



TITLE:

Vertical profiles of methane concentration above and within the canopy of a temperate Japanese cypress forest

AUTHOR(S):

Takahashi, Kenshi; Sakabe, Ayaka; Kanazawa, Akito; Kosugi, Yoshiko

CITATION:

Takahashi, Kenshi ...[et al]. Vertical profiles of methane concentration above and within the canopy of a temperate Japanese cypress forest. *Atmospheric Environment: X* 2021, 12: 100143.

ISSUE DATE:

2021-12

URL:

<http://hdl.handle.net/2433/269103>

RIGHT:

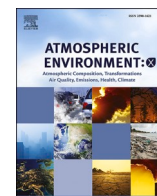
© 2021 The Authors. Published by Elsevier Ltd.; This is an open access article under the Creative Commons Attribution-NonCommercial-NoDerivatives 4.0 International license.



Contents lists available at [ScienceDirect](https://www.sciencedirect.com)

Atmospheric Environment: X

journal homepage: www.journals.elsevier.com/atmospheric-environment-x



Vertical profiles of methane concentration above and within the canopy of a temperate Japanese cypress forest

Kenshi Takahashi^{a,*}, Ayaka Sakabe^{b,c}, Akito Kanazawa^c, Yoshiko Kosugi^c

^a Research Institute for Sustainable Humanosphere, Kyoto University, Gokasyo, Uji, 611-0011, Japan

^b Kyoto University Hakubi Center, Yoshida-honmachi, Sakyo-ku, Kyoto, 606-8501, Japan

^c Graduate School of Agriculture, Kyoto University, Kitashirakawa Oiwake-cho, Sakyo-ku, Kyoto, 606-8502, Japan

ARTICLE INFO

Keywords:

Methane
Vertical profiles
Seasonal variations
Forest ecosystem
Continental outflow

ABSTRACT

Methane (CH₄) is one of the major greenhouse gases, and therefore its source identification and quantification are quite important. So far water-unsaturated soils of upland forests are usually recognized as sinks, but recent increasing evidence supports tree stems in certain environments can be a source of CH₄, and thereby research on the dynamics of CH₄ in forest ecosystems has entered a new phase. In this study, we measured vertical distributions of CH₄ concentrations within and above a forest canopy to examine how they are influenced by local ecological processes and synoptic-to local-scale atmospheric processes. Profile measurements were conducted using a meteorological tower in a temperate Japanese cypress forest with a 30-min time resolution. Time series data of CH₄ concentrations above the canopy at our site showed occasional spikes relative to background levels, and these spikes corresponded with temporal variations in CO₂ concentrations. Backward trajectory analyses showed that most of the air masses associated with the spikes in CH₄ and CO₂ concentrations had continental origins. Seasonal-mean diurnal properties in CH₄ concentrations above and within the canopy were weaker than those for CO₂ concentrations. It was considered likely that nighttime accumulation of CH₄ in a stable surface layer and its convective diffusion into a mixed layer accounted for the diurnal properties in CH₄, while ecological processes played a major role in the diurnal properties observed in CO₂ levels. Vertical distributions in CH₄ concentration within the canopy showed a monotonic decrease from the canopy to the forest floor, with variations depending on the season. The profiles agreed with soil-atmosphere CH₄ fluxes measured using a closed-chamber method at our site, indicating that the surface soil in upland areas plays a major role for determining the within-canopy distributions of CH₄. Also, it was inferred that at least at our site, CH₄ emissions from stems and leaves, which has become a hot topic over the recent years, was not significant for cypress trees in upland area enough to influence the within-canopy distributions of CH₄ during the study period.

1. Introduction

Atmospheric methane (CH₄) is an important greenhouse gas. Fluctuations in atmospheric CH₄ concentrations are largely determined by biogenic and abiotic emissions from the Earth's surface, reduction by gas-phase OH reactions in the atmosphere, and microbial oxidation in aerobic soils (Reeburgh, 2006). Although large annual fluctuations in atmospheric CH₄ concentrations have been reported (e.g., Bousquet et al., 2006; Rigby et al., 2008), the underlying mechanisms for these fluctuations remains controversial. Furthermore, although the major contributors to the global CH₄ budget have generally been identified, the relative contribution of individual sources has not yet been accurately

clarified.

Our group has attempted to clarify the dynamics of CH₄ in forest ecosystems. Specifically, we have been conducting long-term *in-situ* measurements of CH₄ fluxes using closed-chamber systems (Itoh et al., 2009; Takahashi et al., 2012; Sakabe et al., 2015, 2016) and a relaxed eddy accumulation system (Sakabe et al., 2012) at a Japanese cypress (*Chamaecyparis obtusa* Sieb. et Zucc) forest site in Japan. Our findings have shown that 1) the CH₄ flux over the forest canopy clearly shows seasonal fluctuations with daily-scaled changes that are particularly sensitive to Asian monsoon rainfall; 2) CH₄ exchange between the atmosphere and forest soils at the forest study site are heterogeneous at both temporal and spatial scales and show a complex response to

* Corresponding author.

E-mail address: takahashi.kenshi.2v@kyoto-u.ac.jp (K. Takahashi).

<https://doi.org/10.1016/j.aeaoa.2021.100143>

Received 20 April 2021; Received in revised form 20 October 2021; Accepted 19 November 2021

Available online 20 November 2021

2590-1621/© 2021 The Authors.

Published by Elsevier Ltd.

This is an open access article under the CC BY-NC-ND license

(<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

rainfall; and 3) no significant emission or uptake of CH₄ by intact leaves or trunks of individuals occurs in amounts that are detectable with our experimental system occurs in any season. In this study, we report the results of continuous, *in-situ*, one-year measurements of vertical distributions in CH₄ concentration within and above the forest canopy at the same site.

Measurements of the vertical distributions in CH₄ concentrations in forests provide insights into the CH₄ budget in these environments. Simpson et al. (1997) reported tower-based profile and micrometeorological flux measurements of CH₄ in a boreal aspen forest over a 5-month period, based on *in-situ* quantification of CH₄ concentrations with a lead-salt laser. In that study, although CH₄ emission was recorded above the canopy, uptake was evaluated from footprints closer to the tower where the soil was drier. de Carmo et al. (2006) performed vertical profile and closed-chamber measurements for CH₄ at three forest sites in Amazonia. A total of 75 profiles obtained on 19 dates by canister sampling followed by laboratory analysis estimated that net CH₄ emissions in upland forests ranged from 2 to 21 mg CH₄ m⁻² d⁻¹ in both dry and wet seasons at three sites. Bowling et al. (2009) studied tower profiles for CH₄ in a subalpine conifer forest in the Rocky Mountains. Their measurements were made at 2 h intervals over 6 weeks in summer and used *in-situ* quantification by a near-infrared diode laser spectrometer. They proposed that their CH₄ profiles were rationalized by net uptake of CH₄ by upland soils. Querino et al. (2011) undertook tower-based profile and eddy flux measurements of CH₄ in a tropical rainforest. Air was collected at different heights over a period of 6 h, and samples were analyzed in the laboratory. Their profiles reflected the net CH₄ production from the soil surface. Ueyama et al. (2014) inferred CH₄ fluxes in an upland forest based on the vertical profiles of CH₄ concentrations estimated using inverse multilayer models, in which tower-based observations were made in a Japanese larch forest in Yamanashi Prefecture, Japan. Their results showed that the forest acted as a CH₄ sink during the summer study period.

On the other hand, it should be noted that in recent years unique phenomenon of CH₄ release from tree trunks has attracted a great deal of attention as a potential impact for atmospheric CH₄ (Barba et al., 2019; Covey and Magonigal, 2018). In addition, multiple processes have been recognized to contribute in promoting CH₄ emission (Flanagan et al., 2021), and some studies have shown that they weaken the role of CH₄ sinks in forest ecosystems (Shoemaker et al., 2014; Pitz and Magonigal, 2017; Pitz et al., 2018; Ueyama et al., 2018), though it is well known that upland forests are generally CH₄ sinks largely due to surface soil in an aerobic environment (Ishizuka et al., 2000; Le Mer and Roger, 2001; Sakabe et al., 2015; Yu et al., 2017). Within-canopy profiles would provide insight into those issues of whether soil uptake and stem emission offset one another. In our previous chamber flux study (Takahashi et al., 2012), we did not significantly detect CH₄ emission from the stem of a single selected cypress tree, but it cannot be ruled out that some other cypress trees release CH₄ from the stems depending upon the growing environment, since our site is characterized by hillslopes and the activity of methanogens and methanotrophs in soil is spatially heterogeneous.

In this paper, we aimed to investigate how such plant ecophysiological processes and microbial activity in soil determine the within-canopy vertical distributions of CH₄ in a Japanese cypress forest and their seasonal properties. In order to accomplish the aim of this study, we conducted continuous one-year measurements of vertical distributions in CH₄ concentration along a meteorological tower, using a near-infrared laser spectroscopic technique for *in-situ* CH₄ detection. In addition, air-mass origin and the synoptic-scale weather systems may influence the CH₄ concentrations observed at the tower, because reactivity of hydroxyl (OH) radical toward CH₄ is low (Atkinson et al., 2004), resulted in a long lifetime in the troposphere. Therefore, we also aimed to extract the impacts of synoptic scale transport of CH₄ on the temporal variations in local CH₄ concentrations on a several days scale at our forest site.

2. Materials and methods

2.1. Study site

Vertical profiles of CH₄ concentrations were measured in the Kiryu Experimental Watershed (KEW) at 34°58'N, 136°00'E in Shiga Prefecture, central Japan, from August 2009 to August 2010. A detailed description of the site and a topographic map has been reported elsewhere (Kosugi et al., 2007; Ohkubo et al., 2007; Sakabe et al., 2015). The watershed has an area of 5.99 ha (1 ha = 10⁴ m²) and is mainly covered by Japanese cypress trees that were planted in 1959. The elevation range of the study site was 190–255 m. The canopy height was about 17 m. Our site also includes sparsely distributed Japanese red pine trees (*Pinus densiflora*) and several broadleaf tree species. Some wetlands are located in the riparian zones, which are either permanently or periodically submerged, along the streams within the watershed. The wetlands cover an area of approximately 0.2 ha (3.3% of the watershed). The meteorological tower was located in upland area. Air temperatures, which ranged from - 4.4 to 33.7 °C during the study period, were measured with a platinum thermometer (HMP45C, Vaisala, Helsinki, Finland) at a height of 29 m above the ground. Total precipitation was 1982 mm during the study period, as measured by a tipping bucket rain gauge (RT-5, Ikeda Keiki, Tokyo, Japan).

2.2. Experimental design

The vertical profiles of CH₄ and CO₂ concentrations were measured at five heights: 0.3, 1, 10, 20 and 29.5 m. Air was drawn continuously from each of the inlets through a polyfluoroacrylate tube (inner diameter: 4 mm) using a diaphragm pump at a flow rate of approximately 1.8 L min⁻¹. To analyze the CH₄ concentration of the sampled air, some of the main flow (0.7 L min⁻¹) was passed through a membrane dryer (PD-50T-48, Perma Pure, NJ, USA) to remove moisture before being diverted to a CO₂/H₂O analyzer (LI-840, LI-COR Biosciences, NE, USA) and a CH₄ analyzer (FMA-100, Los Gatos Research, CA, USA). Since the drying efficiency of such dryers has been reported to be dependent upon operating conditions (Leckrone and Hayes, 1997), we measured the residual water vapor exiting the membrane dryer using the LI-840 analyzer, and the data were used to compensate for the observed CH₄ concentrations. The vertical CH₄ and CO₂ concentration profiles were measured for 15 min at 30-min intervals. The remaining 15 min was allotted to closed-chamber measurements (Takahashi et al., 2012). The sampling height switched in order of 28.5, 20, 10, 1 and 0.3 m, using program-controlled solenoid valves. Sampling duration for each height was 42, 36, 28, 22 and 22 s, respectively. This cycle worked 6 times during 15 min. At each height, CH₄ and CO₂ concentrations for 15 s just before switching to the next height were averaged, by taking air flushing time within the measurement system into account, then the obtained averages were used to calculate the 6-times measurement mean. Measurements were conducted continuously from August 2009 to August 2010; however, the concentration data at all heights in July 2010 and at 20 m in August 2010 were not collected because of problems with the measurement system.

The CH₄ analyzer used in this study employed cavity-enhanced absorption spectroscopy with a diode laser at a wavelength of approximately 1.6 μm; the instrument permits interference-free real-time monitoring of CH₄ concentration at atmospheric levels (Hendriks et al., 2008; Smeets et al., 2009). Instrument performance was tested by using the Allan variance method (e.g., Eugster and Plüss, 2010). The mixing ratio of CH₄ in a dry compressed air cylinder (Masuda Medical Instruments, Kyoto, Japan) was measured over several hours with 1-Hz resolution, and resultant time series of the data providing 2496 ± 0.9 ppb (mean ± SD) were subjected to Allan variance analysis. This assessment suggested that the Allan deviation for this analyzer was 0.7 ppb with a 1-s integration time (i.e., ±0.03% against a background concentration of 2496 ppb), which is similar to that for a similar

instrument reported previously (Eugster and Plüss, 2010). The measurement precision for CH₄ in the profiling experiments was expected to be 0.24 ppb based on the Allan variance analysis, since our profiling schedule determined CH₄ concentration in one run from 15-s acquisition as described above. In the same manner, the measurement precision for CO₂ was expected to be 0.3 ppm at atmospheric concentrations. The span of our CH₄ analyzer was calibrated against standard gas containing atmospheric levels of CH₄ diluted in synthetic air (Kyoto Teisan, Kyoto, Japan). For the CO₂/H₂O analyzer, the CO₂ span was calibrated using a standard gas cylinder and water vapor using a dew-point generator (LI-610, LI-COR Biosciences, NE, USA), and the zero offset was checked with pure nitrogen gas.

3. Results and discussion

3.1. CH₄ concentration above the forest canopy

Fig. 1 shows the time series of CH₄ and CO₂ concentrations at a height of 29.5 m from September 2009 to August 2010 together with the meteorological data recorded at KEW. The time series of the CH₄ concentrations revealed the existence of several spikes that were superimposed on background measurements. Similar spikes were also observed in the time series obtained for the CO₂ concentration, and most of the spikes in CH₄ and CO₂ concentrations occurred simultaneously. Detailed examination of the data obtained for January and May are expanded and shown in Figs. 2 and 3. In both months, temporal changes in CH₄ and CO₂ concentrations at KEW were very similar on the scale of several days, although in May diel changes in CO₂ concentrations associated with biological processes were superimposed over long-term changes. For comparison, Figs. 2 and 3 include the CH₄ data obtained at two environmental monitoring stations at Kusatsu (35.01°N, 135.96°E; 6.2 km to the north-northwest of KEW) and at Higashiohmi (35.11°N, 136.20°E; 24.8 km to the northeast of KEW). Both stations are located in an urban area and are utilized by the Atmospheric Environmental Regional Observation System in Japan, which records CH₄ measurements at hourly intervals using gas chromatography. Temporal changes in CH₄ concentration at KEW are similar to those at Kusatsu. At Higashiohmi, more pronounced diurnal changes were overlaid on the background fluctuations which were close to those observed at Kusatsu. Spatial similarities in the background distribution of CH₄ concentrations

among three sites are consistent with the long tropospheric lifetime of CH₄ due to its low reactivity toward OH radicals (Atkinson et al., 2004). At Higashiohmi station, the CH₄ concentrations often showed maxima just before or near sunrise, which will be ascribed to the nighttime accumulation in the surface boundary layer. There could be local emissions of CH₄ near the station. Exploring the emission sources are, however, beyond the scope of this paper.

We observed that the spikes in CH₄ and CO₂ concentrations were frequently observed synchronously with decreasing atmospheric pressure and precipitation, particularly during the winter season (Fig. 2). Those observations indicate that low-pressure systems and/or low-pressure troughs pass over the site transporting continental outflows of anthropogenic emissions. Similar features were identified over Narita International Airport (35.8 N, 140.4 E) by Shirai et al. (2012) and at Minamitorishima (24.3 N, 154.0 E) by Wada et al. (2013). A detailed analysis of the mechanisms underlying the transport of surface emissions from continental Asia by low-pressure systems was reported by Bey et al. (2001). The significance of continental outflows reflected by the spikes in CH₄ and CO₂ concentrations at our site was further corroborated by a five-day backward trajectory analysis using NOAA's HYSPLIT model (Stein et al., 2015). For example, on May 21 (Fig. 3), a steep increase followed by a gradual decrease in the CH₄ concentration was observed, which was due to the transport of continental air masses (see Fig. A1 in the supplementary information). On 21–23 May, yellow sand (Kosa aerosol) was observed widely over Japan, which provides further evidence of a continental origin of transported air masses. We found that, including those days, air masses responsible for spiky CH₄ and CO₂ concentrations were mostly attributable to continental origins.

3.2. Vertical distribution of CH₄ concentration within the forest canopy

Fig. 4 shows the seasonal-mean diurnal variations of CH₄ and CO₂ concentrations at all heights of the 29-m tall tower and the friction velocities (u^*). The seasonal mean concentrations of CH₄ at all heights showed a peak in the morning, which was followed by a decrease during the day. Such a pattern could be explained by nighttime accumulation of CH₄ in the stable boundary layer and its daytime diffusion into the mixed layer. Possible causes for the nighttime accumulation include anaerobic CH₄ production in the riparian wetland area within the watershed (Sakabe et al., 2016) and transport of nearby urban

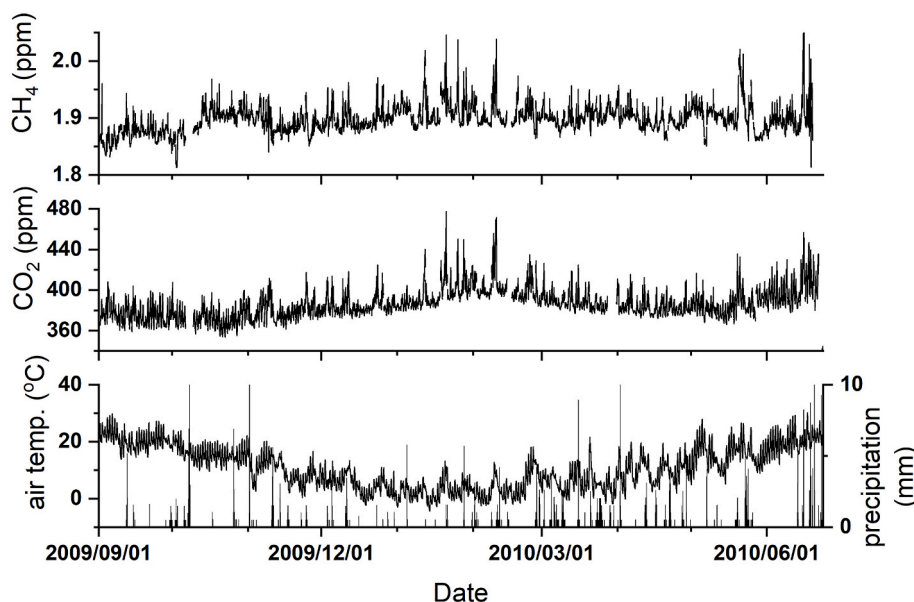


Fig. 1. CH₄ and CO₂ concentrations measured at a height of 29.5 m, air temperature at a height of 29 m above the ground at KEW, and precipitation measured at an open-screen site near the tower. All the data were obtained with 30-min resolution, which are plotted here as they are without any averaging.

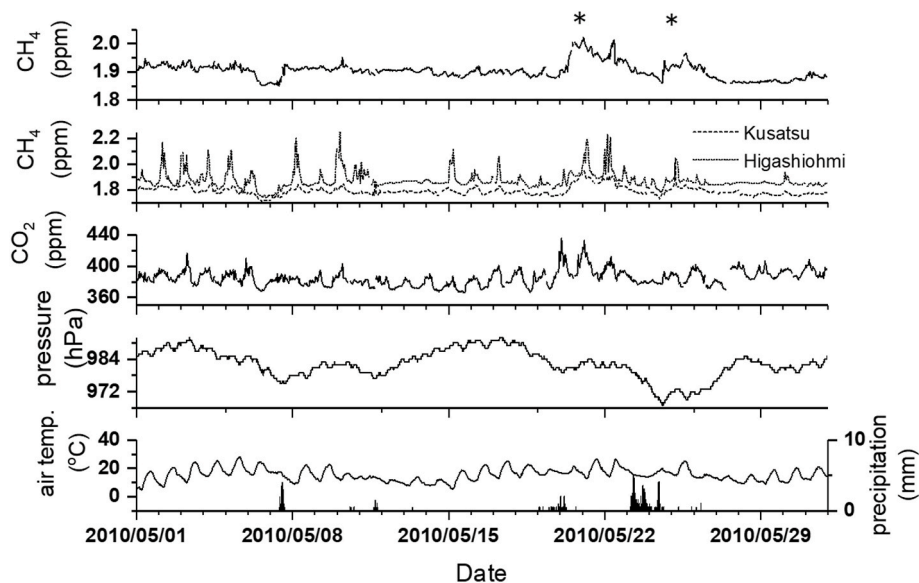


Fig. 2. CH₄ and CO₂ concentrations at a height of 29.5 m, air pressure, air temperature and precipitation measured at KEW, and CH₄ concentrations at the two air-quality monitoring stations of Kusatsu and Higashiohmi in January 2010. CH₄ concentrations at two air-quality monitoring stations were available with 1-h resolution, which are plotted here as they are without any averaging. Asterisks indicate the spiky events (see text).

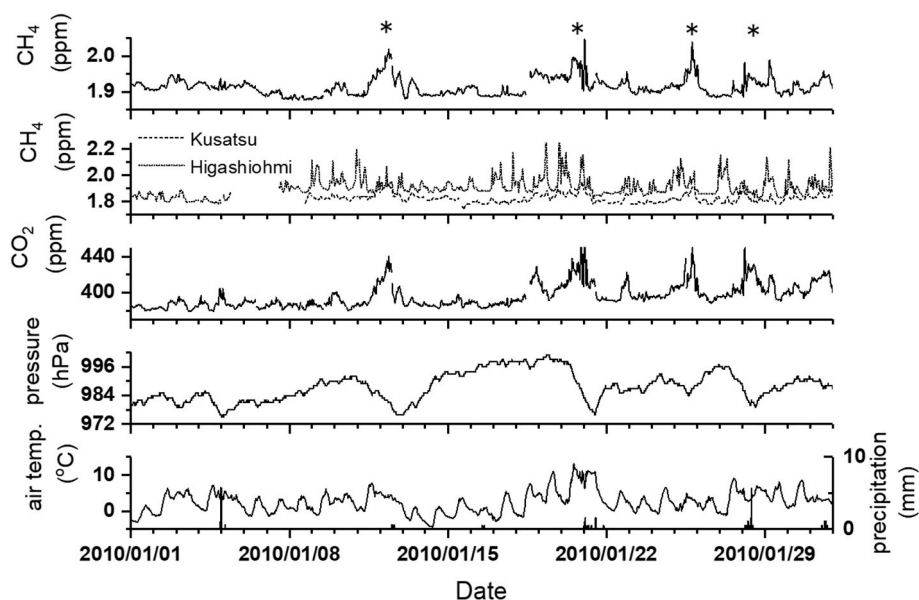


Fig. 3. Same as Fig. 2, for data collected in May 2010.

emissions. On the other hand, in the CO₂ data, seasonally dependent diurnal changes are governed by forest ecological and micrometeorological processes, as discussed in detail by [Ohkubo and Kosugi \(2008\)](#). [Fig. 4](#) also shows that, for all seasons, the CH₄ concentrations were lower near the forest floor than above the canopy, implying that the forest floor around the tower is a net sink for CH₄. The CO₂ concentrations were higher near the forest floor than above the canopy because of the large CO₂ efflux from soil ([Ohkubo et al., 2007](#); [Ohkubo and Kosugi, 2008](#)). To investigate the seasonality in the vertical profiles of CH₄ concentration, we plotted monthly concentration differences at four heights (20, 10, 1 and 0.3 m) relative to the concentration at 29.5 m, as shown in [Fig. 5](#). Our results showed that, (i) negative differences in the CH₄ concentrations were the largest at 0.3 m over the entire observation period, with maximum differences observed in summertime and minimum differences in wintertime, and (ii) such a tendency was

independent of day and night periods. This implies that soil oxidation acts as a sink in the canopy at KEW and that this process is seasonal. Around the observation tower, we measured CH₄ fluxes between the upland soil surface and the atmosphere using a closed-chamber method. The findings showed that uptake was highest in summertime and lowest in wintertime with its large dependence on the soil water contents of the study plot ([Sakabe et al., 2015](#)). Furthermore, as reported by [Sakabe et al. \(2012\)](#), forest-scale CH₄ fluxes measured by the relaxed eddy accumulation (REA) technique were negative (i.e., absorption from the atmosphere to the forest), with an increase in magnitude observed from April 2010 to June 2010. Their results corroborate the present profile data, which implies that microbial CH₄ oxidation in the upland soil surface was highest in summertime and lowest in wintertime.

Here, we assume that the soil CH₄ uptake rate is consistent with the observed concentration distributions. If we assume a scalar similarity

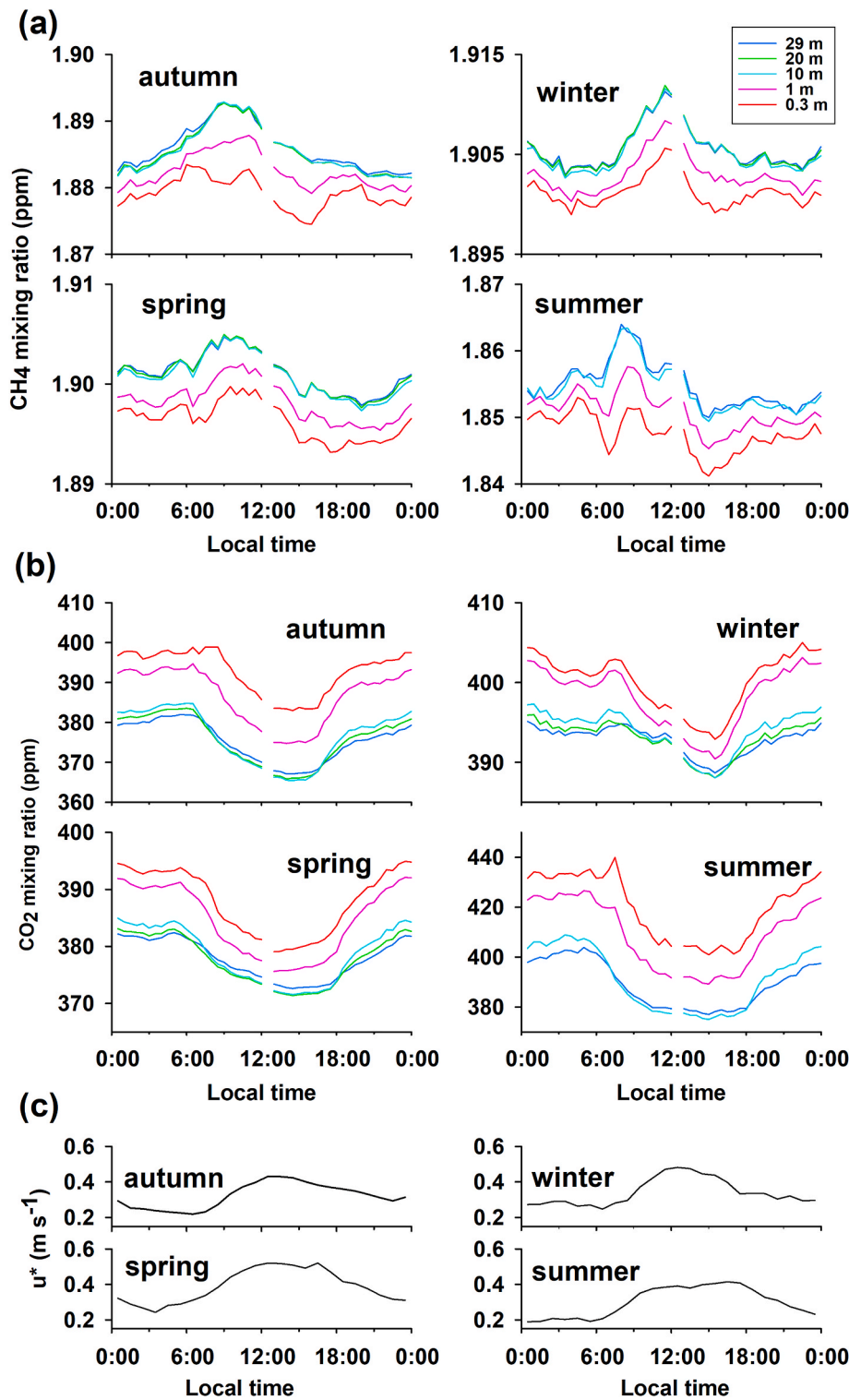


Fig. 4. Diurnal properties of (a) CH₄ and (b) CO₂ concentrations at five different heights along a 29 m-tall tower. Data were acquired in all four seasons: autumn (September 2009 to November 2009), winter (December 2009 to February 2010), spring (March 2010 to May 2010) and summer (June and August 2010). The data in July 2010 were not available due to trouble in the measurement system. Variabilities of the plotted data in panels (a) and (b) are numerically provided in Table A1. Panel (c) shows the diurnal properties in friction velocity (u^*) during the same period.

between CO₂ and CH₄, then the gradients of each gas near the ground should be related to the respective fluxes by the same eddy diffusivity. This is the basis of the flux-gradient method for the estimation of fluxes (Baldochi et al., 1988; Miyata et al., 2000; McMillan et al., 2007), and can be formalized as:

$$F_{CO_2} = -K \frac{d[CO_2]}{dz} \quad (1)$$

$$F_{CH_4} = -K \frac{d[CH_4]}{dz} \quad (2)$$

$$F_{CH_4} = -\frac{F_{CO_2}}{\frac{d[CO_2]}{dz}} \frac{d[CH_4]}{dz} \quad (3)$$

where F_{CO_2} and F_{CH_4} represent the flux of each gas, $d[CO_2]/dz$ and $d[CH_4]/dz$ are the gradients of each gas near the ground, and K is the eddy diffusivity. Equation (3) allows us to estimate the flux of CH₄ from

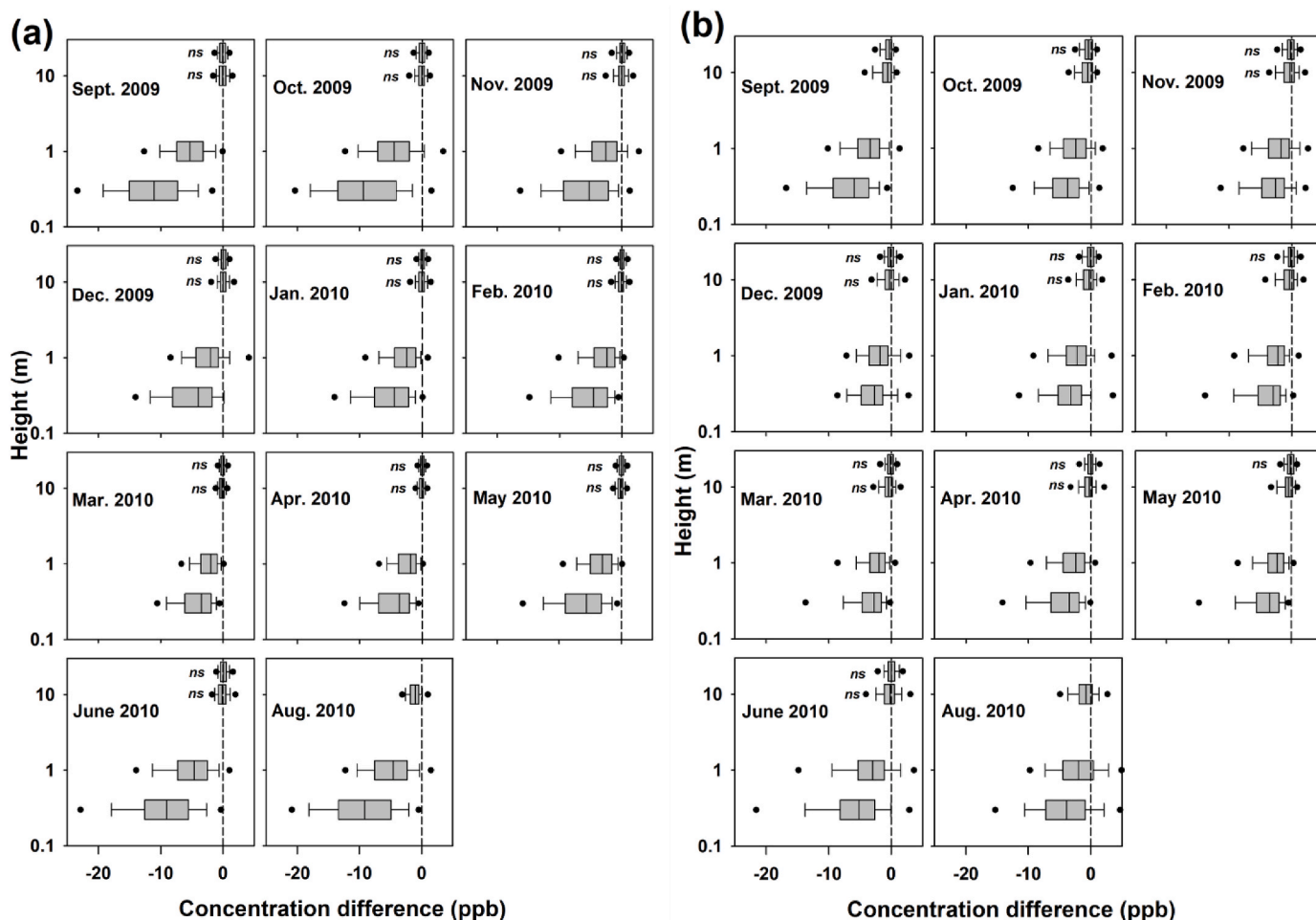


Fig. 5. Differences in average monthly CH₄ concentrations at 20, 10, 1 and 0.3 m relative to that at 29.5 m; (a) daytime data (07:00–18:30 LT), (b) nighttime data (19:00–06:30 LT). Vertical dashed lines are the reference lines at 0 ppb-difference. Boxes enclose all values between the 25th and 75th percentiles, whereas whiskers encompass all values within the 5th to 95th percentile range. Solid vertical bars in the boxes indicate median values. “ns” indicates that concentration differences at each height are not significantly different from 0 ($p > 0.01$). The concentration differences at 0.3 and 1 m were significantly different from zero and from each other for both daytime and nighttime data of all months ($p < 0.01$). The data at 20 m in August 2010 and at all heights in July 2010 were not available due to trouble in the measurement system.

a known CO₂ flux and measured gradients of each trace gas. Ohkubo et al. (2007) reported that the soil CO₂ efflux at KEW ranged from approximately 4 $\mu\text{mol m}^{-2} \text{s}^{-1}$ in summer to approximately 0.6 $\mu\text{mol m}^{-2} \text{s}^{-1}$ in winter. The $d[\text{CH}_4]/dz$ values that were calculated from the daytime and nighttime averages of CH₄ concentrations at heights of 0.3 and 1 m (Fig. 5) ranged from 3.0 $\text{nmol mol}^{-1} \text{m}^{-1}$ during the summertime to 1.7 $\text{nmol mol}^{-1} \text{m}^{-1}$ during wintertime. Similarly, the $d[\text{CO}_2]/dz$ values were -12.3 and -2.2 $\mu\text{mol mol}^{-1} \text{m}^{-1}$ during summertime and wintertime, respectively (Figure A2 in the supplementary information). Using these values with Eq. (3), we obtain the magnitude of the soil CH₄ uptake as 1 $\text{nmol m}^{-2} \text{s}^{-1}$ in summer and 0.4 $\text{nmol m}^{-2} \text{s}^{-1}$ in winter. Although the flux-gradient approach makes certain assumptions, such as horizontal homogeneity of the source and sink distributions of the target gases and the attainment of steady-state conditions, the calculated CH₄ uptake rates are in agreement with those obtained from the chamber flux measurements around the tower conducted in September 2009 to August 2010, and ranged between 0.38 and 2.16 $\text{nmol m}^{-2} \text{s}^{-1}$ with a median of 0.54 $\text{nmol m}^{-2} \text{s}^{-1}$ (Sakabe et al., 2015). As reported in Sakabe et al. (2015), the soil CH₄ fluxes were measured by using three closed chambers which were distributed around the meteorological tower within a radius of 30 m (a topographical map is shown in their paper), accordingly the above referred range of the soil CH₄ fluxes arose both from their seasonality and spatial heterogeneity. The seasonally

dependent uptake of CH₄ by the upland soil surface is directly related to the activity of methanotrophic bacteria, namely, with summer time maxima and winter time minima.

While it has generally been considered that water-unsaturated soils of upland forests can act as sinks for CH₄ (Le Mer and Roger 2001), recent studies support that tree stems in certain environments can emit CH₄ (Barba et al., 2019; Covey and Megonigal, 2018). Stem CH₄ emission may weaken the role of CH₄ sinks in forest ecosystems (Shoemaker et al., 2014; Pitz and Megonigal, 2017; Pitz et al., 2018). Our present results of the within-canopy profiles for CH₄ provided insight into the topic. At heights of 20 and 10 m, no significant differences from zero were mostly observed (Fig. 5), which implies that neither emission nor absorption of CH₄ from the stems and leaves of cypress trees were significant to influence the vertical CH₄ distributions. We note that at the same site closed chamber experiments were conducted to investigate CH₄ fluxes from the stem surface and intact leaves of a Japanese cypress tree from September 2009 to August 2010 (Takahashi et al., 2012), showing that no detectable amount of CH₄ emission or absorption was observed. While a single cypress tree was selected for the chamber experiments, it cannot be ruled out that some other cypress trees release CH₄ from the stems depending upon the growing environment. Because our study site is characterized by hillslopes, and it is generally known that the activity of methanogens and methanotrophs in soil is spatially

heterogeneous depending upon environmental parameters such as soil water content, temperature, and substrates (Le Mer and Roger, 2001). Indeed, a recent work examining CH₄ fluxes from tree stems and soils along a natural moisture gradient has suggested that tree emissions gradually increase from upland forests to forested wetlands (Pitz et al., 2018). At our study site, Sakabe et al. (2015) reported CH₄ fluxes from the surface of upland soils exhibited a high-spatial variability which was largely regulated by soil water content. Nevertheless, from the presently conducted profiling experiments, we conclude that at least at our site, CH₄ emissions from stems and leaves, which has recently been a hot topic, were not significant for cypress trees in upland area to influence the within-canopy distributions of CH₄ during the study period.

4. Conclusions

We have presented the spatial and temporal dynamics of CH₄ concentrations above and within a forest canopy. Background CH₄ concentrations and their temporal variations observed above the canopy at our study site are similar to those at nearby Atmospheric Environmental Regional Observation System stations. Continental outflow frequently influences the background concentrations of both CH₄ and CO₂, and spikes in both gasses were observed over time. Within the forest canopy, the vertical profile of the CH₄ concentrations showed a clear and seasonal influence of soil microbial activity in the surface layer of upland soil. In addition, CH₄ uptake rates estimated from the vertical gradients in CH₄ concentration near the forest floor were consistent with previously reported results obtained by closed-chamber measurements of CH₄ flux between the upland soil surface and the atmosphere at the same study site. The negligible exchange of CH₄ between the tree trunks and foliage of Japanese cypress trees was also inferred from the concentration profiles within the canopy, and the findings were also consistent with previously reported closed-chamber measurements. At our site, CH₄ emissions from stems and leaves, which has become a hot topic over the recent years, was not significant for cypress trees in upland area to influence the within-canopy distributions of CH₄ at least during the study period.

CRediT authorship contribution statement

Kenshi Takahashi: Conceptualization, Project administration, Investigation, Writing – original draft, Funding acquisition. **Ayaka Sakabe:** Writing – original draft, Investigation, Formal analysis. **Akito Kanazawa:** Investigation, Formal analysis. **Yoshiko Kosugi:** Conceptualization, Methodology, Software, Writing – review & editing, Funding acquisition.

Declaration of competing interest

The authors declare that there are no conflicts of interest regarding the publication of this work.

Acknowledgements

The authors would like to thank the Lake Biwa Environmental Research Institute for providing us with the data from air-quality monitoring stations. They are grateful to Dr. M. Ueyama (Osaka Prefecture University) for fruitful discussions. They are also grateful to anonymous reviewers for careful and constructive comments to improve the manuscript. This work was financially supported by JSPS KAKENHI (21H03576, 18H03356, 23310006, 23248023), JSPS DG-RSTHE Joint Research Program for FY 2018–2021, and the Mission Research Project of Research Institute for Sustainable Humanosphere, Kyoto University.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.aeoa.2021.100143>.

[org/10.1016/j.aeoa.2021.100143](https://doi.org/10.1016/j.aeoa.2021.100143).

References

- Atkinson, R., Baulch, D.L., Cox, R.A., Crowley, J.N., Hampson, R.F., Hynes, R.G., et al., 2004. Evaluated kinetic and photochemical data for atmospheric chemistry: volume 1–gas phase reactions of Ox, HOx, NOx, and SOx, species. *Atmos. Chem. Phys.* 4 (6), 1461–1738.
- Baldocchi, D.D., Hicks, B.B., Meyers, T.P., 1988. Measuring biosphere-atmosphere exchanges of biologically related gases with micrometeorological methods. *Ecology* 69, 1331–1340.
- Barba, J., Bradford, M.A., Brewer, P.E., Bruhn, D., Covey, K., van Haren, J., Megonigal, J.P., Mikkelsen, T.N., Pangala, S.R., Pihlatie, M., 2019. Methane emissions from tree stems: a new frontier in the global carbon cycle. *New Phytol.* 222, 18–28.
- Bey, I., Jacob, D.J., Logan, J.A., Yantosca, R.M., 2001. Asian chemical outflow to the Pacific in spring: origins, pathways, and budgets. *J. Geophys. Res.* 106 (D19), 23097–23113.
- Bousquet, P., Ciais, P., Dlugokencky, J. E., et al., 2006. Contribution of anthropogenic and natural sources to atmospheric methane variability. *Nature* 443, 439–443. <https://doi.org/10.1038/nature05132>.
- Bowling, D.R., Miller, J.B., Rhodes, M.E., Burns, S.P., Monson, R.K., Baer, D., 2009. *Biogeosci.* 6, 1311–1324, 2009.
- Covey, K.R., Megonigal, J.P., 2018. Methane production and emissions in trees and forests. *New Phytol.* 222, 35–51.
- de Carmo, J.B., Keller, M., Dias, J.D., de Camargo, P.B., Crill, P., 2006. *Geophys. Res. Lett.* 33, L04809. <https://doi.org/10.1029/2005GL025436>.
- Eugster, W., Plüss, P., 2010. A fault-tolerant eddy covariance system for measuring CH₄ fluxes. *Agricultural and Forest Meteorology* 150 (6), 841–851. <https://doi.org/10.1016/j.agrformet.2009.12.008>.
- Flanagan, L.B., Nikkel, D.J., Scherloski, L.M., Tkach, R.E., Smits, K.M., Selinger, L.B., Rood, S.B., 2021. Multiple processes contribute to methane emission in a riparian cottonwood forest ecosystem. *New Phytol.* 229, 1970–1982.
- Hendriks, D.M.D., Dolman, A.J., van der Molen, M.K., van Huissteden, J., 2008. A compact and stable eddy covariance set-up for methane measurements using off-axis integrated cavity output spectroscopy. *Atmos. Chem. Phys.* 8, 431–443.
- Ishizuka, S., Sakata, S., Ishizuka, K., 2000. Methane oxidation in Japanese forest soils. *Soil Biol. Biochem.* 32, 769–777. [https://doi.org/10.1016/S0038-0717\(99\)00200-X](https://doi.org/10.1016/S0038-0717(99)00200-X).
- Itoh, M., Ohte, N., Koba, K., 2009. Methane flux characteristics in forest soils under an East Asian monsoon climate. *Soil Biol. Biochem.* 41, 388–395. <https://doi.org/10.1016/j.soilbio.2008.12.003>.
- Kosugi, Y., Takanashi, S., Tanaka, H., Ohkubo, S., Tani, M., Yano, M., Katayama, T., 2007. Evapotranspiration over a Japanese cypress forest. I. Eddy covariance fluxes and surface conductance characteristics for 3 years. *J. Hydrol.* 337, 269–283.
- Le Mer, J., Roger, P., 2001. Production, oxidation, emission and consumption of methane by soils: a review. *Eur. J. Soil Biol.* 37, 25–50.
- Leckrone, K.J., Hayes, J.M., 1997. Efficiency and temperature dependence of water removal by membrane dryers. *Anal. Chem.* 69, 911–918.
- McMillan, A.M.S., Goulden, M.L., Tyler, S.C., 2007. Stoichiometry of CH₄ and CO₂ flux in a California rice paddy. *J. Geophys. Res.* 112, G01008. <https://doi.org/10.1029/2006JG000198>.
- Miyata, A., Leuning, R., Thomas, O., Kim, J., Harazono, Y., 2000. Carbon dioxide and methane fluxes from an intermittently flooded paddy field. *Agric. For. Meteorol.* 102, 287–303. [https://doi.org/10.1016/S0168-1923\(00\)00092-7](https://doi.org/10.1016/S0168-1923(00)00092-7).
- Ohkubo, S., Kosugi, Y., 2008. Amplitude and seasonality of storage fluxes for CO₂, heat and water vapour in a temperate Japanese cypress forest. *Tellus B* 60, 11–20. <https://doi.org/10.1111/j.1600-0889.2007.00321.x>.
- Ohkubo, S., Kosugi, Y., Takanashi, S., Mitani, T., Tani, M., 2007. Comparison of the eddy covariance and automated closed chamber methods for evaluating nocturnal CO₂ exchange in a Japanese cypress forest. *Agric. For. Meteorol.* 142, 50–65.
- Pitz, S., Megonigal, P., 2017. Temperate forest methane sink diminished by tree emissions. *New Phytol.* 214, 1432–1439.
- Pitz, S.L., Megonigal, J.P., Chang, C.H., Szlavecz, K., 2018. Methane fluxes from tree stems and soils along a habitat gradient. *Biogeochemistry* 137, 307–320.
- Querino, C.A.S., Smeets, C.J.P.P., Vignani, I., Holzinger, R., Moura, V., Gatti, L.V., Martinowski, A., Manzi, A.O., de Araújo, A.C., Röckmann, T., 2011. Methane flux, vertical gradient and mixing ratio measurements in a tropical forest. *Atmos. Chem. Phys.* 11, 7943–7953. <https://doi.org/10.5194/acp-11-7943-2011>.
- Reeburgh, W.S., 2006. Global methane biogeochemistry. In: Keeling, R.F. (Ed.), *The Atmosphere*. Elsevier, Netherland, pp. 65–89.
- Rigby, M., Prinn, R.G., Fraser, P.J., Simmonds, P.G., Langenfelds, R.L., Huang, J., Cunnold, D.M., Steele, L.P., Krummel, P.B., Weiss, R.F., O'Doherty, S., Salameh, P.K., Wang, H.J., Harth, C.M., Muhle, J., Porter, L.W., 2008. Renewed growth of atmospheric methane. *Geophys. Res. Lett.* 35, L22805. <https://doi.org/10.1029/2008GL036037>.
- Sakabe, A., Hamotani, K., Kosugi, Y., Ueyama, M., Takahashi, K., Kanazawa, A., Itoh, M., 2012. Measurement of methane flux over an evergreen coniferous forest canopy using a relaxed eddy accumulation system with tuneable diode laser spectroscopy detection. *Theor. Appl. Climatol.* 109, 39–49. <https://doi.org/10.1007/s00704-011-0564-z>.
- Sakabe, A., Kosugi, Y., Takahashi, K., Itoh, M., Kanazawa, A., Makita, N., Ataka, M., 2015. One-year of continuous measurements of soil CH₄ and CO₂ fluxes in a Japanese cypress forest: temporal and spatial variations associated with Asian monsoon rainfall. *J. Geophys. Res.* Biogeosci. 120, 585–599. <https://doi.org/10.1002/2014JG002851>.

- Sakabe, A., Kosugi, Y., Okumi, C., Itoh, M., Takahashi, K., 2016. Impacts of riparian wetlands on the seasonal variations of watershed-scale methane budget in a temperate monsoonal forest. *J. Geophys. Res. Biogeosci.* 121, 1717–1732. <https://doi.org/10.1002/2015JG003292>.
- Shirai, T., Machida, T., Matsueda, H., Sawa, Y., Niwa, Y., Maksyutov, S., Higuchi, K., 2012. Relative contribution of transport/surface flux to the seasonal vertical synoptic CO₂ variability in the troposphere over Narita. *Tellus B* 64, 19138. <https://doi.org/10.3402/tellusb.v64i0.19138>.
- Shoemaker, J.K., Keenan, T.F., Hollinger, D.Y., Richardson, A.D., 2014. Forest ecosystem changes from annual methane source to sink depending on late summer water balance. *Geophys. Res. Lett.* 42, 673–679.
- Simpson, I.J., Edwards, G.C., Thurtell, G.W., den Hartog, G., Newmann, H.H., Staebler, R.M., 1997. Micrometeorological measurements of methane and nitrous oxide exchange above a boreal aspen forest. *J. Geophys. Res.* 102 (D24), 29331–29341.
- Smeets, C.J.P.P., Holzinger, R., Vigano, I., Goldstein, A.H., Röckmann, T., 2009. Eddy covariance methane measurements at a Ponderosa pine plantation in California. *Atmos. Chem. Phys.* 9, 8365–8375. <https://doi.org/10.5194/acp-9-8365-2009>.
- Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D., Ngan, F., 2015. NOAA's HYSPLIT atmospheric transport and dispersion modeling system. *Bull. Am. Meteorol. Soc.* 96, 2059–2077. <https://doi.org/10.1175/BAMS-D-14-00110.1>.
- Takahashi, K., Kosugi, Y., Kanazawa, A., Sakabe, A., 2012. Automated closed chamber measurements of methane fluxes from intact leaves and trunk of Japanese cypress. *Atmos. Environ.* 51, 329–332. <https://doi.org/10.1016/j.atmosenv.2012.01.033>.
- Ueyama, M., Takahashi, S., Takahashi, Y., 2014. Inferring methane fluxes at a larch forest using Lagrangian, Eulerian, and hybrid inverse model. *J. Geophys. Res. Biogeosci.* 119, 2018–2031. <https://doi.org/10.1002/2014JG002716>.
- Ueyama, M., Yoshikawa, K., Takagi, K., 2018. A cool-temperate young larch plantation as a net methane source - a 4-year continuous hyperbolic relaxed eddy accumulation and chamber measurements. *Atmos. Environ.* 184, 110–120, 2018.
- Wada, A., Matsueda, H., Murayama, S., Taguchi, S., Hirao, S., Yamazawa, H., Moriizumi, J., Tsuboi, K., Niwa, Y., Sawa, Y., 2013. Quantification of emission estimates of CO₂, CH₄ and CO for East Asia derived from atmospheric radon-222 measurements over the western North Pacific. *Tellus B* 65, 18037. <https://doi.org/10.3402/tellusb.v65i0.18037>.
- Yu, L., Huang, Y., Zhang, W., Li, T., Sun, W., 2017. Methane uptake in global forest and grassland soils from 1981 to 2010. *Sci. Total Environ.* 607–608, 1163–1172.