One-way analysis of variance was conducted to compare the averages (SigmaStat 3.11, Systat Software, Inc.). A factor analysis (ValiMax method) was conducted to analyze the ruling factors for the different methods of deposition analysis (MULTI2, SKSoft, Tokyo).

4 Results and Discussion

4.1 Ammonium Deposition

Stemflow and throughfall for oak and cedar both showed higher NH₄⁺-N concentration compared to

that of open bulk deposition (Fig. [1](#page-3-0)b, c). This indicates capturing of NH_4^+ -N by forest canopy as reported in many proceeding studies (Lindberg et al. [1986](#page-8-0); Chiwa et al. [2003](#page-8-0)). Stemflow and throughfall both showed peaks at the same sampling data. There was a positive correlation between the NH_4^+ -N concentration of stemflow and throughfall, which was significant for both oak ($r=0.727$, $p<0.001$) and cedar ($r=0.373$, $p<0.05$). The cumulative amount of atmospheric N deposition was 8.7, 8.7, and 24.2 kg N ha^{-1} year^{-1} for open bulk deposition and total deposition in oak and cedar forest, respectively. The precipitation within oak forest was 70% of the open bulk precipitation, while that of the cedar forest was 98% of the open bulk precipitation. Previous studies have shown that this area is characteristic for its high N deposition rate (Baba et al. [1995](#page-8-0)). The deposition rate of ammonia seems to be even higher than the study from 1990–1992, where it was 7.1, 11.4, and 19.5 kg N ha⁻¹ y⁻¹ for open bulk deposition and total deposition in oak and cedar forest, respectively. Since the deposition was only monitored at one point, these values should be considered with restriction, but it confirms that the area still obtains a high N input due to atmospheric deposition.

Since oak leaves fall down in winter, no oak leaves samples were available between November 18, 2005 and May 25, 2006 (Fig. [1](#page-3-0)d). Cedar leaves showed significantly higher (p <0.05) NH₄⁺-N deposition per leaf area than oak throughout the year. Two episodes of large deposition for cedar were found on April 20 and June 27, 2006, with a value of 4.0 and 4.3 mg N m⁻², respectively. The average NH_4^+ -N deposition on oak and cedar leaf surface was $0.29\pm$ 0.38 (average \pm standard deviation) and 0.75 \pm 1.06 mg N m−² , respectively. The deposition rate on leaves cannot be calculated, since deposition on leaves was not washed off to reset the deposition. The unit is not the same as for the throughfall method, but the foliar rinsing method also showed higher amount of deposition for cedar than oak. Comparing oak and cedar may give us an idea how differences in surface structure and chemical condition influence dry deposition on leave surface and the different sampling condition may clarify, how rain wash offs N deposition from leaf surface. It will be further discussed in Sections [4.3](#page-5-0) and [4.4](#page-6-0).

Fig. 1 a Weekly precipitation, ammonium concentration of open bulk deposition, stemflow, throughfall in b oak forest, c cedar forest, and d ammonium deposition on oak and cedar leaves

4.2 Nitrate Deposition

Nitrate concentration in stemflow and throughfall was also significantly higher $(p<0.01)$ than that in open bulk deposition for both oak and cedar (Fig. [2](#page-4-0)b, c). The stemflow of oak had significantly higher $(p<$ 0.01) NO_3^- -N concentration than throughfall of oak, while there was no significant difference between $NO₃⁻-N$ concentration in stemflow and throughfall of cedar. This finding indicates that leaching from leaves and stem (Lovett and Lindberg [1984](#page-8-0)) had occurred especially in stem of oak. Like NH_4^+ -N, a significant positive correlation was found between the $NO₃⁻-N$ concentration of stemflow and throughfall for both oak $(r=0.533, p<0.01)$ and cedar $(r=0.689, p<$ 0.001). The cumulative amount of atmospheric N deposition was 11.7, 11.9, and 32.5 kg N ha⁻¹ year⁻¹ for open bulk deposition and total deposition in oak and cedar forest, respectively. As described before, this study site is known to get high amount of N due to deposition (Baba et al. [1995](#page-8-0)). While there was no difference between the amount of NH_4^+ -N and NO₃⁻-N deposition by Baba et al. [\(1995](#page-8-0)), Ueno and Komine [\(2005](#page-8-0)) reported high amount of NO_3 ⁻-N dry deposition in Tokyo region, which accounted for 7.6– 34% of the wet deposition in 2005. In this study, the amount of $NO₃⁻-N$ deposition was larger than NH4 + -N for deposition in open bulk deposition, and Fig. 2 a Weekly precipitation, nitrate concentration of open bulk deposition, stemflow, throughfall in b oak forest, c cedar forest, and d nitrate deposition on oak and cedar leaf

total deposition in oak and cedar forest. Nitrate deposition in cedar forest was found to have a higher value than open area and oak forest. It might be attributed to the efficiency of capturing dry deposition on the leaf surface as described in Sections [4.3](#page-5-0) and [4.4](#page-6-0). Similar to NH_4^+ -N deposition, the amount was higher compared to the amount in 1990–1992; 8.1, 9.2, and 16.4 kg N ha^{-1} year⁻¹ for open bulk deposition and total deposition in oak and cedar forest, respectively (Baba et al. [1995](#page-8-0)).

Similar to NH_4^+ -N deposition, NO_3^- -N deposition on cedar leaves was significantly larger $(p<0.01)$ than that on oak leaves (Fig. 2d). No extremely large deposition was recorded for cedar unlike NH_4^+ -N deposition and all samples were lower than 1.5 mg N m⁻². Like NH_4^+ -N deposition, higher NO₃⁻-N deposition was found for cedar than oak. The average $NO₃⁻-N$ deposition on oak and cedar leaf surface was 0.15 ± 0.18 (average \pm standard deviation) and $0.35\pm$ 0.28 mg N m⁻², respectively. The average NO_3 ⁻-N deposition on leaf surface was less than NH_4^+ -N deposition for both oak and cedar. However, while the proportion of $NO₃⁻-N$ deposition was same or higher than NH₄⁺-N deposition using the throughfall method, $NO₃⁻-N$ deposition was lower than $NH₄⁺-N$ deposition in case of the foliar rinsing method. The apparent accumulation of NH_4^+ -N deposition is higher than NO_3 ⁻-N on leaf surface.

4.3 Influence of Precipitation on Leaf Surface Deposition

Due to the high seasonal variability of dry deposition found on leaf surface, the amount should not only be

Fig. 3 Relation of ammonium and nitrate nitrogen deposition on oak and cedar leaf surfaces and the cumulative amount of precipitation during the following intervals before sampling: 1 $(a, b), 2 (c, d), 3 (e, f),$ and 7 days (g, h)

discussed with the average value but analyzed in relation to the climate condition. Chiwa et al. [\(2003](#page-8-0)) analyzed the relation of the numbers of days without rain event to the amount of deposition on leaf surfaces. Positive linear relationships were found

between the amount of deposition on unit surface area of pine foliage and the duration of nonprecipitation for all the ions except for potassium and hydrogen (Chiwa et al. [2003](#page-8-0)). In the present study, the cumulative amount of precipitation in terms of the number of days before sampling was compared to the amount of NH_4^+ -N and NO_3^- -N deposition on the leaf surface. Both N compounds of both tree species showed low deposition values as there were more than 30 mm rain 1 day before sampling (Fig. [3](#page-5-0)a, b). NH_4^+ -N seems to be more sensitive to the amount of precipitation than NO_3^- -N for both species, while it accumulated much more than $NO₃⁻-N$ on the leaf surface if there is no rain. The relation became weaker as the amount of precipitation increased according to increased number of days before sampling (Fig. [3](#page-5-0)c–h). As the precipitation within 7 days before sampling were compared, high deposition were still found at low cumulative precipitation; however, for high amount of precipitation, low deposition were found for both N compounds for both tree species. Cedar showed higher deposition amounts per unit leaf surface area than oak under the same amount of rain, indicating that N deposition on cedar leaves remains for a longer time on the leaf surface and are less easily washed off than those on oak leaves. These results confirm that dry deposits accumulate on the pine foliage during the duration of nonprecipitation (McColl and Bush [1978](#page-8-0); Lovett and Lindberg [1984](#page-8-0); Puckett [1990](#page-8-0); Chiwa et al. [2003](#page-8-0)). The amount of dry deposition remaining on the leaf surface could be explained by the cumulative amount of rain before sampling. The analysis showed that dry deposition on leaf surface is easily washed off with high amount of precipitation especially within 3 days before sampling.

Ammonium and $NO₃⁻-N$ deposition on leaf surface was higher for cedar than oak (Fig. 4). Significant correlation was found for NO_3 ⁻-N (y=1.31x+ 0.17, $R^2 = 0.66$, $p < 0.001$, whereas y stands for cedar, x for oak). If the high NH_4^+ -N deposition on the June 27, 2006 was excluded (statistical analysis based on Thomson's method), the relation between NH_4^+ -N deposition of oak and cedar was also significantly related ($y=1.09x+0.20$, $R^2=0.47$, $p<0.001$, whereas y stands for cedar, x for oak). Dry depositions on oak surface are more easily washed off than those on cedar leaves. The pockets formed by the overlapping of the leaflets from the cedar might be also one reason for the efficient deposition on cedar leaf surface. The

Fig. 4 Relation of ammonium and nitrate deposition on leaf surface of oak and cedar. Nitrate: $y=1.318x+0.17$ ($R^2=0.66$); ammonium: $y=1.09x+0.20$ ($R^2=0.47$). Outlier of NH4 deposition marked in the bracket was excluded by Thomson's method

difference between cedar and oak is mainly found for $NO₃⁻-N$, which is more difficult to be washed off by rain. These results indicate that not only the threedimensional feature of the cedar trees (Lindberg et al. [1986](#page-8-0)) but also the higher amount of N deposition per unit leaf area was the reason for the higher deposition to cedar trees than to oak trees.

4.4 Relation of Leaf Surface Deposition, Throughfall Concentration, and Total Deposition per Land Area

Even the unit and analysis condition is different for foliar rinsing method and throughfall method, the influencing factors might be common for both methods and analyzing the ruling factors would explain the deposition mechanism. Thus, a factor analysis (ValiMax method) was conducted to analyze the relation among the amount of precipitation, deposition of NH_4^+ -N, and NO_3^- -N on leaf surface, NH_4^+ -N and NO_3^- -N concentration of open bulk deposition and throughfall, and amount of NH_4^+ -N and NO_3^- -N deposition per unit area of land (Table [1](#page-7-0)).

Ammonium N and $NO₃⁻-N$ on foliar surface was both influenced by the first factor loading for both oak and cedar. Ammonium N tended to be more

strongly correlated to this factor than $NO₃⁻-N$. An interesting finding was that the concentration as well as the amount of NH_4^+ -N of open bulk deposition was also strongly influenced by the first factor. Increase in deposition of foliar leaves indicate longer dry period as found by Fig. [3](#page-5-0). Ammonia volatilization might be enhanced by increase in solar radiation for the period (Sommer and Hutchings [2001](#page-8-0)) and its concentration in rain water is increased during this dry period, which led also to the higher amount of NH_4^+ -N deposition on the open bulk deposition. The second factor clearly described the influence precipitation amount before sampling on the amount of N deposition for open bulk deposition and total deposition in oak and cedar forest. Deposition of $NO₃⁻-N$ for open bulk deposition and total deposition in oak and cedar forest was more strongly influenced by the amount of rain before sampling than NH_4^+ -N. This finding is in agreement with the results of Fig. [3](#page-5-0), where NO_3 ⁻-N was found more difficult to be washed off than NH_4^+ -N. The amount of NO_3^- -N washed off by rains will increase more as the amount of rain increase than NH_4^+ -N, which will be washed off with little amount. The third factor influenced mainly the concentration of N in open bulk samples and throughfalls. There is no significant relation between this factor and the precipitation. Increase in N concentration in open bulk deposition and throughfalls in forests is influenced by a more complex factor than the mere amount or timing of precipitation. However, it can be said that both NH_4^+ -N and NO_3^- -N concentrations in open bulk deposition and throughfalls in forests are driven by the same factor.

5 Conclusion

The present study examined a simple method to investigate dry deposition on leaf surface. The prerinsing was not conducted as in previous study, which enabled the analysis of the influence of precipitation events and the amount on the N deposition on leaf surface. The different amount of precipitation from 1 to 7 days before sampling influenced the amount of deposition on leaf surface of oak and cedar. Comparing oak and cedar showed that amount of deposition on leaf surface as well as throughfall deposition was higher for cedar than oak.

Table 1 Results of factor analysis of precipitation, NH_4^+ -N and NO_3^- -N concentration in throughfall and amount of NH_4^+ -N and $NO₃⁻-N$ deposition per leaf and land area

			1st factor	2nd factor	3rd factor
Precipitation (mm)	1 day before sampling		-0.209	$0.838***$	0.037
	2 days before sampling		-0.010	$0.671***$	-0.018
	Total amount during sampling times		0.014	0.087	0.259
Foliar deposition (mg N m ⁻²)	NH ₄	Oak	$0.933***$	-0.065	-0.066
		Cedar	$0.550**$	0.072	-0.127
	NO ₃	Oak	$0.682***$	-0.123	0.012
		Cedar	$0.546**$	-0.143	0.119
Concentration (mg $N l^{-1}$)	NH ₄	Open bulk	$0.952***$	-0.066	0.164
		Throughfall of oak	-0.081	0.260	$0.421*$
		Throughfall of cedar	0.057	0.063	$0.413*$
	NO ₃	Open bulk	-0.016	0.121	$0.941***$
		Throughfall of oak	-0.146	-0.074	$0.922***$
		Throughfall of cedar	-0.211	-0.050	$0.937***$
Amount (mg N ha^{-1})	NH ₄	Open bulk	$0.912***$	0.078	-0.233
		Oak forest	-0.113	$0.646***$	0.115
		Cedar forest	0.299	0.233	0.076
	NO ₃	Open bulk	0.015	$0.833***$	-0.057
		Oak forest	-0.126	$0.632**$	0.158
		Cedar forest	-0.111	$0.776***$	-0.143

 $*_{p}=0.1;$ $*_{p}=0.05;$ $*_{p}=0.01$

The foliar rinsing method clarified that the higher amount of N deposition on cedar forest compared to oak forest was due to the higher amount of N deposition per unit leaf area. Dry deposition on oak leaf surface was more easily washed off by rain than deposition on cedar leaves. For N species, $NO₃⁻-N$ was more easily washed off than NH_4^+ -N. The interactions of amount of precipitation duration of dry period are complicated. Further, application of the foliar rinsing method with special attention on the uptake and leaching mechanism on leaf surface would give more insights to the dry deposition on leaf surfaces.

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