大気降下物が針葉樹林に与ぼす影響

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Atmospheric Deposition and its Effects on Coniferous Forests

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Abstract

Chapter 1 General introduction

There have been many reports concerned about the possible effects of atmospheric deposition on forest decline in Japan as well as in Europe and United States. Japanese red pine (*Pinus. densiflora*) has been severely damaged especially since the 1970's in Hiroshima Prefecture. Mt. Gokurakuji (altitude 693 m), located at the coastal area of Seto Inland Sea, Hiroshima Prefecture was selected, where the mortality of P. *densiflora* at Mt. Gokurakuji was correlated with NO_2 concentration. However, atmospheric concentration of NO_2 is not enough to cause direct cause of pine damage. Therefore, NO_2-related and/or other air pollutants, including atmospheric deposition, are expected to be responsible for the pine decline at Mt. Gokurakuji.

In this thesis, atmospheric deposition, including gaseous air pollutants, was measured at several sites at Mt. Gokurakuji with respect to the areas of the Japanese red pine forest declines in order to investigate the involvements of atmospheric deposition with respect to pine decline. In addition, throughfall chemistry was also studied with polluted mist treatment in spruce plantation in Scotland in order to investigate the canopy interactions with atmospheric deposition.

Chapter 2 Concentrations of SO_2, NO_x and O_3 and their diurnal variation at Mt.Gokurakuji and its vicinity, Hiroshima Prefecture, western Japan

Measurements of sulfur dioxide (SO_2), nitrogen oxides (NO_x), ozone (O_3), and meteorological parameters like wind speeds/directions were carried out at 4 sites of Mt. Gokurakuji in order to investigate the distribution patterns and diurnal variations of air pollutants with respect to areas of the Japanese red pine forest declines. SO_2 concentrations were low (less than 10 ppb) at every observation site. Higher concentrations of NO_x (80-330 ppb) were observed during the nighttime and early morning at the sites (altitude 120 m, 130 m) near the city

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area. On the other hand, NO_x concentration near the mountain summit of Mt. Gokurakuji was as high as 20 ppb at daytime and a few ppb at the nighttime and showed the different patterns of diurnal variation from those at the sites near the city area. Concentration of O_3 at the summit was lower during north wind and increased during south wind, which suggests that photochemically produced O_3 was transported from the city area in which NO_x was emitted. Emission rates of NO_x and the number of vehicles at the surrounding cities of Mt. Gokurakuji have been increased by a factor of 2.5 and 2.0, respectively from 1980 to 1996. Furthermore, annual concentrations of NO_x at the surrounding cities have been increasing slightly from 1985 and O_x concentrations have also been increasing every year.

Chapter 3 Rainfall, stemflow, and throughfall chemistry at declined and non-declined areas of Japanese red pine (*Pinus densiflora*)

Rainfall, stemflow, and throughfall were collected from 1996 to 1999 at the seaward side, the mountain summit, and inland side of Mt. Gokurakuji. Rainfall deposition showed small differences among three sites. The NO_3^- and SO_4^{2-} concentrations in stemflow, however, were higher at seaward side (34.4 and 50.0 μ eq l^{-1}, respectively) than at inland side (4.8 and 28.8 μ eq l^{-1}, respectively), and were several times higher compared to those of rainfall collected along with stemflow. Furthermore, throughfall deposition of NO_3^- and SO_4^{2-} were also higher at the seaward side, whereas deposition of rainfall collected simultaneously showed small differences between study sites. Net throughfall (NTF) deposition of NO_3^- and SO_4^{2-} accounted for 77 and 50 % of total throughfall deposition at seaward side, respectively, while 44 and 23 % at inland side, respectively. These results suggested that there are small spatial changes for fluxes of wet deposition, while dry deposition is higher at seaward side compared to at inland side at Mt. Gokurakuji. Inorganic N (NO_3^- + NH_4^+) deposition was estimated to be around 17-26 kg N ha^{-1} yr^{-1} at seaward side, which was greater than the threshold of nitrogen deposition that could cause nitrogen leaching in Europe and United States. It was suggested that high N deposition may affect the decline of pine forests at the seaward side of Mt. Gokurakuji.

Chapter 4 Dry deposition washoff and dew on the surfaces of pine foliage at Mt. Gokurakuji, western Japan

Dry deposition and dew components on the pine needle were measured during 1998 to 2000 in seaward and inland side of Mt. Gokurakuji. Leaf wash experiment was employed to determine the dry deposition rates on the pine foliage and dry deposition fluxes to the forest floor. Nitrate (NO_3^-) and sulfate (SO_4^{2-}) deposition rates were 1.47 and 0.28 μ mol m-2h-1 respectively in the seaward side of Mt. Gokurakuji, declined area of pine and 0.32 and 0.09 μ mol $m^{-2}h^{-1}$ in inland side, non-declined area of pine. Dry deposition fluxes of inorganic N (NO_3^- + NH_4^+) and sulfate (SO_4^{2-}) to forest floor were 8.4, kg N ha^{-1} yr^{-1} and 2.8 kg S ha^{-1} yr^{-1} in seaward, and were 3.3 kg N ha^{-1} yr^{-1}, 1.8 kg S ha^{-1} yr^{-1} in inland side respectively. Higher dry deposition rants and fluxes of N and S in seaward site could be attributed to the proximity to the urban area. Dew concentration showed higher in seaward than in inland side for most of the ions. NO_3^- and SO_4^{2-} concentrations in dew at the seaward side were 802 and 428 μ eq.l^{-1}, respectively, while at inland side they were 199 and 222 μ eq.l^{-1}, respectively, suggesting that higher dry deposition rates in seaward side enhanced their concentrations in dew in seaward side. The mean pH for the dew showed a small difference between two sites (4.6 in seaward and 4.8 in inland), which may be

explained by cations leaching from pine needles.

Chapter 5 Throughfall chemistry in a sitka spruce plantation in response to six different simulated polluted mist treatments

A Sitka spruce plantation, planted in 1986 on a drained deep peat, has been exposed to 6 different simulated mist treatments in 4 replicated blocks since 1996. Treatments provided N and/or S at a concentration of 1.6 mol m^{-3} [A: NS Acid (NH_4NO_3 + H_2SO_4 at pH 2.5), B: S only (Na_2SO_4), C: 2NS Acid (double dose by application at twice frequency), D: N only (NH_4NO_3), E: control (additional rainwater only), F: no treatment]. Throughfall collected at all the replicate plots in 2000, was analyzed for all major ions (Cl^-, NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, H^+).

Canopy interactions and dry deposition through the canopy were calculated by subtracting rainfall deposition and additional mist deposition from throughfall deposition under the assumption that SO_4^{2-} was not retained by the canopy, but acted as a conservative tracer. The calculation confirmed that thirty to thirty five percent of the applied N was retained by the canopy. Acidity in the applied mist was partly neutralized by the canopy. Linear relationships between the loss H^+ and the increased K^+, Mg^{2+} and Ca^{2+} deposition in throughfall were observed. However these increases in K^+, Mg^{2+} and Ca^{2+} deposition accounted for only about 50 % of total neutralization of the acidity. The involvements of organic anions were suggested for the neutralization of the acidity in throughfall.

Chapter 6 General discussion and conclusion

N deposition at the seaward side of Mt. Gokurakuji was in relatively higher level compared to that of Europe, United States, and Japan. Furthermore those were in the range of the higher level among several sites in Hiroshima Prefecture. Atmospheric N deposition at Mt. Gokurakuji is expected to be more than throughfall N deposition, because nitrogen is retained by the canopy. Considering the sensitivity of Japanese red pine trees to high N loads, high atmospheric N deposition, including dry deposition, was possible to explain the recent decline of pine trees (P. *densiflora*) in the coastal area of Seto Inland Sea in Hiroshima Prefecture. The inducement of the decline by high atmospheric N deposition was (1) high nitrogen load in soil and (2) the source of OH radical on the surface of the pine needle that induces the lower photosynthesis of Japanese red pine tree. It can be concluded that other factors like severe drought and insect cause the dieback of pine trees in combination with excess N deposition and OH radical photoformation.