C-7 Study on Impacts of Acidic and Oxidative Substances on Vegetation and Establishment of Tentative Critical Level for Protecting East Asian Vegetation (Abstract of the Final Report)

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1. Introduction

In European countries, scientists have discussed about critical loads for reducing emission of air pollutants and its deposition in ecosystems. Critical loads handle soil chemistry such as soil acidification associated with Al toxicity in plant root systems due to acid deposition, but not on responses in aerial plant parts to pollutants. In contrast, a concept of critical level, which is based on a dose-response relationship has been proposed. Current 'Level I' critical level for European forests has been proposed to be 20 $\mu g/m^3$ (7-8 ppb) of sulfur dioxide (SO_2) as annual mean concentration or 10 ppm-h of AOT40 of ozone (O_3) for 6 months[2, 3]. However, that for East Asian forests has not been discussed.

Ambient concentration of sulfur dioxide in Japan has extremely decreased by environmental countermeasures in emission sources. While concentration of nitrogen oxides (NOx) in many monitoring stations is over the national standard, direct harmful effects of NOx on vegetation has not been reported. However, increasing N deposition may have a long-term ecological and physiological modification in plant responses under a potential threat of ambient ozone for the vegetation.

Forest monitoring in Japan revealed that there were some distribution of forest declined sites, however, relationships between air quality and forest health has not been analyzed statistically. Thus, publics has still great concerns with effects of acid and oxidative substances on the vegetation through transboudary transport and deposition in associated with a rapid increasing emission of air pollutants in the East Asian countries.

2. Research Objective

Main objectives of this research will establish and propose tentative critical levels of air pollutants for protecting and/or alleviating East Asian vegetation from effects of acid and oxidative substances.

Exposure experiments were conducted to collect and evaluate plant sensitivity and thresholds to induce growth reduction for establishing primary critical levels. In addition, basic information about air quality monitoring data, vegetation types and other environmental information in Japan were transformed into GIS database system and distribution maps of concentration and deposition of pollutants was prepared for visual and quantitative analysis.

As most parts of East Asian countries are not covered with automatic active monitoring network systems, it is necessary to introduce a numerical model analysis of air quality and deposition in regional and local scale. Also, a development of simple method using different indicator plants to pollutants will help to evaluate air quality in the East Asian countries.

3. Results and Discussion

(1) Plant sensitivity to acid and oxidative substances

① Sensitivity of trees to sulfur dioxide or ozone under nitrogen load

The normal linear regression equation of growth response to SO_2 or O_3 in 30 tree species was derived from the dataset obtained in the multiple-season exposure experiments in the potted young trees using the open-top chambers. The total dose (ppm-h) of SO_2 or the AOT40 (accumulated exposure over a threshold of 40 ppb, ppm-h) of O_3 for daylight hours (global radiation > 50 W/m^2) was calculated between April and November during the period of exposure. The whole-plant biomass per tree and a 10% reduction in the increment (from the initial harvest to the final harvest) as compared to the filtered-air treatment were used as indicative parameters. A value of dose corresponding to a 10% growth reduction of a tree species from the slope of the regression line with different experimental period was converted to a mean concentration (ppb) of SO_2 or an AOT40 (ppm-h) of O_3 for a growing season. Mean concentration of SO_2 corresponding to a 10% growth reduction was 5-6 ppb in *Betula platyphylla var. japonica* and *Pinus strobus*, which were the most sensitive species to SO_2 among the species examined. However, it was over 39 ppb for less sensitive tree species. The AOT40 of O_3 corresponding to a 10% growth reduction was 11-12 ppm-h in the most sensitive poplars (*Populus maximowiczii* and *P. nigra var. italica*) and was over 45 ppm-h in less sensitive species.

Seedlings of six tree species (*Quercus serrata*, *Fagus crenata*, *Castanopsis cuspidata var. sieboldii*, *Larix kaempferi*, *Pinus densiflora* and *Cryptomeria japonica*) were exposed to four levels of SO_2 (charcoal-filtered air [CF], CF added at a constant concentration of 10, 20 and 40 ppb [24 hours per day]) or four levels of O_3 (CF, CF added in proportion to 1.0, 1.5 and 2.0 times the ambient O_3 concentration [24 hours per day]) for two growing seasons from April in 2004 using open-top chambers. Each SO_2 or O_3 treatment was split into three nitrogen loads (0, 20 and 50 kg N per hectare per year [N0, N20 or N50 respectively]) by adding NH_4NO_3 solution to the surface of soil.

Mean SO_2 concentrations in the ambient air and CF treatment were lower than 1 ppb over the whole exposure period. Those in targeted 10, 20 and 40 ppb of SO_2 treatments for the whole period were 10, 18 and 35 ppb, respectively. Ambient O_3 at the experimental site was 41 ppb for 24-hour seasonal mean for the whole exposure period. Trees in targeted CF, 1.0, 1.5 and 2.0 times the ambient O_3 treatments were exposed to 32 %, 98 %, 146% and 193 % of O_3 in the ambient air in 24 hour, respectively.

In the N0 treatment, SO_2 at 20 ppb or higher induced leaf visible injuries and/or accelerated defoliation of *Q.serrata*, *F. crenata, C. cuspidata* and *P.densiflora.* Concentration at 40 ppb of SO_2 induced reductions in growth of *F. crenata* and *P.densiflora*. Growth of *L. kaempferi* reduced by the exposure to SO_2 at 20 ppb or higher as compared with CF treatment. At the same N load, O_3 at 1.5 times the ambient concentration and higher level induced leaf visible injuries, accelerations in defoliation or reductions in growth of all tree species. Growth of *C. cuspidata* and *L. kaempferi* reduced by the exposure to O_3 at 1.0 times the ambient concentration as compared with CF treatment.

On the other hand, growth of *Q. serrata*, *F. crenata*, *C. cuspidata* and *C. japonica* increased with increasing load of nitrogen at CF treatment. Significant interactions between SO_2 or O_3 and N treatment were found on the growth of *F. crenata* and *L. kaempferi*. While higher nitrogen treatment exacerbated the negative O_3 effect on *F. crenata*, that treatment counteracted the effect of SO_2 or O_3 on *L. kaempferi*. This result suggests that there may be a need to weight critical levels for the protection of *F. crenata* to account for synergistic effects of O_3 with nitrogen load.

② Physiological and biochemical indicators for plant sensitivity to pollutants

To evaluate the sensitivity of Japanese evergreen broad-leaved trees to N load, 2-year-old seedlings of *Castanopsis cuspidata, Lithocarpus edulis, Quercus glauca* and *Quercus acuta* were grown for two growing seasons in brown forest soil treated with N as NH_4NO_3 solution at a rate of 0 (N-0), 10 (N-10), 50 (N-50), 100 (N-100) or 300 (N-300) kg ha^{-1} $year^{-1}$. The whole-plant dry mass of *C. cuspidata* and *Q. glauca* were significantly reduced by the N-100 and N-300 treatments, and by the N-300 treatment, respectively. In *L. edulis* seedlings, the whole-plant dry mass was significantly increased by the N-50 treatment, while it was significantly reduced by the N-300 treatment. The whole-plant dry mass of *Q. acuta* seedlings increased with increasing amounts of N added to the soil. In *C. cuspidata, L. edulis* and *Q. glauca* seedlings, the whole-plant dry mass was negatively correlated with the ratio of Mn concentration to Mg concentration (Mn/Mg) in the leaves.

There was a great difference in the sensitivity to N load among the four evergreen broad-leaved tree species, *C. cuspidata* being relatively sensitive and *Q. acuta* being tolerant to high N load. Threshold values of N load for the appearance of growth reduction in *C. cuspidata, L. edulis* and *Q. glauca* seedlings were estimated to be 50-100, 100 and 200-300 kg ha^{-1} $year^{-1}$, respectively. Concentration ratio of Mn/Mg in the leaves was a useful indicator for evaluating the negative effects of N load on the whole-plant dry weight growth of Japanese evergreen broad-leaved trees.

To clarify physiological and biochemical indicators for Japanese forest tree species to O_3 or SO_2, net photosynthetic rate, concentration and activity of RuBP carboxylase/oxygenase (Rubisco), active oxygen scavenging system and leaf nutrient status of 6 representative Japanese forest tree species (*F. crenata*, *Q. serrata, Ca. cuspidate, Cr. japonica*, *P. densiflora* and *L. kaempferi*) under different N loads were investigated.

When O_3 reduced whole-plant dry mass of the seedlings, it reduced net photosynthetic rate and concentration and/or activity of Rubisco in the leaves. There was a positive correlation between the whole-plant dry mass and net photosynthetic rate of the seedlings exposed to O_3. However, no clear relationships were obtained between the whole-plant dry mass and enzyme activity and antioxidant concentration in the active oxygen scavenging system or leaf nutrient status.

When SO_2 also reduced the whole-plant dry mass of the seedlings, it reduced net photosynthetic rate and Rubisco concentration, but increased S concentration in the leaves. There was a positive correlation between the whole-plant dry mass and net photosynthetic rate of the seedlings exposed to SO_2. However, there were no clear relationships between the whole-plant dry mass and enzyme activity and antioxidant concentration in the active oxygen scavenging system or leaf nutrient status. In the O_3- or SO_2-experiments, positive correlation was obtained between the whole-plant dry mass and the product of leaf area (*F. crenata*, *Q. serrata* and *Ca. cuspidate*) or leaf dry mass (*Cr. japonica*, *P. densiflora* and *L. kaempferi*) and net photosynthetic rate. From these results, net photosynthetic rate and leaf growth parameter were suitable indicators for the sensitivity of the whole-plant dry weight growth of Japanese forest tree species to O_3 or SO_2.

(2) Establishment of air quality monitoring system using indicator plants

To determine the relationships between the degree of air pollution and the plant response, 29 cultivars/varieties/species of the widely cultivated, fast-grown annual agricultural crops in the East Asian region were tested in Japan using the open-top chambers. Radish (*Rhaphanus sativus* cv. Red Chime) and Chinese vegetable (*Brassica campestris var. rosularis* cv. ATU171) were selected as an indicator plant for O_3 and SO_2, respectively.

Raphanus sativus cv. Red Chime and *B. campestris var. rosularis* cv. ATU171 were exposed to non-filtered ambient air (NF) and activated charcoal-filtered air (CF) in mini-open-top chamber systems at different locations in the East Asia. Total of 29 experiments were conducted in 9 locations: Shanghai in China, Ha Noi in Vietnam, Lampang, Phitsanulok and Pathumtani in Thailand, Hiratsuka, Kisai, Abiko and Akagi in Japan. Although simple linear regression analysis failed dose response relationships between growth responses expressed as NF/CF in dry mass of plant and mean concentration of ambient O_3, multiple linear regression analysis suggested that cumulative daily mean O_3 (CDMO, ppb/days) could be predicted by NF/CF in dry weight ratio of biomass, mean temperature and relative humidity as describe as below;

Equation from *R. sativus* cv. Red Chime:

CDMO (ppb/Days) = 5.8903-0.8756x(NF_{DW}/CF_{DW})-0.4557x(RH/T)-0.0044x(TxDays)

Equation from *B. campestris var. rosularis* cv. ATU171:

CDMO (ppb/Days) = 5.2547-0.2682x(NF_{DW}/CF_{DW})-0.5280x(RH/T)-0.0039x(TxDays)

Where

NF: Dry mass of plants grown in the non-filtered air chamber (NF)

CF: Dry mass of plants grown in the activated charcoal-filtered air chamber (CF)

DW: Total dry weight of top and hypocotyl for *Raphanus* or top for *Brassica.*

Days: Days for cultivation of plants after germination of seeds in the chamber

RH: Mean relative air humidity (%) during cultivation of plants grown in the chamber

T: Mean air temperature (C) during cultivation of plants grown in the chamber

This exposure system and predicting model may support environmental activities in developing countries[1].

(3) Establishing provisional critical levels and mapping of exceedance

① Estimation and mapping of pollutant concentration and deposition in East Asia

A chemical transport model developed by US EPA, CMAQ, was applied to East Asia and Japan in order to estimate spatial distributions of ground-level concentrations of acidic and acidifying substances. The distribution fields of SO_2 concentrations were estimated for a vast Asian domain including South, East and Southeast Asia from March 2001 to March 2002.

The distribution fields were remarkably influenced by emission distributions of SO_2, with higher concentrations in the winter and lower concentrations in the summer. These results were submitted to an air quality model intercomparison project for East Asia, MICS-Asia. In comparisons with measurements provided in MICS-Asia, our performance was relatively high among the eight participating models. Our estimates were more accurate in lower concentration areas but less in higher concentration areas. Similar tendencies were also found in the other models.

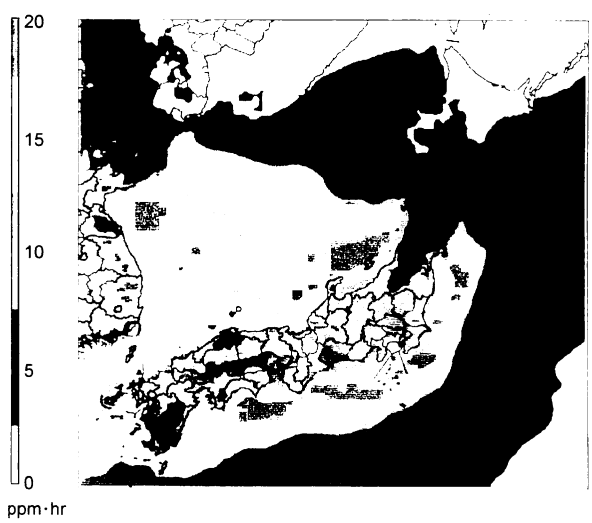


Fig. 1. Estimated spatial distribution of AOT40 in 2000

For the mainland of Japan, ozone concentrations, AOT40 and nitrogen depositions were estimated for the warmer months of April to September, 2000. The estimates well agreed with measurements. Mean concentrations of ozone were relatively high in other areas than large cities, except for summertime. However, AOT40, which evaluates ozone concentrations in daytime, was relatively high in large cities and industrial areas of Kanto, Chukyo and Seto Inland Sea coast (Fig. 1). Total (wet+dry) nitrogen depositions from total ammonia and total nitrate were relatively large in urban areas and south Kyusyu, where dry depositions of ammonia were very large.

② Construction of GIS database on air quality and vegetation

Forest health monitoring data, long term monitoring data of air pollutants, the vegetation survey data and other related information were collected and transformed into GIS storage system. Concentration of photochemical oxidants (Ox) in Japan is officially monitored. In addition, excess hours over the standards were recorded but 24-hour data was not presented the tables. Relationship between total excess hours and observed AOT40 (Ox) was analyzed and an equation to calculate AOT40 from excess hours was introduced as described in Fig.2. Total excess hours are the sum of the excess hours of 0.06 ppm and the over hours of 0.12 ppm in April to September.

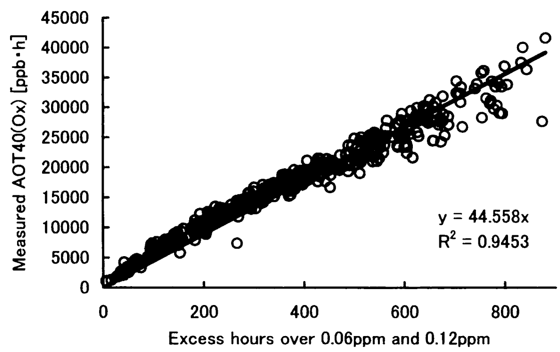


Fig.2 Relationship between total excess hours of photochemical oxidants over 0.06 and 0.12 ppm, and measured AOT40 (Ox)

Average AOT40(Ox) was calculated from the GIS data base transformed from nationwide air quality monitoring data from 1975 to 2003. Recent nationwide average of SO_2 was about 5ppb and AOT40 (Ox) was 12ppm-h which was about three times the early 1980's (Fig. 3). GIS analysis also revealed trend of nationwide AOT40(Ox) as shown in Fig.4.

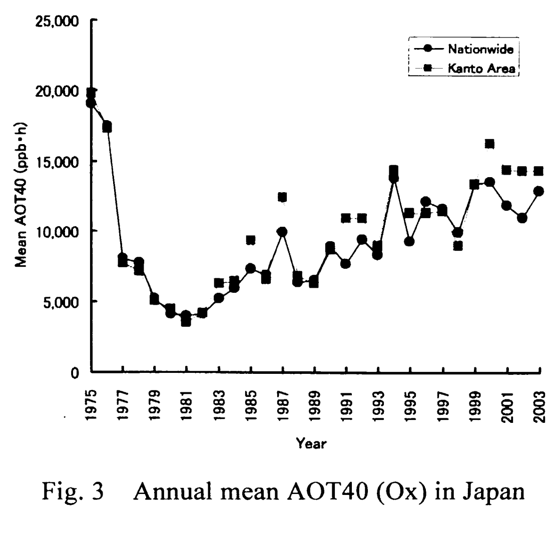


Fig. 3 Annual mean AOT40 (Ox) in Japan

Individual critical level of O_3 for the representative sensitive species in Japanese forest trees was summarized about 17 ppm-h of AOT40 based on the exposure experiments. Assuming that this would be applicable to natural conditions and mature forest stands or trees, GIS overlay analysis in the distribution of AOT40(Ox) and *P. densiflora*, *L. kaempferi* or *F. crenata* were conducted. It suggested that areas exceeding such a critical level expanded gradually in recent years.

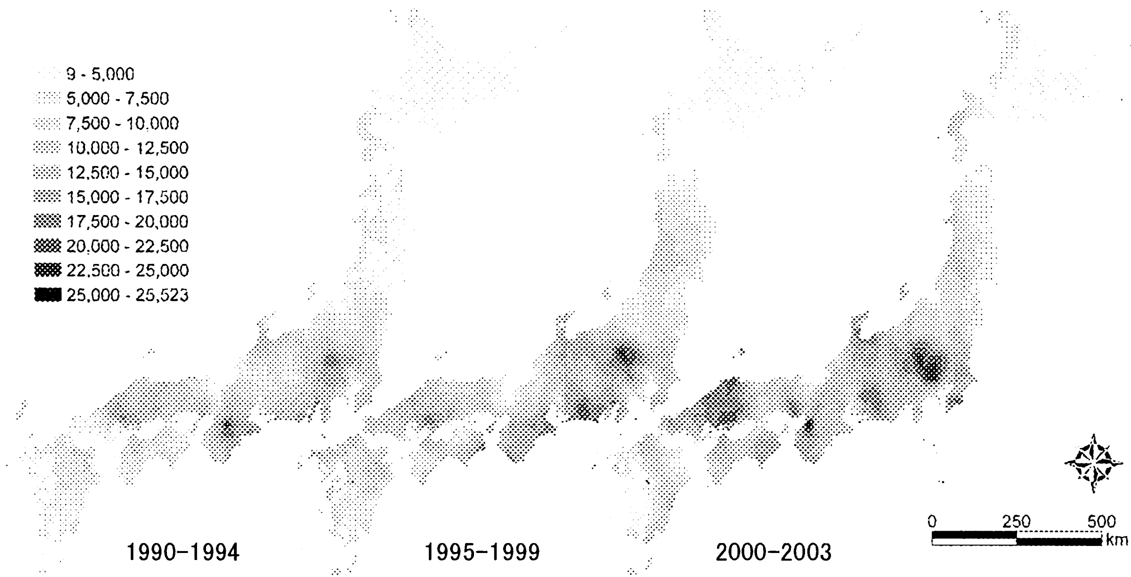
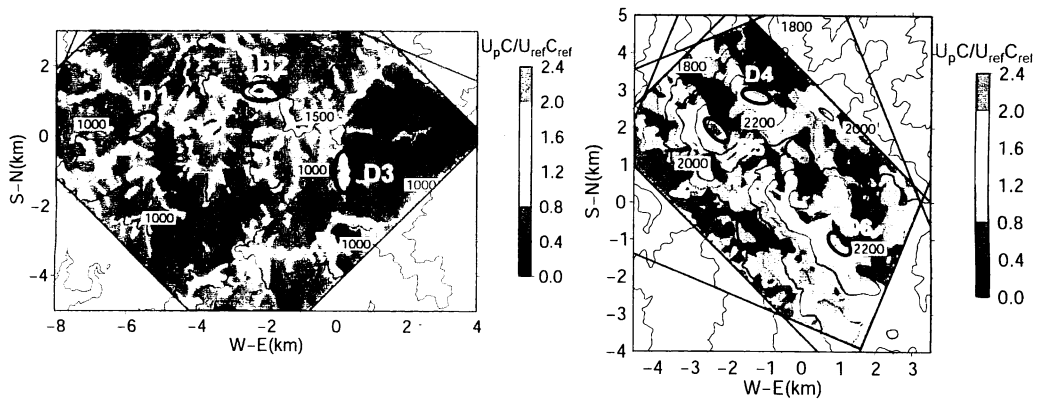


Fig. 4 Trend of mean AOT40 (Ox) in Japan

③ Numerical analysis of effects of wind and oxidative substances on high mountains

Dry deposition model of ozone (O_3) for evaluating in complex terrains was newly designed, and the numerical simulation of wind and O_3 transport was performed in Tanzawa Mountains, Kita-Yatsugatake Mountains and Mt. Tsurugi. As a result, wind velocity and O_3 concentration fields were found to be decisively affected by complex terrains. Areas of strong wind corresponded well with the forest (tree) decline areas at high altitudes, and O_3 concentration also tended to become higher there. Consequently, trees near the forest decline areas in high elevation sites were suggested to be considerably affected by strong wind and background O_3 (Fig. 5). Advection flux will be a good indicator for predicting possible forest/tree decline areas in the high elevation sites. However, experimental verification of advection flux will be required.



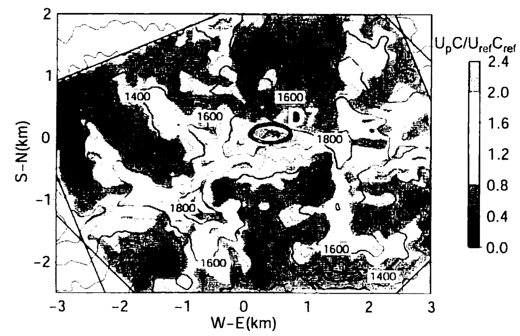


Fig. 5 Distribution of O_3 advection flux at a ground height of 10m
Upper left: Tanzawa Mountains
Upper right: Kita-Yatsugatake Mountains
Lower left: Mt. Tsurugi
Solid line: contour line [m]
Ellipse: ascertained tree decline area

④ Provisional critical levels of air pollutants

Europe has already established tentative critical levels of air pollutants based on the dose responses[2, 3]. Current ozone level has a potential adverse impact to plants. Trials to present exceeding maps of pollutants would help a risk assessment of the impacts on forests health and to design environmental regulations for protecting vegetation[4].

Current program summarized experimental results of exposure to SO_2 and O_3 in tree species with or without N treatments. Assuming that individual critical level of O_3 in sensitive species would be 17 ppm-h of AOT40 for 6 months from April to September as a critical level and it would be equivalent to Ox, rounded value of 20 ppm-h was proposed as a provisional critical level of O_3 (Table 1). Area exceeding this value in *P. densiflora* was indicated as an example in Fig. 6. It suggested that exceeding area increased recently and pine stands in such areas might have a greater threat of Ox. *Populs maximowiczii* and *Poplus nigra* were quite sensitive to O_3, however, they would be minor domestic or not native species in Japan. Critical level for such a specific sensitive species should be 10 ppm-h of AOT40. In contrast to the low elevation sites, mountain areas may have an extremely high dose of O_3 due to high concentration in nighttime as well as daytime. As a whole day dose should be considered to evaluate the effects of O_3[5], we also should take account of whole day value of AOT40 for high elevation sites.

Critical level for sensitive tree species to SO_2 ranged from 5 to 20 ppb as an annual mean. That for the most sensitive species: eastern white pine (*P. strobus*) and *Betula* *platyphylla var. japonica* was 5-6 ppb, however, considering distribution of such forests and ambient quality in Japan, we ignored to set such a strict level. In contrast, East Asian countries except Japan, the regional model revealed areas exceeding 20 ppb and 10 ppb of annual mean. Consequently we proposed annual mean SO_2 of 20 ppb as a primary provisional critical level and 10ppb as secondary level.

Nitrogen load may bring advantage for many species for their growth performance, however, *F.crenata* may receive adverse effect of increased input of N under O_3 stress. Current analysis suggested that most of species do not need to concern about combined effect of current N load with SO_2 or O_3 except *F. crenata*. As long-term load in the higher range of N input may have a potential ecological risk, tentatively we proposed 20 kg ha^{-1} $year^{-1}$ as a provisional critical level of the total N load for East Asian countries.

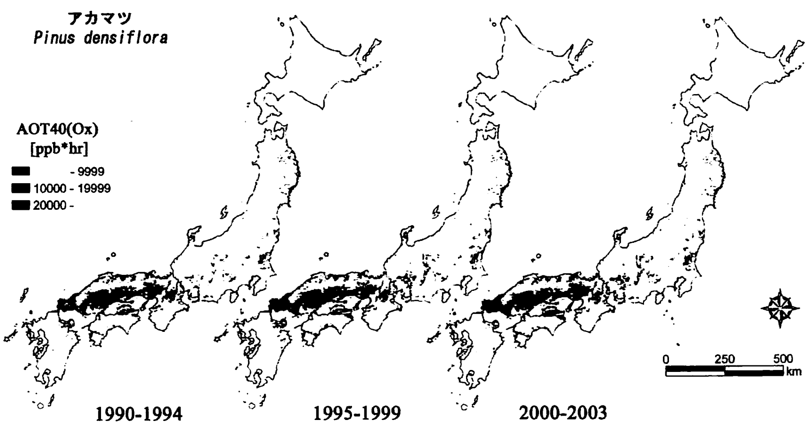


Fig-6 Distribution of *Pinus densiflora* and exceeded areas of provisional critical level of ozone
Provisional critical level of ozone: AOT40(Ox)=20ppm•h
Red area exceeds a provisional critical level of ozone.

Table 1 Provisional critical levels for forests and trees for East Asian region

Pollutant	Category	Critical level	
SO_2	Primary level	Annual mean	20 ppb
	Secondary level	Annual mean	10 ppb
	Third level	Annual mean	5 ppb
O_3	Low elevation sites	AOT40(12h)	20 ppm-h
	High elevation sites	AOT40(24h)	20 ppm-h
N deposition		Total-N(Wet+Dry)	20 kg/ha/year

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