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Atmospheric Measurement Techniques Discussions

A new disjunct eddy-covariance system for BVOC flux measurements – validation on CO_2 and H_2O fluxes

R. Baghi[1], P. Durand[1], C. Jambert[1], C. Jarnot[1], C. Delon[1], D. Serça[1], N. Striebig[2], M. Ferlicoq[3], and P. Keravec[3]

[1]Laboratoire d'Aérologie, UMR5560, CNRS – Université de Toulouse and Centre National de la Recherche Scientifique, Toulouse, France
[2]Observatoire Midi-Pyrénées, Toulouse, France
[3]Centres d'Etudes Spatiales de la BIOsphère (CESBIO), UMR5126, Toulouse, France

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Correspondence to: R. Baghi (romain.baghi@aero.obs-mip.fr)

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Abstract

The disjunct eddy covariance (DEC) method is an interesting alternative to the conventional eddy covariance (EC) method, because it allows the estimation of turbulent fluxes of species for which fast sensors are not available. A new disjunct sampling system (called MEDEE) was developed and validated. This system was built with chemically inert materials. Air samples are grabbed quickly and alternately in two cylindrical reservoirs, whose internal pressures are regulated by a moving piston. It was designed to be operated either on ground or aboard an airplane (the French ATR-42 research aircraft). It is also compatible with most analysers since it transfers the air samples at a regulated pressure. For validating the system, DEC and EC measurements of CO_2 and latent heat fluxes were performed concurrently during a field campaign. EC fluxes were first compared to simulated DEC (SDEC) fluxes and then to actual DEC fluxes. The EC fluxes were in agreement with both the simulated and actual DEC fluxes. The EC fluxes compare well to SDEC fluxes ($R^2 = 0.92$ and 0.68 for latent heat and CO_2 fluxes, respectively) and to actual DEC fluxes ($R^2 = 0.91$ and 0.67 for latent heat and CO_2 fluxes, respectively), in spite of low fluxes experienced during the campaign. This good agreement between the two techniques demonstrates that MEDEE is suitable for DEC measurements and highlights the DEC method as a reliable alternative to EC for slower sensors. A first field campaign focused on biogenic volatile organic compound (BVOC) emissions was done to measure isoprene fluxes above a downy oak (*Quercus Pubescens*) forest in the southeast of France. The measured emission rates were in good agreement with the values reported in earlier studies. Further analysis will be conducted from ground-based and airborne campaigns in the forthcoming years.

1 Introduction

Chemistry in the lower atmosphere is mostly driven by sources and sinks of trace species at the earth surface; reciprocally, the biosphere is affected by changes in atmospheric properties (Pielke et al., 1998). Flux measurements are essential for quantifying atmosphere-biosphere exchanges and understanding physical and chemical processes in the atmosphere, but they are often difficult to obtain. There are two widely used flux measurement techniques: enclosure and micrometeorological techniques.

Enclosure technique is relatively inexpensive and easy to install as it can be placed over ground, water, or vegetation. Enclosures allow monitoring of emission rates of the enclosed subject by following the evolution of concentration therein by different analysers (gas chromatography coupled to a mass spectrometer, to a flame ionization detector or to other analysers). However in most cases, enclosure measurements are representative of a very small area and cause disturbance to their sampling area (Dabbert et al., 1993).

Micrometeorological techniques consist of measuring the vertical turbulent flux near the surface. They provide integrated fluxes over, e.g., crop fields or forest canopies. Among these micrometeorological techniques, eddy covariance (EC) is the most direct method to estimate surface-atmosphere exchanges (Baldocchi et al., 1988; Aubinet et al., 1999). It relies on measurement of both vertical velocity (w) and scalar of interest concentration (c) at a high rate to characterize the mass carried by eddies of all sizes. The flux is then estimated from the covariance of w and c fluctuations averaged over a period of time. This averaging period must be long enough to be statistically representative but short enough to assume steady state conditions. Unfortunately, fast response gas analysers are available for a limited number of trace species, and therefore, restrain EC measurements to only those species, though recent mass spectrometers are on the edge of the method applicability, for gas or even aerosol flux (Müller et al., 2010; Farmer et al., 2011). Alternative approaches bypassing this limitation have been proposed to extend the use of micrometeorological flux measurements to other

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species measured with relatively slow analysers. For relatively short-lived species, turbulent flux provided by micrometeorological techniques is similar to the surface flux as long as the measurement height is only a few meters away from the surface; otherwise chemical reactions occur and affect the flux (Kristensen et al., 2009).

The gradient method is used as an alternative to EC and relies on the similarity theory of the surface layer (flux-profile relationships). It consists of measuring mean concentrations at different heights in the surface layer. The flux is then estimated from the concentration profile and the stability parameters (Obukhov length and friction velocity). This method is indirect since it requires an empirical parameterization. Furthermore, when the chemical reaction time is not much longer than the turbulent diffusion characteristic time, the impact of chemistry on profiles has to be taken into account through a coupled chemistry-dynamics model (Kristensen et al., 2009).

The eddy accumulation (EA) method initially proposed by Desjardins (1977) aimed at increasing sample analysis time but was a technical challenge. In this method, air is sampled in two separated reservoirs depending on the sign of the vertical wind velocity. The sampling rate has to be proportional to the vertical velocity, which is very difficult to set in practice. Businger and Oncley (1990) simplified the EA technique by introducing an empirical calibration and named this method relaxed eddy accumulation (REA). The REA method requires samples to be collected in separated downdraft and updraft reservoirs like in the EA method, but with a constant flow. The flux is then proportional to (1) the concentration difference between the two reservoirs over the same period as for the EC technique (e.g. 30 min), (2) the standard deviation of w, and (3) a scaling parameter β, which is dependent upon the statistical properties of the time series and the w threshold values beyond which the air is collected (Businger and Oncley, 1990; Andreas et al., 1998; Fotiadi et al., 2005). While the REA method is relatively easy to implement in the field, it cannot be used for species whose characteristic reaction time is not much longer than the accumulation period.

Finally, the disjunct eddy covariance method (DEC) is an EC method from which it only differs by the number of samples captured. This method allows a rate

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approximately down to one sample every ∼10–30 s, whereas data are acquired at high rate for EC measurements (10–20 Hz). Among alternative techniques of EC, the DEC approach does not rely on any parameterization and does not involve further assumptions. However, air samples must be acquired in a very short time (few 0.1 s at most) and precisely dated to capture the turbulent transport (Rinne et al., 2001; Turnipseed et al., 2009). When measuring fluxes of reactive compounds, characteristic chemical time scales have to be larger than (or at least comparable to) turbulent transport times (Vila-Guerau de Arellano and Duynkerke, 1992). Flux measurements of short-lived species are an issue when using EC alternative techniques as they often require intermediate storage of samples before analysis. Thus, the DEC method offers an advantage with a storage time that is two orders of magnitude lower than required for the REA technique. Advancements in PTR-MS technology, a fast response analyser capable of measuring sequentially a wide array of organic compounds present in our atmosphere, have extended the range of species whose flux can be measured with EC or DEC technique. A variant of DEC, named virtual DEC (VDEC), was specifically designed for the PTR-MS instrument (Karl et al., 2001). With this method, it is possible to achieve flux measurements of several trace gases simultaneously, with only one PTR-MS instrument without the use of intermediate reservoirs.

Several disjunct eddy samplers (DES) are described in the literature (Rinne et al., 2000, 2008; Grabmer et al., 2004; Turnipseed et al., 2009). These DESs have been used either with infra-red gas analyser (IRGA) or PTR-MS for biogenic volatile organic compounds (VOCs) measurements. Some of them present limitations: Rinne et al. (2008) demonstrated the validity of the DEC technique through direct comparison of DEC latent heat flux with EC ones; however, in their DES, the air was re-injected into the reservoirs once analysed by the IRGA in order to avoid a pressure drop during the analyses. Such a system is only suitable for certain analysers which neither destroy nor contaminate the sample during the analysis.

In this study, we present the design of an innovative disjunct eddy sampler named MEDEE for tower-based and airborne DEC measurements. MEDEE stands for “Mesure

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par Échantillonnage Disjoint des Échanges d'Espèces en trace" (translation: trace gas exchange measurements by disjunct sampling). Our primary objective in developing this instrument was to measure biogenic VOC (BVOC) fluxes but the design of MEDEE offers compatibility to a wide range of compounds and analysers. As aforementioned, the DEC method offers many advantages over other micrometeorological techniques for compounds for which fast response analysers are not available.

The paper is organized as follows: in the next section, a brief theoretical background of EC and DEC techniques will be given. Then we will describe the MEDEE instrument in detail. In the following section, we will report the results from a field campaign set up for the validation of the MEDEE system. Latent heat and CO_2 fluxes were measured concurrently with MEDEE coupled to an IRGA, and with an EC system, over a winter wheat plot during summer 2011. In the last section, the results from a field test campaign focused on BVOC emissions will be reported.

2 Theoretical background

2.1 Eddy covariance method

Trace gases and energy are transported between the Earth surface and the atmosphere via upward and downward eddies of air. In the eddy covariance technique, fast rate monitoring of these turbulent motions allow for the determination of the net exchange between the surface and the atmosphere. The complete turbulent flux of a scalar is described as the mean product of the vertical wind component w and the scalar concentration c:

$$F = \overline{wc}. \tag{1}$$

Equation (1) requires the time series of w and c to be defined at a rate and over a duration that allows sampling of all the scales which contribute to the covariance (i.e.

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the scales at which the (w, c) cospectrum is significant). In general, a 10–20 Hz sampling frequency over a 20–30 min period of time is adequate for surface flux estimates. Equation (1) is solved using Reynolds decomposition of w and c into their mean and fluctuation values. Taking into account that $\overline{w}$ is non-zero (even over flat and homogeneous terrain) when a buoyancy flux exists (Webb et al., 1980; Foken and Wichura, 1996), there are two ways to estimate F from the fluctuations of w and scalar concentration. The first way is to compute F from the covariance $\overline{w'c'}$ and to correct the value according to the buoyancy flux (the so-called "Webb correction", see e.g. Webb et al., 1980; Fuehrer and Friehe, 2002; Lee and Massman, 2011). The second way is to compute the covariance between w' and the scalar mixing ratio χ'. F is thus estimated as:

$$F = \rho_a \overline{w'\chi'}, \tag{2}$$

where ρ_a is the mean density of dry air.

2.2 Disjunct eddy covariance method

DEC was derived from EC as a mean for the use of slower analysers. Vertical wind is measured at high rate, but unlike the EC method, samples are separated by a constant time interval Δt. Samples are captured quickly (e.g. 0.2 s) and analysed until the next sample. The turbulent flux is thus determined as the average on the flux calculation period of a "discrete" number of samples n:

$$F = \rho_a \frac{1}{n} \sum_{i=1}^{n} (w'\chi')_i . \tag{3}$$

It has been shown that as long as the time interval Δt between two measurements is less than the integral time scale of the turbulence, the flux can be estimated with only a small increase in random error (Lenshow et al., 1994). The number of samples n depends on Δt, which is generally between 1 s and 30 s.

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Rinne et al. (2002, 2008) estimated the uncertainty on the DEC flux (random error) according to the number of measurements available for an averaging period of half an hour by two ways (Fig. 1). If N is the number of points of the high rate time series, and n the number of points of the sub-sample (assumed statistically independent), then a theoretical estimate of the uncertainty of the covariance computed on the n values is given by (Rinne et al., 2008):

$$\varepsilon_{w'\chi'} = \sqrt{\frac{\overline{(w'\chi')^2}}{n}\frac{N-n}{N-1}} \approx \frac{\sigma_{w'\chi'}}{\sqrt{n}}, \quad \text{if } N \gg n. \tag{4}$$

In the above equation, $\sigma_{w'\chi'}$ is the standard deviation of the instantaneous covariance $w'\chi'$. The error $\varepsilon_{w'\chi'}$, normalised by $\sigma_{w'\chi'}$, is represented by the continuous line in Fig. 1. The uncertainty can also be estimated from measured high-rate time series, by generating from a complete time series several sub-sampled time series, and computing the corresponding covariances. The standard deviation of the covariance values for a given number of disjunct samples n gives an estimate of $\varepsilon_{w'\chi'}$. Rinne et al. (2002, 2008) reported the results of such simulations on ground and airborne measurements. In Fig. 1 we reproduced their results obtained with observations above an alfalfa field (Rinne et al., 2008). The uncertainty simulated by Eq. (4) follows well the behaviour of the data points with a small overestimation. This figure illustrates that as long as n is of the order of, or larger than ∼100, the statistical uncertainty on the covariance estimate remains acceptable. The blue arrow on the figure indicates the number of samples of the MEDEE system in the two field campaigns of the present study. The expected uncertainty is thus no larger than 8 %.

Other sources of uncertainty for the DEC system are the sample carry-over and the period of time required for filling the reservoirs. Sample carry-over results from the remaining air in the reservoir because of the partial vacuum before the capture of the next sample, and from the air remaining in the "dead volume" between the reservoir and the on-line analyser. The filling time is in general longer than the vertical wind measurement rate (~0.1 s), which acts as a filter for higher frequencies. The consequences of these two error sources will be analysed in the following sections.

3 Disjunct eddy sampler

MEDEE was designed to be operated on the ground as well as aboard an airplane. It was built to fit in a 19″ airplane rack. Sampling parts are mounted on a sturdy anodized aluminium plate and electronic modules are embedded in several rack compartments. MEDEE was developed to collect a small air sample promptly at ~0.2 s, and to ensure its transfer at a constant regulated pressure to an on-line gas analyser. The latter feature is new and has not been reported in literature before. This enables the use of pressure sensitive analysers pumping in a closed reservoir. A scheme of the MEDEE system is shown in Fig. 2.

MEDEE comprises two stainless-steel cylindrical reservoirs. Each cylinder is connected to a stainless-steel piston moved by an electric actuator making volumes variable. This technology allows real-time compensation of any pressure drop in the reservoirs. Airtightness is ensured by Teflon seals and Teflon guides prevent the pistons from misalignments. Each cylinder is connected to the fluid circuit and to a pressure transducer (A-10, WIKA Instruments, Cergy Pontoise, France) allowing pressure measurement within the cylinder. These cylinders are 130 mm long with an inner diameter of 100 mm, which corresponds to a maximum volume of 1 l. High flow conductance solenoid valves (72B11DCM, Peter Paul Electronics, New Britain, CT, USA) with stainless-steel bodies serve for sample inlets. Teflon bodied solenoid valves (Type 121, Burkert, Ingelfingen, Germany) are used to direct the flow towards the analyser or towards a vacuum pump (80110131, Thomas, Wayne, PA, USA). All solenoid valves present a fast response time (<50 ms). Stainless-steel connectors and Teflon pipes (9.6 mm inner diameter) are used to connect the cylinders to sample inlets, while 4 mm inner diameter Teflon pipes are used for the analyser and vacuum circuits. To reduce the errors resulting from sensor separation (Moore, 1986), inlet solenoid valves are deported most

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closely to the wind measurement point, which increases the reservoir volume while minimizing the capture lag time with respect to the vertical wind measurement. For the validation field campaign described below, the total reservoir volume is thus extended to 1.78 l. In the same way, on the line towards the analyser, the solenoid valves could be placed in such a position that the dead volume before the analyser would be at minimum.

This sampling system can be represented as two mechanical syringes. A 3-D model of one mechanical syringe is depicted in Fig. 3. Reservoir walls were made transparent to unveil the piston. A Teflon bellows seals hermetically the connection between the electric actuator arm and the piston and prevents from grease degassing.

MEDEE is operated by a LabVIEW (National Instruments) program running on an embedded box PC, and a micro-controller chip that gives the rhythm of the operating phases and triggers the solenoid valves. A servomechanism implemented in the program drives the piston through the electric actuator according to the pressure value inside the reservoir. Pressure values and on-line gas analyser data are stored on a data-logger (CR1000, Campbell Scientific Inc., Logan, UT, USA), the former at a rate of 20 Hz and the latter at 10 Hz. Extra entries available on this data acquisition system are used for sonic anemometer data storage if the system is operated on the ground or synchronisation parameters if the system is on board the aircraft.

The two "mechanical syringes" work alternately. With respect to a single system, this allows doubling the number of samples and continuously feeding air samples to the analyser. A full cycle of operation for one system is divided into three main phases: (1) vacuuming, (2) sampling, and (3) transferring to the analyser at a constant regulated pressure (see Fig. 4). During the first step, the reservoir pressure is brought down to ~20–40 hPa with a vacuum pump and by pulling forward and backward the piston in the cylinder. The sample grab is then triggered by opening the inlet valve. Once captured, the sample is transferred to the analyser by opening the analysis valve. At the end of this sequence, the second reservoir is ready to transfer a new sample. The

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synchronisation between the valves on the two reservoirs allows the analyser to receive samples following one another without any interruption.

The pressure regulation is ensured by the electric actuator in response to the measured pressure in the reservoir. The set point pressure used for the regulation is the value measured in the reservoir when the instrument is in starting phase (all valves open), and thus corresponds to the atmospheric pressure at this time. This regulation allows the analyser to draw sample air without experiencing a pressure drop that would disturb flow rate and analyser response. Pressure cycles inside the reservoirs are illustrated in Fig. 5. The three main phases of operation are emptying, sampling, and analysing. The figure highlights the pressure regulation during the analysis periods, illustrated by the horizontal arrows. Given that the pressure signal resolution is not better than 1 hPa, we observe a quite stable value (±2 hPa) at least during the last ~70 % of the analysis period. In the switching periods and during the beginning of the analysis, the pressure fluctuations are somewhat higher but remain within ±5 hPa.

In the cycle shown on Fig. 4, 11.5 s are dedicated to the analysis of an air sample. This is also the time interval Δt between two consecutive samples. This value can be adjusted between 10 and 30 s with this instrument. For a Δt of 11.5 s, 155 samples are analysed during half an hour, which would correspond to a low uncertainty (~8 %) on the covariance estimate (see Fig. 1).

When switching on MEDEE, a pre-loading phase starts. Inlet solenoid valves are open to equilibrate reservoirs to ambient pressure, followed by the opening of analysis valves allowing air to freely flow from the inlet to the analyser. In this standby setup, the analyser can be turned on for pre-heating. Next, a manual switch-on will trigger the disjunct sampling sequence. A first reservoir will be emptied before sampling while air is flowing to the analyser through the second reservoir, and the operational cycle starts with the first capture. When stopped, MEDEE returns to a standby mode similar to the one in the beginning. The current analysis is achieved and both inlet valves are open to ambient air. The second analysis valve is then open and the air can flow freely to the analyser. The system can be run for several days without interruption.

4 Validation campaign

4.1 Site description

A field campaign was carried out from 9 June to 17 June 2011 on a field site located at Lamasquère, a country side area 12 km away from Toulouse (southwest France). The Lamasquère site is characterized by a cultivated plot of 0.32 km^2 in a flat terrain. It is part of the Carbo-Europe network (Dolman et al., 2006) and has been instrumented for meteorological and micrometeorological measurements since March 2005. CO_2 and water vapour fluxes are monitored continuously at the site. The altitude is 180 m, and the mean annual wind speed is 1.79 $m\,s^{-1}$. Main wind directions are from west and east-southeast with a fetch of 200 and 140 m, respectively. Crop management consists of a rotation of winter wheat, and maize. An exhaustive description of the field site is given in Béziat et al. (2009). During the validation campaign, CO_2 and water vapour turbulent flux measurements were performed concurrently by the conventional EC method and the DEC method over a senescent winter wheat (*Triticum aestivum*) crop. The instruments were installed in the middle of the plot for an optimal fetch in the main wind directions. The average air temperature during the measurement period was 18.5 °C. The hottest day was 15 June with a maximum of 29 °C reached in the beginning of the afternoon. Weather conditions were mostly sunny, but 10 and 16 June were cloudy with some short rain events. The experiment occurred during the ripening stage of winter wheat when plants became photosynthetically less active and were drying. CO_2 uptake as well as plant transpiration were thus expected to be low. This crop has been harvested on 2 July, two weeks after the end of this experiment.

4.2 Eddy covariance measurements

The eddy covariance instrumentation setup is composed of a three-dimensional sonic anemometer (CSAT 3, Campbell Scientific Inc, Logan, UT, USA) for wind components and speed of sound (from which is deduced the "sonic" temperature), and an open-path

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infrared gas analyser (LI-7500, LI-COR, Lincoln, NE, USA) for CO_2 and water vapour concentrations. Both sensors were installed on a mast at 3.65 m above the ground. Data were recorded with MEDEE's datalogger at 10 Hz. EC fluxes of CO_2 and H_2O were calculated using MATLAB routines with a linear detrending. No density correction (Webb et al., 1980) was needed as concentration measurements were converted to mixing ratios and fluxes computed according to Eq. (2). H_2O fluxes were multiplied by the latent heat of vaporisation of water L_v and thus converted into latent heat flux expressed in W m^{-2}.

4.3 Disjunct eddy covariance measurements

During this experiment, the same sonic anemometer was used for the EC and DEC measurements. MEDEE was coupled to a closed-path infrared gas analyser (LI-6262, LI-COR, Lincoln, NE, USA) for disjunct eddy covariance measurement of CO_2 and water vapour fluxes. Both instruments were installed next to the EC mast. MEDEE's inlet solenoid valves were deported from the sampling system and brought closer to the wind measurement point (0.2 m). Two-metre long 9.6 mm inner diameter Teflon tubing was used between the inlet valves and the sampling system. Same type of Teflon tube of 60 cm in length was used as inlet from the solenoid valves toward the sonic anemometer. In this configuration, the dead volume contained in the tubing upstream of the inlet solenoid valves represents 2.4 % of the reservoir volume (43 ml). Longer tubing slightly impaired the sampling duration that increased to ∼0.3 s during which the reservoir filling was observed to be almost linear. All sensors signals were stored on MEDEE's data-logger to avoid synchronization issues.

DEC flux calculations are done by a MATLAB routine where samples are dated precisely using reservoir pressure time series. The vertical wind values falling during the reservoir linear filling period were averaged and used with the concentration values for covariance calculation. Concentration measurements were averaged over 6 s during the analysis period with the rejection of the 6 first seconds, taking into account the stabilisation of the pressure in the reservoir, and the sample transfer time from the

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reservoir to the analyser. EC and DEC fluxes were computed according to Eqs. (2) and (3), respectively.

This study does not intend to provide fluxes for annual budget or other use but to proof the agreement of the two measurement techniques. Since both techniques used the same wind data, further processing steps (e.g. sonic coordinate rotation) were not needed.

4.4 Data quality and processing

The time series of the main parameters measured during half an hour (05:30 to 06:00 UTC on 15 June) are presented in Fig. 6. During this half-hour period, the *w* fluctuations do not exceed 0.5 $m\,s^{-1}$ because of the moderate (yet increasing) turbulence in the growing boundary layer. CO_2 concentration starts at 600 ppm, resulting from night time accumulation, then it decreases with time. The decrease in CO_2 concentration is explained by the vertical mixing in the growing boundary layer. The humidity mixing ratio is increasing in the same time, because energy (from the net radiation) is available for evaporation. The LI-7500 humidity time series clearly exhibits the increase with time of both the amplitude of fluctuations and the size of eddies. The time series of the CO_2 and H_2O mixing ratios measured by the LI-6262 closed-path analyser connected to MEDEE reproduce well the slow variations of the 0.1 s open-path time series. On the contrary, the high frequency variations are smoothed since the same air sample is analysed during 11.5 s. The vertical winds corresponding to the times at which the samples were captured are indicated by the red dots on the figure: the value of *w* for each dot has been computed as the average of the 0.1 s values during the period of the capture.

The lower part of Fig. 6 displays a zoom on a 2-min period of the 30-min time series. Each of the two circuits of MEDEE is discriminated with the red and blue colours, on the *w* time series (capture dots) and on the closed-path analyser signal (only the CO_2 values are represented here). The central times of the captures are indicated by the vertical dotted lines. The coloured CO_2 values are those retained for DEC flux

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computation. Precautions were made to avoid contamination by previous sample. As a result, concentrations are constant for a given sample. The amplitudes of the variations in concentration are well reproduced by the open-path and closed-path analysers; for example, the abrupt ~20 ppm decrease observed at 27 min 26 s by the Li 7500 is seen by the Li 6262 but only once a capture (red dot) has been done after the decrease.

In the following sections, the EC fluxes of CO_2 and H_2O will be used as the reference for the evaluation of the DEC fluxes. The EC fluxes have been calculated from the 0.1 s time series as described in Sect. 1. In a first step, EC fluxes will be compared to simulated DEC (SDEC) fluxes. SDEC time series are defined as a sub-sample of the 0.1 s time series, at a time interval equal to that of the MEDEE system (11.5 s), and with a "capture" averaging corresponding to the effective response of the solenoid valves, i.e. the vertical wind as well as the H_2O and CO_2 time series are averaged on 2–3 consecutive values. The covariances are then computed identically on the complete (10 Hz) and on the sub-sampled time series. This step must be regarded as a theoretical evaluation of the DEC system, aiming at the validation of critical parameters like the time interval between the captures and the duration of the capture. In a second step, EC fluxes will be compared to DEC fluxes computed from the concentrations measured through MEDEE. In this case, the vertical wind is the only common element between the two systems, and the performance of the DEC prototype can thus be evaluated objectively.

4.5 Simulated disjunct eddy covariance – eddy covariance inter-comparison

The diurnal course of EC and SDEC fluxes are presented on Fig. 7 for the 15 June case. The weak amplitude of the fluxes is noteworthy. Latent heat flux does not exceed 110 W m^{-2} in the middle of the day. Such values are 3–4 times lower than what could be observed above this kind of vegetation but in an active phase (spring). Similarly, CO_2 fluxes does not reach considerable values nor exhibits a clear diurnal cycle, translating as the absence of photosynthetic activity of the wheat in its ripening phase. On the same site with similar vegetation type, Béziat et al. (2007) reported large changes in net

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CO_2 assimilation, respiration and daily gross ecosystem production (GEP) throughout the season (see their Fig. 4). In April and May, for example, GEP reached daily values of -10 to $-15\,gC\,m^{-2}\,day^{-1}$ ($1\,\mu mol\,m^{-2}\,s^{-1} \approx 1.04\,gC\,m^{-2}\,day^{-1}$). From these daily means we could expect higher values (i.e. -30 to $-45\,gC\,m^{-2}\,day^{-1}$) in the middle of the day. Despite the low values of the fluxes, the two methods agree to reproduce their diurnal course in general, though some discrepancies are sometimes present, larger (in relative value) on the CO_2 flux than on the latent heat flux.

The comparison for the whole campaign is presented in Fig. 8 through scatter diagrams. The results are very good for the latent heat flux, with a determination coefficient R^2 of 0.93. There is no bias between the two techniques, which demonstrates that the capture time is short enough and turbulent fluctuations are not significantly damped. The scatter is small; the difference between the two methods is not larger than the random error on EC estimates (see e.g. Lambert and Durand, 1998). The CO_2 flux have larger scattering ($R^2 = 0.69$), which is possibly due to the weakness of the flux. In such conditions, the turbulent part of the concentration signal is reduced with respect to larger scale variations, and the integral scale of the instantaneous covariance $w'c'$ is increased accordingly, which degrades the performance of both the DEC and EC methods.

4.6 Disjunct eddy covariance – eddy covariance inter-comparison

The EC and DEC fluxes are compared in the same way as done for the EC and SDEC fluxes. Figure 9 presents the diurnal course on 15 June. The evolution of the latent heat flux is well reproduced by the two methods, whereas the weakness and small evolution of the CO_2 flux give rise to some discrepancies on few estimates. It is interesting to note that the observed significant differences on the CO_2 flux occur at the same time and are of the same order as for the EC-SDEC comparison (see Fig. 7). That means that the CO_2 variations observed by the open path and closed path analysers are quite identical, and that the discrepancies between the fluxes cannot be attributed to the

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MEDEE system, but are related to the behaviour of the DEC method itself in such conditions.

The comparison for the whole campaign is given in Fig. 10. We note that the determination coefficients are quite identical to those obtained in the EC-SDEC comparison (see Fig. 8), though the two methods were based on different analysers (open-path for SDEC and closed-path for DEC). That means that the MEDEE system behaves as expected theoretically. However, we observe a slight underestimation of the DEC latent heat fluxes, which can be attributed to a different response of the two analysers to H_2O fluctuations. Instruments were calibrated separately before the field measurements and in the absence of an external reference during the field campaign, we chose not to calibrate one analyser against the other, because we were not able to define which one should have been adjusted. Anyway, we should keep in mind that the H_2O and CO_2 fluxes were weak during the campaign, and the DEC results must be considered as satisfactory. Higher correlations between EC and DEC fluxes have thus to be expected for larger flux conditions.

5 BVOC measurements

5.1 Site description

The first field campaign for measuring BVOC emissions with MEDEE was carried out at the Oak Observatory at "Observatoire de Haute-Provence" (O3HP) site from 24 July to 6 August 2010. The site is located south east of France, 70 km north of Marseilles, and its vegetation is a forest of downy oak (*Quercus pubescens*) and Montpellier maple (*Acer monspessulanum*) for 90 and 10 %, respectively, with large patches of smoke tree (*Cotinus coggygria*) as undergrowth vegetation. Downy oak is a major tree species in the Mediterranean region and is known as a strong isoprene emitter (Keenan et al., 2009). In this forest, the trees are about 70 yr aged, with a small younger parcel located west of the measurement site. The main wind direction is from north-west (Mistral wind)

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and the terrain presents a 2 % slope in this direction. The climate is Mediterranean with a dry season somewhat shortened due to the altitude (680 m a.s.l.). The temperature during the measurement period varied between 15 °C at night and 25 °C in daytime. An 8 m scaffolding tower was set up in the forest with the fast sensors installed above the upper platform at 10 m above the ground (4.5 m above the top of the canopy). Among the sensors, a 3-D sonic anemometer (CSAT 3, Campbell scientific) and a LICOR 7500 (LICOR) measured the three wind components, the sonic temperature and the CO_2 and H_2O concentrations. The MEDEE system was installed on the highest platform of the tower with sample inlets 20 cm away from the sonic transducers. Isoprene concentration was measured with a Fast Isoprene Sensor (Hills scientific, CO) (Guenther and Hills, 1998), with 4 l min^{-1} flow rate coupled to MEDEE. The scaffolding tower was also equipped with radiometers (incoming photosynthetically active radiation (PAR) measured by a Licor LI-190SA, incoming and outgoing short- and long-wave radiation measured by a Kipp and Zonen CNR1). Ozone and NO_x analysers were installed on a small cabin 30 m away from the tower with inlets at 7 m above ground. Leaf area index (LAI) for the area surrounding the site was measured during the month of August 2010 thanks to the O3HP monitoring activity, and was found to be 2.5.

5.2 Results and discussion

DEC isoprene fluxes measurements were performed as well as EC CO_2 and H_2O fluxes during the measurement period. EC fluxes calculations were similar as described in Sect. 4.1 and DEC data were processed as described in Sects. 4.2 and 4.3. The "sonic" temperature is converted into air temperature T after correction of moisture contamination, and the sensible heat flux (in W m^{-2}) is computed as the kinematic heat flux ($\overline{w'T'}$) multiplied by $\rho_a C_p$, where C_p is the heat capacity of air at constant pressure. Figure 11 presents the 30-min time series of EC sensible heat flux, CO_2 flux and latent heat flux, and DEC isoprene flux, for 5 and 6 August. 5 August was characterized by a moderate mistral wind with a diurnal average wind speed of 5.4 m s^{-1}. Because of the wind, temperature did not rise above 22 °C for that day. The mistral wind was

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still present on 6 August but with a lower average wind speed of 3.3 m s^{-1}. Maximum temperature for 6 August was 25 °C. The conditions are characteristic of a sunny dry period, with the sensible heat flux reaching during the day 3–4 times the values of the latent heat flux. As expected, the CO_2 uptake occurred during the day due to vegetation photo-synthesis activity, whereas it reversed during the night due to the respiration.

A clear diurnal isoprene emission cycle was observed with values in the range 1.5–2.8 µg m^{-2} s^{-1} around midday. In order to compare these results with previous observations done on the same kind of vegetation, the isoprene fluxes were then reduced at the standard conditions (temperature of 30 °C and PAR of 1000 µmol m^{-2} s^{-1}), using the G93 algorithm for isoprene (Guenther et al., 1993; Guenther, 1997). The air temperature measured on the scaffolding tower was used for the conversion instead of leaf temperature because this latter was not available. However, using the surface layer flux-profile relationships to estimate the temperature profile revealed that the day time mean temperature at the top of the canopy did not differ from the air temperature on the scaffolding tower by more than 1 °C. The wind conditions probably explained this small difference, and allowed us to use this temperature as an acceptable approximation of the leaf temperature. The reduced fluxes were then converted into emission rates, using the measured LAI on the site and a leaf mass area value of 134.3 g m^{-2} given by Simon et al. (2005) for a downy oak forest in the same area and presenting very similar morphology. Normalized isoprene emission rates were averaged from 07:00 to 14:00 UTC for the two days. The resulting emission rate was of 39.7 µg g^{-1} h^{-1}. Simon et al. (2005), Owen et al. (1998), Steinbrecher et al. (1997) and Kesselmeier et al. (1998) report for *Quercus Pubescens* isoprene normalized emission rates of 134.7, 92, 90.7 and 42 µg g^{-1} h^{-1}, respectively. In these studies measurements were performed using branch enclosures. The value found in the present study is in the lower range compared to the literature but represents an integrated flux at the canopy scale. The lower emission rates observed might be due to the windy conditions (Loreto and Sharkey, 1993), and/or to chemical transformation of isoprene between the leaf and the above canopy areas (Fuentes et al., 2000). We are willing to improve the

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parameterization of BVOC emission rates, including key parameters other than temperature and PAR, but this improvement is beyond the scope of this paper.

6 Conclusions

The goal of this study was to develop, validate, and deploy a new DEC flux measurement system known as MEDEE. This system was made of chemically inert materials to avoid sample contamination and built to meet airplane (the French ATR-42 in a first step) requirements. The MEDEE system is able to quickly grab 1 l air samples and ensure their continuous transfer at constant pressure to an online analyser. The system is built as twin mechanical syringes, whose alternate functioning ensures a continuous alimentation of the on-line analyser. MEDEE was operated with a switching period from a sample to the other of 11.5 s, which fulfils the requirements for keeping the covariance estimate in a good accuracy in spite of the reduced number of data. With such intervals and the reservoir volume, the system can be connected to on-line analysers with flow rates up to 4 l min^{-1}. The stability of the pressure (of the order of 1–2 hPa) is in favour of a better functioning of most analysers. These features make MEDEE suitable for measurements of different trace gases and compatible with a wide array of analysers. For example, quantum cascade laser absorption spectrometer (QCLAS) (Joly et al., 2011) or cavity ringdown spectrometer (CDRS) (Crosson, 2008; Langridge et al., 2008; Fiddler et al., 2009) that monitor trace gas such as CO, CH_4, N_2O or NO_x.

The validation of the system was done through a field campaign during which H_2O and CO_2 fluxes were simultaneously measured with EC and DEC techniques with different analysers. The EC fluxes were used as a reference. In a first step, which can be considered as a theoretical validation, the EC fluxes were compared to the simulated DEC fluxes calculated from the sub-sampled high-rate time series, but taking into account the averaging resulting from non-instantaneous filling time. Despite the weak CO_2 and latent heat fluxes observed during the campaign, a good agreement was observed between the two methods with determination coefficients R^2 of 0.93 and 0.69

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for H_2O and CO_2 fluxes, respectively. The comparison between EC and "real" DEC fluxes was quite similar, with R^2 coefficients quite identical, but a slight underestimation of DEC H_2O fluxes was observed, attributed to a difference in the calibration of the two analysers. The differences of flux estimates with the two techniques were within a satisfactory range of uncertainty for micrometeorological methods.

MEDEE was also tested for BVOC flux measurements over a downy oak forest. The results showed a reduced (30 °C, 1000 µmol m^{-2} s^{-1}) isoprene emission rate of 39.6 µg g^{-1} h^{-1} which was found to be of the same order but rather lower than the values reported in the literature for comparable vegetation. Such measurements will be realized in the coming years in the frame of the Chemistry-Aerosol Mediterranean Experiment (ChArMEX) project, in order to improve the parameterization of the BVOC emission rates. It is planned to improve the parameterization of the emission by taking into account other parameters than the PAR and temperature.

MEDEE is also scheduled for airborne measurement on the French ATR-42 aircraft. This aircraft is equipped for turbulence measurements (Saïd et al., 2010), and the use of MEDEE with a PTR-MS will allow the estimation of BVOC fluxes at the landscape scale and throughout the whole atmospheric boundary layer.

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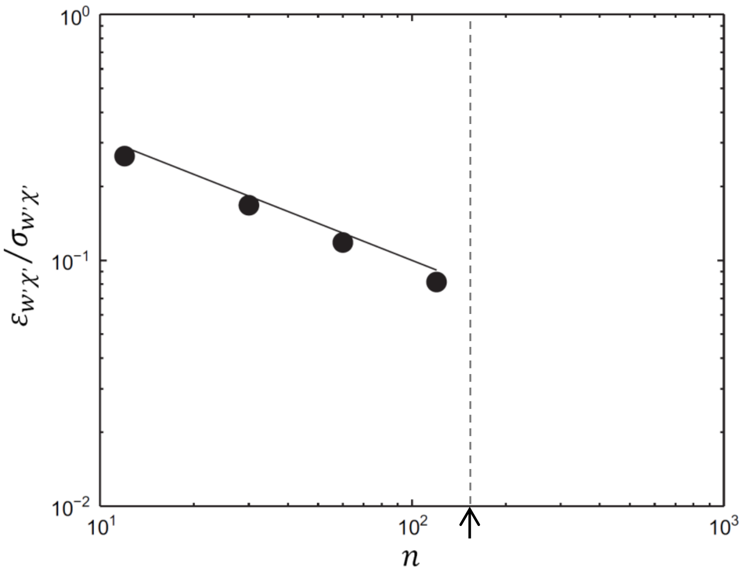

Fig. 1. Evolution of the accuracy of the DEC flux estimate according to the number of values over the averaging period. The solid line is the theoretical flux uncertainty according to Eq. (4). The filled circles represent the uncertainties on DEC fluxes simulated from sub-sets of 12, 30, 60 and 120 samples. The dotted line indicates the number of sample produced by MEDEE with a sample capture every 11.5 s. Reproduced and adapted from Rinne et al. (2008).

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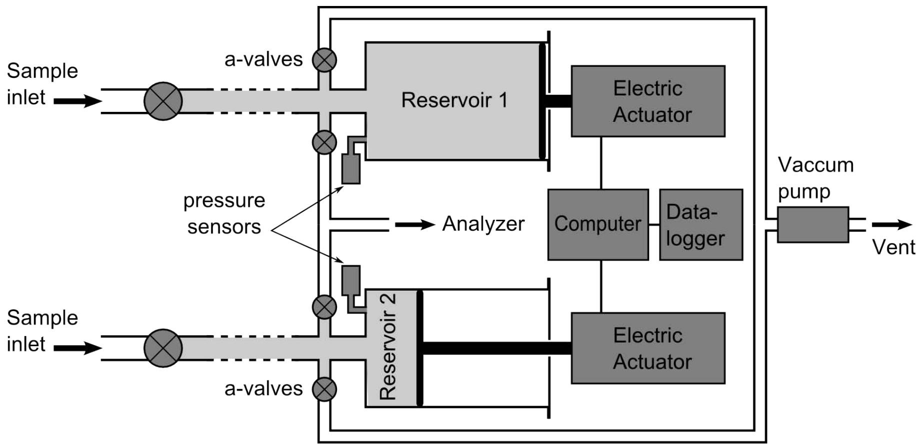

Fig. 2. Scheme of the MEDEE system.

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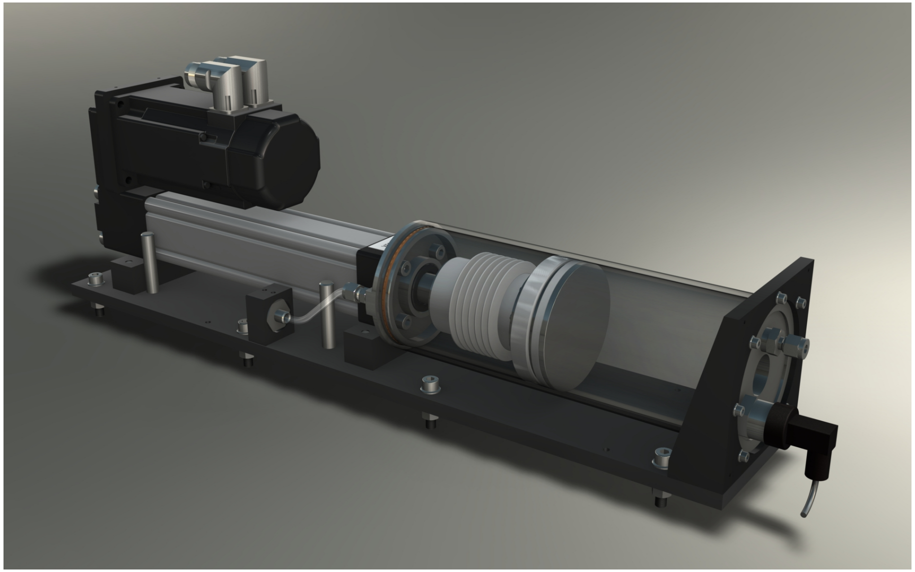

Fig. 3. 3-D drawing of one of the two "mechanical syringes" of the MEDEE system. The cylindrical reservoir is made transparent on the illustration in order to exhibit the moving piston. The grey rectangular box on the left houses the jack screw which is moved by the engine inside the black compartment above it.

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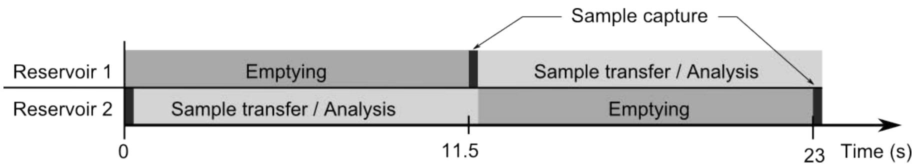

Fig. 4. The three steps of the MEDEE operating cycle (emptying, capture and analysis), for each of the two reservoirs.

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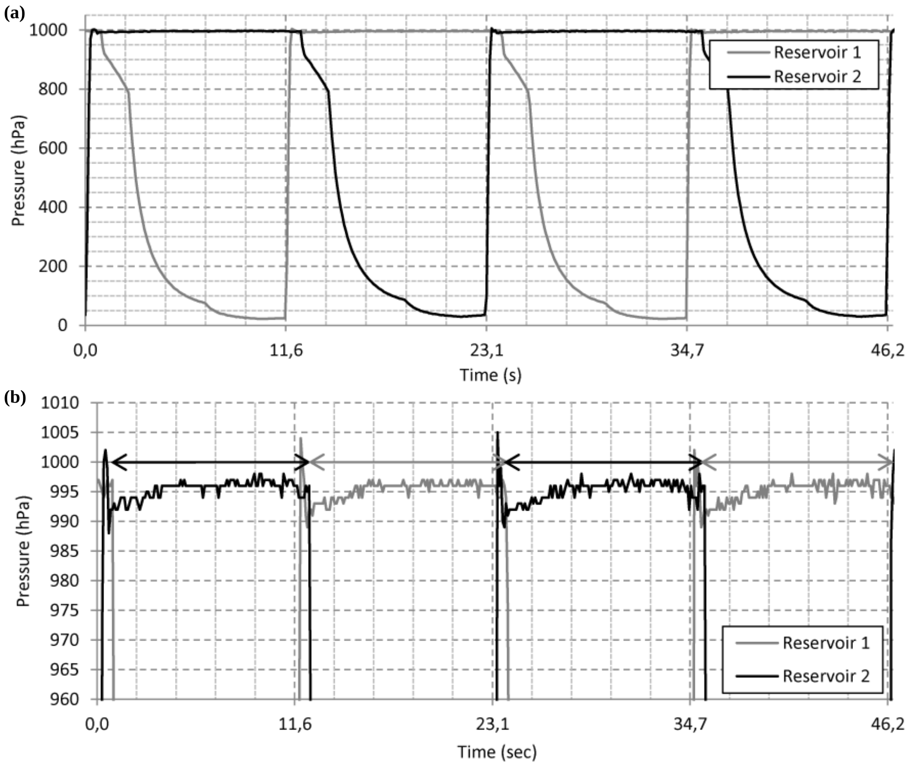

Fig. 5. Pressure (hPa) cycles inside MEDEE's reservoirs displayed full scale **(a)**. The bottom diagram **(b)** is identical but the pressure scale is zoomed around atmospheric value (here 996 hPa), the grey and black signals correspond to the two reservoirs. The horizontal arrows indicate the analysis periods of each sample. Pressure peaks of some hPa are observed at the end of the captures but the analyses starts later on.

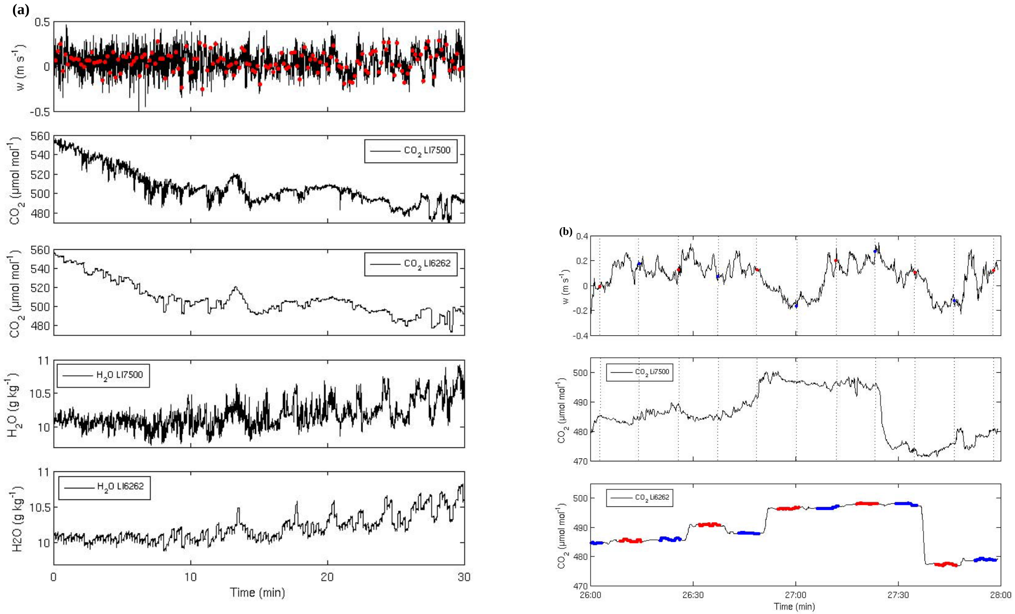

Fig. 6. (a) From top to bottom: 30-min time series of w ($m\,s^{-1}$), open-path analyser CO_2 mixing ratio ($\mu mol\,mol^{-1}$), closed-path analyser CO_2 mixing ratio ($\mu mol\,mol^{-1}$), open-path analyser H_2O mixing ratio ($g\,kg^{-1}$) and closed-path analyser H_2O mixing ratio ($g\,kg^{-1}$). Red dots are averaged w values during the captures. **(b)** Same as above, but restricted to a 2-min period, and without the H_2O signals. The coloured parts on the CO_2 closed path measurements represent the data points used for mixing ratio estimates. The vertical dotted lines represent the sampling times, and the blue and red colours refer to each of the two reservoirs.

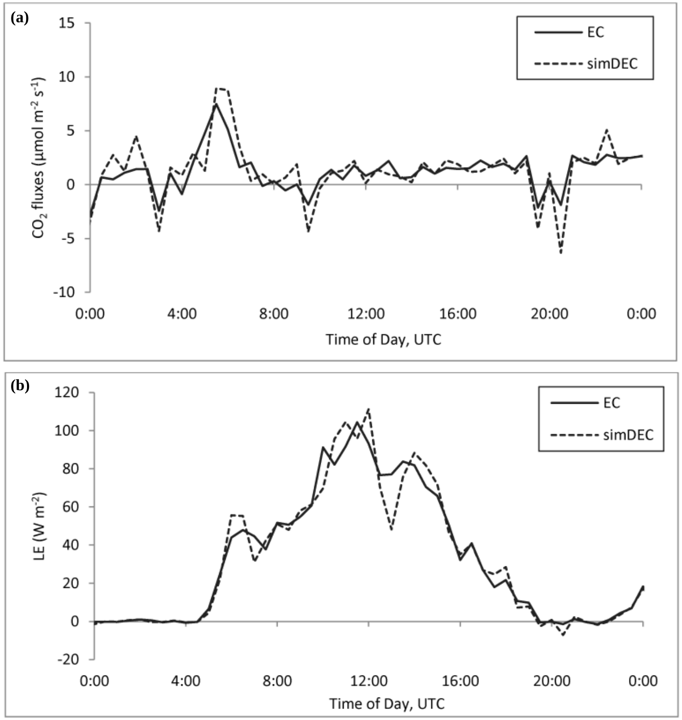

Fig. 7. Diurnal course of CO_2 flux (µmol m^{-2} s^{-1}) **(a)** and latent heat flux (W m^{-2}) **(b)** computed on 15 June by eddy covariance (solid line) and simulated disjunct eddy covariance (dashed line) methods.

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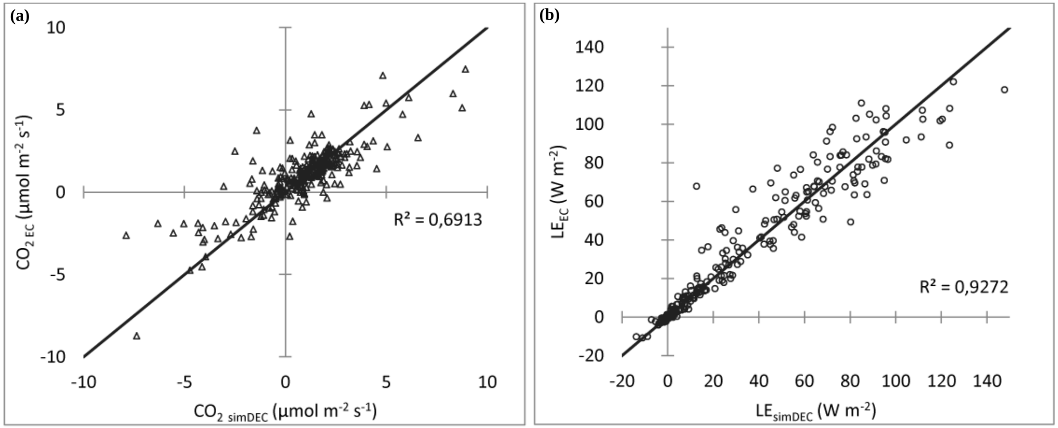

Fig. 8. Simulated disjunct eddy covariance vs. eddy covariance CO_2 fluxes (µmol m^{-2} s^{-1}) **(a)** and latent heat (W m^{-2}) **(b)**. Both graphs report data from the entire field campaign (i.e. from 9 June to 17 June 2011). Solid line is the 1:1 line.

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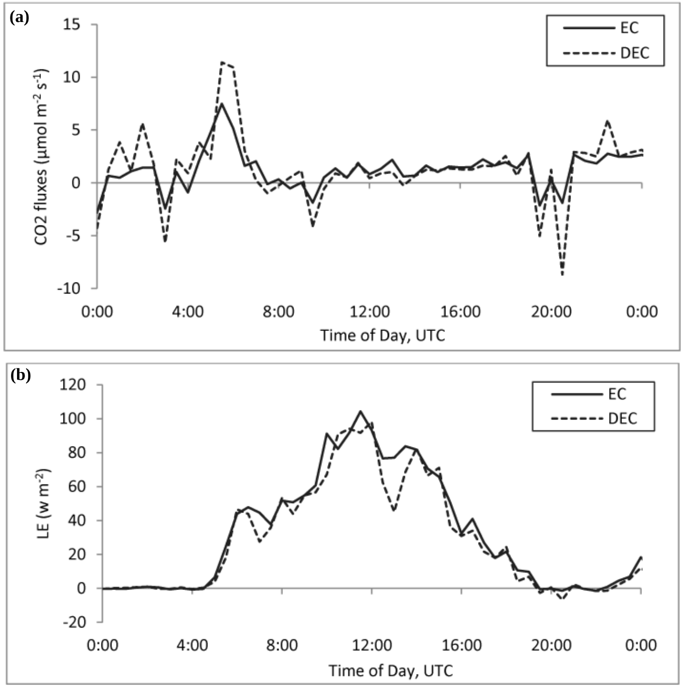

Fig. 9. Same as Fig. 7, but for "real" DEC instead of simulated DEC fluxes.

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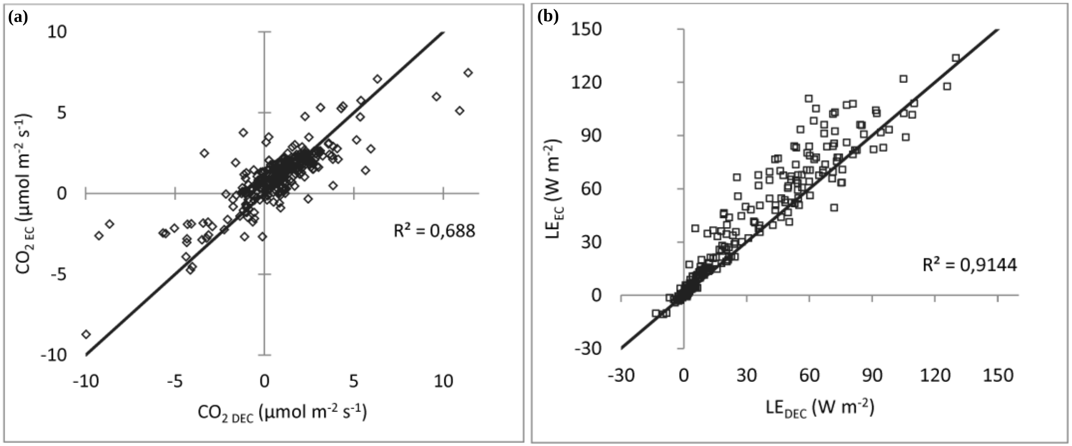

Fig. 10. Same as Fig. 8, but for "real" DEC instead of simulated DEC fluxes.

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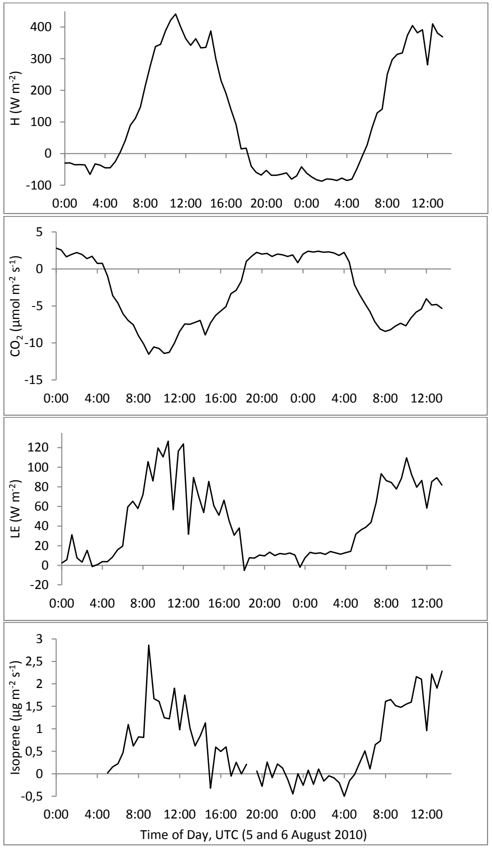

Fig. 11. From top to bottom: sensible heat flux ($W m^{-2}$), CO_2 flux ($\mu mol\, m^{-2} s^{-1}$) and latent heat flux ($W m^{-2}$) computed with the eddy covariance method, and Isoprene flux ($\mu g\, m^{-2} s^{-1}$) computed with the disjunct eddy covariance method. The measurement period is on 5 and 6 August 2010.

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