



Ecosystem-scale biosphere–atmosphere interactions of a hemiboreal mixed forest stand at Järvelja, Estonia

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ABSTRACT

During two measurement campaigns, from August to September 2008 and 2009, we quantified the major ecosystem fluxes in a hemiboreal forest ecosystem in Järvelja, Estonia. The main aim of this study was to separate the ecosystem flux components and gain insight into the performance of a multi-species multi-layered tree stand. Carbon dioxide and water vapor fluxes were measured using the eddy covariance method above and below the canopy in conjunction with the microclimate. Leaf and soil contributions were quantified separately by cuvette and chamber measurements, including fluxes of carbon dioxide, water vapor, nitrogen oxides, nitrous oxide, methane, ozone, sulfur dioxide, and biogenic volatile organic compounds (isoprene and monoterpenes). The latter have been as well characterized for monoterpenes in detail. Based on measured atmospheric trace gas concentrations, the flux tower site can be characterized as remote and rural with low anthropogenic disturbances.

Our results presented here encourage future experimental efforts to be directed towards year round integrated biosphere–atmosphere measurements and development of process-oriented models of forest–atmosphere exchange taking the special case of a multi-layered and multi-species tree stand into account. As climate change likely leads to spatial extension of hemiboreal forest ecosystems a deep understanding of the processes and interactions therein is needed to foster management and mitigation strategies.

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1. Introduction

Forest ecosystems are a major part of the biosphere and control as such land surface–atmosphere interactions. They influence atmospheric composition and climate significantly because of their importance as sources and sinks of trace gases and energy on both, local and regional scales (Högberg, 2007; Magnani et al., 2007; Misson et al., 2007). To better understand both, how forest ecosystems will respond to changes in climate and how they will feed back to the atmosphere, we need to gain quantitative insight into the energy and matter fluxes between the biosphere and atmosphere.

Forests themselves form complex ecosystems, exhibiting a great variability in their composition and structure in horizontal and vertical directions. Major fluxes of carbon dioxide, methane and water vapor are exchanged between soil, plants and the atmo-

sphere at several distinct layers. Those layers are characterized by very different functional properties and they also differ in their flux contributions (Baldocchi, 1997; Misson et al., 2007).

Reactive trace gases such as ozone or nitrogen oxides (NO_x) and sulfuric compounds for example represent abiotic stressors to the plants in the forest ecosystem, causing reductions in biomass production (Wallin et al., 1990; Laurence et al., 2001; Manning et al., 2003). Together with biogenic volatile organic compounds (BVOC) that are emitted by many plant species to substantial amounts, these reactive trace gases determine the local, regional and global air quality. Their reaction products play a major role in the formation and growth of aerosol particles, ozone and organic acids (Atkinson and Arey, 2003; Bonn et al., 2008; Hewitt et al., 2009; Mentel et al., 2009).

Hemiboreal forests, located in the transition zone between boreal and temperate forest biomes, are characterized by mixed stands of both coniferous and deciduous tree species, where deciduous species can dominate the initial stages of succession, being replaced by coniferous species at later stages of succession. Especially in mixed stands, the presence of deciduous tree species exerts a great variability on the forest microclimate, canopy shape and

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density by closing the canopy in spring and opening it up again in autumn when the leaves drop off.

Typically, the presence of multiple leaf layers in the canopy leads to gradients of environmental variables such as light, temperature, wind speed and turbulent mixing. These gradients in turn affect the magnitude of the ecosystem fluxes throughout the canopy. Furthermore, differences in energy availability and mixing properties of the air below and above canopy result in within-canopy variation in concentrations of carbon dioxide and reactive trace gases impacting on the growth and biomass production of the trees differently at different canopy layers.

So far, integrated forest ecosystem scale studies, covering leaf, soil and ecosystem fluxes of reactive and non-reactive trace gases have not yet been conducted for hemiboreal forest ecosystems in Estonia. Our goal was to assess a first overview on fluxes and concentrations of carbon dioxide, water vapor and reactive trace gases under the influence of a hemiboreal forest ecosystem. Specifically the occurrence of a major and suppressed tree layer and consequences thereof on the fluxes and concentrations are of particular interest. Therefore, we present below and above canopy eddy covariance fluxes, leaf level net photosynthesis rates, soil trace gas fluxes, within and above-canopy trace gas concentrations, and BVOC emissions measured during two field campaigns from August to September 2008 and 2009 at the Experimental Forestry Station in Järvelja, Estonia.

2. Materials and methods

2.1. Site description

The measurements were carried out at a site located in Järvelja Experimental Forest of Estonian University of Life Sciences, located in southeastern Estonia (58°25'N, 27°46'E). Järvelja is situated in the hemiboreal forest zone with a moderately cool and moist climate. Mean annual temperature is 4–6 °C. Annual precipitation is 500–750 mm, about 40–80 mm of this total is snow. The length of the growing season (daily air temperature above 5 °C) averages between 170 and 180 days. A scaffolding tower of 20 m height

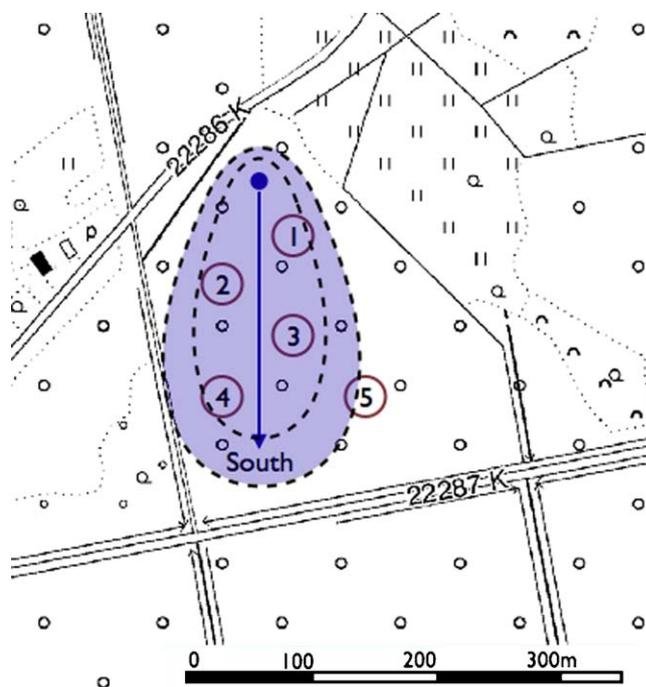


Fig. 1. Flux tower site at Järvelja, the blue dot marks the location of the tower and the numbered circles represent the five permanent sample plots. The broken lines denote the flux footprint in southern direction. The measurement cottage is located two meters in western direction at the base of the tower.

was used for the flux measurements above the forest canopy and the cumulative contribution (Horst and Weil, 1992) was 78% at a distance of 200 m upwind of the tower. The measurement cottage to house the sensors for the trace gas analysis and auxiliary equipment was established beside the base of the tower in 2008. For an overview of the location see Fig. 1. Both campaigns were covering the time of late summer and in 2008 the measurements started from 6th of August and ended at 27th August while 2009

Table 1
Overview on the measurement series conducted throughout both campaigns. In 2008 we conducted already several measurements, denoted by “preview”, which have been used to set up the measurements in 2009.

	2008		2009	
	Time	Height over ground	Time	Height over ground
Flux measurements ^a	6th to 21st August	20 m and 2 m	12th to 19th August 19th to 25th August 25th Aug. to 9th September 9th to 21st September	20 m 2 m 20 m 2 m
Trace gases	6th to 21st August	20 m and 2 m	12th August to 21st September	20 m and 2 m
Particles	6th to 20th August	3 m	–	–
Leaf level BVOC and photosynthesis	11th to 17th August	16 m <i>B. pendula</i> 18–16 m <i>P. abies</i> 0–2 m understory Preview	24th to 27th August 2nd and 5th September	16 m <i>B. pendula</i> 0.5 m <i>T. cordata</i> 16–18 m <i>P. abies</i> 2 m <i>Q. robur</i> 1 m <i>B. pubescens</i> 1.5 m <i>P. tremula</i>
Soil fluxes CO ₂ automatic chamber manual chamber Soil BVOC	–14th to 15th August –	– –	14th to 16th September 7th October/23rd November 7th to 11th September	0 m 0 m 0 m
Auxiliary ^b	6th to 21st August	20 m and 2 m	12th August to 21st September	20 m and 2 m

^a Data of the year 2009 have been measured with one Eddy system on timely separated heights and were therefore not included into Fig. 1.

^b Auxiliary measurements are air temperature, pressure, relative humidity, and global solar radiation.

the measurement campaign started at 12th August and ended at 23rd September. Table 1 gives an overview on the measurements that have been conducted during those times.

To characterize the forest ecosystem structure five circular permanent sample plots with 30 m diameter were established in the vicinity of the flux tower. These were located along a virtual line to the south at distances ranging from 50 to 200 m which distributes them such that they are located within the footprint area of the tower according to the main wind direction during that time of the year. Within those permanent sample plots all trees with a diameter at breast height (dbh) more than 6 cm were recorded to account for the tree number by species. Height measurements were conducted for 20% of trees per plot which have been randomly chosen. Each species has been handled as a separate element in that procedure. The age was determined by counting the rings on increment cores for one average tree per major species. The stand characteristics were calculated separately for the dominant (Table 2) and suppressed tree layers. The forest around the flux tower site is secondary growth managed with clear cuts of various sizes (1–5 ha) resulting in quite fine mosaic of stands. The regeneration of this type of stands is usually a combined method with planted coniferous and naturally established hardwoods in mixture. The young stands are treated to reduce the proportion of hardwoods in favor of the economically more valuable coniferous species (both planted and advance regeneration). This type of management which employs a natural regeneration after stand-replacing disturbances and following population control is the characteristic management according to medium intensity forestry (Metsläid et al., 2007) and a common practice in Estonian forestry. The plots were dominated by Norway spruce (*Picea abies* (L.) Karst.) and the forest site type according to the Estonian classification is the *Oxalis-Myrtillus* (Löhmus, 2004) type. We found as co-dominant species silver birch (*Betula pendula* Roth.) and black alder (*Alnus glutinosa* L.) and due to the presence of other species such as *Populus tremula* L., *Alnus incana* (L.) Moench, *Tilia cordata* Mill., and *Fraxinus excelsior* L. the stand mixture is fairly complex. Of particular importance is the presence of a suppressed tree layer (mean height of 6.4 ± 0.6 m over the five plots) which affects turbulent air flows in the stand. The main components of ground vegetation are *Oxalis acetosella* (L.), *Vaccinium myrtillus* (L.), *Calamagrostis arundinacea* (L.), *Convallaria majalis* (L.), and *Melampyrum pratense* (L.). The moss layer consists of several species and partly at humid spots we found *Sphagnum* spp. The mean N concentration in the aboveground litter was $1.53 \pm 0.10\%$, and the mean C concentration $48.65 \pm 0.72\%$.

As the site is located in the Lake Peipsi depression (Arold, 2005) and bordered by wetland massifs the soils of the site are strongly groundwater influenced, and employed a high groundwater table due to the vicinity to Lake Peipsi and the low altitudes of forest grounds. The soil around the flux tower showed a thick raw humus horizon with an average thickness of 24 cm, and the underlying sediment material was characterized by blue gleyic spots. The soil type was Haplic Gleysol (eutric) (WRB, 2006). Due to high clay content of the soil, the soil hydraulic conductivity is low; implying that the soils will stay wet longer times after snowmelt in spring. If affected by drought, even heavy rainfalls poorly penetrate into the soil and much of the precipitation is lost as runoff (Niinemets et al., 1999). To obtain data representing the area at larger scale, additional 4 sampling plots were established to the east direction of the flux tower. The soil types were Haplic Podsol and Haplic Stagnosol (WRB, 2006) and the forest site type *Oxalis-Myrtillus*.

2.2. Eddy covariance system

Eddy covariance (Baldochi et al., 1988; Aubinet et al., 2000) CO₂, H₂O and energy flux measurements were conducted above

the forest canopy on a 20 m tall tower (referred to as overstory flux measurements) and below the canopy at 1.5 m height above the ground (referred to as understory flux measurements). The three wind components and the speed of sound were measured by three-dimensional sonic anemometers (2008: CSAT3, Campbell Scientific, Logan, UT, USA; 2009: Metek USA-1, Metek GmbH, Elmshorn, Germany), CO₂ and H₂O mol densities/fractions in 2008 (over- and understory) by an open-path infrared gas analyzer (IRGA) (Li-7500) and in 2009 (over- and understory, but not at the same time) by a closed-path IRGA (Li-7000, both Li-Cor, Lincoln, NE, USA). In the closed-path system air was sucked from the inlet, mounted 0.1 m below the centre of the sensor volume of the sonic anemometer, through a 35 m Teflon tube of 0.008 m inner diameter through a filter (Acro 50, Gelman, Ann Arbor, MI, USA) to the closed-path IRGA deployed in a measurement cottage at the base of the tower at a flow rate of 15.2 L min^{-1} (KNF Laboport, KNF GmbH Freiburg, Germany). The closed-path IRGA was operated in the absolute mode, flushing the reference cell with dry N₂ from a gas cylinder at 0.1 L min^{-1} . The open-path IRGAs were mounted 0.2 m below the centre of the sensor volume of the sonic anemometer in a horizontal position. Sonic anemometers, the intake of the closed-path inlet tube, and the open-path IRGAs were oriented into the main wind direction to minimize flow distortion and longitudinal sensor separation to the sonic anemometers. In 2008, data from the sonic anemometers and the open-path IRGAs were acquired at 20 Hz and transferred via the SDM protocol to data loggers (CR3000, Campbell Scientific, Logan, UT, USA). In 2009, the data from the sonic anemometer was transferred by serial protocol with a frequency of 10 Hz to a computer and closed path IRGA data were transferred as analog signals with the same frequency and captured by a datalogger (Pico ADC20, Pico Technology, St. Neots, UK) connected to the same computer. Half-hourly average eddy fluxes were calculated from quality controlled flux data as the covariance between the turbulent departures from the mean of the vertical wind speed and the CO₂ and H₂O mixing ratios as described in detail in Haslwanter et al. (2009) and will thus not be repeated here. Net CO₂ and H₂O fluxes were calculated as the sum of the corrected vertical eddy term and the storage fluxes, the latter being estimated from the time-rate-of-change in scalar density at the respective reference height. Negative fluxes represent transport from the atmosphere towards the surface, positive ones the reverse.

2.3. Leaf level photosynthesis

We used a portable infrared gas analyzer (IRGA) cuvette system (Walz, GFS3000, Walz GmbH, Effeltrich, Germany) for leaf level photosynthesis measurements. Broad leaved species were measured using a clamping cuvette with parameter settings: flow rate of 1 L min^{-1} , $380 \mu\text{mol mol}^{-1} \text{ CO}_2$, 75–80% humidity and 25 °C. Conifers (*P. abies*) were measured using the conifer chamber (provided by Walz for GFS3000), where small twigs can be fitted in. To avoid clamping stress, twigs were selected and prepared 3 days before the measurements by removing a portion of the needles and wrapping the site of needle removal airtight in Teflon tape. Trees in the top part of the canopy (*P. abies* and *B. pendula*) were accessed from the scaffoldings located at the measurement site (2 towers of 16 and 20 m height) and were measured at a height of 16 and 18 m (maximal canopy height about 20 m). To represent sun and shade conditions, we choose leaves that are exposed to south, west and north directions. Leaves of trees in the understory (*P. abies*, *T. cordata*, *B. pendula* and *B. pubescens*) were measured at a height between 0.1 and 1.5 m and are supposed to be all under shade condition during summer time where the canopy is densely closed by the leaves of the deciduous trees. Leaves of trees in forest gaps were measured at heights between 0.4 and 1.5 m and we choose mostly

Table 2
Description of the dominant canopy layer of sample plots at the atmosphere–forest measurement campaign site. The proportion of the tree species is calculated on volume basis of stand total wood volume.

Plot number	Stand volume (m ³ ha ⁻¹)	Number of trees (ha ⁻¹)	Proportion (%)					
			<i>Picea abies</i>	<i>Betula pendula</i>	<i>Alnus glutinosa</i>	Other	Age	Height
1	171.4	1789	46.6	46.0	0.6	7.4	28	14.3
2	72.8	2395	39.8	34.5	25.3	0.4	21	10.0
3	54.2	2621	46.7	41.1	12.0	0.2	29	8.9
4	116.7	2621	43.9	28.7	–	27.4	23	10.2
5	210.8	2015	57.6	39.6	1.7	1.1	27	14.4

sun exposed leaves to assess if there is difference to the sun exposed leaves in the top canopy.

2.4. Trace gas analyses

The trace gas analyzers have been installed within the measurement cottage. We took their inlet airstreams from above canopy by a Teflon tube that reached to the top of the tower and a bypass inlet at 2 m height to allow sampling also below canopy. The ozone was detected using a Thermo (Model 49i) ozone analyzer and NO/NO₂/NO_x were detected with a Thermo (Model 42i), both Thermo Scientific, Waltham, MA, USA. During the campaign in 2008 we had additionally an air quality measurement transportable laboratory, equipped with instruments for NO_x (Horiba APNA-370), SO₂ (Horiba APSA-370) both from Horiba Jobin Yvon GmbH, Unterhaching, Germany and particulate matter (PM₁, PM_{2.5} and PM₁₀, Grimm EDM107, Grimm Aerosol Technik GmbH & Co. KG, Ainring, Germany) sensors, placed on the side of a small forest road 100 m west of the flux tower.

2.5. Manual soil chamber measurements of CH₄, N₂O and CO₂ soil fluxes

One plot was selected near to the understory eddy covariance system and two circular stainless steel collars with a diameter of 305 mm were inserted about 6 cm into the top soil. The placement of the collars took place in the beginning of the campaign in 2008 and measurements conducted refer to the campaign in 2009 (Table 1). The effective chamber volume (23.8 L) of the cylindrical stainless steel chambers was calculated from the average height from the soil surface to the top of the chamber. The chamber atmosphere was mixed by a small built-in fan to prevent the formation of gradients (Frenzel and Karofeld, 2000) and sealed air tight with water. Pre-installed thermocouple sensors connected to a thermocouple reader (Comark KM330) registered the temperatures in soil (–0.1 m, –0.15 m, –0.25 m of depth), inside the chambers, and in the atmospheric air (0 m, 0.5 m, 2 m of height).

Samples were drawn into 100 mL gas-tight syringes from the top of the chamber via a capillary (diameter 2 mm). Thirty seconds before each sampling, the capillary air was mixed for four times using the syringe. The samples were injected into 7 mL non-evacuated vials (Labco Exetainer 768W). The vials were flushed with 80 mL of sample air and over-pressurized with 20 mL of sample air. The chambers were operated in closed static mode and closed for a maximum of 30 min. 6–10 samples were collected during this time. These were stored in darkness at –18 °C and analyzed within 2 months after sampling by gas chromatography with a GC-FID/ECD (model HP 6890) and a GC-TCD (model HP 7890) as described by Syväsalo et al. (2004). All flux rates were calculated from the exponential change in gas concentrations in the chamber and temperature corrected (Appendix A). Air temperature inside the chambers was taken each time a sampling was conducted.

2.6. Automatic chamber measurements of CO₂ soil flux

Within the five circular sampling plots and at four reference locations outside of the sample plots, two stainless steel collars were placed in each, 7 m north and 7 m south from the plot center or at similar distance if outside a plot. The placement of the collars took place in the beginning of the measurement campaign 2009. In total, 18 collars of the size of 230 mm in diameter and 80 mm in height were installed and inserted about 2–3 cm into the top soil in order to eliminate diffusion through the soil away from the measurement area.

Soil respiration was measured using an automatic CO₂ Exchange System ACE (ADC BioScientific Limited, UK) equipped with a soil chamber, an infra-red gas analyzer (IRGA), a sensor for detecting photosynthetic active radiation, a soil moisture sensor (SM200, Delta-T Devices Ltd., Cambridge, England) and two temperature sensors. The soil chamber was equipped with a small fan for mixing the air. Volume of the lid of the soil chamber was 2.6 L. Upon measurement the lid was automatically placed tightly on the collar. Tight closing of the soil chamber was guaranteed by a rubber ring-seal in the contact surface between the rims of the lower part of the chamber (collar inserted under a special rim of the frame of the station) and its upper (guided) counterpart. The height of the collars from the soil surface was measured at every occasion. The ACE station was operated in closed mode with zero option. In this mode the increase of CO₂ in the chamber from soil activity is measured. The zero measurement is made by passing CO₂-free air through the chamber. In that mode, the chamber is operated as a closed static chamber (Pumpanen et al., 2004) and details about the data handling are given in Appendix A. Continuous measurements were conducted in one collar using the interval of 15 min between each measurement cycle of 5 min. Single repeated measurements were conducted upon the time of collar installments, during the main campaign, and later in October and November 2009. 2–10 sequential measurements were conducted. Simultaneously with the soil flux measurements, soil temperature and moisture were measured at the depth of 5 cm, and air temperature on the ground level.

Litter samples were collected in each plot. To distinguish the share of respiration originated from the fallen litter and from the soil, upon measurement the litter was removed in one collar (always the southern one) of each plot. The respiration from the decomposition of litter would result from the difference between the collars. The litter was taken to the lab, dry weight measured and analyzed for C and N contents.

2.7. BVOC sampling and analyses

We measured volatile organic compounds (VOC) emitted from leaves in parallel with leaf physiological parameters by diverting a part of the Walz leaf and conifer cuvette outflow. The VOC's were adsorbed onto multibed stainless steel cartridges (10.5 cm length, 3 cm inner diameter, Supelco, Bellefonte, USA) filled with Carbotrap C 20/40 mesh (0.2 g), Carbopack C 40/60 mesh (0.1 g) and Carbotrap X 20/40 mesh (0.1 g) adsorbents (Supelco, Bellefonte, USA) at

a flow rate of 250 mL min⁻¹ for 10 min. (altogether 2.5 L air). Background air samples were collected from the empty chamber before and after the measurements. Adsorbent cartridges were analyzed with a combined Shimadzu TD20 automated cartridge desorber and Shimadzu QP2010 plus GC–MS instrument (Shimadzu Corporation, Kyoto, Japan). The TD20 parameters, GC–MS conditions and compounds identification were presented in detail in Copolovici et al. (2009) and Toome et al. (2010).

From the manually operated soil chambers VOC samples were taken analogously as from the leaves using the same flow rates of 250 mL min⁻¹ but sampling for 20 min. After sampling a background air sample, the chamber was closed for a period of 30 min and then the sample was taken onto the steel cartridge. To avoid underpressure in the water-sealed chamber, we allowed background air to enter via a small Teflon tube to the bottom of the chamber during sampling using a valve sealing that allows to open an inlet while sampling. That led to a change in the operation mode from a closed static chamber to an open dynamic chamber. The data handling is described in detail in Appendix A. Subsequent GC–MS analysis of the VOC soil emissions have been conducted as described above.

3. Results and discussion

3.1. Net ecosystem CO₂ exchange

The net ecosystem CO₂ exchange (NEE) above the canopy exhibited a clear diurnal cycle with net uptake during daytime and net loss of CO₂ during nighttime (Fig. 2 and Table 3). These fluxes fall in between those observed for European deciduous (Granier et al., 2003) and coniferous (Bernhofer et al., 2003; Ceulemans et al., 2003) forest ecosystems and likely reflect the mixed nature of the stand. In contrast, the NEE measured in the understory exhibited hardly any diurnal cycle and was positive for almost all of the time (Fig. 2 and Table 3). Consistent positive daytime understory NEE for summertime conditions were also reported by Misson et al. (2007) for 8 out of 11 study sites. The contribution of the storage flux to the NEE was negligible in the understory (Misson et al., 2007), while an appreciable negative and positive storage flux (Fig. 2) in the morning and evening, respectively, was detected with the overstory system indicative of venting and accumulation of canopy air, respectively (Aubinet et al., 2000). Flux divergence was observed during nighttime, when the respiration measured above the canopy was lower than in the understory (Fig. 2). This may reflect differences in the footprint between the over- and understory eddy covariance systems which generally differ greatly in extent (Baldochi, 1997) and in addition sometimes differed in direction due to occasional decoupling of flow directions above and below the canopy (data not shown). The differences in nighttime over- and understory respiration may also be indicative of advection (Aubinet et al., 2000), part of the CO₂ being respired by the soil and understory vegetation potentially being advected (horizontally and/or vertically). In order to confirm the latter hypothesis, however, detailed measurements of within-canopy horizontal and vertical gradients in CO₂ concentration and wind speed would be required (Feigenwinter et al., 2008).

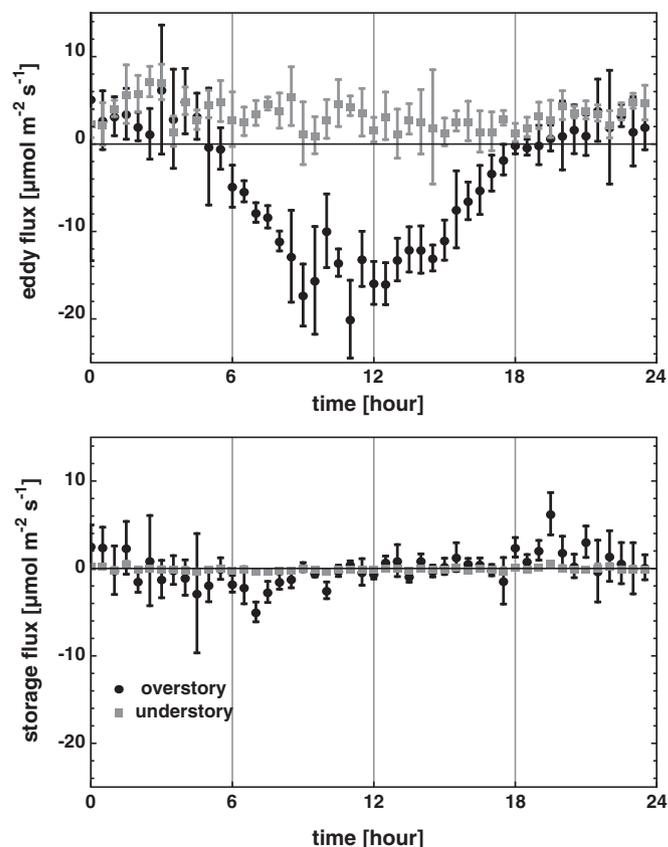


Fig. 2. Eddy fluxes (upper panel) and storage fluxes (lower panel). Overstory fluxes are black dots and understory fluxes gray squares. Median daily fluxes were compiled from the measurements in 2008 and error bars denote standard errors.

3.2. Reactive trace gases

3.2.1. Ozone and nitrogen oxides

The formation of tropospheric ozone is bound to the availability of sunlight and NO_x which provide the energy and the primary source of oxygen atoms, respectively. VOCs act as catalysts of this reaction (Atkinson, 2000; Atkinson and Arey, 2003). As seen in Fig. 3, ozone varied in a clear diurnal cycle above and below the canopy. Lowest values were found under nighttime conditions, while the highest values occurred during daytime. The peak in ozone concentrations above canopy was broader and somewhat skewed towards the afternoon, while below canopy ozone concentrations peaked around noon and reached nighttime levels about

Table 3

Mean values and standard errors of the ecosystem CO₂ fluxes (August 2008). All fluxes are expressed in μmol m⁻² s⁻¹.

		Overstory			Understory		
			Min	Max		Min	Max
Daily total	f_{cn}	-4.59 ± 1.01	-19.83	4.56	3.60 ± 0.23	1.19	7.15
	f_{sn}	0.1 ± 0.3	-4.97	6.26	0.03 ± 0.03	-0.35	0.66
Daytime	f_{cn}	-10.28 ± 0.99			2.93 ± 0.99		
	f_{sn}	-0.5 ± 0.3			-0.04 ± 0.03		
Nighttime	f_{cn}	2.92 ± 0.25			4.34 ± 0.34		
	f_{sn}	0.74 ± 0.41			0.09 ± 0.04		

f_{cn} , net CO₂ flux; f_{sn} , net storage flux.

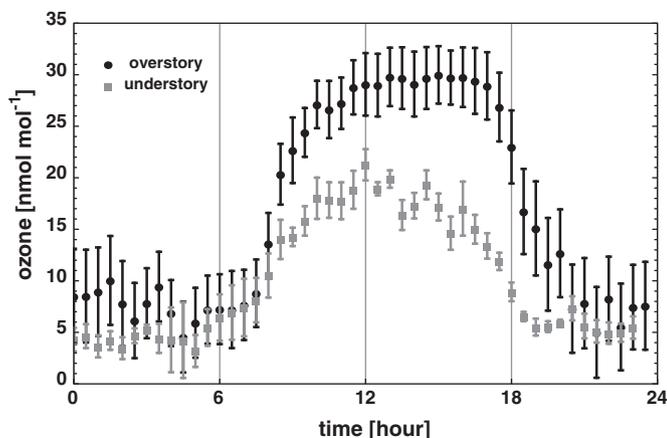


Fig. 3. Comparison between the ozone mixing ratios above and below the canopy. The data sets have been combined from both measurement campaigns. Ozone data was acquired with 1 s intervals, and half hourly averaged. From the combined data sets, medians have been calculated and error bars denote standard errors.

1 h earlier. Typical daily variations in ozone concentration above forest canopies have been reported to span the range from 10 to 70 nmol mol⁻¹ (Mikkelsen et al., 2000), which is well represented by our findings as well.

NO and NO₂ concentrations showed as well diurnal cycles where NO ranged between 0.05 and 0.3 nmol mol⁻¹ and NO₂ between 0.2 and 1.5 nmol mol⁻¹ with lower values during nighttime. A slight canopy gradient with smaller values below was found. Nitrogen oxides, as originated from burning processes and soil efflux, were found to correlate with wind direction (Fig. 4). Westerly winds caused higher NO and NO₂ concentrations, while smallest concentrations were associated with easterly winds. Cities and settlements as well as an access road to the region are located to the West of the measurement station. Ozone concentrations followed that same pattern and were highest on days with West to South-West winds and lowest with winds from the East (Fig. 4).

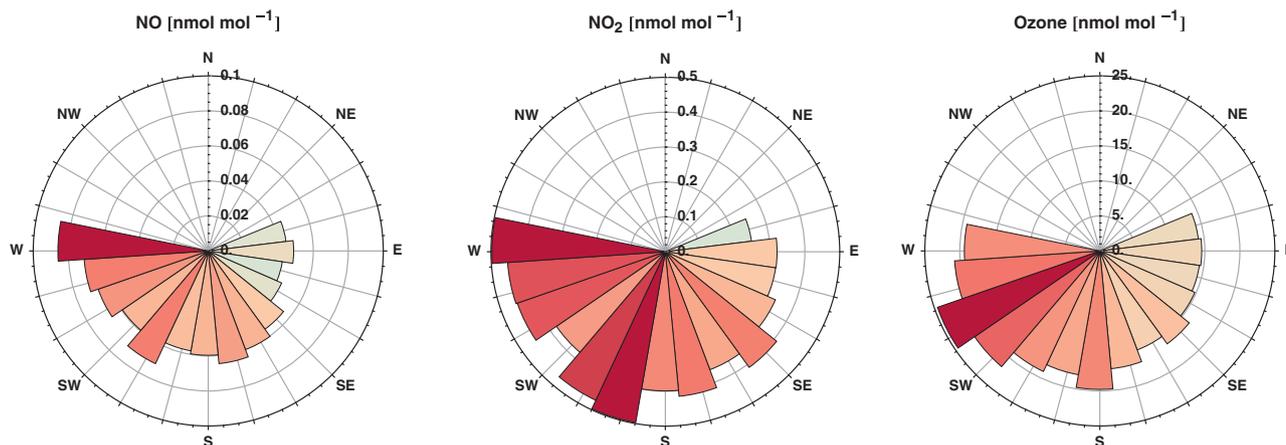


Fig. 4. Relationship between the wind direction and the mixing ratios of nitrogen oxides (NO and NO₂) and ozone during the measurement campaign in 2009. Darker color refers to higher mixing ratios.

Table 4

Comparison of trace gases and particles ($\mu\text{g m}^{-3}$) measured at the roadside (bus) and the flux tower in the forest (cottage).

Location	NO	NO ₂	SO ₂	O ₃	PM1	PM2.5	PM10
Instrument	Bus APNA 370	Cottage TEI 42i	Bus ASNA 370	Cottage TEI 49i	Bus Grimm monitor N 180		
Median	0.1–0.7	0.2–0.9	0	43.33	4	6	14.50
95%	1.2–1.5	1.2–2.3	0.4	78.2	12	20.0	75.0
5%	0–0.2	0–0.2	0	5.71	2	2.90	5.50

Ozone uptake to forest canopies has been reported for temperate and boreal ecosystems (Mikkelsen et al., 2004; Rannik et al., 2009). That uptake is mostly reported to occur through stomata, surface deposition and also by reaction with emitted BVOC. While the difference in the ozone concentrations above and below the canopy are suggesting that there should be a net ozone uptake by the canopy the question remains to what extend the different environmental conditions above and below the canopy influence that flux.

3.2.2. Particulate matter and anthropogenic influences

In order to assess possible anthropogenic influences on the location of the flux tower we compared the measurements of the air quality measurement bus and the measurements within and above the canopy. Table 4 summarizes the results. If compared to the reported Estonian yearly background values (Pajuste et al., 2004), the situation found in Järvselja is about 2–3 times lower. Sulfur dioxide (SO₂) is predominantly of anthropogenic origin and plays a role in acid rain formation, which has a negative impact on forest ecosystems and tree growth. The values found indicate, that the measurement site is not much influenced by SO₂ emissions. The same picture was found for NO_x. Especially the low NO values, direct tracers of nearby combustion, support the conclusion that Järvselja is located in an area with low human impact on air pollution.

Particulate matter concentrations (PM1, PM2.5 and PM10) also remained far below the national threshold (50 $\mu\text{g m}^{-3}$, averaged over 24 h for a period of 20 years) and characterize the site as between “remote” and “non-urban continental” (Seinfeld and Pandis, 2006). In conclusion, the measurement site at the flux tower in Järvselja can be graded as an atmospherically clean and low anthropogenic impacted site.

3.2.3. BVOC emissions

To assess the state of the atmosphere and the flux of carbon within the forest ecosystem at Järvselja, we also conducted measurements of leaf level and soil BVOC fluxes. These were

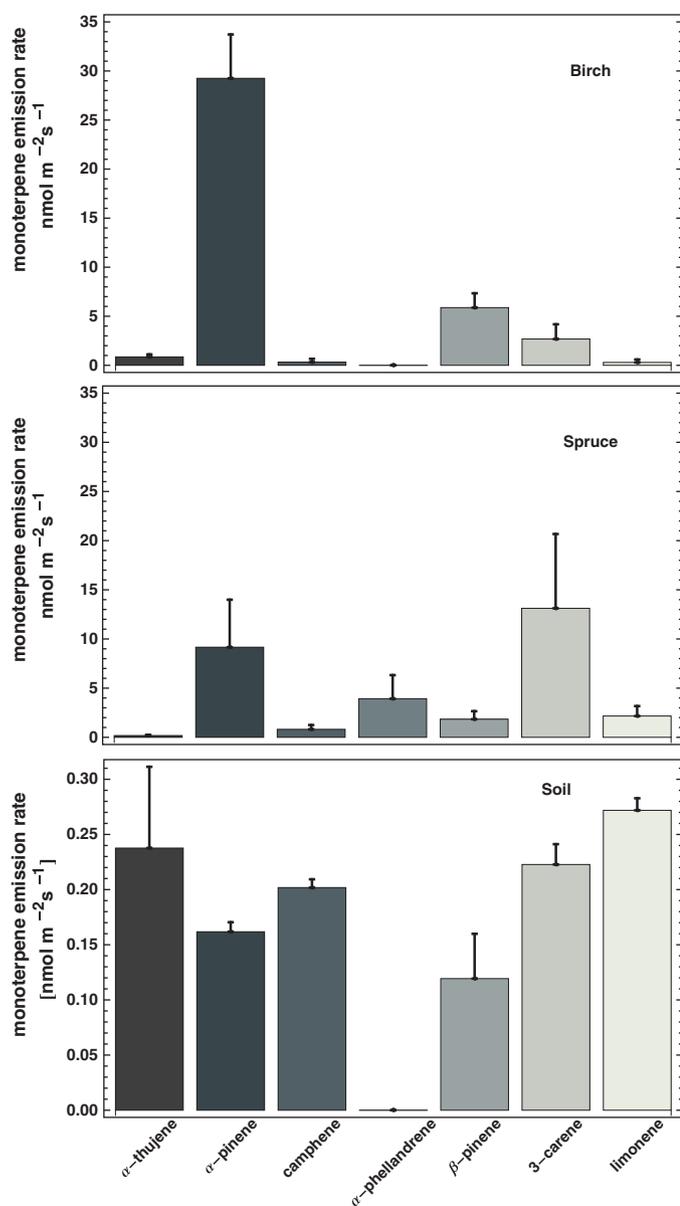


Fig. 5. Monoterpene emissions from the main tree species *B. pendula* and *P. abies*, averaged over leaves and branches differently exposed to sunlight. We used in both cases West, South and North exposed leaves or branches. The lower panel shows monoterpene emissions from soil and leaf litter. Error bars indicate standard errors.

focused on isoprene and monoterpenes. Largest monoterpene emission rates (sum of monoterpenes, $46 \pm 9 \text{ nmol m}^{-2} \text{ s}^{-1}$) were found from *Betula* with α -pinene as the major monoterpene emitted. *P. abies* emitted a total monoterpene flux of $33 \pm 11 \text{ nmol m}^{-2} \text{ s}^{-1}$, where Δ^3 -carene was the main contributing monoterpene followed by α -pinene and α -phellandrene (Fig. 5). The isoprene fluxes for *B. pendula* ($1.2 \pm 0.6 \text{ nmol m}^{-2} \text{ s}^{-1}$) and *P. abies* ($1.02 \pm 0.36 \text{ nmol m}^{-2} \text{ s}^{-1}$) showed no substantial differences. Compared to the total monoterpene emissions, isoprene emissions were about 45 times smaller.

Total soil monoterpene fluxes were found to be 40 times smaller ($1.22 \pm 0.16 \text{ nmol m}^{-2} \text{ s}^{-1}$) than leaf emission rates. As isoprene is synthesized from recently fixed carbon by plants, the very small flux from the soil ($0.05 \pm 0.01 \text{ nmol m}^{-2} \text{ s}^{-1}$) may be caused by the enclosed small plants and mosses which are known to emit isoprene as well (Tiiva et al., 2009). Another possibility is that soil bacteria are also involved in the isoprene fluxes – release and consumption of isoprene by soil bacteria has been reported

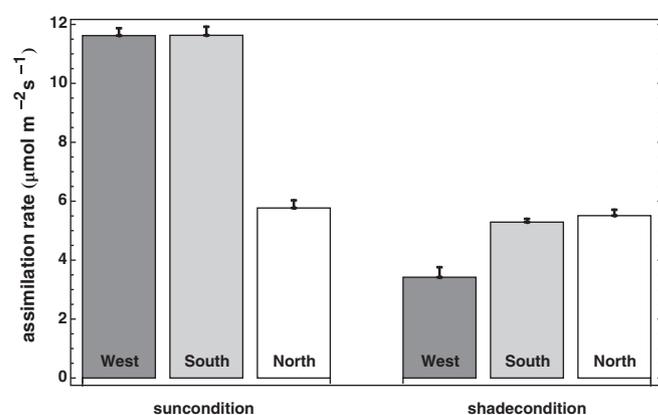


Fig. 6. Photosynthetic net assimilation rate of *B. pendula* on leaf level, averaged over 3–5 leaves. To simulate high light conditions (denoted sun condition) the measurement was conducted with a quantum flux of $1000 \mu\text{mol m}^{-2} \text{ s}^{-1}$ and shade conditions were simulated by a quantum flux of $200 \mu\text{mol m}^{-2} \text{ s}^{-1}$. Directions given in the bars refer to the direction in which the leaves measured have been exposed. Error bars denote standard errors.

(Cleveland and Yavitt, 1998; Scholler et al., 2002). The composition of monoterpene soil fluxes (Fig. 5) shows some interesting patterns as compared to the monoterpene emissions from the leaves of the species mainly contributing to the leaf litter in the soil O-horizon. We found, that α -phellandrene is lacking completely and that the main contributing monoterpenes at the leaf level, α -pinene and Δ^3 -carene, are less abundant in the soil monoterpene fluxes. Instead, α -thujene and limonene, which have been found less abundant at the leaf level, are main contributors to the soil monoterpene fluxes. It is known, that monoterpene emission patterns are changing with the seasons and that stored monoterpenes, i.e. from resin ducts, exhibit different patterns than emission from newly synthesized monoterpenes (Hakola et al., 2003; Vuorinen et al., 2005; Holzke et al., 2006; Räisänen et al., 2008). Our result suggest, that α -thujene and limonene may be stored to greater amounts in the leaf tissues related to their synthesis when still attached to the branches and that α -phellandrene seems not to be stored in substantial amounts. Given the fact, that deciduous leaves mostly do not build specialized storage organs we can speculate that soil monoterpene emissions are caused by the coniferous litter. The contribution of the different litter parts, needles, leaves, cones etc. to the terpene fluxes remains to be investigated for our measurement site. As it is known that beside the leaf litter soil fungi and roots also emit different volatiles (Leff and Fierer, 2008) their contribution to the total emitted soil fluxes presented here have to be assessed in future.

3.3. Leaf level photosynthesis

Betula pendula is the most abundant, deciduous tree species present in both over- and understory of the experimental site. Leaf level net photosynthetic rates showed a clear dependency on and adaptation to light availability. *Betula* leaves at the top of the canopy (about 16–18 m) showed highest net assimilation rates when exposed to light fluxes of $1000 \mu\text{mol quanta m}^{-2} \text{ s}^{-1}$. Leaves that exposed to south and west directions were found to exhibit twice as high net assimilation ($11.6 \pm 0.3 \mu\text{mol m}^{-2} \text{ s}^{-1}$) as compared to north exposed leaves ($5.7 \pm 0.4 \mu\text{mol m}^{-2} \text{ s}^{-1}$; Fig. 6). To simulate also shade conditions, the same leaves were measured with a light flux of $200 \mu\text{mol quanta m}^{-2} \text{ s}^{-1}$. In this case, lowest net assimilation rates were measured in west exposed ($3.5 \pm 0.6 \mu\text{mol m}^{-2} \text{ s}^{-1}$) and highest in north exposed ($5.5 \pm 0.3 \mu\text{mol m}^{-2} \text{ s}^{-1}$) leaves (Fig. 6). Photosynthetic photon flux density (PPFD) in the top of the canopy was between 130 and $1230 \mu\text{mol quanta m}^{-2} \text{ s}^{-1}$ for south- and west-exposed leaves

Table 5

CO₂ assimilation and transpiration from tree leaves in the understory at a photon flux density of 200 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$. Values shown are mean \pm standard error of 3 different trees (3–7 measurements per tree).

Species	CO ₂ assimilation ($\mu\text{mol m}^{-2} \text{s}^{-1}$)	Transpiration ($\text{mmol m}^{-2} \text{s}^{-1}$)	N
<i>Betula pendula</i> ^a	2.18 \pm 0.13	0.78 \pm 0.10	7
<i>Betula pubescens</i>	1.97 \pm 0.14	1.03 \pm 0.10	5
<i>Tilia cordata</i>	3.79 \pm 0.27	1.12 \pm 0.10	3

^a Overstory assimilation values measured under 200 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$ (shade condition) are inserted from data given in Fig. 5 for comparison.

and between 261 and 77 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$ for north-exposed leaves. Interestingly, the photosynthesis of north-exposed *Betula* leaves in the top of the canopy seemed to be light-saturated already at a rather low photon flux density of 200 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$. Increasing light to 1000 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$ increased photosynthesis only marginally, while the photosynthesis rate of west- and south-exposed leaves was about doubled (Lichtenthaler et al., 2007). Adaptations of the photosynthetic apparatus to light intensities are explaining that behavior (Niinemets et al., 1998, 1999, 2004).

P. abies is the most abundant tree species in the overstory of the measuring site. Net assimilation rates measured at 16–18 m height and a light flux of 1000 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$ were 7.9 \pm 1 $\mu\text{mol m}^{-2} \text{s}^{-1}$. Although *P. abies* net assimilation rate is lower than that of *B. pendula* (as was to be expected), it is still appreciable and in the normal to high range (Wallin et al., 1990; Marek et al., 1997). The expositions of the branches measured have been West, South and North.

Net assimilation rates of *B. pendula* leaves in the understory (apparent light was between 6 and 69 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$) were about 2 $\mu\text{mol m}^{-2} \text{s}^{-1}$ when measured with a light flux of 200 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$ (Table 5) and therefore much lower than overstory leaves measured under the same conditions (Fig. 6). For *B. pendula*, a light-requiring species, only small seedlings were found in the understory. Another typical pattern in forest ecosystems are gaps due to fallen or removed trees. We measured also in such gap situations. Light conditions there were much improved compared with the understory (apparent light was between 7 and 824 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$) and due to that we measured again with 1000 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$. The net assimilation rate of *Betula* leaves were very variable, ranging between 5.6 and 9.6 $\mu\text{mol m}^{-2} \text{s}^{-1}$, at average about 8 $\mu\text{mol m}^{-2} \text{s}^{-1}$, almost as high as in the overstory (Table 6). Of the species measured in the understory (3 species shown in Table 6), *Tilia* had higher net assimilation rates than the two *Betula* species. The gap offered a niche for those species, which were not found in the understory, like *Quercus robur* and *Populus tremula*. In general, the canopy discriminates the irradiative energy input strongly. In particular, the photosynthetic active radiation (PAR) input that is absorbed more strongly than near infra-red light can be even less than 1–2% of above-canopy value in these types of forests (Niinemets et al., 1998).

Table 6

CO₂ assimilation and transpiration from tree leaves in a forest gap at a photon flux density of 1000 $\mu\text{mol quanta m}^{-2} \text{s}^{-1}$. Values shown are mean \pm standard error of 3 different trees (4–8 measurements per tree).

Species	CO ₂ assimilation ($\mu\text{mol m}^{-2} \text{s}^{-1}$)	Transpiration ($\text{mmol m}^{-2} \text{s}^{-1}$)	N
<i>Quercus robur</i>	5.14 \pm 1.22	1.46 \pm 0.32	4
<i>Populus tremula</i>	8.24 \pm 0.88	2.97 \pm 0.09	5
<i>Betula pendula</i>	8.03 \pm 1.23	2.29 \pm 0.25	8

Table 7

Mean soil CO₂ effluxes and key environmental variables at Järvelja, measured with the automatic chamber system during the campaign in 2009. Time of measuring was between 12 and 17 h. Data given as mean (of 2–10 measurements) \pm standard error.

Date	16 September	7 October	23 November
Soil efflux, $\mu\text{mol CO}_2 \text{ m}^{-2} \text{ s}^{-1}$	8.40 \pm 0.82	9.60 \pm 0.04	5.45 \pm 0.53
$T_{\text{air}}, ^\circ\text{C}$	13.3 \pm 0.3	8.4 \pm 0.0	6.3 \pm 0.1
$T_{\text{soil}}, ^\circ\text{C}$	12.6 \pm 0.1	8.4 \pm 0.1	5.7 \pm 0.0
PPFD, $\mu\text{mol m}^{-2} \text{ s}^{-1}$	5.30 \pm 1.14	21.50 \pm 1.50	5.00 \pm 1.10
Soil moisture % (v/v)	67.0 \pm 5.8	69.1 \pm 0.5	81.9 \pm 10.9
Number of collars used	3	1	3

3.4. Soil chambers

3.4.1. Soil CO₂ efflux

Manual chamber measurements resulted in a mean soil CO₂ efflux of 3.54 \pm 0.62 $\mu\text{mol m}^{-2} \text{s}^{-1}$. Compared to boreal soil fluxes ranging between 2 and 6 $\mu\text{mol m}^{-2} \text{s}^{-1}$ in summer (Rayment and Jarvis, 2000; Kolari et al., 2006, 2009) and cool temperate forest ecosystem soil fluxes ranging between 2 and 8 $\mu\text{mol m}^{-2} \text{s}^{-1}$ (Lee et al., 2002; Yuste et al., 2005) depending on the species contribution the values found in Järvelja fit into that picture. The average air temperature recorded on the soil surface was 17.7 \pm 1.3 $^\circ\text{C}$ and the average soil temperature at -0.1 m was 13.6 \pm 0.5 $^\circ\text{C}$. Mean soil CO₂ efflux was 4.29 \pm 0.11 $\mu\text{mol m}^{-2} \text{s}^{-1}$ over the continuous measurement period of 72 h with the automatic chamber system. Average volumetric soil moisture was 51.36% (max 56.97%, min 45.71%), the average air temperature 10.2 \pm 0.2 $^\circ\text{C}$, and the average soil temperature 11.7 \pm 0.1 $^\circ\text{C}$ during the days of measurements. The average daytime temperature from 9 to 18 h was very stable at 11.9 \pm 0.3 $^\circ\text{C}$. Results of the additional single measurements are presented in Table 7. In both cases, manual and automatic chamber measurements, we found rather stable emissions over the course of the day which is in accordance with the stable soil temperature during the measurements. As the chambers have been operated under shade conditions in the understory, the average temperature change during measurements have been 0.9 \pm 0.5 $^\circ\text{C}$. The data, obtained on both plots, give hints about the plasticity which occurs through changes in soil type and canopy structure and may be reflected by substantial changes in soil CO₂ efflux.

Even though soil respiration is known to correlate positive with soil temperature which is influenced by soil moisture and PAR (Lloyd and Taylor, 1994; Ruehr and Buchmann, 2010), we found only weak correlations which is mainly caused by the limited number of the measurements during the campaign in 2009.

Comparison of the two methods of continuous measurement cycles (Fig. 7) revealed that greater values of CO₂ efflux were recorded with the automatic chamber system. Even though, the mean daytime air temperature was by 5.8 $^\circ\text{C}$ lower and there was one week difference between the first conducted manual chamber measurements. Given the temperature dependency of soil respiration, this is a contradictory result, which may be reconciled based on differences in the sampling and analysis techniques employed. Among the technical explanations, site heterogeneity, different depth or the time of collar installation might have played a role as well.

Forest floor soil respiration measured may represent more than 75% of total ecosystem respiration (Solondz et al., 2008). Compared with the results of the understory eddy covariance fluxes as given by Table 3 our chamber fluxes exceeded the daytime eddy fluxes as measured in 2 m height by about 20% for the manual chamber measurements and on the whole day basis about 19% as com-

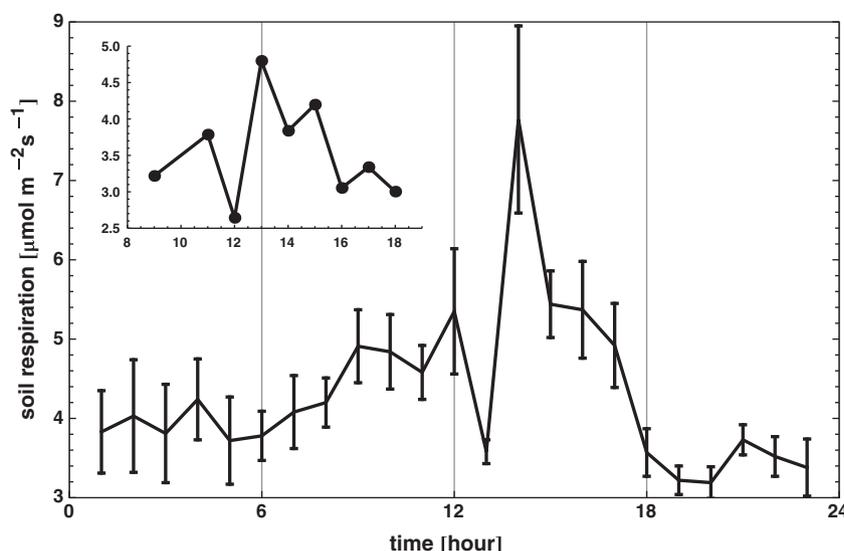


Fig. 7. Daily mean soil respiration measured over 3 days during the 2009 campaign by the automatic chamber system. Error bars denote standard errors of hourly measurements ($N=2-9$). The inset shows a one day data set of soil respiration fluxes measured with the manual chamber system.

pared to the mean fluxes from the automatic chamber over the 24 h period. Given the differences in footprint between both methods (Balducchi, 1997), the random and systematic uncertainties associated with each method (Hollinger and Richardson, 2005), and finally the differing time frames during which these measurements were made, we consider these results as very encouraging.

3.4.2. CH_4 and N_2O soil fluxes

Fluxes of methane and nitrous oxide were measured using the manual chamber system. We found negative methane fluxes during the campaign in 2009, which indicates a possible net consumption of CH_4 in soil (Chan and Parkin, 2001; Mori et al., 2008) which was also reported for boreal forest ecosystems (Whalen et al., 1992) or a possible atmospheric reaction of methane with hydroxyl radicals given sufficient high levels of NO_x (Wuebbles and Hayhoe, 2002) enclosed into the chamber. It is further known, that dry soils are sinks and wet soils act as sources for atmospheric methane as well as that tree species affect the source and sink properties of forest soils (Saari et al., 1998; Borken and Beese, 2006). The daytime average CH_4 flux was $-0.69 \pm 0.36 \text{ nmol m}^{-2} \text{ s}^{-1}$. If compared to boreal soil methane fluxes in Hyytiälä, Finland ($61^\circ 84'N$, $24^\circ 29'E$) of about $-1.4 \text{ nmol m}^{-2} \text{ s}^{-1}$ (Pihlatie et al., 2009a) we found a slightly lower sink flux. The inverse relationship between the CO_2 and CH_4 fluxes may support the idea of methane consumption by methanotrophic bacteria in the soil as a reason for the flux.

We found daytime nitrous oxide emissions that averaged $0.18 \pm 0.09 \text{ nmol m}^{-2} \text{ s}^{-1}$ which are about the same compared to soil N_2O fluxes at Hyytiälä of $0.15 \text{ nmol m}^{-2} \text{ s}^{-1}$ (Pihlatie et al., 2009b). N_2O is mainly produced by denitrification process in soils. In deeper layers, given wet and anaerobic conditions denitrification may occur frequently (Ball et al., 1999; Yamulki and Jarvis, 2002). In our groundwater influenced study site, the soil temperature should positively correlate with the N_2O efflux (Schindlbacher et al., 2004). Due to the limited number of measurements we observed only a weak positive correlation.

4. Conclusions and outlook

One might argue, and rightfully so, that the two measurement campaigns conducted at the Järvelja Experimental Forest station during the summers 2008 and 2009 only allowed us to take a snapshot of the multitude of trace gas fluxes which occur on a continuous basis between this forest and the atmosphere. While

this is likely to be correct, we strongly believe that the two measurement campaigns already unearthed a number of encouraging results which may serve as building blocks upon which we aim to continue research at this site. We have characterized the structure and composition of the forest and the soil, we conducted above- and below-canopy eddy covariance CO_2 and H_2O flux measurements, quantified leaf photosynthesis, transpiration and isoprene/monoterpene emissions, soil CO_2 , methane, nitrous oxide and monoterpene fluxes, as well as the above- and within-canopy concentrations of several reactive trace gases and particulate matter. Our major findings are that (i) in terms of the magnitude of the CO_2 exchange this mixed forest ranged in between boreal coniferous and temperate broad-leaved forests, that (ii) understory eddy covariance CO_2 fluxes and manual/automated soil respiration rates agreed reasonably, (iii) leaf and soil monoterpene emissions had contrasting signatures, (iv) soils were sinks for methane and sources for nitrous oxides, and (v) from an air quality point of view the study site was characterized as remote/rural. Future experimental efforts will be directed towards year-round flux measurements of a larger number of trace gases, of which some sources/sinks, such as of methane, nitrous oxide, isoprene and monoterpenes, were already characterized at the leaf and soil level. In a next step, these measurements both at the ecosystem and leaf/soil scale should be assimilated into process-oriented models of forest-atmosphere exchange in order to test our quantitative ability of simulating these processes. Combining these two lines of information, experimental field data and simulation analysis, will be critical in order to finally assess the role this and similar forests in this region are playing in modulating climate and how sensitive these ecosystems are to likely future climate.

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Appendix A.

We describe the concentration change in time by the flux from soil into a chamber by an exponential relation (Pedersen et al., 2001; Pumpanen et al., 2001, 2004):

$$C(t) = C_0 e^{kt} \quad (1)$$

where C_0 denotes the background concentration that is enclosed inside the chamber at the beginning of the closing period. In case of the manual chamber, the sampling procedure led to a series of data points $d = \{t_0, C_0, \dots, t_n, C_n\}$, which have been used to estimate the flux rate k by non-linear least-square fit of Eq. (1) to d . To calculate the molar flow F_c , we apply:

$$F_c = C(t_n) \frac{V}{A} \cdot \frac{1}{V_m} \quad (2)$$

with a given chamber volume V in m^3 , the covered area A in m^2 , and the molar volume V_m of 22.4 L mol^{-1} . We further assumed that the pressure stayed constant throughout the sampling period and corrected for the temperature changes by multiplying Eq. (2) with:

$$\xi(T) = \frac{T_0 - T}{T_0} \quad (3)$$

where T_0 is 273.15 K and T the temperature as measured from the chamber sensor in Kelvin.

We conducted also the BVOC sampling by usage of the manual chamber system. In this case, it is only possible to apply a data set of two measurement points $d = \{t_0, C_0; t_1, C_1\}$, with the sample at t_0 given the background concentration and at t_1 the sample which is sucked onto the Carbotrap cartridge. Rearranging equation 1 and application of the logarithm led to:

$$\log(C(t_1) - C(t_0)) = k \quad (4)$$

and we applied again Eqs. (1)–(3) to achieve the temperature corrected molar flow of BVOC emitted from the top soil layer including leaf litter at the start of the BVOC sampling procedure. While the amounts of aliquot we took from the manual chambers volume (23 L) with the syringes are neglectable if compared to the total chamber volume that is not true in case of the BVOC measurements where we took an aliquot of five liters per sample over a period of 20 min. To prevent underpressure we let stream in background air to the bottom of the chamber with a small tube while sampling and by that changing instantaneously the concentration within the chamber. That resembles the open flow chamber system as described by Pumpanen et al. (2001) and can be described by a budget function assumed that there were no leak flows and the sample was taken with a flow q_1 :

$$\frac{dC_i}{dt} = Q + q_1(C_0 - C_i) \quad (5)$$

where Q denoted the flow from the soil into the chamber while sampling and C_0 , the background air replacing the air drawn into the cartridge. Integration of Eq. (5) and rearranging with respect to Q leads to a timely description of the flux added while sampling:

$$Q(t) = q_1(-C_0 + C_i - e^{-q_1 t}) \quad (6)$$

With that, we could account for the portion of soil efflux that have been generated during the 20 min sampling period and have been added to chamber.

The automatic chamber system measured the fluxes directly relative to a CO_2 free air standard and resembles in its operation therefore also a closed static system (Pumpanen et al., 2004). The software of the automatic chamber system used the method as given by Pedersen et al. (2001) to calculate the chamber flows. Measurements that did not reach a stable fluxes have been discarded by the system.

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