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Evaluation of light dependence of monoterpene emission and its effect on surface ozone concentration

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HIGHLIGHTS

- Monoterpene emission from Japanese conifers was evaluated by growth camber method.
- Dependence of monoterpene emission rates on light intensity was clearly observed.
- Effect of light-dependent monoterpene emission on O_3 was evaluated by WRF/CMAQ.
- Monoterpene generally played a role of reducing O_3 concentration.
- Daytime monoterpene emission increase slightly reduced daily maximum O_3.

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ABSTRACT

This study evaluated the effect of light intensity on monoterpene emission from the three dominant coniferous tree species in Japan (*Cryptomeria japonica*, *Chamaecyparis obtusa* and *Pinus densiflora*). Monoterpene emission experiments were conducted by using a growth chamber where temperature and light intensity can be controlled. In the experiments, air temperature was set at 30 °C and light intensity was set at 0, 500, 700, 850, 1200, and 1400 μmol m^{-2} s^{-1}. Because monoterpene emissions from the three tree species similarly increased with increasing light intensity, a new empirical equation considering light dependence was proposed to estimate monoterpene emission. In addition, monoterpene emission in the Kinki region of Japan was estimated with and without light dependence using meteorological field produced by the Weather Research and Forecasting model (WRF) in summer 2010. The monoterpene emissions estimated with light dependence were larger than those without light dependence in the daytime under clear sky conditions and consistently smaller in the nighttime. In order to evaluate the effect of light dependence of monoterpene emission on ozone concentration in the Kinki region, two cases of air quality simulations by the Community Multiscale Air Quality model (CMAQ) were conducted using the monoterpene emission data estimated with and without light dependence. Comparisons of the two cases showed that the monoterpene emission changes due to light dependence slightly but systematically affected ozone concentrations. Monoterpene generally played a role of reducing ozone concentration in the CMAQ simulations. Consequently, because of the light dependence, the mean daily maximum ozone concentrations decreased by 0.3 ppb on average with a maximum of 2.2 ppb, and the mean daily minimum values increased by 0.4 ppb on average with a maximum of 1.8 ppb in the Kinki region in summer 2010.

1. Introduction

It is recognized that much amount of biogenic volatile organic compounds (BVOC) are emitted from plants to the atmosphere.

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Among various species of BVOC, isoprene and monoterpenes are known as the dominant species. BVOC play an important role in atmospheric chemistry controlling ozone (O_3) and secondary organic aerosols in the troposphere (Yokouchi and Ambe, 1985; Goldstein and Galbally, 2007). Therefore, better understanding of atmospheric processes of BVOC, including emissions and chemical reactions, is required for reliable assessment of air quality.

BVOC emissions from trees are affected by various environmental factors, such as leaf temperature (Tingey et al., 1980; Guenther et al., 1993; Schuh et al., 1997), light intensity (Schuh et al., 1997; Demarcke et al., 2010), relative humidity (Vallat et al., 2005), water content of the leaf (Lamb et al., 1985) and ozone concentration (Peñuelas et al., 1999). Among these environmental factors, temperature and light intensity are known to be the most important (Staudt and Lhoutellier, 2011). In the most frequently used equations for BVOC emission estimates by Guenther et al. (1993), isoprene emission depends on temperature and light intensity, and monoterpene emission depends on only temperature. Meanwhile, Bao et al. (2008) showed the possibility of monoterpene emission dependence on light intensity for Japanese dominant coniferous tree species.

There are several approaches to estimate light-dependent monoterpene emissions. Bertin et al. (1997) and Ciccioli et al. (1997) directly applied the light-dependent isoprene algorithm by Guenther et al. (1993) to estimate of monoterpene emission. Bäck et al. (2005) proposed a close relationship between photosynthesis and terpenoid emissions using carbon dioxides (CO_2) concentrations. Hybrid monoterpene algorithms that combine traditional light-independent algorithms with light-dependent components have also been used (Schuh et al., 1997; Schurgers et al., 2009; Guenther et al., 2012).

Many studies have been carried out to estimate BVOC emissions in various areas, including global scale (Guenther et al., 1995), the United States (Sakulyanontvittaya et al., 2008), Europe (Zemankova and Brechler, 2010), China (Zhihui et al., 2003) and Japan (Bao et al., 2008). In addition, Guenther et al. (2006, 2012) developed the Model of Emissions of Gases and Aerosols from Nature (MEGAN) designed for both global and regional emission modeling, which is widely used to estimate natural emissions for air quality modeling (Itahashi et al., 2013; Kajino et al., 2013; Chatani et al., 2014).

Various modeling studies have been conducted to evaluate the effect of BVOC on the atmospheric environment. Vogel et al. (1995) showed that the contribution of BVOC emission to ozone concentration reached 18 ppb (v/v) during high air temperature episodes in Germany. Solmon et al. (2004) found an increase in simulated ozone concentrations by 18–30% after including summertime biogenic emissions in France. Bao et al. (2010) showed that summertime BVOC emissions contributed to an increase in the daily maximum O_3 concentration by 6 ppb on average in Japan. Situ et al. (2013) reported that the impact of BVOC emissions on the surface ozone peak was 3 ppb on average with a maximum of 25 ppb in autumn, and 10 ppb on average with a maximum value of 34 ppb in summer in southern China.

In general, BVOC emissions are measured by outdoor field experiments, such as branch enclosure techniques (Matsunaga et al., 2013) and open-top chamber methods (Llusià et al., 2002). While such experiments can be conducted in natural forests, it is difficult to evaluate the effect of environmental factors. In contrast, a growth chamber method can control environmental factors although the method can measure emissions only from small trees.

In our previous studies, Bao et al. (2008) employed a growth chamber method to measure BVOC emissions from the nine most dominant tree species in the Kinki region of Japan (*Quercus serrata*, *Quercus crispula*, *Fagus crenata*, *Quercus acutissima Carruthers*, *Quercus glauca* and *Quercus myrsinaefolia*, *Cryptomeria japonica*, *Chamaecyparis obtusa* and *Pinus densiflora*) at the standard condition (air temperature: 30 °C; photosynthetically active radiation (PAR): 1000 μmol m^{-2} s^{-1}). Isoprene and monoterpene were dominantly emitted from the broadleaf and coniferous tree species, respectively. Bao et al. (2010) estimated BVOC emissions using the experimental data of Bao et al. (2008), conducted air quality simulations to evaluate the effect of BVOC emissions, and showed the large contribution of BVOC to ozone production in the region in summer 2002.

This study focused on light dependence of monoterpene emission and its effect on surface ozone concentration. First, growth chamber experiments were conducted in order to evaluate the effect of light intensity on monoterpene emission from the dominant coniferous trees (*C. japonica*, *C. obtusa* and *P. densiflora*). Second, air quality simulations by the Community Multiscale Air Quality model (CMAQ) (Byun and Schere, 2006) with the Weather Research and Forecasting model (WRF) (Skamarock and Klemp, 2008) were conducted in order to evaluate the effect of change in monoterpene emission due to light dependence on ozone concentration in the Kinki region in summer 2010.

2. Materials and methods

2.1. Procedure of growth chamber experiment

2.1.1. Target trees

The three most dominant coniferous trees in Japan (*C. japonica*, *C. obtusa* and *P. densiflora*) were selected as target trees. The trees used in the experiments were 3- to 5-year-old saplings with 0.8–1.2 m heights that were planted in 10-L plastic pots. They were grown in open air and regularly watered and fertilized during the growing season to provide optimal growth conditions.

2.1.2. Monoterpene sampling procedure

The measurement of monoterpene emissions were performed by using an 8800-L closed growth chamber (ESPEC MIC, TGE-3), in which air temperature and light intensity can be controlled. Six plants of each species were grown outside and individually transferred 24 h before the experiments to the chamber that is ventilated until the beginning of the experiments. During the experiments, the growth camber was closed, and air temperature and light intensity in the chamber were set to specific conditions. Air temperature was set to 30 °C, which is the standard temperature of an equation for monoterpene emission estimate proposed by Guenther et al. (1993). Light intensity was measured with a photometer (PREDE, PAR-01) and was set to constant values during each experiment (PAR flux: 0, 500, 700, 850, 1200, and 1400 μmol m^{-2} s^{-1}). The experiment with PAR flux higher than 1400 μmol m^{-2} s^{-1} could not be conducted due to the limitation of the equipment. Air inside the growth chamber was sampled by using a 200 mg Tenax TA adsorbent tube (Supelco, mesh 60/80) and a vacuum pump (GL Science, SP208-1000Dual) with a flow rate of 100 mL min^{-1}. Air samples of 12 L were collected at every two hours for *C. japonica* and *C. obtusa*, and 6 L at every hour for *P. densiflora*.

2.1.3. Chemical analysis procedure

This study used Gas Chromatograph Mass Spectrometer (Shimadzu, GC/MS-QP 2010) equipped with Thermal Desorber (PerkinElmer, ATD-50) to quantify nine kinds of monoterpenes in the growth chamber: α-Pinene, β-Pinene, β-Myrcene, α-Phellandrene, α-Terpinene, p-Cymene, d-Limonene, γ-Terpinene, and Terpinolene. The trapped compounds into adsorbent tubes were thermally desorbed at 280 °C by ATD connected to GC/MS. The separation of monoterpenes was performed by a capillary column (30 m × 250 μm × 0.25 μm, J&W Scientific). The carrier gas was