



Effects of soil water content and elevated CO₂ concentration on the monoterpene emission rate of *Cryptomeria japonica*

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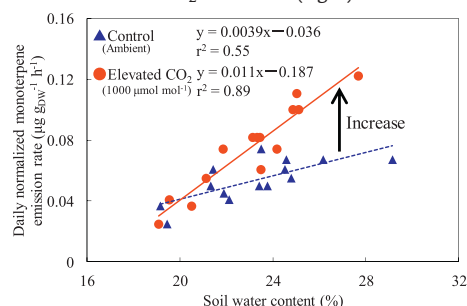


HIGHLIGHTS

- *C. japonica* clone saplings were grown under ambient and elevated CO₂ conditions.
- No difference in monoterpene compositions was observed between the two treatments.
- Monoterpene emission depended on both temperature and soil water content.
- Degree of soil moisture effect on monoterpene emission differed by CO₂.
- Considering combined effects of CO₂ and soil moisture improved the emission estimate.

GRAPHICAL ABSTRACT

The experimental setup (left) and the relationships between the daily normalized monoterpene emission rate and soil water content of *C. japonica* under control and elevated CO₂ conditions (right).



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ABSTRACT

Monoterpenes emitted from plants contribute to the formation of secondary pollution and affect the climate system. Monoterpene emission rates may be affected by environmental changes such as increasing CO₂ concentration caused by fossil fuel burning and drought stress induced by climate change. We measured monoterpene emissions from *Cryptomeria japonica* clone saplings grown under different CO₂ concentrations (control: ambient CO₂ level, elevated CO₂: 1000 µmol mol⁻¹). The saplings were planted in the ground and we did not artificially control the SWC. The relationship between the monoterpene emissions and naturally varying SWC was investigated. The dominant monoterpene was α-pinene, followed by sabinene. The monoterpene emission rates were exponentially correlated with temperature for all measurements and normalized (35 °C) for each measurement day. The daily normalized monoterpene emission rates ($E_{s0,10}$) were positively and linearly correlated with SWC under both control and elevated CO₂ conditions (control: $r^2 = 0.55$, elevated CO₂: $r^2 = 0.89$). The slope of the regression line of $E_{s0,10}$ against SWC was significantly higher under elevated CO₂ than under control conditions (ANCOVA: $P < 0.01$), indicating that the effect of CO₂ concentration on monoterpene emission rates differed by soil water status. The monoterpene emission rates estimated by considering temperature and SWC (Improved G93 algorithm) better agreed with the measured monoterpene emission rates, when compared with the emission rates estimated by considering temperature alone (G93 algorithm). Our results demonstrated that the combined effects of SWC and CO₂ concentration are important for controlling the monoterpene emissions from *C. japonica* clone saplings. If these relationships can be applied to the other coniferous tree species, our results may be useful to improve accuracy of monoterpene emission estimates from the coniferous forests as affected by climate change in the present and foreseeable future.

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1. Introduction

Biogenic volatile organic compounds (BVOCs) are emitted by vegetation. They may act against abiotic stress such as oxidative and heat stresses and biotic stress including insect herbivory (Niinemets et al., 2010 and references therein). Annual BVOC emissions on a global scale (about 1000 Tg) are estimated to be several times greater than annual anthropogenic VOC emissions (Boucher et al., 2013). Monoterpene emissions account for about 15% of the total BVOC emissions (160 Tg year⁻¹) (Guenther et al., 2012). Monoterpenes react greatly with OH radicals and O₃ in the atmosphere and produce biogenic secondary organic aerosols (BSOAs) (e.g., Mochizuki et al., 2015a). BSOAs are highly water-soluble and act as cloud condensation nuclei to form cloud droplets (Engelhart et al., 2008), which contribute to direct and indirect radiative forcing and affect the radiation budget of the Earth's atmosphere (Kanakidou et al., 2005; Boucher et al., 2013). Monoterpenes contribute to the formation of the photochemical oxidant in the presence of NO_x. High levels of photochemical oxidants degrade air quality and negatively affect human health (Ebi and McGregor, 2008).

Before industrial revolution, atmospheric CO₂ concentration is estimated to be about 280 μmol mol⁻¹. Since industrial revolution, the global atmospheric CO₂ concentration has been increasing and reached about 400 μmol mol⁻¹ in 2016 (Cubasch et al., 2013). Monoterpene emissions from *Quercus ilex* L. (Loreto et al., 2001) and *Larix gmelinii* var. *japonica* × *Larix kaempferi* (hybrid larch F₁) (Mochizuki et al., 2017) have been reported to decrease under elevated CO₂ concentrations. Loreto et al. (2001) have reported that the decreased monoterpene emission rate of *Q. ilex* under elevated CO₂ concentration (700 μmol mol⁻¹) was correlated with monoterpene synthase activity. However, elevated CO₂ concentration had no effect on the monoterpene emission of *Pinus sylvestris* (Räsänen et al., 2008). The effects of elevated CO₂ concentration on the monoterpene emission of trees seem to differ among species.

Simpraga et al. (2011) and Wu et al. (2015) performed laboratory experiments and reported that monoterpene emissions from non-storing species were positively correlated with soil water content (SWC). They suggest that either monoterpene synthase activity or production of the monoterpene precursor is inhibited by drought. Mochizuki et al. (2014) performed a field experiment and reported that monoterpene fluxes in a *Larix kaempferi* forest were increased after rain events and monoterpene fluxes had a strong positive correlation with SWC. Lin et al. (2007) suggest that drought conditions may decrease monoterpene synthesis in tree roots and by soil microbes, resulting in a decrease in monoterpene emission rates. Therefore, soil moisture conditions may be an important factor that controls monoterpene emissions from trees. Since precipitation is expected to change in the foreseeable future (Dai, 2013), soil water status may be an important factor for predicting monoterpene emissions.

It is unclear whether elevated CO₂ concentration and SWC independently affect monoterpene emissions or affect it in combination with each other. The BVOC emission models proposed to date, such as MEGAN2.1 (Guenther et al., 2012) and the model based on DGVM LPJ-GUESS (Smith et al., 2001; Schurgers et al., 2009), consider the effects of SWC and CO₂ concentration on isoprene emission only. For monoterpene emission, the models do not consider the single and combined effects of SWC and CO₂ concentration. Addressing these effects can contribute to the improvement of BVOC emission models.

A coniferous tree species *Cryptomeria japonica* is endemic to Japan tree species and occupies 21% of the total forest area in Japan (Forest Agency, 2009). The monoterpenes emitted by *C. japonica* have been reported to depend on temperature (Matsunaga et al., 2011), as do most monoterpene emitters. Okumura et al. (2013) performed a laboratory experiment and reported that light intensity (100–500 photosynthetic photon flux density) has little effect on monoterpene emission from *C. japonica*. The effects of SWC and CO₂ concentration on the monoterpene emission rates of *C. japonica* have not yet been addressed. In this

study, to investigate effect of CO₂ concentration on monoterpene emissions from *C. japonica*, we grew *C. japonica* clone saplings by using open-top-chambers (OTCs) (see graphical abstract) at different CO₂ concentrations (ambient CO₂ level and 1000 μmol mol⁻¹). The CO₂ concentration of 1000 μmol mol⁻¹ was chosen following the RCP scenarios 6 and 8.5. In addition, we measured the monoterpene emission rates of the saplings under different soil water status to investigate the effect of naturally varying SWC on the monoterpene emissions from *C. japonica*.

2. Materials and methods

2.1. Experimental design

Two-year-old *C. japonica* clone saplings that produce a small amount of pollen were purchased from a local nursery. We prepared eight plots for the OTC experiments (60 cm × 60 cm) at the University of Shizuoka (34°59' N, 138°26' E, 110 m above sea level). The OTC has an air-mixing chamber next to the fumigation chamber. Four *C. japonica* clone saplings were planted in the ground in each of the experimental plots on March 21, 2013. The saplings were enclosed by the OTC on April 11. The experimental setup has been described by Mochizuki et al. (2015b) and Tani et al. (2017).

Two experimental treatments were established: ambient CO₂ conditions (control) and elevated CO₂ conditions (elevated [CO₂]) (target: 1000 μmol mol⁻¹ during the daytime). Four OTCs were used as replicates for each treatment. For elevated [CO₂], pure CO₂ was supplied from a CO₂ cylinder into the air-mixing chamber for dilution with ambient air. The CO₂ concentration was continuously monitored with a non-dispersive infrared gas analyzer (ZFP9, Fuji Electric, Japan). We did not measure CO₂ concentration in control chamber during the fumigation period. Before the start of the fumigation experiment (from April 21 to 28), it was measured to be 378 ± 20 μmol mol⁻¹. The daytime average CO₂ concentration under elevated [CO₂] ranged from 790 to 1140 μmol mol⁻¹ during the fumigation period. The saplings under elevated [CO₂] were fumigated with high CO₂ air for one growing season, from April 30 to October 7, 2013. The flow rates of the ambient air and high CO₂ air into the OTC were controlled at 1000 L min⁻¹ by adjusting the voltage of the power supply into two axial flow fans (MU1238A-11B, Oriental motor, Japan). The air temperature inside and outside the OTC, photosynthetic photon flux density (PPFD) at the top of the OTC, and SWC inside the OTC (depth: 10–30 cm) were continuously measured with a thermistor (VT3, CHINO, Japan), photodiode (DS-SR, KONA, Japan), and time-domain reflectometer (10HS, DECAGON DEVICES Inc., USA), respectively.

We did not artificially control the SWC. For our study site, rainfall occurred frequently during the measurement period. SWC varied from 19 to 30% during our OTC experiments except for rainfall events. The observed SWC range in the periods of monoterpene sampling was 19 to 29% (Table 1), which was sufficient to investigate the relationship between monoterpene emissions and SWC under both control and elevated [CO₂].

2.2. Monoterpene measurement

Measurements of the monoterpene emission rate of the trees under control and elevated [CO₂] were conducted from July 9 to August 10 (Table 1). Monoterpene sampling was conducted with a newly developed dynamic enclosure system by using the existing OTC (Mochizuki et al., 2015b). To prevent an inflow of ambient air, the open side (top) of the fumigation chamber was covered by a transparent acrylic plate with a cylinder-shaped ventilation duct (10 cm in diameter) in the center. Air from a compressed air cylinder was passed through a platinum catalyst heated to 400 °C to remove VOCs. The flow rate of the purified air into the OTC was adjusted to 10 L min⁻¹ using a mass flow controller (SEC-E40, HORIBA STEC, Japan). Before monoterpene sampling, fumigation chamber was flushed for >50 min at a flow rate of 10 L min⁻¹ with

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