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Title

Risk assessment of ozone impact on the carbon absorption of Japanese representative conifers

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Abstract

A risk assessment of ozone (O_3) impact on the annual carbon absorption (*ACA*) of Japanese representative conifers was conducted based on the results of an experimental study, monitoring data of oxidant concentrations and vegetation surveys. The areas with high O_3-induced reduction in *ACA* did not necessarily correspond to the areas with relatively high O_3-exposure. Widespread distribution of O_3-sensitive tree species such as *P. densiflora* and *L. kaempferi*, and high *ACA* were important factors that induced a high risk of O_3 impact on the *ACA*. Therefore, we concluded that not only the accumulated O_3-exposure but also the variety of tree habitat, the tree sensitivity to O_3 and the *ACA* among the tree species must be taken into account to assess the risk of O_3 impact on the *ACA* of Japanese conifers. The O_3-induced reduction in the total *ACA* of the three tree species in Japan was estimated to be 0.8%.

1. Introduction

Ozone (O_3) in the troposphere is recognized as a widespread phytotoxic gaseous air pollutant and its concentrations have been increasing in the Northern Hemisphere (Akimoto 2003; ADORC 2006). The annual average concentration of photochemical oxidant, whose main component is O_3, during the daytime throughout Japan increased from 1985 to 1999 with the rate of increase being 0.33 nmol mol^{-1} year^{-1} and was about 31 nmol mol^{-1} as an average from 1999 to 2002 (Ohara and Sakata 2003; ADORC 2006). Furthermore, relatively high concentrations of O_3

above 100 nmol mol^{-1} have been frequently detected 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 et al. 2007).

Assimilation of CO_2 by forest tree species reduces atmospheric CO_2 concentration and contributes to the mitigation of global warming. However, many experimental studies have indicated that the ambient levels of O_3 reduce the dry matter production and net photosynthesis of forest tree species (Matsumura et al. 2001; Matyssek and Sandermann 2003; Oksanen 2003; Karnosky et al. 2005; Watanabe et al. 2006, 2007; Karlsson et al. 2007). Sitch et al. (2007) suggested that the O_3-induced reduction in the capacity of CO_2 absorption of terrestrial vegetation could contribute more to global warming than the radiative forcing of O_3 itself.

The extent of the negative impact of O_3 is considerably different among tree species (Kohno et al. 2005; Watanabe et al. 2006; Karlsson et al. 2007), which is one of the most important factors affecting the risk of O_3 impact on the carbon absorption capacity of forest tree species. The risk of O_3 impact in the habitats of O_3-sensitive tree species is greater than that in the habitats of O_3-tolerant tree species, even when the O_3 levels are the same. Furthermore, the difference in the degree of carbon absorption among the areas is an important factor. A reduction in the atmospheric concentration of O_3 in areas with a high degree of carbon absorption would be effective in mitigating global warming.

Cryptomeria japonica, *Pinus densiflora* and *Larix kaempferi* are important tree species for forestation in Japan. *C. japonica* and *P. densiflora* are evergreen coniferous species and are widely distributed in the temperate region of Japan (Hirai 1980a, 1980b). *L. kaempferi* is a deciduous coniferous species and is distributed in the northern part and in the highland of the central part of Japan (Hirai 1980a).

Although the three tree species were planted for the production of timber during the period between the 1950s and the 1970s, presently, the planted forests play an important role in carbon absorption under the Kyoto Protocol (Forestry and Forest Products Research Institute 2004). Watanabe et al. (2006) reported that the extent of O_3-induced reduction in the whole-plant growth of *L. kaempferi* and *P. densiflora* seedlings was greater than that of *C. japonica* seedlings. There are several variations in the habitats, the sensitivity of the trees to O_3 and the amount of carbon absorption among *C. japonica*, *P. densiflora* and *L. kaempferi* grown in Japan. Therefore, we hypothesized that areas with a high risk of O_3 impact on the carbon absorption of the three tree species would not necessarily correspond to areas with a high O_3-exposure. To test this hypothesis, we assessed the risk of O_3 impact on the annual carbon absorption (*ACA*) of *C. japonica*, *P. densiflora* and *L. kaempferi* in Japan based on the results of an experimental study and statistical data such as atmospheric O_3 concentrations and vegetation surveys.

2. Materials and methods

2.1 Experimental evaluation of the impact of ozone on the carbon absorption

2.1.1 Experimental design

In the present study, the O_3 exposure-response relationships of annual carbon absorption (*ACA*) of *C. japonica*, *P. densiflora* and *L. kaempferi* were evaluated based on the experimental study reported by Watanabe et al. (2006). In this experiment, the seedlings of *C. japonica*, *P. densiflora* and *L. kaempferi* were grown in 12 experimental treatments, which were comprised of 4 gas treatments [charcoal-filtered air (CF) and 3 levels of O_3 at 1.0 (O_3×1.0), 1.5 (O_3×1.5) and 2.0

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times the ambient concentration ($O_3 \times 2.0$)] in combination with 3 soil nitrogen (N) treatments with NH_4NO_3 (0, 20 and 50 kg N ha^{-1} $year^{-1}$) in open-top chambers during the 2 growing seasons. The average 24-h concentration of O_3 in the CF, $O_3 \times 1.0$, $O_3 \times 1.5$ and $O_3 \times 2.0$ treatments from April to September of the 2 growing seasons were 11.7, 42.6, 63.3 and 83.7 nmol mol^{-1}, respectively.

The average annual wet deposition rate of inorganic N (sum of NO_3^- and NH_4^+) throughout Japan from 1998 to 2002 was 7.2 kg N ha^{-1} $year^{-1}$ (Ministry of Environment 2004). Because the dry deposition rate of N is considered to be similar to the wet deposition rate of N (Matsuda et al., 2001), the total annual N deposition (sum of wet deposition and dry deposition) in Japan can be estimated to be around 15 kg N ha^{-1} $year^{-1}$. This deposition rate was the closest to the treatment with the N supply at 20 kg N ha^{-1} $year^{-1}$ in Watanabe et al. (2006). Therefore, we evaluated the effects of O_3 on the *ACA* of *C. japonica*, *P. densiflora* and *L. kaempferi* based on the results obtained from the seedlings grown in the soil supplied with N at 20 kg N ha^{-1} $year^{-1}$.

2.1.2. Calculation of annual carbon absorption of the seedlings

The experiment was conducted during the 2 growing seasons from April 2004 to November 2005. At the end of the first and second growing seasons, the seedlings were harvested to determine the dry mass and carbon concentration in the plant organs. The harvested samples were separated into needles, stems and roots. The plant organs were dried at 80 °C for 1 week and weighed. Dried samples were ground to a fine powder with a sample mill. The concentration of carbon in each plant organ was determined with a C/N analyzer (MT-700, Yanako, Japan). The whole-plant carbon content of the seedlings was calculated as a sum of the product of

the dry mass and the carbon concentration in each plant organ. The *ACA* of the seedlings was calculated as the difference between the whole-plant carbon content of the seedlings at the end of the first growing season and that at the end of the second growing season.

2.1.3. Analyses of ozone exposure-response relationships

The AOTX (accumulated exposure over a threshold of X nmol mol^{-1}, X = 0, 20, 40, 60, 80 and 100) of O_3 from April to September of the second growing season in 2005 were calculated based upon the monitoring data of atmospheric O_3 concentration in the OTCs. The AOTX was accumulated for 12 h (0600-1800 hours). The analyses of the O_3 exposure-response relationships for the *ACA* were performed by the methods described below. A regression line was obtained from the relationship between AOTX and the *ACA*. The theoretical *ACA* at zero AOTX was determined to be the y-axis intercept of the regression line. The theoretical *ACA* at zero AOTX was used as a reference (100%) to calculate the relative *ACA* for each gas treatment. The slope and coefficient of determination values (R^2) were calculated from the regression line between AOTX and the relative *ACA*.

Table 1 indicates the slope and coefficient of determination (R^2) values of the linear regression between the O_3-exposure indices and relative reduction in the *ACA* of *C. japonica*, *P. densiflora* and *L. kaempferi* seedlings. The degree of O_3-induced reduction in the *ACA* of *P. densiflora* and *L. kaempferi* was greater than that in *C. japonica*. In AOT60 and AOT80, significant regression lines were found in both of *C. japonica* and *P. densiflora* seedlings. Although no significant regression line was found for *L. kaempferi* seedlings, highest R^2 was detected in AOT60 ($P = 0.067$). Therefore, we employed the AOT60 as common O_3-exposure index for the

risk assessment of O_3 impact on the carbon absorption of the three tree species.

2.2 Estimation of AOT60 in habitat of Japan

The concentrations of photochemical oxidants are officially monitored at approximately 1200 monitoring stations in 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 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 Environment 1996). In the present study, therefore, the concentration of photochemical oxidants was regarded as that of O_3.

The number of hours with a concentration of O_3 above 0.06 μmol mol^{-1} (N_{60}) and that above 0.12 μmol mol^{-1} (N_{120}) at all the monitoring stations in Japan are officially tabulated and can be obtained from the web site of National Institute for Environmental Studies (http://www.nies.go.jp/igreen/index.html). However, hourly data of O_3 concentration were available in limited number (approximately 40%) of prefectures. Ishii et al. (2007) developed the method for the estimation of AOT40 from the N_{60} and N_{120}. Based on the available hourly data of O_3 concentration, they found the high correlation between the sum of the N_{60} and N_{120}, and observed the AOT40 for 12 h (0600-1800 hours) from April to September. Therefore, we

estimated the AOT60 of O_3 at all the monitoring stations in Japan by using the modified method of Ishii et al. (2007).

Original formula of Ishii et al. (2007) expressed the AOT40 as follows:

$$AOT40 = a \times (N_{60} + N_{120}).$$

Because N_{120} contributes more to AOT60 than N_{60}, we added the weighting factor for N_{120}:

$$AOT60 = a \times (N_{60} + b \times N_{120}).$$

The coefficients that indicated best correlation between estimated AOT60 and observed AOT60 were calculated by solver function of spreadsheet software (Excel 2003, Microsoft, Redmond, WA, USA). We used the data of N_{60}, N_{120} and available O_3 concentration from 2000 to 2004 for the estimation of AOT60 (μmol mol^{-1} h) and obtained the coefficients: a = 12.86 and b = 11.05.

The map of spatial distribution of AOT60 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 AOT60 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).

The habitats of *C. japonica*, *P. densiflora* and *L. kaempferi* in Japan were determined based on the vegetation raster data (45" × 30" per mesh) of the National

Survey on the Natural Environment, carried out by the Ministry of the Environment. These raster data were obtained from the web site of the Japan Integrated Biodiversity Information System (http://www.biodic.go.jp/J-IBIS.html). The geographical meshes that contain the vegetation code of *C. japonica*, *P. densiflora* and *L. kaempferi* were extracted as their habitat, as shown in Fig. 1. The AOT60 of O_3 in each habitat of the three tree species was extracted from the above-mentioned AOT60 map and was averaged in each prefecture.

2.3 Estimation of ozone-induced reduction in the annual carbon absorption

The calculation method for the *ACA* of *C. japonica*, *P. densiflora* and *L. kaempferi* was according to Forestry and Forest Products Research Institute (2004) and is described below. The calculation was conducted in each prefecture and for each tree species. We obtained the data of the forest resource assessment from the web site of the Forestry Agency of Japan (http://www.rinya.maff.go.jp/toukei/genkyou/index.htm). The data contain the basal area (ha) and stand volume (m^3) of *C. japonica*, *P. densiflora* and *L. kaempferi* in each forest age class from planting seedlings. The forest age class was delimited in 5 years, i.e. Class 1 = 1-5 years, Class 2 = 6-10 years etc. Stem volume per basal area ($m^3\ ha^{-1}$) was calculated and converted to the carbon content of each organ (leaf, stem + branches, and root) per basal area (kg carbon ha^{-1}) according to the following formula:

Carbon content = Stem volume $\times$ Basic density $\times$ Conversion coefficient

$\times$ Carbon concentration.

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Basic densities (the ratio of stem dry mass to stem volume) were according to the data from the Forestry Experiment Station (1982), i.e. 316 kg m^{-3} for *C. japonica*, 405 kg m^{-3} for *P. densiflora* and 444 kg m^{-3} for *L. kaempferi*. The conversion coefficient was the ratio of the dry mass of the each organ to that of stem; values were obtained from the Forestry and Forest Products Research Institute (2004). The carbon concentration of each tree organ obtained from the experimental study described above was used for this calculation of carbon content. Because the exposure to O_3 little affected the carbon concentration of the organs in the three tree species as shown in Table 2, the carbon concentrations in each gas treatment study were averaged and used for the calculation.

The carbon content of the whole-tree per basal area was calculated as the sum of the carbon contents of leaf, stem + branch and root. The Gompertz curve was fitted to the relationship between forest age class and carbon content of the whole-tree per basal area (Fig. 2, Forestry and Forest Products Research Institute, 2004). The fifth part of the difference between the carbon content of the whole-tree per basal area of an age class and that of the next age class was calculated based on the regression curve, and regarded as the *ACA* per basal area in each forest age class. The O_3-induced reduction in the *ACA* per prefecture (C_{red}, Gg carbon year^{-1}) of each forest age class was calculated by the following formula:

$$C_{red} = C_{abs} \times R \times \text{AOT60} \times \text{Basal area},$$

where C_{abs} is the *ACA* per basal area (Gg carbon ha^{-1} year^{-1}) and R is the rate of reduction in the *ACA* per unit AOT60, which is the absolute value of the slope of the regression line between AOT60 and *ACA*, obtained from the experimental study

(Table 1). This C_{red} indicated the reduction in the *ACA* at an AOT60 compared to that at zero AOT60. Finally, the C_{red} of all forest age classes was summed up.

3. Results

3.1 Estimation of AOT60 in Japan

The relationship between the estimated AOT60 and observed AOT60 is shown in Fig. 3. High correlation along the 1:1 line was observed although a little over estimation and under estimation were found in relatively low and high AOT60, respectively. Figure 4 illustrates the map of the estimated AOT60 of O_3 in Japan. The highest AOT60 was estimated in the western part of the Kanto region. Relatively high AOT60 values were estimated not only in the metropolitan areas of Tokyo and Osaka but also in the countryside such as the northern part of the Chubu region and the northern part of the Chugoku region.

3.1 O_3-induced reduction in annual carbon absorption

The estimated C_{red} in each prefecture is shown in Table 3. The prefectures that indicated relatively high C_{red} were different among the three tree species. Relatively high C_{red} was estimated in Gunma, Akita and Miyazaki Prefectures for *C. japonica*; Shimane, Iwate and Gunma Prefectures for *P. densiflora*; and Nagano, Gunma and Yamanashi Prefectures for *L. kaempferi*. The average and maximum rates of the C_{red} in Japan were estimated to be 0.7% and 2.1% for *C. japonica*, 1.2% and 3.6% for *P. densiflora* and 1.4% and 3.4% for *L. kaempferi*, respectively. The total C_{red} of the three tree species in all prefectures was estimated to be 0.8%.

Figure 5 illustrates the map of the total C_{red} of the three tree species in Japan. The total C_{red} of the three tree species in Nagano, Gunma, Akita and Iwate

Prefectures were relatively high as compared with that of the other prefectures. The areas with relatively high AOT60 of O_3 did not necessarily correspond to areas with high C_{red}. Although the AOT60 in Nagano, Akita and Iwate Prefectures were not as high as that in the other prefectures, the extents of the estimated C_{red} in these prefectures were relatively high (Figs. 4 and 5). In Nagano and Iwate Prefectures, this phenomenon was mainly attributed to the widespread distribution of O_3 sensitive species *L. kaempferi* and *P. densiflora* (Fig. 1 and Table 1). In Akita Prefecture, on the other hand, *C. japonica* was the primary tree species. The rate of the C_{red} of *C. japonica* in this prefecture was relatively low because of the relatively low AOT60 and good O_3 tolerance of *C. japonica* (Fig. 4 and Table 1). However, the *ACA* of *C. japonica* in Akita Prefecture was highest among all the prefectures, as shown in Fig. 6. Therefore, we conclude that the high C_{red} in this prefecture was mainly due to the accumulation of small effect on individual trees of *C. japonica*. This accumulation effect also contributed to the high C_{red} of *P. densiflora* in Iwate and that of *L. kaempferi* in Nagano Prefectures (Fig. 6 and Table 3).

4. Discussion

In Europe, the risk assessment of O_3 impact on forest trees had been conducted based on the concept of critical level (Mills 2004). The critical level of O_3 for forest trees was evaluated from the relationship between the growth of seedlings of O_3-sensitive tree species and accumulated exposure (e.g. AOTX) or accumulated stomatal flux (e.g. $AF_{st}Y$) of O_3 by the analysis of 99% confidence limits of the regression line (Karlsson et al. 2004; Mills 2004; Karlsson et al. 2007). The areas exceeding the critical level of O_3 were detected as high-risk areas (Simpson et al. 2007). This risk assessment is easy to understand and useful, for example, in the

formulation of policy aimed at a reduction in the precursors of O_3. In the present study, on the other hand, we focused not only on accumulated O_3-exposure but also on the variety of tree habitat, the sensitivity of tree species to O_3 and the *ACA* among the three tree species for evaluating the risk of O_3 impact on the *ACA*. As a result, we estimated a relatively high risk of O_3 impact on the *ACA* in the areas where the AOT60 of O_3 was relatively low and where the O_3-tolerant species, *C. japonica*, was primary species (Figs. 1, 4 and 5). Our results indicate that the variety of tree habitat, tree sensitivity to O_3 and the *ACA* among the tree species must be taken into account to assess the risk of O_3 impact on the carbon absorption of forest tree species.

Recently, the risk of O_3 has been mainly assessed based on the index of the accumulated stomatal flux of O_3, while the exposure index was retained as the recommended method for calculating critical levels for forest tree species (Emberson et al. 2000; Mills 2004; Matyssek et al 2007; Simpson et al. 2007). This change from the assessment by exposure basis to that by flux basis is reasonable because the main impact was induced by the O_3 that enters the plants' leaves through the stomata (Reich 1987). The stomatal uptake of O_3 reduces under dry conditions such as the summer of the Mediterranean region, whereas relatively high O_3 concentration is observed in this region (Ferretti et al. 2007). On the other hand, the O_3 uptake of the trees grown in northern latitudes of Europe becomes high due to high air humidity and long days even the O_3 concentration is relatively low (Simpson et al. 2007). Unfortunately, we could not use the data of hourly O_3 concentrations from many monitoring stations in Japan. Currently, it is therefore difficult to assess the risk of O_3 impact in Japan based on the accumulated stomatal flux of O_3. However, it is to be noted that since Japan is a country where precipitation ranging from 1100 to 2300 mm and air humidity is generally high, drought-induced stomatal closure would not

be as frequent as that in European countries (National Astronomical Observatory 2007; Ministry of Land, Infrastructure, Transport and Tourism 2008).

To evaluate the changes in the risk of O_3 impact on the *ACA* of *C. japonica*, *P. densiflora* and *L. kaempferi* with the increasing concentrations of O_3 in Japan (Ohara and Sakata 2003; ADORC 2006), we compared the estimated C_{red} based on the AOT60 of O_3 from 2000 to 2004 and that from 1990 to 1994. The ratio of the total C_{red} of the three tree species in all the prefectures from 2000 to 2004 (91.0 Gg carbon year^{-1}) to that from 1990 to 1994 (69.2 Gg carbon year^{-1}) was calculated to be 136%. One of the main reasons for this rapid increase in the risk of O_3 impact on the C_{red} in Japan is considered to be transboundary air pollution from other East Asian countries (Ohara and Sakata 2003; Tanimoto et al. 2005; ADORC 2006; Yamaji et al. 2006). Many researchers have predicted that the increase in O_3 concentration in Japan will continue with the increasing emissions of precursors of O_3 such as NO_x and volatile organic compounds from other East Asian countries in the near future (Prather et al. 2001; Stevenson et al. 2006; Ohara et al. 2007). Therefore, the development of control policies for lowered emissions of precursors of O_3 in East Asian countries is crucially important for reducing the risk of O_3 impact on Japanese forest tree species. This reduction leads to the mitigation of global warming not only by reduction in radiative forcing of O_3 but also by the stimulation of CO_2 absorption by forest trees (Sitch et al. 2007).

Many reports indicate O_3-induced change of structural and metabolic carbon allocations of tree species. It has been well documented that the O_3 increased the shoot/root ratio (Matyssek et al. 1992; Landolt et al. 2000; Witting et al. 2009). In the leaves exposed to O_3, more carbon would be distributed to the functions of detoxification for O_3 and its derivations, and repair for injured organs (Fuhrer and

Booker 2003; Castagna and Ranieri 2009). On the other hand, several researchers reported that O_3-induced increase of dark respiration, which leads to carbon loss back to the atmosphere (Wallin et al. 1990; Maurer et al. 1997). These structural and metabolic changes of carbon allocation induced by O_3 will affect the net carbon uptake of the trees, especially as long-term accumulated effects. In the future, therefore, we must develop the estimation model with structural and metabolic processes for the risk assessment of O_3 impact on the *ACA* of Japanese coniferous forest tree species.

In conclusion, the results obtained from the present study support our hypothesis that the area with a high risk of O_3 impact on the *ACA* of the three tree species does not necessarily correspond to the area with high O_3-exposure. Widespread distribution of O_3-sensitive tree species such as *P. densiflora* and *L. kaempferi* and high *ACA* were important factors that induced a high risk of O_3 impact on the *ACA*. Therefore, we must take into account not only the accumulated O_3-exposure but also the variety of tree habitat, tree sensitivity to O_3 and the *ACA* among the tree species to assess the risk of O_3 impact on the carbon absorption of Japanese conifers.

Acknowledgments

We have used the data file of photochemical oxidants from the 'Numerical database for environment' of the National Institute for Environmental Studies, Japan.

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Table 1 The slope and coefficient of determination (R^2) values for the linear regression between AOTX (µmol mol^{-1} h) and the relative reduction in the annual carbon absorption per one growing season (%) of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi* seedlings.

		AOT0	AOT20	AOT40	AOT60	AOT80	AOT100
Slope	*C. japonica*	-0.096	-0.116	-0.152	-0.209	-0.301	-0.462
	P. densiflora	-0.147	-0.184	-0.249	-0.355	-0.526	-0.826
	L. kaempferi	-0.158	-0.200	-0.271	-0.387	-0.568	-0.879
R^2	*C. japonica*	0.895	0.944*	0.983**	0.988**	0.949*	0.887
	P. densiflora	0.736	0.813	0.899	0.959*	0.968*	0.936*
	L. kaempferi	0.657	0.735	0.818	0.871	0.857	0.789

The intercept of each the regression line was adjusted to 100% (see Materials and Method).

Single regression analysis: *p<0.05; **p<0.01.

Table 2 Effects of O₃ on carbon concentrations (%) of plant organs of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi* seedlings at the end of the second growing season.

		Needle	Stem	Root
C. japonica	CF	50.5 (0.1)	48.5 (0.1)	48.4 (0.4)
	O₃×1.0	50.8 (0.1)	48.3 (0.0)	48.6 (0.1)
	O₃×1.5	50.8 (0.2)	48.3 (0.1)	48.5 (0.3)
	O₃×2.0	51.0 (0.1)	48.5 (0.2)	48.6 (0.3)
	ANOVA	*	n.s.	n.s.
P. densiflora	CF	52.8 (0.3)	49.6 (0.2)	47.8 (0.1)
	O₃×1.0	52.8 (0.0)	49.7 (0.1)	46.4 (0.9)
	O₃×1.5	52.7 (0.1)	49.6 (0.1)	46.1 (1.0)
	O₃×2.0	52.4 (0.1)	49.5 (0.1)	46.5 (0.5)
	ANOVA	n.s.	n.s.	n.s.
L. kaempferi	CF	49.5 (0.6)	50.3 (0.0)	49.2 (0.5)
	O₃×1.0	49.9 (0.4)	50.4 (0.4)	48.4 (0.1)
	O₃×1.5	49.1 (0.0)	50.7 (0.1)	48.4 (0.2)
	O₃×2.0	50.1 (0.6)	50.5 (0.0)	48.6 (0.5)
	ANOVA	n.s.	n.s.	n.s.

Each value is the mean of 3 replicates, and the standard deviation is shown in parentheses.

The seedlings were exposed to charcoal-filtered air (CF) or three levels of O₃ at 1.0, 1.5 and 2.0 times ambient concentration.

ANOVA: *$p < 0.05$; n.s. not significant.

Table 3 The estimated O_3-induced reduction in the annual carbon absorption (C_{red}, Gg carbon year^{-1}) of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi*. Values in the parentheses show the reduction rate (%) compared to the annual carbon absorption at zero AOT60.

Region	Prefecture	C. japonica	P. densiflora	L. kaempferi	Total
Hokkaido	Hokkaido	0.12 (0.23)		0.49 (0.19)	0.61 (0.19)
Tohoku	Aomori	1.18 (0.42)	0.26 (0.70)	0.21 (0.82)	1.64 (0.48)
	Iwate	1.76 (0.38)	1.54 (0.63)	0.77 (0.70)	4.07 (0.50)
	Miyagi	0.71 (0.32)	0.25 (0.54)	0.04 (0.67)	1.01 (0.36)
	Akita	3.80 (0.55)	0.21 (1.00)	0.15 (0.90)	4.15 (0.57)
	Yamagata	1.44 (0.52)	0.13 (0.90)	0.22 (0.83)	1.78 (0.57)
	Fukushima	2.07 (0.49)	1.06 (0.79)	0.35 (1.29)	3.48 (0.60)
Kanto	Ibaraki	0.54 (0.72)	0.34 (1.61)		0.97 (1.01)
	Tochigi	1.62 (1.19)	0.25 (1.98)	0.31 (2.23)	2.18 (1.34)
	Gunma	4.26 (1.86)	1.25 (3.06)	2.02 (3.12)	7.53 (2.25)
	Saitama	1.33 (2.09)	0.01 (3.64)	0.15 (3.29)	1.49 (2.18)
	Chiba	1.01 (1.25)	0.03 (1.97)		1.04 (1.27)
	Tokyo	0.57 (1.72)	0.02 (3.35)	0.02 (3.39)	0.61 (1.78)
	Kanagawa	0.28 (0.93)	0.02 (1.40)		0.30 (0.94)
Chubu	Niigata	1.53 (0.52)	0.08 (0.72)	0.12 (1.95)	1.73 (0.55)
	Toyama	1.14 (1.02)	0.01 (1.79)	0.02 (1.81)	1.17 (1.03)
	Ishikawa	1.57 (0.95)	0.29 (1.84)		1.86 (1.03)
	Fukui	1.17 (0.59)	0.11 (1.20)		1.28 (0.62)
	Yamanashi	0.49 (1.18)	1.03 (1.88)	1.82 (2.19)	3.35 (1.86)
	Nagano	1.09 (1.02)	1.17 (1.73)	5.80 (1.98)	8.05 (1.72)
	Gifu	1.67 (0.86)	0.07 (1.38)	0.58 (1.71)	2.32 (0.99)
	Sizuoka	1.53 (1.06)	0.22 (1.88)	0.10 (1.86)	1.84 (1.15)
	Aichi	2.05 (1.05)	0.10 (1.48)		2.15 (1.06)
Kinki	Mie	1.24 (0.94)	0.16 (1.59)		1.40 (0.98)
	Shiga	0.59 (0.95)	0.03 (1.55)		0.62 (0.97)
	Kyoto	0.83 (0.88)	0.17 (1.41)		1.00 (0.94)
	Osaka	0.07 (1.39)	0.06 (1.93)		0.14 (1.60)
	Hyogo	2.00 (0.79)	0.37 (1.51)		2.38 (0.85)
	Nara	1.92 (1.19)	0.03 (1.95)		1.95 (1.20)
	Wakayama	1.47 (1.02)	0.13 (1.98)		1.60 (1.06)
Chugoku	Tottori	1.03 (0.79)	0.62 (1.31)		1.65 (0.93)
	Shimane	1.92 (1.20)	1.78 (2.07)		3.70 (1.51)
	Okayama	0.54 (0.75)	0.35 (1.28)	0.02 (1.27)	0.91 (0.90)
	Hiroshima	0.69 (1.27)	0.41 (2.26)		1.10 (1.51)
	Yamaguchi	0.96 (0.85)	0.39 (1.36)		1.35 (0.96)
Shikoku	Tokushima	2.37 (0.99)	0.15 (1.38)		2.52 (1.01)
	Kagawa	0.01 (0.61)	0.09 (0.90)		0.11 (0.85)
	Ehime	1.67 (0.77)	0.12 (1.26)		1.79 (0.79)
	Kochi	2.45 (0.69)	0.10 (1.09)		2.54 (0.70)
Kyusgu	Fukuoka	1.00 (0.67)	0.02 (0.79)		1.03 (0.67)
	Saga	0.67 (0.62)	0.01 (0.97)		0.68 (0.62)
	Nagasaki	0.40 (0.63)	0.01 (0.91)		0.42 (0.64)
	Kumamoto	1.76 (0.43)	0.08 (0.75)		1.84 (0.44)
	Oita	2.12 (0.50)	0.04 (0.76)		2.17 (0.50)
	Miyazaki	3.62 (0.67)	0.12 (1.16)		3.73 (0.68)
	Kagoshima	1.66 (0.51)	0.09 (0.86)		1.75 (0.52)
Total		63.91 (0.73)	13.82 (1.23)	13.27 (1.35)	91.00 (0.84)

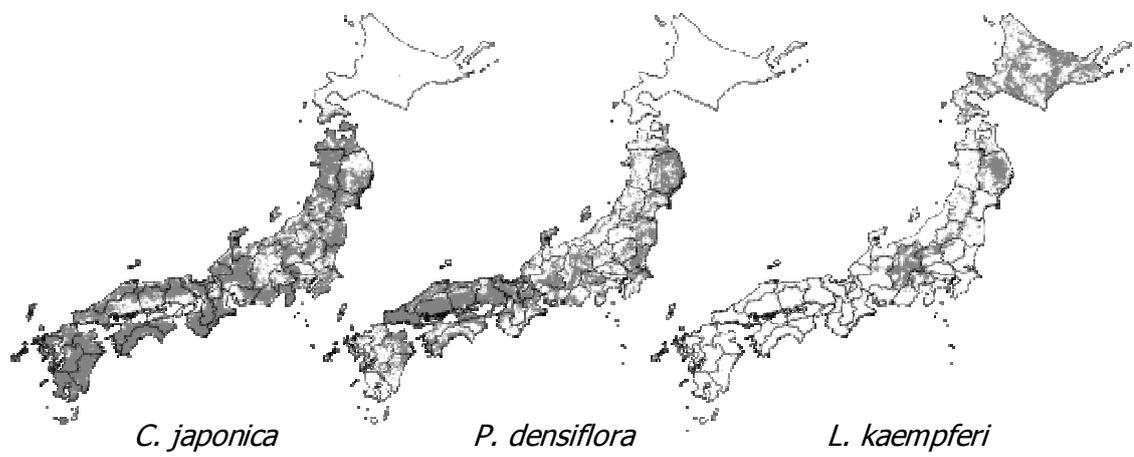C. japonica   P. densiflora   L. kaempferi

Fig. 1 The habitats of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi* in Japan
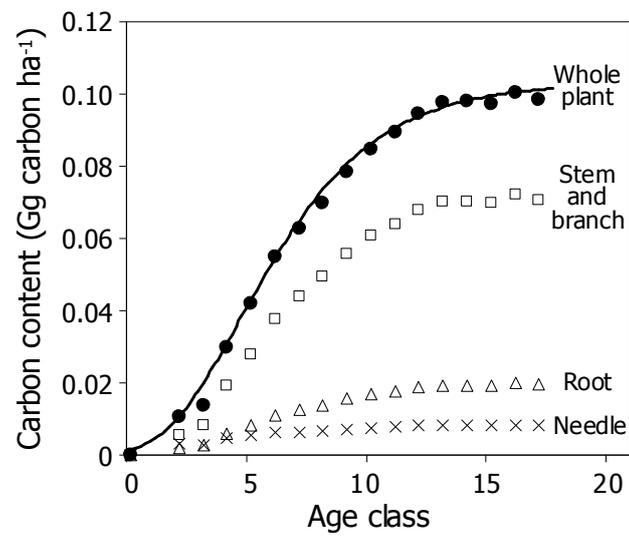
Fig. 2 Example of the relationship between forest age class (Class 1 = 1-5 years, Class 2 = 6-10 years etc.) and carbon content of trees per area (*Cryptomeria japonica* in Tokyo Prefecture)
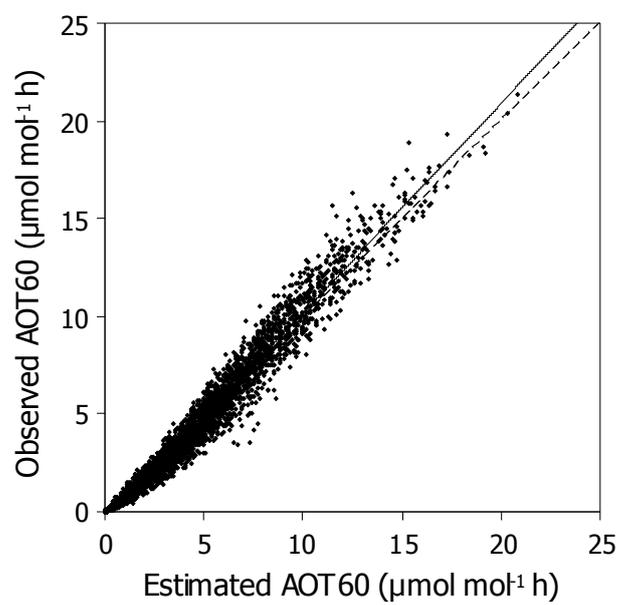
Fig. 3 Comparison between estimated and observed AOT60. The solid and dashed lines indicate the regression line (y = 1.071x − 0.526, R^2 = 0.964) and 1:1 line, respectively
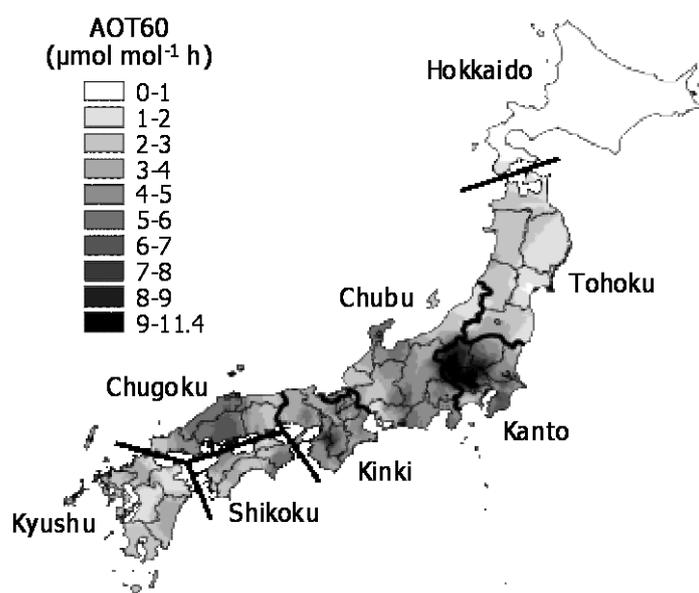
Fig. 4 The estimated AOT60 in Japan. The AOT60 data was accumulated during 0600-1800 hours from April to September and averaged from 2000 to 2004
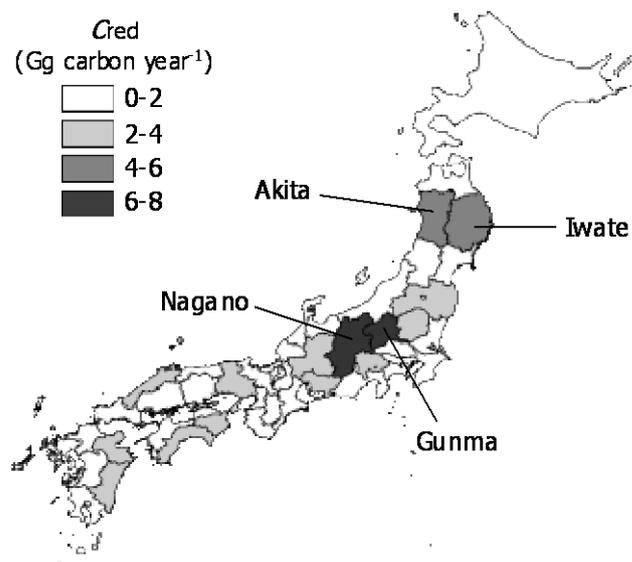
Fig. 5 The estimated O$_3$-induced reduction in the annual carbon absorption (C_{red}) in Japan. The values were the sum of the C_{red} of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi*
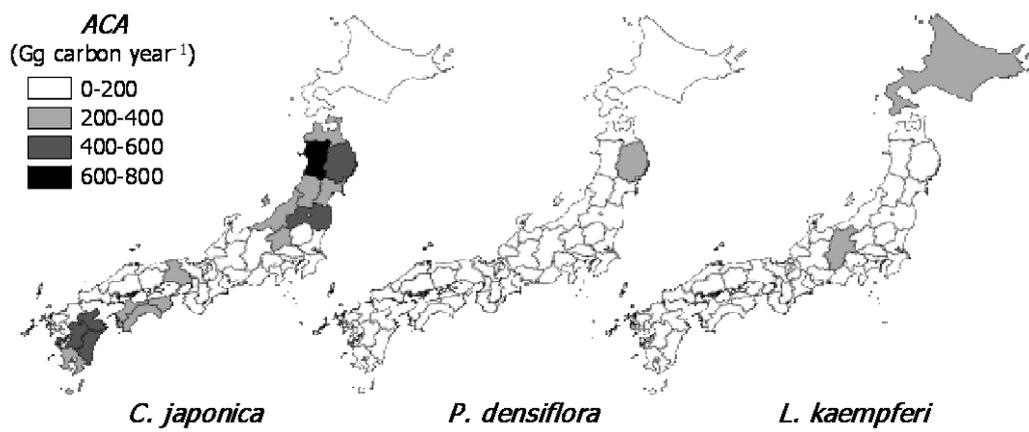
Fig. 6 The annual carbon absorptions (*ACA*) of *Cryptomeria japonica*, *Pinus densiflora* and *Larix kaempferi* in Japan