Title

Risk assessment of ozone impact on *Fagus crenata* in Japan: Consideration of atmospheric nitrogen deposition

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Abstract

Tropospheric ozone (O_3) is considered to be the air pollutiant relating to the decline of *Fagus crenata* forest in Japan. In the present study, we assessed a risk of O_3 impact on the growth of *F. crenata* in Japan, giving consideration to the effects associated with atmospheric nitrogen (N) deposition based on the experimental study, national monitoring data for oxidant concentration and atmospheric N deposition, and a national vegetation survey. The average and maximum O_3-induced relative growth reduction (RG_{red}) of *F. crenata* across Japan were estimated to be 3.2% and 9.7%, respectively. Current levels of atmospheric N deposition were found to significantly affect the sensitivity of *F. crenata* to O_3. When the N deposition was assumed as zero, the estimated average and maximum RG_{red} were 2.3% and 5.7%, respectively. The inclusion of atmospheric N deposition data thus increased the estimated values for average and maximum RG_{red} (by 38% and 71%, respectively). Our results demonstrate that a change in the sensitivity to O_3 associated with atmospheric N deposition is an important consideration in the risk assessment of O_3 impact on the growth of *F. crenata* in Japan.

Keywords

Ozone, Nitrogen deposition, Risk assessment, Fagus crenata, Growth reduction

1. Introduction

Tropospheric ozone (O_3) is recognized as a widespread phytotoxic gaseous air pollutant, and the concentration has been increasing in the Northern Hemisphere (Akimoto 2003; Matyssek and Sandermann 2003; ADORC 2006). In Japan, relatively high concentrations of O_3 (above 100 nmol mol^{-1}) have been frequently recorded not only in the suburbs of big cities such as Tokyo and Osaka, but also in several mountainous areas (Wakamatsu et al. 1998; Yoshikado 2004; Network Center for EANET 2007; Takeda and Aihara 2007). In Europe, the risk assessment of O_3 impact on forest tree species has been conducted based on the concept of critical level, which was evaluated from experimental studies (Kärenlampi and Skärby 1996; Mills 2004; Simpson et al. 2007). On the other hand, although the current levels of O_3 in Japan could adversely affect forest tree species, risk assessments of O_3 impact were limited (Kohno et al. 2005; Watanabe et al. 2010).

The atmospheric deposition of nitrogen (N) to terrestrial ecosystems has been increasing in line with elevated anthropogenic emissions of N (Ohara et al. 2007; Galloway et al. 2008). Several researchers reported relatively high amount of N deposition at 10 to 20 kg N ha^{-1} $year^{-1}$ by wet N deposition (bulk precipitation) and 10 to 50 kg N ha^{-1} $year^{-1}$ by throughfall and stemflow in the forested areas of Japan (Baba and Okazaki, 1998; Baba et al., 2001; Okochi and Igawa, 2001; Sase et al. 2008; Kimura et al. 2009).

In East Asia, emission of air pollutants such as N compounds and precursors of O_3 has been increased rapidly since 1980s owing to the increased energy demands due to rapid economic growth, industrialization and urbanization (Ohara et al. 2007; International Energy Agency 2008). Several researchers have implicated transboundary air pollution (O_3 and NO_X) from East Asian countries other than Japan as contributing to the recent increases in the concentration of O_3 and atmospheric N deposition in Japan, especially in the areas along the Sea of Japan (Holloway et al. 2002; Ministry of the Environment 2004; Tanimoto et al. 2005; Yamaji et al. 2006; Han et al. 2007). In fact, the annual average daytime O_3 concentration has been increased at a rate of 0.27 nmol $mol^{–1}$ $year^{–1}$ between 1985 and 2007, whereas the emissions of precursors in Japan has been decreased during the same period (Ohara

2011). The median value of the annual wet N deposition (sum of NO_3^- and NH_4^+) at 20 monitoring stations throughout Japan has been increased at a rate of 0.11 kg ha^{-1} year^{-1} between 1991 and 2007 (Ministry of the Environment 2009). Furthermore, continuous increase in the emission of air pollutants in the near future are predicted in East Asian countries (Ohara et al. 2007; International Energy Agency 2008). Therefore, tropospheric O_3 concentration and atmospheric N deposition in Japan will increase through the transboundary air pollution if emissions of air pollutants will not be strictly controlled (Yamaji et al. 2008).

Since the increases in the O_3 concentration and atmospheric N deposition are spatially correlated in general (Holland et al. 1997; Ollinger et al. 2002), we should consider interactive effects of atmospheric N deposition and O_3 on forest tree species. There have been several experimental studies on the combined effects of O_3 and the supply of N to the soil on the growth of various tree species. Utriainen and Holopainen (2001b) and Yamaguchi et al. (2007) reported that the supply of N to the soil increased the growth sensitivity of *Pinus sylvestris* and *Fagus crenata* seedlings to O_3, respectively. However, opposite responses were reported for seedlings of *Larix kaempferi* and *Populus tremula* × *Populus tremuloides* (Watanabe et al. 2006; Häikiö et al. 2007). The effect of N supply on the sensitivity to O_3 has not found to be significant for *Picea abies* seedlings (Utriainen and Holopainen 2001a; Thomas et al. 2005). These results indicate that atmospheric N deposition must be taken into account when we conduct a risk assessment of O_3 impact on forest tree species whose sensitivity to O_3 is affected by changes in the supply of N to the soil.

Fagus crenata is the most common and widely distributed deciduous broad-leaved tree species in the cool temperate forests of Japan (Nakashizuka and Iida 1995). *F. crenata* is an important tree species in Japan because its forests help to conserve forest soil and to maintain biodiversity, and is planted for afforestation as well as for ceremonial plantations (Murai et al. 1991; Nakashizuka 2004; Terazawa and Koyama 2008). Virgin natural forests of *F. crenata* on the Shirakami Mountains (northeast Japan) were registered by UNESCO as a World Natural Heritage sites in December 1993. Several researchers have implicated O_3 as an important factor in the decline and dieback of *F. crenata* forests in Japan (Yonekura et al. 2001; Takeda and Aihara 2007; Kume et al. 2009). As discussed, the growth sensitivity of *F. crenata* seedling to O_3 was found to be directly related to increases in the supply of N to the soil (Yamaguchi et al. 2007; Yamaguchi et al. 2010a). Therefore, the increase in the atmospheric N deposition in Japan may correspondingly increase the sensitivity of *F. crenata* to O_3, thereby negatively affecting the growth of this tree species. However, a risk assessment of O_3 impact on the growth of *F. crenata* in which atmospheric N deposition-induced changes in the sensitivity to O_3 are considered has not been conducted. We addressed this in the present study, assessing the risk of O_3 impact on the growth of *F. crenata* in Japan, giving consideration to the effects associated with atmospheric N deposition based on the experimental study, national monitoring data for oxidant concentration and atmospheric N deposition, and a national vegetation survey.

2. Methods

2.1 The estimation of O_3-induced growth reduction of *Fagus crenata*

Our methods for estimating the effects of O_3 on the growth of *F. crenata* were based on the results of Yamaguchi et al. (2007). In this study, the seedlings of *F. crenata* were grown under 12 experimental treatment conditions, as determined by the combination of 4 gas treatments (charcoal-filtered air and 3 levels of O_3 at 1.0, 1.5 and 2.0 times the ambient concentration) and 3 soil N treatments with NH_4NO_3 [0 (N0), 20 (N20) and 50 kg N ha^{-1} year^{-1} (N50)] in open-top chambers during the 2 growing seasons. The whole-plant dry mass increment for a single growing season (WDM_{inc}) was calculated as the difference in the whole-plant dry mass of the seedlings between the ends of the first and second growing seasons. We measured the atmospheric concentration of O_3 in the open-top chambers during the second growing season to calculate AOT40 (accumulated exposure over a threshold of 40 nmol mol^{-1}, in μmol mol^{-1} h) of O_3 over 12 h periods (0600–1800 hours) between April and September. The AOT40 is the sum of the differences between the hourly mean O_3 concentration and 40 nmol mol^{-1} for each hour when the O_3 concentration exceeded 40 nmol mol^{-1} (Kärenlampi and Skärby 1996)

The analysis of the O_3 exposure-response relationships for the WDM_{inc} was performed according to Watanabe et al. (2007) as described below. A regression line was obtained from the relationship between AOT40 and the WDM_{inc}. The theoretical WDM_{inc} at zero AOT40 was determined to be the y-axis intercept of the regression line, and was used as a reference (100%) to calculate the relative WDM_{inc} for each gas treatment. The slope and coefficient of determination values (R^2) were calculated from the regression line between AOT40 and the relative WDM_{inc}. Because the sensitivity to O_3 of *F. crenata* is reported to increase with an increase in N supply to the soil (Yamaguchi et al. 2007), this procedure was conducted separately for each N treatment. We regarded the absolute value of slope in the regression line as the O_3-induecd relative growth reduction (RG_{red}, %) per unit AOT40 for each N treatment. The relationship between the amount of N supply and the RG_{red} per unit AOT40 was analysed to estimate the RG_{red} per unit AOT40 in the area with different depositions of N. We calculated the RG_{red} in each *F. crenata* habitat as a product of

RG_{red} per unit AOT40 and AOT40.

2.2 Estimation of AOT40 of O_3 in Japan

The concentrations of photochemical oxidants are officially monitored at approximately 1200 monitoring stations throughout Japan. Originally, photochemical oxidants have been measured by absorption spectrophotometry using a neutral potassium iodide solution (AS-NPI). The atmospheric concentration of O_3 can be tabulated as that of photochemical oxidants under the Air Pollution Control Law Enforcement Regulations in Japan from 1996, because of following reasons: a) the concentration of peroxi-acetyl nitrate (PAN), main component of photochemical oxidant without O_3, was very low and b) the sensitivity of AS-NPI to PAN concentration was low (Ministry of the Environment 1996). In fact, the 1-year field measurement indicated that the little difference between the concentrations of O_3 measured by UV absorption photometry and chemiluminescence method and that of photochemical oxidant measured by AS-NPI (Ministry of the Environment 1996). In the present study, therefore, the concentration of photochemical oxidants was regarded as that of O_3.

The number of hours in which the concentration of O_3 is above either 0.06 μmol mol^{-1} (Num_{60}) or 0.12 μmol mol^{-1} (Num_{120}) is recorded by all of the monitoring stations in Japan and made available by the National Institute for Environmental Studies. However, hourly data concerning O_3 concentrations were available in approximately 40% of prefectures. Ishii et al. (2007) reported a high correlation (r = 0.97) between the sum of Num_{60} and Num_{120} and the AOT40 over 12 h periods (0600–1800 hours) based on the monthly data between April and September, as calculated from available hourly O_3 concentration data. Therefore, we use the method of Ishii et al. (2007) to estimate the AOT40 for all of the monitoring stations in Japan from 1999 to 2001.

The map of spatial distribution of AOT40 in Japan was created using the Geostatistical Analyst Extension of the ArcGIS 9.0 software (ESRI inc. USA). The kriging interpolation was applied for the estimation of AOT40 among the monitoring stations. The cell size in the kriging interpolation was set as 0.05º. The Gaussian model was used as a semivariogram model in the kriging interpolation because the kriging variance was lower than that of the other semivariogram models (Spherical, Circular, Exponential and Linear).

2.3 Estimation of atmospheric N deposition in Japan

In general, atmospheric N deposition is classified into wet deposition and dry deposition. We obtained wet deposition data for NO_3^- and NH_4^+ from the Ministry of the Environment and Environmental Laboratories Association of Japan, which had monitoring station numbers of 97, 98 and 98 in 1999, 2000 and 2001, respectively (Environmental Laboratories Association 2003; Ministry of the Environment 2004). The distribution of wet deposition of N in Japan was estimated from these data. Flux of wet deposition of N (F_w) was calculated from the concentration of N in precipitation (C_p) and the amount of precipitation (P) as follows:

$$F_w = C_p \cdot P. \tag{1}$$

Our method for estimating the dry deposition of N was based on that of Fujita (2004). The flux of dry deposition of N (F_d) was calculated as the product of dry deposition velocity (V_d) and atmospheric N concentration (C_a):

$$F_d = V_d \cdot C_a \,. \tag{2}$$

Puxbaum and Gregori (1998) reported the V_d of N compounds for several forests based on inferential models, and the average values across these forests were used in our calculations. The average V_d of gaseous HNO_3, gaseous NH_3, and particulate matter with NO_3^- and NH_4^+ were 2.72, 0.74, and 0.18 cm s^{-1}, respectively. C_p was considered as proportional to C_a, and Formula 1 can be rewritten as follows:

$$F_w = K \cdot C_a \cdot P \tag{3}$$

where K is the ratio of C_p to C_a (washout ratio). From Formulas 2 and 3, F_d can be described follows:

$$F_d = V_d \cdot F_w/(K \cdot P). \tag{4}$$

The dry deposition flux of N can thus be estimated by determining K. C_a values for gaseous HNO_3 and NH_3, and for NO_3^- and NH_4^+ originating from particulate matter, were obtained alongside measurements of C_p values for NO_3^- and NH_4^+ made by the 24–27 monitoring stations of the Environmental Laboratories Association of Japan between 1999 and 2001 (Environmental Laboratories Association 2003). K values were estimated from these data as the ratio of C_p to the sum of C_a values for gaseous and particulate forms. Extremely high K values were observed at several monitoring stations. To avoid any spurious results potentially associated with their inclusion, we averaged K values across the frequency distribution from the 10th to 90th percentile. This resulted in K values that were averaged for each month (Fig. 1). For monitoring stations where C_a was not measured, its value was used. The commonly-used ratio of gaseous form C_a to particulate forms C_a (Gas/Particle) was estimated from data provided by the Environmental Laboratories Association (2003). Extremely high Gas/Particle values were observed at several monitoring stations, and, as for K values, we used Gas/Particle values that were averaged across the frequency distribution from the 10th to 90th percentile. The F_d for all monitoring stations was estimated using Formula 2, and in to which V_d from Puxbaum and Gregori (1998) and measured or estimated C_a values were entered. To avoid change in C_a and C_p associated with the eruption of the Miyake volcano in August 2000, data from this month were omitted from the analyses.

The inverse distance weighted (IDW) method was applied for the estimation of values of wet and dry deposition of N among the monitoring stations. Cell size in the IDW interpolation was set as 0.2º. Total N deposition (TN_{dep}) was calculated as sum of the wet and dry depositions of N in the GIS software.

2.4 Habitats of *Fagus crenata* in Japan

The habitats of *F. crenata* in Japan were determined from vegetation raster data (45" × 30" per mesh) of the National Survey on the Natural Environment, conducted by the Ministry of the Environment. These data were obtained from the Japan Integrated Biodiversity Information System (http://www.biodic.go.jp/J-IBIS.html). Geographical meshes containing the vegetation code for *F. crenata* were taken to be *F. crenata* habitats. Figure 2 shows the habitats of *F. crenata* across various geographical regions of Japan. The AOT40 and TN_{dep} for each *F. crenata* habitat were extracted from the above-mentioned AOT40 and TN_{dep} map and were used to the calculation.

3. Results

3.1 Estimation of RG_{red} per unit AOT40

Figure 3a shows the relationship between AOT40 and the relative WDM_{inc} of *F. crenata* seedlings. The slopes of the regression line were decreased with increasing the amount of N treatment. Although we found high values of the determination coefficient in the regression line for N20 and N50, that for N0 was low and not significant. The RG_{red} per unit AOT40 increased linearly with increases in N supply (Fig. 3b). Therefore, we calculated the RG_{red} per unit AOT40 with different TN_{dep} by the formula of a regression line. With differing AOT40 (μmol mol^{-1} h) and TN_{dep} (kg ha^{-1} $year^{-1}$), the RG_{red} for each habitat of *F. crenata* was calculated as follows:

$$RG_{red} = (0.0055 \cdot TN_{dep} + 0.230) \cdot \text{AOT40}$$

3.2 Estimations of distribution in AOT40 and nitrogen deposition

The highest AOT40 was estimated in the western part of the Kanto region (Fig. 4a). Relatively high AOT40 values were estimated not only for the areas along the Pacific Ocean where there are many big cities, and also for the areas along the Sea of Japan, including the northern parts of the Chubu and Chugoku regions. As shown in Fig 4b, relatively high TN_{dep} was estimated in the western parts of the Kanto and Chubu regions. The average TN_{dep} for Japan was 14.8 kg ha^{-1} $year^{-1}$ and average ratio of dry deposition to wet deposition was 0.88.

3.3 Estimated O_3-induced relative growth reduction of *Fagus crenata* in Japan

Relatively high RG_{red} values for *F. crenata* were estimated across a relatively wide area comprising the northern part of the Chubu region and the northwestern part of Kanto region (Fig. 5a). The estimated RG_{red} values for the western part of the Kanto region, the southern parts of the Chubu and Kinki regions, and the central part of the Chugoku region were also higher than for other areas. The average and maximum estimated RG_{red} values for Japan were 3.2% and 9.7%, respectively. As shown in Figure 6, the RG_{red} for *F. crenata* increased with increasing AOT40. However, there was a large variation in the RG_{red} across the range of AOT40 values, with the maximum value of the RG_{red} per unit AOT40 20–30% greater than the minimum. When the TN_{dep} was assumed to be zero, the average and maximum estimated RG_{red} values for Japan were 2.3% and 5.7%, respectively (Fig. 5b). Thus, the average and maximum estimated RG_{red} values were increased when atmospheric N deposition was considered (by 38% and 71%, respectively).

4. Discussion

For *F. crenata* habitats, areas with relatively high AOT40 of O_3 did not completely correspond to those with relatively high TN_{dep} (Fig. 7): Relatively high AOT40 was estimated even where the TN_{dep} was relatively low. This result differs from the previous reports by Holland et al. (1997) and Ollinger et al. (2002). The incomplete correspondence between TN_{dep} and AOT40 was not explained by differences in the accumulation period. As mentioned above, transboundary air pollution from East Asian countries is considered to be a significant factor that affects O_3 concentration and atmospheric N deposition in Japan (Holloway et al.

2002; Ministry of the Environment 2004; Tanimoto et al. 2005; Yamaji et al. 2006; Han et al. 2007). The contribution of transboundary air pollution to the AOT40 of O_3 in Japan is likely to be higher than that of NO_X to N deposition, because the life time of O_3 in the air is longer than that of NO_X. It is, therefore, possible that the incomplete correspondence between areas with relatively high TN_{dep} and those with relatively high AOT40 is a result of increases in the AOT40 associated with transboundary air pollution.

Increase in the estimated RG_{red} values associated with consideration of atmospheric N deposition was especially high (40–60%) for the relatively wide area comprising the northern part of the Chubu region and the northwestern part of Kanto region. In Europe, a critical level of O_3 for sensitive forest tree species, such as European beech and birch, was determined to be 5 μmol mol^{-1} h of daylight AOT40, value associated with a 5% reduction in seedling growth (Karlsson et al. 2004; Mills 2004). The present study found the ratio of *F. crenata* habitats with RG_{red} above 5% to all *F. crenata* habitats to be 0.3% when the TN_{dep} was assumed to be zero. However, this ratio increased to 16.9% when atmospheric N deposition was considered. These results suggest that atmospheric N deposition-induced changes in the sensitivity to O_3 must be taken into account in conducting a risk assessment of O_3 for *F. crenata* in Japan.

The increase in RG_{red} for *F. crenata* associated with an increase in the sensitivity to O_3 induced by atmospheric N deposition may be important in terms of competition with other tree species. Kume et al. (2009) reported that O_3 was an important contributor to the dieback of *F. crenata* in the mixed *F. crenata* and *Cryptomeria japonica* forest of the Toyama Prefecture in the northern part of the Chubu region (Fig. 2). Our risk assessment also indicates relatively high risk of O_3

impact in this area. Relatively high concentrations of O_3 were observed in this forest, while the concentrations of other air pollutant such as NO_2 and SO_2 were low (Kume et al. 2009). Although the total stem cross-sectional area at breast height of *C. japonica* increased, that of *F. crenata* decreased across 1999 to 2006 research period. As indicated by Kume et al. (2009), a difference in the sensitivity to O_3 of *F. crenata* and *C. japonica* would explain this phenomenon. Indeed, numerous experimental studies have shown that while *F. crenata* is relatively sensitive to O_3, *C. japonica* is tolerant (Izuta 2003; Kohno et al. 2005; Watanabe et al. 2006; Yamaguchi et al. 2007). On the other hand, the TN_{dep} in this area estimated by the present study is about 19 kg ha^{-1} year^{-1} (Fig. 4b). Because the supply of N to the soil stimulates growth of *C. japonica* and *F. crenata* to a similar extent (Watanabe et al. 2006; Yamaguchi et al. 2007), the effect of atmospheric N deposition on growth stimulation would not influence the competition between *F. crenata* and *C. japonica*. In contrast, the sensitivity of *C. japonica* to O_3 does not change with changes in the amount of N supplied to the soil (Watanabe et al. 2006). The estimated RG_{red} per unit AOT40 for *F. crenata* in this area was 45% higher than that with the TN_{dep} assumed to be zero. It is, therefore, possible that by inducing a change in the sensitivity to O_3, atmospheric N deposition in this region negatively affects the ability of *F. crenata* to compete with *C. japonica*.

The O_3 concentration and atmospheric N deposition in Japan has been increasing through the transboundary air pollution and this trends will continue in the near future (Ohara et al. 2007; Yamaji et al. 2008; Ohara et al. 2011). In fact, the trends of increase in the O_3 concentration and atmospheric N deposition in Japan have been observed after 1999-2001, which is the period that we assessed the risk of O_3 impact in the present study (Ministry of the Environment 2009). Therefore, the risk of O_3 impact on *F. crenata* in Japan at present time may be higher than that in the present study and will be serious by the increase in the O_3 concentration and atmospheric N deposition in the near future.

In the present study, we combined data from an experiment study, field monitoring and vegetation survey, and produced a strong evidence that atmospheric N deposition should be included in the risk assessment of O_3 impact on *F. crenata*. Because the present study is relatively extensive, we consider main uncertainties in the estimations of O_3 sensitivity of *F.* crenata, AOT40 of O_3 and atmospheric N deposition should be discussed. The relationship between AOT40 and WDM_{inc} in the N0 treatment was not significant because of small extent of O_3-induced reduction in WDM_{inc}, the limitation of the number of plot and relatively large variations (Fig. 3a). However, the value for the slope of the N0 treatment was reasonable to express the relationship between TN_{dep} and RG_{red} per unit AOT40 (Fig. 3b). Furthermore, when we used the data of each replication (i.e. n=12, Yamaguchi et al. (2007) applied 3 chamber replication), the relationship between TN_{dep} and RG_{red} per unit AOT40 in the N0 treatment was significant (p=0.027, data not shown). Yamaguchi et al. (2007) evaluated the effects of N supply to the soil on *F. crenata* seedlings. However, the actual N cycle in the forest ecosystem is more complex. For example, because plant can directly absorb the water from canopy leaves (Limm et al. 2009), the N deposition through fog and dew would be significant especially in the mountainous environment. There is a possibility that the effects of N uptake from leaves on O_3 sensitivity is different as compared to the process from soil owing to a skip of soil chemical process. The growth sensitivity of *F. crenata* to O_3 in the present study was evaluated by the experiment with seedlings in open-top chamber. There is a concern of differences in the sensitivities between seedlings and mature trees, and in the environmental conditions between open-top chamber and field (Karnosky et al. 2003; Matyssek et al. 2007). Therefore, we need further experimental studies to evaluate actual sensitivity of *F. crenata* in future for developing the risk assessment of O_3 impact in Japan. Meanwhile, Pretzsch et al. (2009) reported a comparable growth sensitivity of mature *Fagus sylvatica* trees to O_3 as compared to that of juvenile seedlings although the mechanism of O_3-induced growth reduction may differ. Variations such as the area of individual leaf and physiological traits have been reported among *F. crenata* genotypes in Japan (Hagiwara 1977, Koike 1998). The provenance of *F. crenata* seedlings in the experiment of Yamaguchi et al. (2010a) is Nagano prefecture, belonging to the clade that distributes in the widest area (Chubu, western part of Tohoku and Hokkaido regions) in Japan (Fujii et al. 2002). These genetic variations of *F. crenata* may have an uncertainty for risk assessment because Paludan-Müller et al. (1999) reported that the sensitivities to O_3 of *F. sylvatica* seedlings differed among the provenances in Europe. We did not have any information on the genetic variability in the sensitivities to O_3 among *F. crenata* genotypes. In the near future, therefore, the comparison of O_3 sensitivity among the *F. crenata* genotypes is needed to improve the quality of the risk assessment of O_3 impact. Monitoring stations for O_3 in Japan have been mainly located in the urban areas because the aim of monitoring is the protection of human health. There are limited number of monitoring station in the mountain and the rural areas. However, there are several phenomena that O_3 concentration in mountain and rural areas were higher than that in urban region (Yamaguchi et al. 2010b). Furthermore, atmospheric concentration of O_3 in mountainous areas sometimes show different diurnal variation as compared to urban areas (mainly flatland). Especially, little change in O_3 the concentration under inversion layer is typical phenomenon in mountainous areas.

This phenomenon make a concern when we estimate AOT40 based on the Num_{60} and Num_{120} according to Ishii et al. (2007). For example, two constant O_3 concentrations at 70 and 100 nmol mol^{-1} show the same Num_{60} and Num_{120}, but different AOT40. Reconsidering of the location of monitoring station for O_3 and availability of hourly data are needed for accurate assessment of O_3 impact on forest trees in Japan. At the present time, rutine method for measuring dry deposition of gases and particles has not yet established, while wet deposition continuously monitored throughout Japan. We needed the information on atmospheric dry deposition of N in large-scale because our aim is to clarify the extent of atmospheric N deposition-induced change in O_3 risk for *F. crenata* throughout Japan. In the present study, therefore, we applied simple method for estimating atmospheric dry deposition of several N compounds with the data obtained from nationwide researches and constant value of V_d (Puxbaum and Gregori 1998; Environmental Laboratories Association 2003; Fujita 2004; Ministry of the Environment 2004). As a results, we clarified the importance of the change in the sensitivity to O_3 associated with atmospheric N depsotion. In addition, similar amount of total dry N deposition as compared to that of total wet N deposition in the present estimation partly supports the validity of our estimation (Matsuda et al. 2001). The applied V_d values for gaseous HNO_3 and particulate matter with NO_3^- and NH_4^+ (2.72 and 0.18 cm s^{-1}, respectively) are relatively lower than those in other studies, whereas the V_d for gaseous NH_3 (0.74 cm s^{-1}) is similar (Hanson and Lindberg 1991; Endo et al. 2011). Therefore, there is a possibility that actual dry deposition of N is higher than our estimation. Although the estimation of parameters for dry deposition of N in Japan is difficult owing to the complex geography with monsoonal climate, the development of ideal method for estimating dry deposition of N that apply to nation wide scale is needed.

5. Conclusion

The results of the present study lead us to conclude that the current level of atmospheric N deposition-induced change in the sensitivity to O_3 is an important factor in the risk assessment of O_3 impact on *F. crenata* in Japan. The average and maximum estimated RG_{red} values increased when atmospheric N deposition was considered (by 38% and 71%, respectively). Increases in the estimated RG_{red} values were especially high, ranging from 40% to 60%, in the wide areas comprising the northern part of the Chubu region and the northwestern part of Kanto region.

As reported previously, there are several tree species for which changes in the supply of N to the soil alters their sensitivity to O_3 (Utriainen and Holopainen 2001b; Watanabe et al. 2006; Häikiö et al. 2007). For protecting these tree species, future risk assessments of O_3 impact must be conducted with consideration of atmospheric N deposition.

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Captions of figures

Fig. 1 Monthly variations of estimated *K* value (the ratio of concentration in the precipitation to that in the atmosphere) of NO_X and NH_Y. Each value is the mean of the data in 1999, 2000 and 2001, and the standard deviation is shown by vertical bars

Fig. 2 The habitats of *Fagus crenata* and classification of the regions in Japan

Fig. 3 Relationships between AOT40 and relative whole-plant dry mass increment (WDM_{inc}) (a) and between nitrogen supply and relative growth reduction per unit AOT40 (b) of *Fagus crenata* seedlings grown in the soil supplied nitrogen at 0 (N0), 20 (N20) and 50 kg ha^{-1} $year^{-1}$ (N50). The relative WDM_{inc} and AOT40 were recalculated from the results of Yamaguchi et al. (2007). Regression line of (b): y = $0.0055x + 0.230$; $R^2 = 0.967$

Fig. 4 The distribution of the estimated AOT40 of O_3 and annual deposition of the total nitrogen (TN_{dep}) in Japan. The AOT40 was accumulated during 0600–1800 hours from April to September and averaged across 1999 to 2001. The TN_{dep} was average across 1999 to 2001

Fig. 5 The distributions of O_3-induced relative growth reduction (RG_{red}) of *Fagus crenata* in Japan with consideration of nitrogen deposition (a) and without consideration of nitrogen deposition (b), which was estimated based on the RG_{red} per unit AOT40 at 0 kg ha^{-1} year^{-1} of annual deposition of the total nitrogen

Fig. 6 The relationship between the AOT40 and O_3-induced relative growth reduction (RG_{red}) of *Fagus crenata* in Japan

Fig. 7 The relationship between the total nitrogen deposition (TN_{dep}) and AOT40 in habitats of *Fagus crenata* in Japan

Figure 1

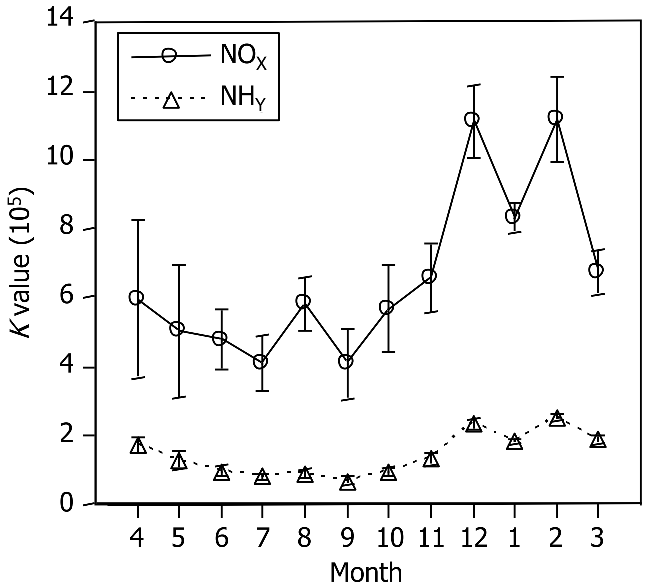
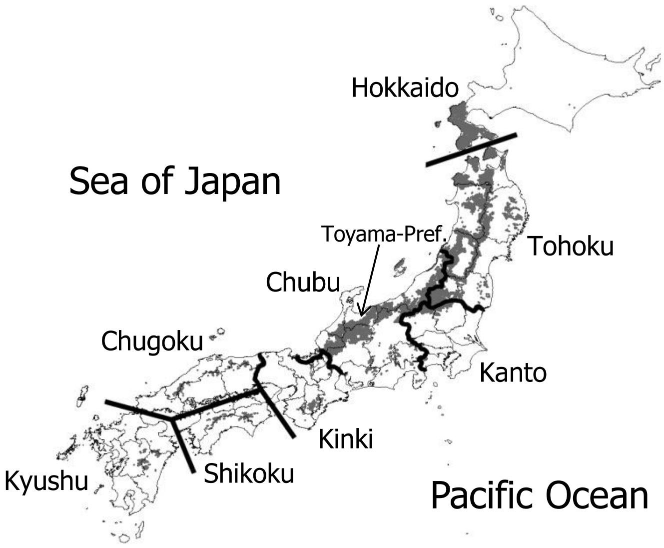
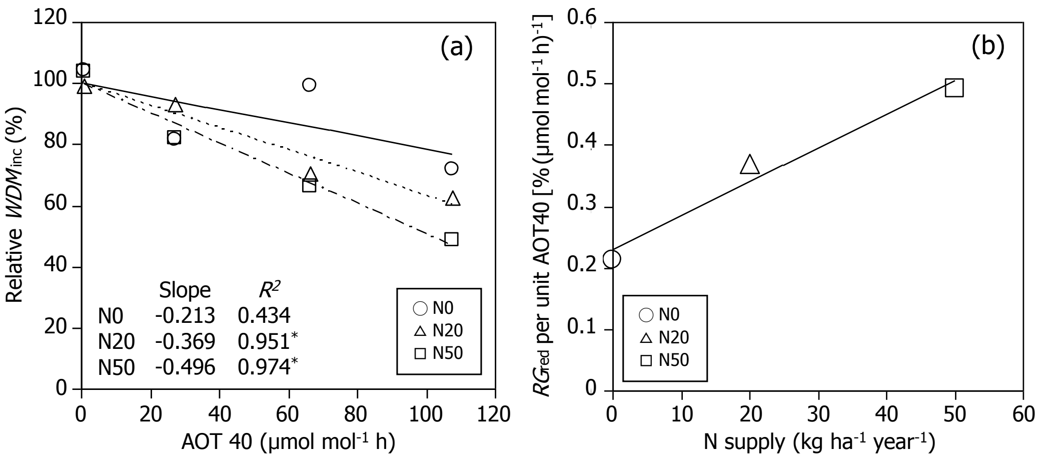


Figure 2



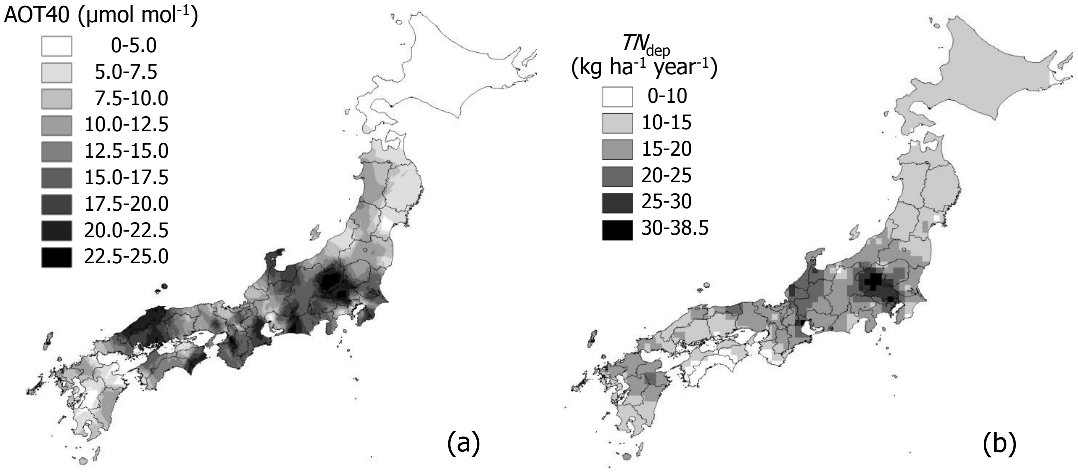
Hokkaido
Sea of Japan
Toyama-Pref.
Tohoku
Chubu
Chugoku
Kanto
Kinki
Kyushu
Shikoku
Pacific Ocean

Figure 3



(a)
Relative WDM_{inc} (%)
AOT 40 (μmol mol^{-1} h)
Slope R^2
N0 -0.213 0.434
N20 -0.369 0.951*
N50 -0.496 0.974*
N0
N20
N50
(b)
RG_{red} per unit AOT40 [% (μmol mol^{-1} h)$^{-1}$]
N supply (kg ha^{-1} year^{-1})
N0
N20
N50

Figure 4



AOT40 (µmol mol-1)
0-5.0
5.0-7.5
7.5-10.0
10.0-12.5
12.5-15.0
15.0-17.5
17.5-20.0
20.0-22.5
22.5-25.0
(a)
TNdep
(kg ha-1 year-1)
0-10
10-15
15-20
20-25
25-30
30-38.5
(b)

Figure 5

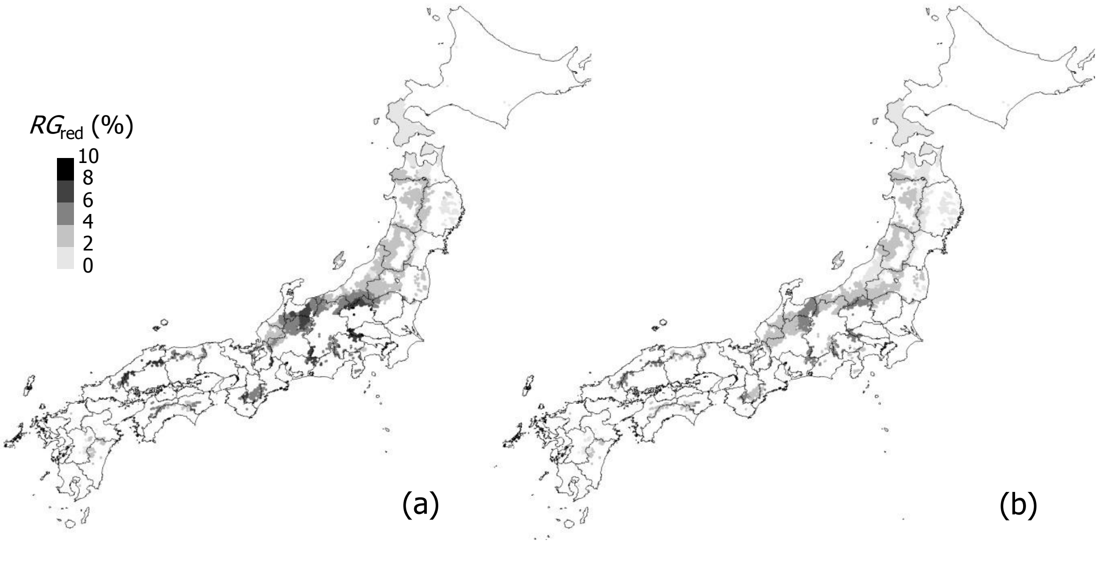


Figure 6

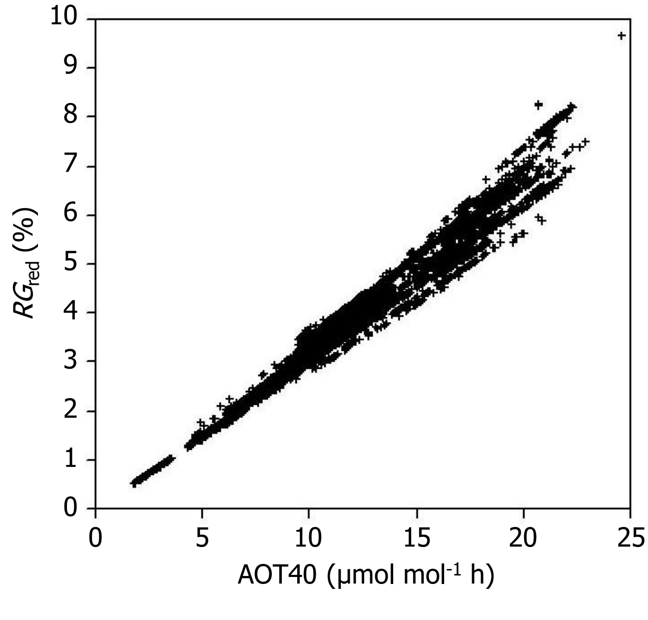
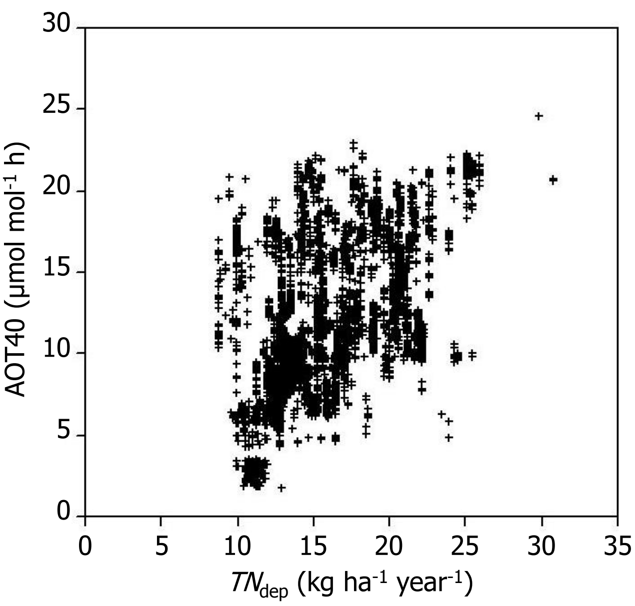


Figure 7



30
25
20
15
10
5
0
AOT40 (µmol mol-1 h)
0
5
10
15
20
25
30
35
TNdep (kg ha-1 year-1)