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A true eddy accumulation system for trace gas fluxes using disjunct eddy sampling method

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Abstract. A new true eddy accumulation flux measurement system, based on the disjunct sampling approach, has been developed and tested. In disjunct sampling, short, separate samples are taken instead of continuously sampling the air as in traditional relaxed eddy accumulation and eddy covariance systems. This method reduces the number of samples but allows more time to process them. Simulation shows that the fluxes, calculated using disjunct data, are close to those calculated using continuous data. The disjunct true eddy accumulation instrument was successfully deployed to measure monoterpene fluxes at Niwot Ridge, Colorado. The ability of the system to measure the sample flow accurately, critical for any eddy accumulation system, was tested using atmospheric CFC-113 as a tracer, with good results. The system was capable of measuring relatively low α-pinene fluxes, below 10 $ng\ m^{-2}\ s^{-1}$ at $T = 8° - 18°C$, over a subalpine forest.

1. Introduction

The most direct method for measurement of vertical fluxes of atmospheric constituents in the surface layer is the eddy covariance (EC) method. This method, however, requires fast sensors as the timescales of the smallest flux-carrying eddies are below 1 s. Most commercial acoustic anemometer-thermometers are capable of measuring three-dimensional wind and temperature at 10-20 Hz frequency. Fast commercial sensors also exist for water vapor, carbon dioxide and ozone. Some instruments capable of measuring the concentrations of certain other trace gases fast enough for EC applications have also been developed [e.g., *Guenther and Hills*, 1998].

For measuring the vertical fluxes of those trace gases for which no fast sensor exists, two approaches are used. The first and traditional approach is the gradient method, in which the flux is inferred from the mean concentration difference between two or more measurement heights. Variants of this method have been widely used for measurements of biogenic hydrocarbon fluxes [e.g., *Fuentes et al.*, 1996; *Guenther et al.*, 1996; *Schween et al.*, 1997; *Rinne et al.*, 2000]. More direct flux measurement techniques are variants of the eddy accumulation (EA) method. These techniques sample air into two reservoirs depending on the sign of the vertical wind velocity. In the original, or true eddy accumulation method, introduced by *Desjardins* [1977], the air is sampled proportionally to the vertical wind velocity. It is, however, difficult to control the sample flow accurately and fast enough. The relaxed eddy accumulation method (REA), proposed by *Businger and Oncley* [1990], bypasses this difficulty by using constant sample flow rate. The lost information is substituted by similarities of atmospheric behavior of the trace gas and other scalars, for example, temperature. The REA method was tested by *Oncley et al.* [1993] using carbon dioxide and has been successfully used for measurement of biogenic hydrocarbon fluxes by for example, *Guenther et al.* [1996], *Bowling et al.* [1998] and *Ciccioli et al.* [1999].

Traditional flux measurement methods sample air in a continuous manner. It is, however, also possible to sample noncontinuously and still have enough samples to calculate the fluxes [*Haugen*, 1978; *Kaimal and Gaynor*, 1983; *Lenschow et al.*, 1994]. In this approach, called disjunct sampling by *Lenschow et al.* [1994], quick samples are taken with a relatively long time-interval between them (Figure 1). This gives more time to process the samples, but increases the statistical uncertainty of the fluxes calculated using such disjunct data. As a large part of the fluxes in the atmospheric surface layer is carried by relatively large eddies, continuously sampled samples are not totally independent. Thus the statistical uncertainty of the fluxes does not increase as much as they would in the case of independent samples. According to *Lenschow et al.* [1994], the disjunct sampling does not increase the statistical uncertainty of the flux values more than 8% if the time interval between samples is shorter than the appropriate integral timescale.

In this paper we present a system for disjunct true eddy accumulation (DEA) flux measurements. We first

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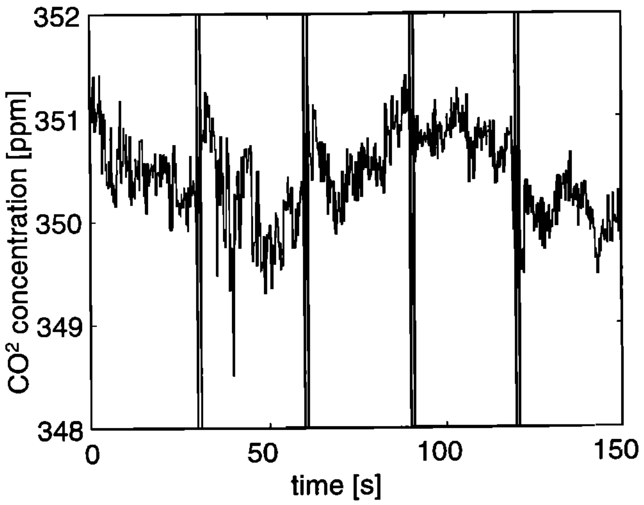

Figure 1. Concept of disjunct sampling. Short samples are taken from continuous data.

describe the method and the instrument and then present results of simulations and field test. Finally, we discuss further possibilities of the disjunct sampling strategy.

2. Discourse on Method

2.1. Concept of Disjunct Eddy Sampling

Conventional eddy flux measurement methods sample air nearly continuously. The flux is calculated as a time average. In the case of eddy covariance measurement

$$F_c = \overline{w'c'} = \frac{1}{t_2 - t_1} \int_{t_1}^{t_2} w'(t)c'(t)dt, \qquad (1)$$

where w' and c' are the eddy components of the vertical wind velocity and the concentration of a scalar, respectively. In the disjunct eddy sampling the flux is calculated as an average of a smaller subset of samples:

$$F_c = \langle w'c' \rangle = \frac{1}{N} \sum_{i=1}^{N} w'(t_i)c'(t_i). \qquad (2)$$

In (2) the time interval between samples is longer than in (1), and thus the sum in (2) is an ensemble average rather than a time average.

The concept of disjunct eddy sampling was tested empirically by sampling 10 Hz data of wind and temperature noncontinuously with a 30 s interval between samples, calculating $\langle w'T' \rangle$, and comparing this with the fluxes calculated with the continuous data set $(\overline{w'T'})$. The values of $\langle w'T' \rangle$ and $\overline{w'T'}$ were in good agreement (Figure 2). A few larger deviations occurred during nonstationary conditions. If these nonstationary data are removed, the correlation between disjunct and continuous sampling is very good with $r^2 = 0.94$ and slope of 0.99. Thus it seems that the disjunct sampling gives flux values similar to continuous sampling. The correlation can be further increased, either by making the time step between samples shorter, or using a longer averaging time.

2.2. True Eddy Accumulation by Disjunct Sampling

In the true eddy accumulation method [*Desjardins*, 1977], air is sampled into updraft and downdraft reservoirs proportionally to the vertical wind velocity. The total mass of a compound in the reservoirs is

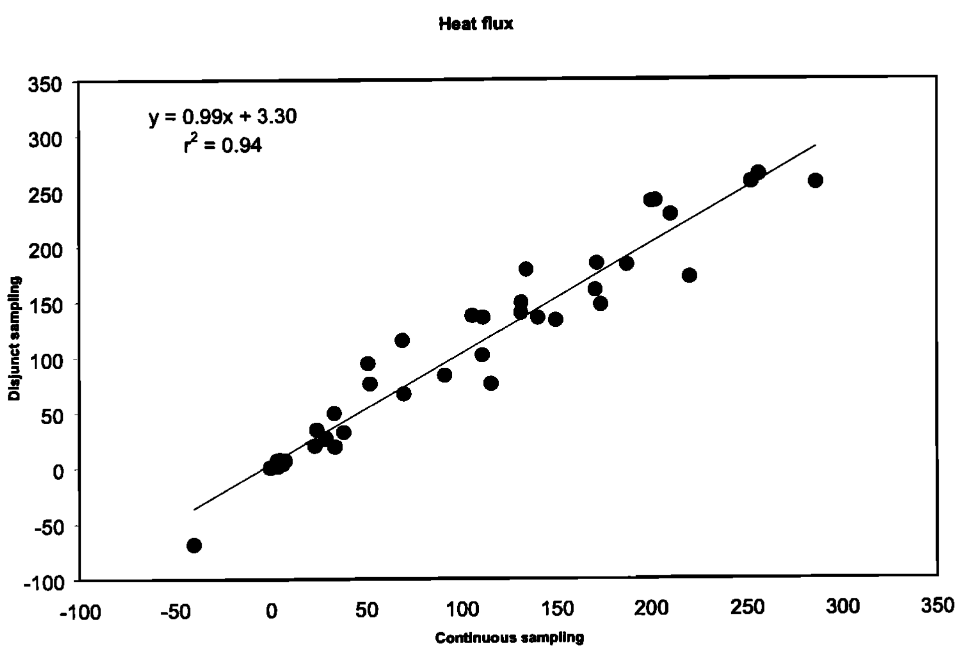

Figure 2. Comparison between heat fluxes ($W\ m^{-2}$) calculated using continuous sampling and disjunct sampling with 30 s sampling interval. The data are collected during the same days as the α-pinene flux data.

$$m^{\uparrow} = \gamma \int_{updrafts} w'c\,dt \quad (3a)$$

$$m^{\downarrow} = \gamma \int_{downdrafts} w'c\,dt, \quad (3b)$$

where γ is the pumping coefficient, defining the relation between sample flow and w', and arrows indicate updraft and downdraft reservoirs. The integration is conducted separately over updrafts and downdrafts. It can be shown that

$$\overline{w^+}c^{\uparrow} + \overline{w^-}c^{\downarrow} = \overline{w'c'} = F_c, \quad (4)$$

where $c^{\uparrow}$ and $c^{\downarrow}$ are concentrations of c in updraft and downdraft samples, respectively, and

$$\overline{w^+} = \frac{1}{t_2 - t_1}\int_{t_1}^{t_2} w'\delta_+ dt \quad (5a)$$

$$\overline{w^-} = \frac{1}{t_2 - t_1}\int_{t_1}^{t_2} w'\delta_- dt, \quad (5b)$$

where δ_+ and δ_- are delta functions ($\delta_+ = 1$ when $w' > 0$ and $\delta_+ = 0$ when $w' < 0$; $\delta_- = 1$ when $w' < 0$ and $\delta_- = 0$ when $w' > 0$). As the true eddy accumulation method does not apply any similarity theory or empirical coefficient, it can be regarded as a direct flux measurement method.

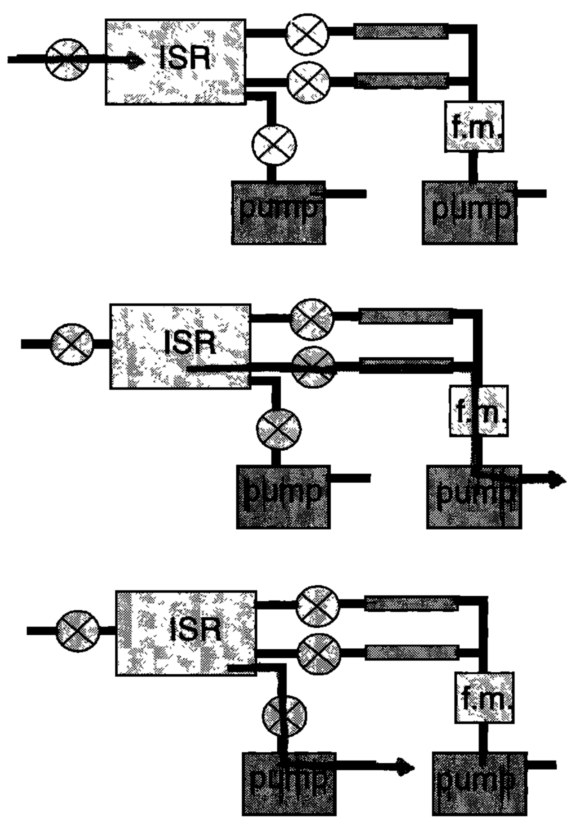

Figure 3. The working sequence of a simple disjunct eddy accumulator. Circles with diagonal crosses indicate valves; ISR is intermediate storage reservoir; long medium gray rectangles indicate updraft and downdraft sample reservoirs, and f.m. is a mass flowmeter.

A disjunct eddy accumulation system quickly grabs samples (sampling time, $t_s < 0.5s$) every 10-60 s into an intermediate storage reservoir (ISR, Figure 3). When the sample is in the ISR, a small part of it is collected into the updraft or downdraft sample container, according to the sign of the vertical wind velocity at the moment the grab sample was taken, and proportionally to the vertical wind velocity. After the sample has been collected, the ISR is evacuated for the next round. The sampling sequence is repeated typically for 30-60 min, long enough to sample eddies of all scales. The equation for true eddy accumulation with disjunct sampling can be written

$$F_c = \langle w^+ \rangle c^{\uparrow} + \langle w^- \rangle c^{\downarrow}. \quad (6)$$

The notation of averages in (4) and (6) is the same as in (1) and (2).

2.3. Description of the Instrument

The instrument described in this paper consists of two ISRs (Figure 4), which are operated by turns. When one is being evacuated, the other one is sampling. This way it is possible to get twice as many samples as with one ISR. The operation sequence of the system is described in Table 1. The time needed to fill the ISR to 80% of the ambient pressure is 0.2 s, which is the opening time of the α-valves. Samples are taken every 30 s giving 60 samples for half an hour measurement period. The ISRs were evacuated to 10% of full pressure in preparation for the aspiration of the next sample. The effect of this 10% carryover was tested by simulation with 10 Hz data of wind, temperature, and carbon dioxide concentration measured above a boreal forest. According to the simulation the underestimation caused by the carryover was around 4% and the correlation coefficient r^2 between fluxes with and without carryover was 0.94-0.99.

The ISRs with 1.5 L volume were made of electropolished stainless steel. The diameter of the reservoirs was 11.3 cm, and the length was 15 cm. This aspect ratio gives relatively low surface to volume ratio. The α-valves (Skinner 7121KBN44V00) are brass-bodied direct-acting solenoid valves since Teflon or stainless steel bodied fast-acting valves with large enough conductance were not available. Since the flow through the α-valves is fast and the residence time in the ISR is short, there is little time for hydrocarbons to react with the surface of the valves. The response time of these valves is less than 20 ms.

All valves downstream from IRS are Teflon-bodied direct-acting valves (Fluoroware 203-1414-415). The sample flow was directed from an ISR into the cartridges by four valves via Teflon and stainless steel tubing. The stainless steel tubing is heated to 50°C to prevent compounds from sticking onto the surfaces. The sample flow is adjusted manually by a needle valve to the desired

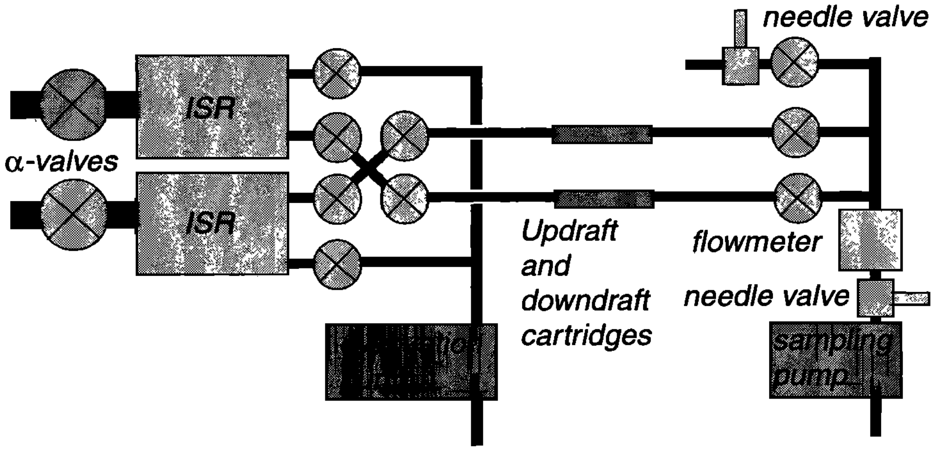

Figure 4. Schematic of the disjunct eddy accumulator with two intermediate storage reservoirs (ISR). Circles with diagonal crosses indicate valves; ISR is intermediate storage reservoir.

range. The flow through the updraft and downdraft reservoirs is measured by the same mass flowmeter (Micro Switch AWM3300V), preventing any biases due to different flow sensors. The volume sampled through cartridges is controlled by integrating the measured sample flow and terminating the sampling when the desired volume has been sampled. This prevents any undesired feedback effects between flow controller and the sample flow rate.

The ISRs, α-valves, and an array of other valves were situated on the end of a 2 m long boom, 1 m below an acoustic anemometer (ATI, SATI 3K), following the suggestions of *Kristensen et al.* [1997]. The valves are operated by a laptop computer through a set of solid state relays. The computer also reads and stores the data from the acoustic anemometer. The computer, most of the electronics, and a few valves were situated in a box within the walk-up tower, and the pump used to evacuate the ISRs was also on the tower. The system is controlled by a LabVIEW-based program, that consists of two modules: one used to acquire the sonic wind data developed in the Finnish Meteorological Institute [*Hatakka et al.,* 1999], and a second used to control the DEA system, developed for this project.

3. Experiment

The field test of the DEA system was conducted at the Ameriflux site near Niwot Ridge, Colorado (40°02'N, 105°33'W, 3050 m above sea level) in August-October 1999. The forest surrounding the measurement tower consists mainly of Engelmann spruce (*Picea engelmannii* Parry ex Engelm.), lodgepole pine (*Pinus contorta* Dougl. ex Loud.), and subalpine fir (*Abies lasiocarpa* (Hook.). Nutt.). The zero plane displacement height at the site is 12 m. The site has 5° slope eastward which was taken into account by rotating the wind vector. The rotation was done using predetermined rotation angles in such a way that the w axis was perpendicular to the mean streamlines. The rotation angles were determined from previous measurements made at the site. The vertical wind data were not high-pass-filtered. High-pass filtering could remove the effect of tilted mean streamlines on the mean vertical wind, and longer-term horizontal gusts on the vertical wind variation, but the effect of short-term variation would remain. The measurements were conducted on a 30 m high walk-up tower. The height of the DEA was 19.3 m, and that of the acoustic anemometer was 20.3 m.

Table 1. A Schematic Working Sequence of a Disjunct Eddy Accumulator

	ISR	
	1	2
Time ⇓	Grab a sample into ISR 1, $t = 0.2$ s	Evacuate ISR 2, $t = 30$ s
	Sample through cartridge, $t \propto w$	
	Wait until 30 s is complete	
	Evacuate ISR 1, $t = 30$ s	Grab a sample into ISR 2, $t = 0.2$ s
		Sample through cartridge, $t \propto w$
		Wait until 30 s is complete

According to *Kristensen et al.* [1997] the underestimation caused by this sensor separation is less than 4%. The booms pointed toward south.

The sample cartridges used were loaded with 350 mg of Carbosieve and 180 mg of Carbotrap (Carbotrap 200, Supelco, Bellefonte, Pennsylvania). The cartridges were analyzed using a gas chromatograph with mass spectrometer. A more detailed discussion on the chemical analysis is given by *Greenberg et al.* [1999]. The cartridges were stored at -30°C until analyzed.

The temperatures at the site during measurements were relatively low, ranging from 2°C to 18°C. The typical winds were westerly, but in the afternoons some easterly upslope winds occurred.

4. Results and Discussion

A critical aspect for a cartridge eddy accumulation system is the ability to measure the sample volume accurately. To test how well the sample volumes were measured, the ratio of CFC-113 concentrations of updraft and downdraft cartridges were compared to the ratios of corresponding sample volumes. The atmospheric CFC-113 concentrations are nearly constant in the remote areas away from anthropogenic sources. As the area in Niwot Ridge is a Climate Monitoring and Diagnostic Laboratory (CMDL) of the National Oceanic and Atmospheric Administration (NOAA, Boulder, Colorado) monitoring station, there are no sources of CFC-113 nearby, since this is one of their target compounds. The ratios of CFC-113 concentrations and ratios of volumes follow very closely the 1:1 line (Figure 5) and the correlation coefficient $r^2 = 0.98$. This gives confidence on the systems ability to integrate the sample flow. The typical sample volumes were in the range of 1.0-2.5 L.

All the components of energy balance were not measured as a part of this experiment. University of Colorado (CU), however, measured the energy balance components at the same tower. These measurements show a good closure (A. Turnipseed, Department of Environmental, Population, and Organism Biology, University of Colorado, Boulder, personal communication, 2000), and thus validity of the micrometeorological assumptions at the site. The heat fluxes as well as the friction velocities measured by the acoustic anemometer used for this study during the hydrocarbon flux measurements agree with those measured by CU, with $r^2 = 0.96$ and slope of 1.06 for friction velocity and $r^2 = 0.80$ and slope of 0.96 for heat flux. Thus we can conclude that the turbulence measurements used by the disjunct eddy accumulator worked well.

As the terrain at Niwot Ridge site is not flat, the tilting of mean wind can influence the flux measurements. The α-pinene fluxes plotted against the absolute value of mean vertical wind velocity after the rotation, normalized by dividing by the standard deviation of vertical wind velocity,

$$w_n = \left|\frac{\overline{w}}{\sigma_w}\right|, \tag{7}$$

shows that large negative flux values occur with high values of w_n (Figure 6). This is due to the tilted mean wind vector, causing w' to be contaminated with horizontal wind. The flux measurements with $w_n > 0.1$ were subsequently excluded from further analysis. Also

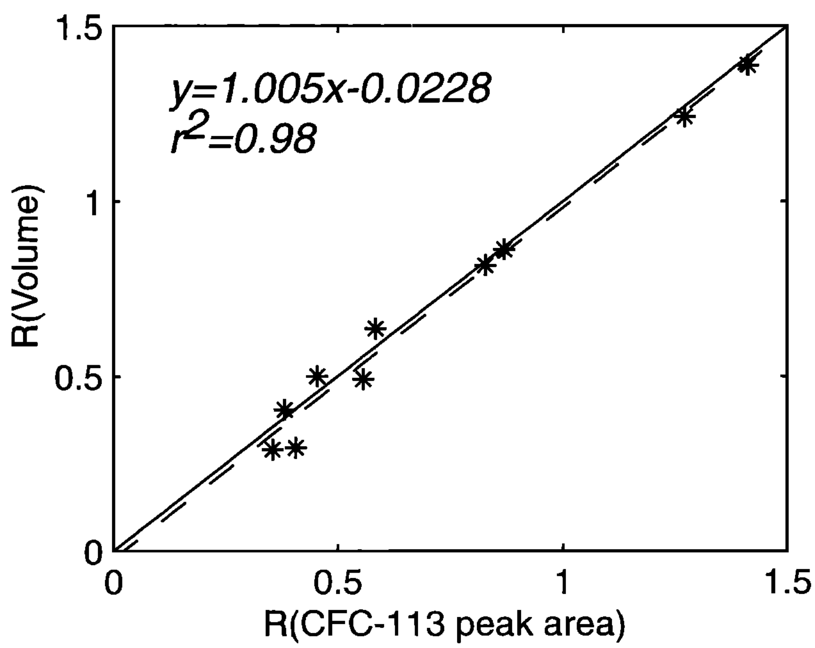

Figure 5. Comparison of updraft to downdraft ratios of measured sample volumes and CFC-113 peak areas ($R_{volume} = V_{up}/V_{down}, R_{peak\ area} = A_{up}/A_{down}, R_{volume} = 1.005R_{peak\ area} - 0.0228, r^2 = 0.98$).

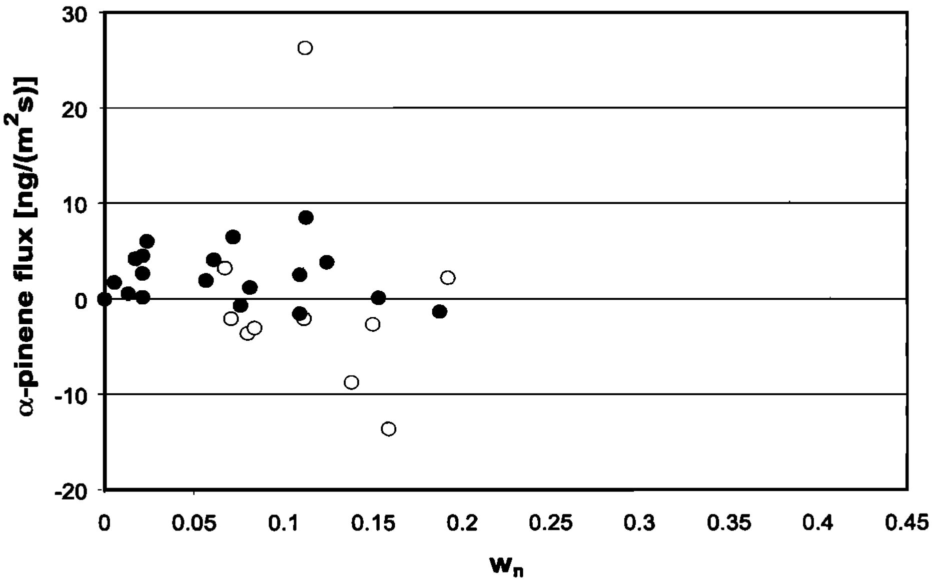

Figure 6. Observed α-pinene fluxes versus normalized vertical wind velocity (w_n, equation (7)). Solid circles are data with normalized sample volume difference (ΔV_n, equation (8)) less than 0.5; open circles are those with more than 0.5. The data with $w_n > 0.1$ and $\Delta V_n > 0.5$ were excluded from further analysis.

the measurements where the normalized difference between updraft and downdraft sample volume,

$$\Delta V_n = \frac{|V_{up} - V_{down}|}{\frac{1}{2}(V_{up} + V_{down})}, \tag{8}$$

was more than 0.5 were excluded as the ensemble average of these measurements may be biased. These limits together excluded more than 50% of the measurements from further analysis. The high fraction of data points rejected is due to the nonideal site for flux measurements. In the eddy covariance technique the wind data could be rotated during postprocessing. This is not possible for the eddy accumulation measurements, leading to higher rejection rate.

The measured α-pinene fluxes during three measurement days are shown in Figure 7. The highest fluxes were measured on August 27, which was the warmest day. On the other days there were some negative values which may be due to the random uncertainty associated with low fluxes. The fluxes were classified into four equal sized temperature classes (Figure 8) to study the temperature dependence of the fluxes using [*Guenther et al.*, 1991]

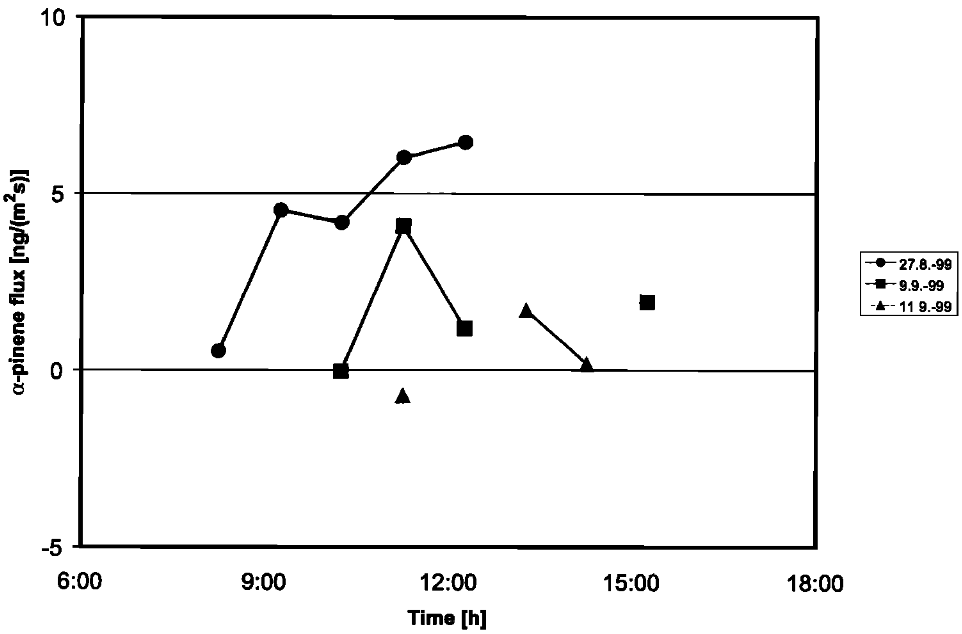

Figure 7. Observed α-pinene fluxes during three measurement days.

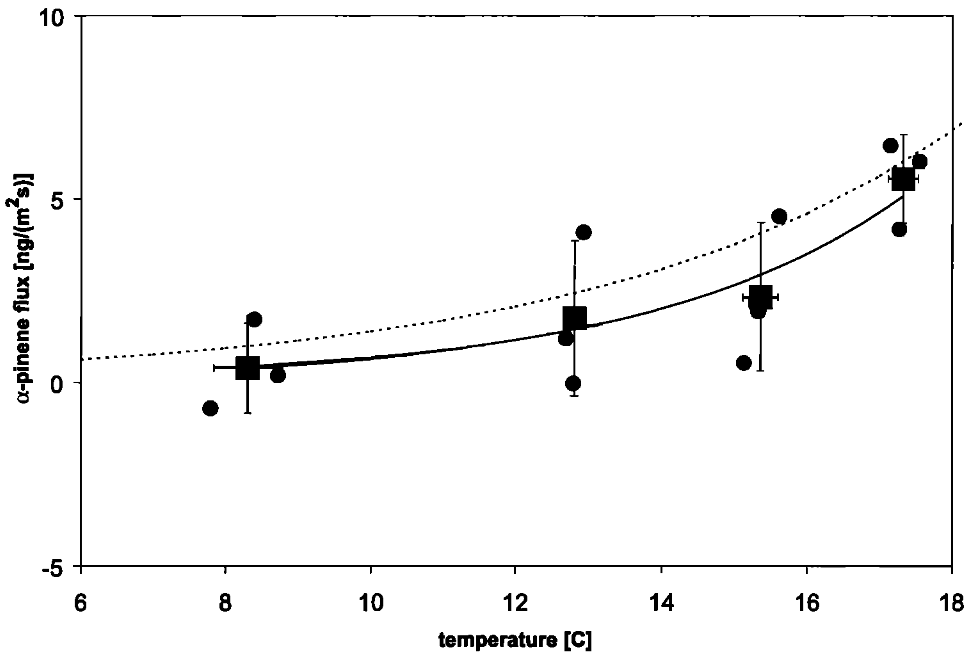

Figure 8. The temperature dependence of α-pinene fluxes. Squares are mean values of each temperature class, and error bars indicate the standard deviation of temperature and flux within the class. Circles are measured half-hourly fluxes. The solid line is the best fit into equation (9), and dashed lines indicate the lower limits of calculated emission.

$$E = E_{30} exp[\beta(T - 30°C)]. \quad (9)$$

The temperature dependence of these classes, β=0.28°C^{-1}, was higher than the leaf level value of β=0.09°C^{-1} [*Guenther et al.*, 1991] as is often the case for canopy scale emissions measured by micrometeorological methods [*Schween et al.*, 1997; *Rinne et al.*, 2000]. The observed fluxes were in the lower range of the emissions predicted by an emission model. This may be associated with model uncertainties in emission factors and biomass density. The emission factors of monoterpenes used in emission models are likely to be biased upward due to the disturbances in the cuvette measurements.

The data provided by DEA method, as well as any micrometeorological methods, need to be filtered to remove the data for which the micrometeorological and other assumptions do not hold. This filtering should be done with objective criteria by setting limiting values for suitable parameters. Generally, friction velocity has been used to exclude the data with too weak turbulence by setting a lower limit of u_*=0.1 m s^{-1}. The use of normalized mean vertical wind w_n to exclude the data with too tilted mean wind is discussed above. The importance of integral timescale is presented by *Lenschow et al.* [1994]. Also, stationarity seems to be an important factor, but an objective filtering parameter needs to be developed.

A major advantage of disjunct true eddy accumulation lies in the fact that the true eddy accumulation is a direct flux measurement method and does not depend on similarity assumptions or empirical relationships. This makes the approach less vulnerable to systematic errors, which can affect gradient and REA measurements.

Future applications of the disjunct eddy sampling approach can include several different flux measurement techniques. The disjunct sampling strategy can enable eddy covariance measurements of trace gas fluxes using semifast sensors with response times between 1-60 s. Also, different variants of eddy accumulation method, such as hyperbolic relaxed eddy accumulation [*Bowling et al.*, 1999], can be incorporated with the disjunct sampling approach. These applications could be used for flux measurements of a wide range of chemical compounds. The disjunct eddy sampling method also has potential for aircraft applications since the wind data, which can be delayed for a few seconds, are easily accommodated by the technique.

5. Concluding Remarks

The disjunct sampling approach can be incorporated into several eddy flux measurement techniques, true eddy accumulation described in this paper being only one of them. The instrument described in this paper was able to measure low fluxes of α-pinene at Niwot Ridge, Colorado. Data filtering using objective exclusion limits was used to ensure data quality. Fluxes measured by the disjunct true eddy accumulation instrument were in the lower range of expected emissions, below 10 $ng\ m^{-2}\ s^{-1}$ at $T = 8° - 18°C$.

The simulations of disjunct sampling by continuous w and T time series confirmed conclusions of *Lenschow et al.* [1994], that disjunct sampling does not greatly increase the statistical uncertainty of flux values. The critical sample volume measurement was tested using atmospheric CFC-113 as a tracer. Results showed very close agreement between updraft to downdraft ratios of CFC-113 peak area and sample volume. The major advantage of the DEA lies in the fact that it is a direct flux measurement method. Further efforts are needed to characterize the accuracy and reliability of this method relative to other flux measurement techniques.

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