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Discussions

Methane emissions from boreal and tropical forest ecosystems derived from in-situ measurements

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Abstract

Methane is a climatologically important greenhouse gas, which plays a key role in regulating water vapour in the stratosphere and hydroxyl radicals in the troposphere. Recent findings that vegetation emits methane have stimulated efforts to ascertain the impact of this source on the global budget. In this work, we present the results of high frequency (ca. $1\,min^{-1}$) methane measurements conducted in the boreal forests of Finland and the tropical forests of Suriname, in April–May, 2005 and October 2005 respectively. The measurements were performed using a gas chromatograph – flame ionization detector (GC-FID). The average of the median mixing ratios during a typical diel cycle were $1.83\,\mu mol\,mol^{-1}$ and $1.74\,\mu mol\,mol^{-1}$ for the boreal forest ecosystem and tropical forest ecosystem respectively, with remarkable similarity in the time series of both the boreal and tropical diel profiles. Night time methane emission flux of the boreal forest ecosystem, calculated from the increase of methane during the night and measured nocturnal boundary layer heights yields a flux of $(3.62\pm0.87)\times10^{11}$ molecules $cm^{-2}\,s^{-1}$ (or $45.5\pm11\,Tg\,CH_4\,yr^{-1}$ for global boreal forest area). This is a source contribution of circa 8% of the global methane budget. These results highlight the importance of the boreal and tropical forest ecosystems for the global budget of methane. The results are also discussed in the context of recent work reporting high methane mixing ratios over tropical forests using space borne near infra-red spectroscopy measurements.

1 Introduction

Methane is the most abundant hydrocarbon (circa $1.8\,\mu mol\,mol^{-1}$) in the atmosphere. It has significant implications for atmospheric chemistry and climate, as it plays a key role in regulating stratospheric water vapour and tropospheric hydroxyl radicals (Khalil, 2000). Atmospheric methane mixing ratios have nearly tripled since pre-industrial times (Prather et al. 2001; IPCC 2001), and while it is generally agreed that approxi-

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mately 600 Tg CH_4 yr^{-1} (1 Tg=10^{12} g) is released to the atmosphere globally, estimates of some of the individual source and sink terms within the global budget are rather uncertain (Lelieveld et al., 1998). For example, the range of the emission estimates for wetlands, the single largest natural methane source, is 92–232 Tg CH_4 yr^{-1} with a best guess estimate of 145 Tg CH_4 yr^{-1} (Wuebbles et al., 2002; Matthews et al., 2000; Prather et al., 2001). With such uncertainties in the existing global budget of methane, the discovery that terrestrial plants emit methane under aerobic conditions (Keppler et al., 2006) and reports of enhanced methane emissions from Siberian thaw lakes (Walter et al., 2006), have reinvigorated efforts to better constrain sources and sinks.

The global estimate of a 62–236 Tg CH_4 yr^{-1} vegetation source by Keppler et al. (2006), was based on an extrapolation of incubation chamber measurements using the metric of net primary productivity (NPP) for various ecosystems, and subsequent studies (Butenhoff and Khalil, 2007; Houweling et al., 2006; Ferreti et al., 2006; Kirschbaum et al., 2006; Parsons et al., 2006), have presented revised upper limit estimates in the range of 60–176 Tg CH_4 yr^{-1}, based on alternative up-scaling approaches. Very recently, using ^{13}C and a laser based measuring technique, Dueck et al. (2007) found no evidence of substantial aerobic methane emissions from terrestrial plants and concluded that the contribution of the new terrestrial plant source is very small at best. As the underlying production mechanism is still unknown, any extrapolation to the global scale is highly speculative. Reanalysis and potential reinterpretation of methane measurements conducted in Venezuela (Crutzen et al., 2006; Sanhueza et al., 2006) concluded that tropical savannah vegetation may also emit methane. Using space borne near infra-red remote sensing by the scanning imaging absorption spectrometer for atmospheric chartography (SCHIAMACHY), Frankenberg et al. (2005) observed almost 40 nmol/mol higher mixing ratios of methane over tropical rainforests compared to values predicted by a global 3D transport model (TM3) for the period October–November, 2003. Space borne measurements provide greater spatial coverage compared to local in-situ measurements at surface sites, which are closer to sources and sinks. On the other hand, the column averaging employed in volume

mixing ratio (VMR) retrievals of space borne measurements, has potential offsets and biases (Bergamaschi et al., 2007; Frankenberg et al., 2006), rendering remote sensing measurements less precise and less accurate than in-situ field measurements. A combination of in-situ and satellite retrieved methane measurements from forested regions therefore appears to be an ideal approach to assess the vegetative methane emission. However, such in-situ data (i.e. real time diel profiles of methane) from the boreal and tropical forests are surprisingly sparse, even though boreal and tropical forests collectively represent almost 76 percent of all forested land and hence have a large potential impact on the global methane budget.

In this study, we present the results from a high frequency gas chromatograph equipped with a flame ionization detector (GC-FID). Measurements were taken approximately every minute within a boreal forest ecosystem in Finland and a tropical forest ecosystem in Suriname in April–May, 2005 and October 2005 respectively. We discuss trends in the diel profiles, calculate the nighttime ecosystem flux for the boreal forest ecosystem and consider potential implications of these results.

2 Experimental

2.1 Site description: Boreal forest

Measurements were conducted from 16 April to 1 May 2005, as part of the BACCI/QUEST III campaign near (<100 m) the SMEAR II measurement station (Station for Measuring Forest Ecosystem- Atmosphere Relations) in Hyytiäla, southern Finland (61°51′ N, 24°17′ E, 170 m a.s.l.). About 600 m to the south west of the measurement station, there is an oblong lake (circa 2 km long). Figure 1a shows a picture of the boreal forest vegetation in the vicinity of the measurement tower. In addition to routinely measured meteorological parameters such as wind direction, wind speed etc., the SMEAR II station also conducts tower based measurements of gases such as CO_2 and O_3 throughout the year, providing a comprehensive suite of supporting

measurements as detailed in Hari and Kulmala (2005) and Vesala et al. (1998). A detailed distribution of the different land use categories in a $1600\,km^2$ area around the SMEAR II station has been described recently in Haapanala et al. (2007) using data derived from satellite photographs and forest inventories of the National Land Survey of Finland. The $1600\,km^2$ area around the SMEAR II station has spruce dominated forest (*Picea abies*; 25.5%), pine dominated forest (*Pinus sylvestris*; 22.7 %), mixed forest (European aspen: *Populus tremula* and Birch: *Betula pendula* and *pubescens*; 20.9%), deciduous forest (2%), agriculture (10.2%), wetlands (0.7%) and water bodies (13%). The Scots pine forest was planted in 1964 and in 2004 the canopy height was around $14\,m$ (Rinne et al., 1999; Hari and Kulmala, 2005).

Figure 2 shows the wind rose plot for the period of our study in Hyytiäla. During the campaign the wind blew mostly from the 0°–135° and 315°–360° wind sectors. Wind speeds were generally less than $1\,m\,s^{-1}$, although occasionally they reached up to $1.5\,m\,s^{-1}$. Our study was conducted in April–May, which marks the transition from winter to spring as the ground begins to thaw and the temperature increases marking the onset of photosynthetic activity (Suni et al., 2003). The minimum and maximum temperatures during the period of our study were around −5°C (typically at circa 06:00 local time LT) and 11°C (typically at circa 14:00 LT), with no well defined increasing trend, from the first to the last day of the study.

The annual mean temperature at the site is 3°C. The warmest month is July with a mean temperature of 16°C and the coldest is February with mean temperature of −8°C. Detailed long term climatic data for the site is available in Drebs et al. (2002).

2.2   Site description: Tropical forest

High frequency methane measurements were also conducted using the same instrument, within the nature reserve of Brownsberg (4°56′ N, 55°10′ W, $512\,m$ a.s.l.), Suriname from 5–14 October, 2005, as part of the Guyana Atmosphere-Biosphere exchange and Radical Intensive Experiment with the Learjet (GABRIEL) campaign. Brownsberg is situated around $120\,km$ inland and is topographically elevated with re-

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spect to the Brokopondo lake (circa $40\,km$ long), which lies south east of the measuring site. Figure 1b gives a view of the vegetation from Brownsberg where the canopy height was typically around $40\,m$. Rainforest vegetation is too diverse to be classified in terms specific plant types, however, based on the United States Geological Survey (USGS) world vegetation maps, the main general vegetation types in the area are lowland evergreen broad leaf trees, semi-evergreen moist broadleaf trees and some schlerophyllus dry forest trees. Upwind from the site is 300–400 km of pristine rainforest before the coast of French Guyana. During October, Suriname experiences the long dry season, in which steady south easterly winds advect clean marine air westwards over the rainforest. Typically, the range of temperatures experienced during this period is between 20°C–32°C.

2.3   Sampling procedure and measurements:

Ambient forest air was sampled within the canopy, at heights of $8.5\,m$ and $35\,m$ for the boreal and the tropical forests respectively. Employing the same sampling procedure for both sites, ambient air was drawn rapidly (circa $6\,L\,min^{-1}$; inlet residence time $<12\,s$) and continuously through $0.64\,cm$ diameter and $25\,m$ long shrouded Teflon tubing using an external pump (Rietschle Thomas, Memmingen GmbH, Memmingen). From this main flow of $\sim6\,L\,min^{-1}$, $1.2\,L\,min^{-1}$ was drawn continuously by an internal pump within the methane instrument, which is a commercial gas chromatograph equipped with a flame ionization detector (GC-FID) (Model 55C Hydrocarbon Analyzer, Thermo Electron Corporation, Massachusetts, USA). Nitrogen was used as carrier gas while hydrogen and zero air were used as fuel for the FID detector. Central to the instrument′s operation is an eight port, two position, rotary valve which is used to introduce the gas sample into the analyzer and to control the flow of gases through the chromatographic column. The operational temperature of this column is 65°C. After separation from other airborne components within the column, the methane peak is measured by the FID and the signal area is converted into a mixing ratio by comparison with the signal produced by a calibration gas ($3.96\,\mu mol\,mol^{-1}$ methane standard

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procured from Westfalen AG, Germany; stated accuracy 2%). The detection limit of the instrument is 20 nmol mol^{-1} methane and the measurement range is 0–20 μmol mol^{-1} methane, over which the detector responds linearly. One measurement was taken approximately every 70 s (frequency of measurements = 0.014 Hz).

The accuracy and precision errors are each ±2% of the measured value. However, for the data presented here, the overall uncertainty has been determined as 2.58%, comprising of the 2% accuracy error (a systematic error which is the same for all measurements) and a reduced precision error of 0.58% for each data point due to the 15 min time averaging employed in preparing the box and whisker plots (Figs. 3 and 4).

In order to check for instrumental drifts, all the basic instrumental operational parameters such as the oven temperature, column temperature and support gas pressures were monitored continuously, in addition to regular in-field calibrations performed at the start, middle and end of the measurement campaigns.

At the SMEAR II station, carbon dioxide was measured using an infrared light absorption analyser (URAS 4 CO_2, Hartmann & Braun, Frankfurt am Main, Germany) while the wind speed and wind direction were measured with an ultrasonic anemometer (Ultrasonic anemometer 2D, Adolf Thies GmbH, Göttingen, Germany).

3 Results

3.1 Boreal forest: trends in methane mixing ratios and diel cycle

Methane mixing ratios were highest when the winds were blowing from the north, which has extensive forest cover and low population (Hari and Kulmala et al., 2005). In order to minimise influences from anthropogenic emissions (from the city of Tampere south east of the site), and from wetlands (primarily south west of the site, Haapanala et al., 2007), the methane data was filtered to include data only from the 315°–45° wind sector. A box and whisker diel plot of the filtered data for the entire campaign is shown in Fig. 3. As per convention, the box encloses all values from the 25–75 percentile

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while the whiskers encompass all values in the 5–95 percentile range. It can be seen that the median and mean values compare closely most of the time, indicating that no sharp bursts of methane emissions were detected. The plot also shows a distinct diel variation in the mixing ratios of methane, with lowest median values of around 1.81 μmol mol^{-1} occurring typically between 15:00–16:00 LT and the highest median mixing ratios of the day ($\sim$1.85 μmol mol^{-1}) occurring at around 06:00 LT. Variability in the methane mixing ratios during the campaign was seen to be least for the measurements between 16:00 and 18:00 LT and higher during the early morning hours. Also evident is a steady increase in the median methane mixing ratios from around 20:00 LT in the evening (just after sunset) till 06:00 LT in the morning (just before sunrise). The average of the median mixing ratios during a typical diel cycle was 1.83 μmol mol^{-1}.

3.2 Tropical forest: Trends in methane mixing ratios and diel cycle

Figure 4 shows a box and whisker plot derived from the continuous 8700 high frequency (0.014 Hz) measurements conducted in Brownsberg from 5–14 October, 2005. The median mixing ratios of methane again show a clear diel variation. The median mixing ratios were highest ($\sim$1.76 μmol mol^{-1}) from 06:30 to 08:00 LT in the morning, and lowest ($\sim$1.72 μmol mol^{-1}) from 14:30 to 16:00 LT in the afternoon. Variability is comparatively higher between 16:30 and 19:00 LT. Median and mean values again differ only slightly, except between 19:30 to 20:30 LT, when the difference is almost 10 nmol mol^{-1}. Remarkably, the median methane mixing ratios show a distinct increase from 19:45 LT in the evening till around 05:45 LT in the morning. The average of the median mixing ratios during a typical diel cycle at Brownsberg was 1.74 μmol mol^{-1}.

3.3 Determining the night time methane flux for the boreal forest ecosystem

In order to calculate the nighttime methane flux for the boreal forest using median mixing ratios in the box and whisker diel profile (Fig. 3), it is necessary to measure the nocturnal boundary layer (NBL) growth. Additionally, to establish whether the NBL is

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well mixed, it is also important to assess whether there is any significant gradient in the vertical profile of methane. In Sect. 3.3.1 we describe how the boundary layer heights were deduced while Sect. 3.3.2 explains how vertical gradients were assessed. Finally, Sect. 3.3.3 details the flux calculation methodology.

3.3.1 Determining the boundary layer height

Balloon soundings with temperature and humidity sensors were launched (approximately thrice a day) during the campaign to determine the height of the boundary layer. As the height of the boundary layer scales directly with the calculated flux (see Sect. 3.3.3), and is therefore an important step of the flux calculation methodology, we have illustrated how the NBLs were deduced using typical examples in Figs. 5a and 5b. Figure 5a shows the vertical profiles of potential temperature and relative humidity measured on 23 April at 07:45 LT while Fig. 5b shows how the boundary layer heights were established from the turning points of the potential temperature vertical profiles at 19:45 LT on 24 April, and 05:45 LT on 25 April, 2005. Table 1 lists the boundary layer height values that were measured in the evening ($\sim$20:00 LT) and the morning ($\sim$06:00 LT), on consecutive days from 24–28 April. From these values the average evening time ($\sim$20:00 LT) boundary layer height was established as 80 m while the average morning time ($\sim$06:00 LT) boundary layer height was established as 180 m. A gradual growth of this magnitude is in keeping with current understanding of nocturnal boundary layer dynamics (Stull, 1988).

3.3.2 Assessing vertical gradients

Vertical gradients within the nocturnal boundary layer (NBL) were assessed using vertical profiles of carbon dioxide made at multiple heights from the SMEAR II measurement tower (heights: 4.2 m, 8.4 m, 16.8 m, 33.6 m, 50.4 m, 67.4 m, 74 m) at the start (17 and 18 April), middle (20 and 21 April) and end (30 April and 1 May) of the study period (16 April to 1 May). Carbon dioxide is emitted from the forest at night and has a

rather long atmospheric lifetime (on the order of several years). Thus within the NBL, the vertical gradients of both methane and carbon dioxide will be driven by physical parameters rather than chemical reactions with other species. Figure 6 shows the carbon dioxide vertical profiles in the evening, when the NBL starts to grow ($\sim$20:00 LT) and in the morning ($\sim$06:00 LT), when it has attained its maximum height. In order to examine gradients with respect to the methane sampling height of 8.5 m, the values in Fig. 6 are represented as a percentage of the carbon dioxide mixing ratio at the 8.5 m level. It can also be seen in Fig. 6, that the difference between the highest point and the 8.5 m level is typically less than 0.3% and in no case more than 0.5%. This shows that the NBL was well mixed and that mixing ratios measured at the 8.5 m level can be taken to be representative of the entire mixed NBL, for both carbon dioxide and methane.

3.3.3 Flux calculation

Consider a box of cross section area A cm^2 and height 80 m at 20:00 LT which grows to a height of 180 m at 06:00 LT. Then using the mass balance approach, the total number of methane molecules in the box at 06:00 LT ($N_{06:00}$) can be expressed as the contribution of the initial number of methane molecules in the box at 20:00 LT ($N_{20:00}$), the increase due to the emission flux (F_{CH4}), assumed to be constant from 20:00–06:00 LT, and the contribution of methane molecules due to mixing of residual air from above ($N_{ResidualLayer}$), as the box grows from 80 m at 20:00 LT to 180 m at 06:00 LT. Expressing the above as an equation we get,

$$N_{06:00} = N_{20:00} + F_{CH4} \times A \times (36000) + N_{ResidualLayer} \qquad (1)$$

Where,

$N_{06:00}$ = CH$_4$ [t=06:00] $\times$ C.F. $\times$ A $\times$ 18 000;

$N_{20:00}$ = CH$_4$ [t=20:00] $\times$ C.F. $\times$ A $\times$ 8000;

$N_{ResidualLayer}$ = CH$_4$ [avg. b/w 16:00 LT and 19:00 LT] $\times$ C.F. $\times$ A $\times$ 10 000;

C.F. is the factor used for converting nmol mol^{-1} to molecules cm^{-3} = 2.69×10^{10};

and from Fig. 3,

CH$_4$ [t=06:00], the median mixing ratio at 06:00 LT = 1850 nmol mol^{-1}
CH$_4$ [t=20:00], the median mixing ratio at 20:00 LT = 1820 nmol mol^{-1}
and CH$_4$ [avg. b/w 16:00 and 19:00], the average of median mixing ratios between 16:00 LT and 19:00 LT (assumed to be the residual layer mixing ratio) = 1819 nmol mol^{-1}

Using appropriate values in Eq. (1), we derive a net night time ecosystem emission flux of 3.62×10^{11} CH$_4$ molecules cm^{-2} s^{-1}.

4 Discussion

4.1 Boreal forest data

Previous work on methane emissions from boreal forest ecosystems has been mainly based in Canada and the United States (Simpson et al., 1999, 1997). At Hyytiäla, a background forest site comprising of more than 80% vegetation in 1600 km^2 of surrounding area (Haapanala et al., 2007), research has been mainly focused on aerosol and emissions of reactive biogenic compounds such as terpenes and oxygenated volatile organic compounds (Kulmala et al., 2004; Tunved et al., 2006; Yassaa and Williams, 2007).

To our knowledge, this is the first time that such high resolution methane diel profiles have been reported from a boreal forest ecosystem, showing night time emission and hence accumulation of methane within the NBL. The atmospheric lifetime of methane is around 8.9 years and so the diel variation at Hyytiäla cannot be explained by the reaction of methane with hydroxyl radicals during daytime. Rather, the diel profile of methane is driven by the dynamics of the boundary layer coupled with biogenic emissions or uptake. After sunset, the ground starts to cool radiatively, causing the growth of a stable nocturnal boundary layer (NBL). Thus there is a decoupling (inhibited gas exchange) of the surface boundary layer from the free troposphere (Stull, 1988). Although during the night the nocturnal boundary layer deepens, mixing ratios

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of methane rise through the night clearly indicating an emission. After sunrise, the ground starts to warm and the surface heat flux causes the boundary layer to grow until it reaches its maximum height in the afternoon. The volume of this daytime well mixed layer is greater than the volume of the well mixed NBL, and the dilution effect of air from above overwhelms the emissions during the day, leading to a daytime decrease in the volume mixing ratio of methane (see Fig. 3). When compared with three month averaged mixing ratios for April–June, 2003 of circa 1.75 μmol mol^{-1} methane over Hyytiäla (Fig. 5 in Bergamaschi et al., 2007) based on SCHIAMACHY measurements, the value of 1.83 μmol mol^{-1} presented here for 2005 is almost 80 nmol mol^{-1} higher. The flux of circa 4 mg methane m^{-2} day^{-1} (1.74×10^{11} molecules cm^2 s^{-1}) reported in the same work is also considerably smaller in comparison to our night time flux of 3.62×10^{11} molecules cm^2 s^{-1}. One explanation for this difference could be that source strengths may have increased in 2005 compared to 2003, as the sink due to OH is unlikely to have changed by factor of 2, between 2003 and 2005. Alternatively, it might be that potential offsets and biases in the SCHIAMACHY (Frankenberg et al., 2006; Bergamaschi et al., 2007) retrieval scheme cause it to underestimate methane over Hyytiäla.

The boreal forest ecosystem has been previously reported as both a net sink and a net source, for methane. However, this has not been well characterized. While Tyler (1991) and Steudler et al. (1989) reported a sink for the boreal forest ecosystem, Simpson et al. (1997) reported a net emission flux of 0.8–1.05 $\times 10^{11}$ molecules cm^{-2} s^{-1} (mixing ratio circa 1.82–1.90 μmol mol^{-1}) from a boreal forest ecosystem in Saskatchewan during springtime (April–September, 1994) using micrometeorological tower measurements. Reports of the boreal forest as a net sink for methane are almost exclusively based on data from chamber measurements (Savage et al., 1997; Schiller and Hastie et al, 1996; Whalen et al., 1992). Simpson et al. (1999) suggested that the apparent discrepancy is most probably because chamber measurements are conducted close to the surface and are influenced strongly by the soil, which is known to uptake methane. Further, Simpson et al. (1997) reported

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that micrometeorological measurements conducted only at the base of the tower indicated the forest to be a sink while overall, the forest was found to be a net source of methane.

As detailed in Khalil (2000) and references therein, methane emissions can vary considerably based on seasonality, land use and environmental parameters such as soil temperature and water table levels. Nevertheless, bottom up estimates using in-situ field measurements are useful to get an idea about the possible contribution of particular ecosystem types to the global budget. Extrapolating the net night time methane emission flux of 3.62×10^{11} molecules $cm^{-2} s^{-1}$ derived in this work, to the global boreal forest area of $1.5 \times 10^{17} cm^2$ (Tunved et al., 2006), we estimate an annual boreal forest ecosystem flux of $45.5 \, Tg \, CH_4 \, yr^{-1}$. This is around 7.5% of the estimated global 600 Tg $CH_4 \, yr^{-1}$ released into the atmosphere. It should be noted however, that this flux calculation methodology is rather sensitive to the NBL estimates. Based on the NBL height variability observed at Hyytiälä (see Table 1), this introduces an uncertainty of up to ±25% for the calculated flux.

From our data, it is not possible to derive an accurate boreal vegetation flux as the measured flux is a net ecosystem flux. However for the planning of future studies that seek to derive the vegetation flux using in situ field measurements, and to illustrate the challenge inherent in obtaining a field estimate of emissions only due to vegetation, we discuss herein a land use based approach which would help to constrain the source contribution of vegetation to the global methane budget, using in-situ field measurements from different forest ecosystem types. This would be especially useful, since the laboratory based up scaling estimates as well as the subsequent model estimates, for the contribution of the new source discovered by Keppler et al. (2006) have considerable uncertainty.

Average fluxes compiled by Bartlett and Harriss (1993) for broad ecosystem types show that values for the boreal wetlands are between $3.7-4.1 \times 10^{12}$ molecules $cm^{-2} s^{-1}$ (or 87–96 ng $m^{-2} s^{-1}$). The average methane emission flux, from southern boreal lakes (located between $60°57'$, $24°27'$ and $61°22'$,

$25°15'$) for the growing seasons of April–September (years 1998–2002), is 2.15×10^{12} molecules $cm^{-2} s^{-1}$ (Bergstrom et al., 2006). Thus a wetland flux of 4.1×10^{12} molecules $cm^{-2} s^{-1}$ and lake emission flux of 2.15×10^{12} molecules $cm^{-2} s^{-1}$ can be considered as generous estimates for wetland and lake emissions in Hyytiälä ($61°51'$ N, $24°17'$ E) during April–May. Knowing that wetlands and water bodies make up 0.7% and 13% of the $1600 \, km^2$ footprint area around the measurement site (Sect. 2.1) a conservative estimate of the boreal vegetation can be calculated as follows:

$$\text{Wetland flux} + \text{Lake flux} + \text{Vegetation flux} = \text{Net ecosystem flux} \tag{2}$$

That is,

0.7% of 4.18×10^{12} molecules $cm^{-2} s^{-1}$ + 13% of 2.15×10^{12} molecules $cm^{-2} s^{-1}$ + Vegetation flux = 3.62×10^{11} molecules $cm^{-2} s^{-1}$

Solving the above yields a boreal vegetation flux of 5.32×10^{10} molecules $cm^{-2} s^{-1}$. Upscaling for the global boreal forest area of $1.5 \times 10^{17} cm^2$ (Tunved et al., 2006), the contribution of boreal vegetation to the total methane budget would be circa 6.7 Tg $CH_4 \, yr^{-1}$. Note that this is a highly uncertain estimate because the contribution of the individual components in Eq. (2) are in themselves quite uncertain, and were not measured concurrently with the methane measurements of this study, and even a circa 1% change in the flux value used for wetlands would render the boreal vegetation flux negligible. It should also be noted that an estimate based on such an approach would probably be a lower range estimate because according to Keppler et al. (2006) plants emit 2–5 fold more methane during the day, so that the net ecosystem flux might actually be higher if day time emissions could be included.

Thus, as reiterated earlier, based on the data obtained in this study, we can neither confirm nor disprove whether boreal vegetation emits methane.

Keppler et al. (2006) reported boreal vegetation emissions to be in the range of $1.1-4.1 \, Tg \, CH_4 \, yr^{-1}$, based on the upscaling metric of net primary productivity (NPP). Using the metrics of leaf mass index and photosynthesis rates for different biomes,

Kirschbaum et al. (2006) upscaled the results of Keppler et al. (2006) and reported boreal vegetation emission estimates of 2.8 Tg yr^{-1} and 0.6 Tg yr^{-1}, respectively. Emulating the Kirschbaum et al. (2006) approach of biome leaf mass for upscaling, Parsons et al. (2006) revaluated the results of Keppler et al. (2006) and proposed an estimate of 3.6 Tg CH$_4$ yr^{-1} for boreal vegetation. Thus based on these previous estimates, it appears that boreal vegetation contributes only circa 1% to the global methane budget. The boreal ecosystem as a whole, however, based the results of this study, contributes almost 7.5%.

4.2 Tropical forest data

Analogous to the boreal methane profile, the diel profile from Brownsberg (Fig. 4) is also characterized by an increase in the median methane mixing ratios during nighttime. Carmo et al. (2006) and Crutzen et al. (2006) have reported similar profiles from upland amazon forests and mixed tropical savannah grasslands, respectively. Frankenberg et al. (2005) observed enhanced mixing ratios of methane over tropical rainforests and called for in-situ measurements to validate their measurements. It is interesting to note that compared to the SCHIAMACHY measurements of 1.76 μmol mol^{-1} methane, and the global chemistry transport model (TM3) value of 1.72 μmol mol^{-1} for the same season in 2003 over Suriname, the average of all our in situ measurements in 2005 is 1.74 μmol mol^{-1}. If we assume that the sources and sinks in 2003 and 2005 were not significantly different, then the in-situ measurements would seem to suggest that the enhanced emissions over the Suriname rainforest are probably not as strong as first reported in Frankenberg et al. (2005). The latter argument is supported by the work of Bergamaschi et al. (2007) that extended the original analysis by Frankenberg et al. (2006), and reported that SCHIAMACHY CH$_4$ retrievals may have some bias (up to ~30 nmol mol^{-1}) that depends on latitude and season.

Krejci et al. (2005) have estimated the NBL over the rainforest in Suriname to be less than 250 m. Brownsberg is situated on a hill (512 m a.s.l.) and the presence of the nearby Brokopondo lake (also called Prof. Dr. Bloomstein lake) may potentially impact

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our methane measurements. So while the measurements at Brownsberg (512 m a.s.l.), resemble tower type measurements with respect to the surrounding rainforest, overall the situation presents a less than ideal site for flux measurements. Still for the sake of discussion, if we assume that methane was well mixed within the NBL and that the NBL height over the rain forest was 100 m at ~19:45 LT and 200 m at ~05:45 LT, then applying the method described in Sect. 3.3.3, we would derive a night time emission flux of 3.77×10^{11} molecules cm^{-2} s^{-1}. Since, vertical potential temperature profiles and meteorological data of the type used for the Hyytiäla site measurements were not available for the tropical forest site, the tropical methane flux has a greater uncertainty and this flux must be interpreted with appropriate care. Previous measurements from the upland forest area (1.5×10^6 km^2) of the Amazon region, by Carmo et al. (2006) have suggested a flux range of 0.96–9.1×10^{11} molecules cm^{-2} s^{-1}(or 4–38 Tg yr^{-1}) which is lower than the flux of ~1.2×10^{12} molecules cm^{-2} s^{-1} reported recently by Miller et al. (2007) referring to the eastern Amazon basin.

5 Conclusion and outlook

The boreal forest ecosystem as a whole is a significant source of methane (~45.5±11 Tg CH$_4$ yr^{-1}) and appears to contribute circa 7.5% to the global methane budget. High temporal resolution measurements, from the boreal forest and tropical forest ecosystems, made over longer time periods are needed to assess methane emissions from these regions more accurately, because based on the short term data presented in this work, it appears that boreal forest ecosystem emissions may be currently underestimated, as SCHIAMACHY data for the same period over Hyytiäla showed methane mixing ratios of 1.75 μ mol mol^{-1}, while our in-situ measurements were 1.83 μmol mol^{-1} and the methane flux reported in Bergamaschi et al. (2007) of 1.74×10^{11} molecules cm^2 s^{-1} was lower than the flux of 3.62×10^{11} molecules cm^2 s^{-1}, obtained in this study. Collectively, the nighttime emissions from boreal and tropical forest ecosystems indicate a global source contribution of circa 100 Tg CH$_4$ yr^{-1}, which

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is almost 17% of the global budget of circa 600 Tg CH_4 yr^{-1}. The similarity in the diel methane profiles at such widely different climates is remarkable. Until the mechanism of methane production from plants under aerobic conditions, is better understood, examining net ecosystem fluxes under in-situ natural conditions, would help in constraining the contribution due to a potential new vegetation source and assessing whether their magnitude is of the order of uncertainties in the existing sources, or higher. Although our methane flux estimates refer to nighttime conditions only, whereas aerobic methane production is expected to be larger during daytime, our observations cannot be reconciled with a very large direct methane source of up to 176–236 Tg/yr from the vegetation.

Furthermore, ^{13}C isotope measurements can help to apportion the methane sources in a better manner and thus a combined approach of in-situ measurements at suitable remote and homogeneous sites, remote sensing retrievals and isotope studies would help to reduce the uncertainties in the global methane budget much more effectively.

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Table 1. Boundary layer heights at Hyytiäla (61°51′ N, 24°17′ E, 170 m a.s.l.)

Date	Time (LT)	B.L.Height (m)
24 April	19:45	80
25 April	05:45	160
25 April	19:30	70
26 April	05:35	190
27 April	20:00	90
28 April	05:50	210

Fig. 1. View of boreal forest vegetation in Hyytiäla (61°51′ N, 24° 17′ E, 170 m a.s.l.) (above) **(a)** and view of tropical forest vegetation in Brownsberg (4° 56′ N, 55°10′ W, 514 m a.s.l.) (below) **(b)**.
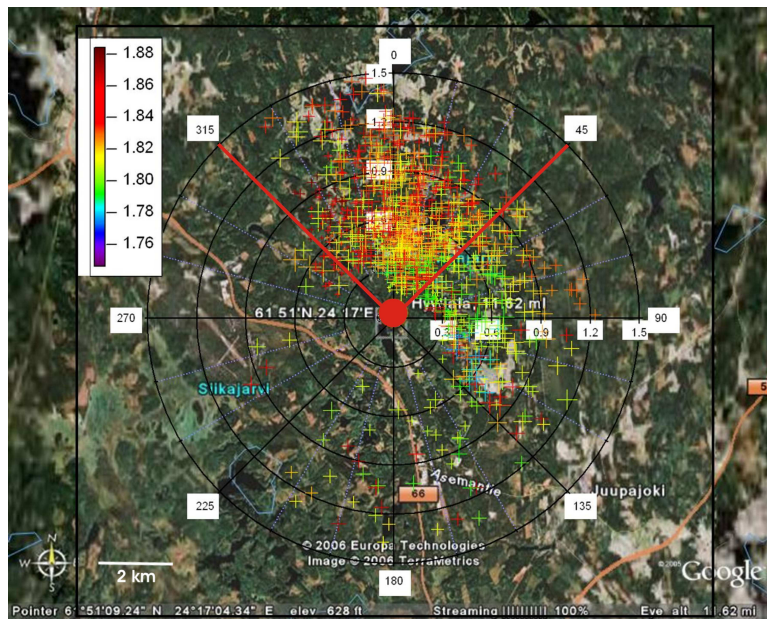
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Fig. 2. Wind rose plot showing methane (coloured markers and scale), wind speed (radii) and wind direction (angle) in Hyytiäla during the campaign (Map courtesy of Google Earth[TM] mapping service)
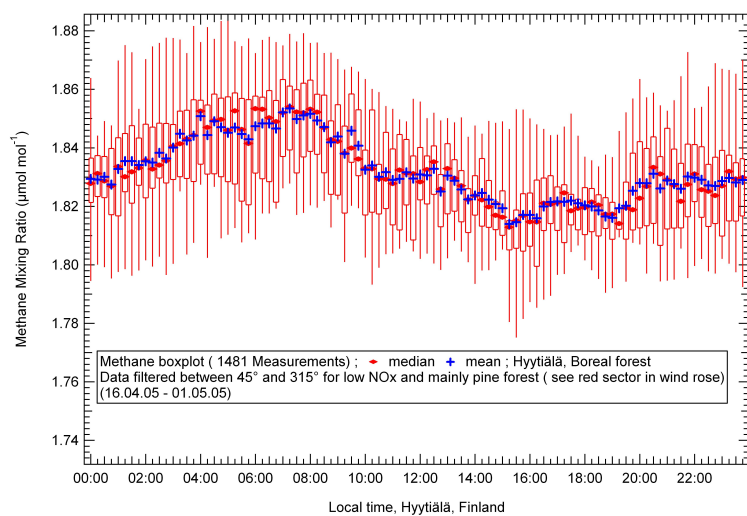
14034

Fig. 3. Box and whisker plot for diel cycle of filtered methane data (315°–45° wind sector) from Hyytiäla (61°51′ N, 24°17′ E, 170 m a.s.l.), Finland during the BACCI/QUEST III Campaign.

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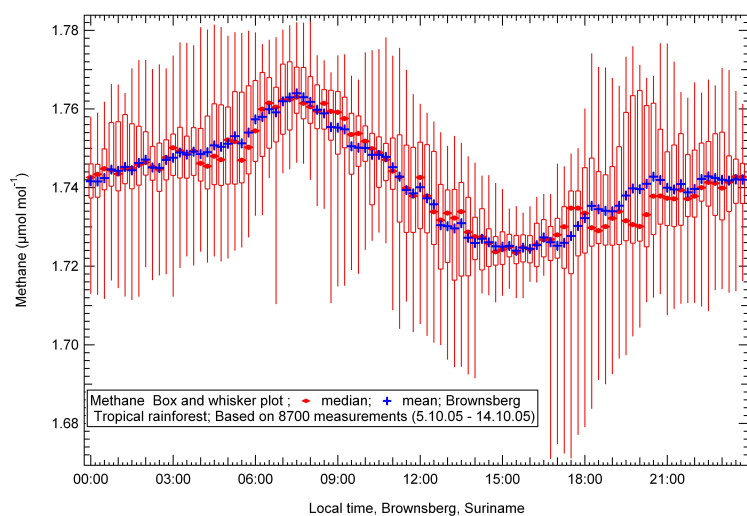
Fig. 4. Box and whisker plot for diel cycle of methane data from Brownsberg (4° 56′ N, 55°10′ W, 514 m a.s.l.), Suriname during the GABRIEL campaign.
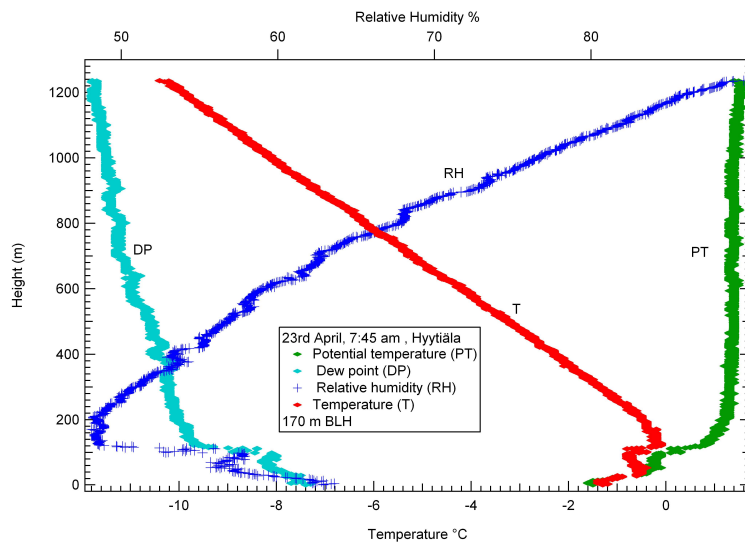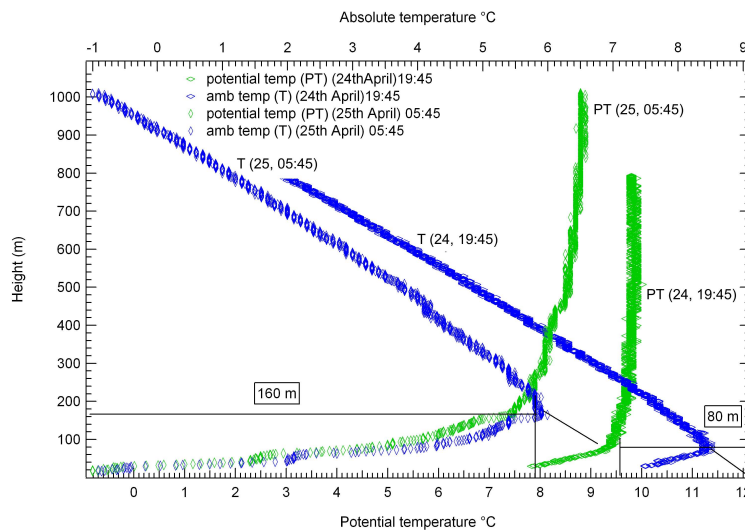
Fig. 5a. Typical example for determination of boundary layer height at Hyytiäla (61°51′ N, 24° 17′ E, 170 m a.s.l.) using several physical parameters (above) and potential temperature profile (below).

Fig. 5b. Typical example for determination of boundary layer height at Hyytiäla (61°51′ N, 24° 17′ E, 170 m a.s.l.) using several physical parameters (above) and potential temperature profile (below).
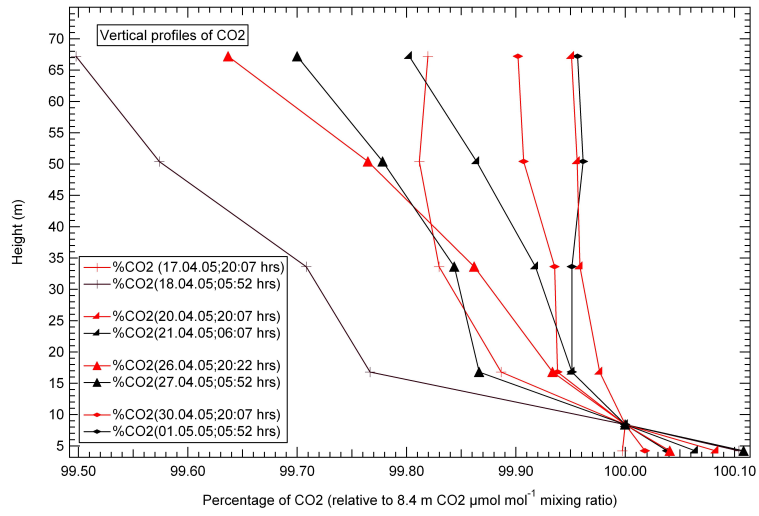
Fig. 6. Assessing vertical gradients in the NBL at Hyytiäla (61°51′ N, 24° 17′ E, 170 m a.s.l.) using CO_2 profiles.