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TELLUS

On the use of the Webb–Pearman–Leuning theory for closed-path eddy correlation measurements

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ABSTRACT

We consider an imperfection of real closed-path eddy correlation systems—the decoupling of the water vapour and CO_2 concentrations—with respect to the application of the Webb–Pearman–Leuning (WPL) theory. It is described why and how the current application of the WPL theory needs to be adapted to the processes in closed-path sensors. We show the quantitative effects of applying the WPL theory in different ways using CO_2 flux measurements taken above the Danish Beech forest CarboEurope site near Sorø, Zealand.

Using the WPL theory in closed-path sensors without taking amplitude damping and decoupling into account, over-corrected the annual flux by 21%, or 31 g m^{-2} yr^{-1}, to which the decoupling effect contributed with 7%. We suggest either converting the raw data point-by-point to mixing ratios or using the uncorrected covariances of water vapour mole fractions with the vertical wind velocity that were calculated with the same time lag as for the scalar concentration when correcting the dilution effect. We showed that the two approaches yielded equivalent flux results. Correct ways of applying spectral corrections to CO_2 fluxes calculated in either way are also shown. The findings reported here do not apply to open-path sensors.

1. Introduction

Webb et al. (1980, henceforth WPL) published a theory that explained the effects of density fluctuations on flux measurements with devices that measure scalar densities as opposed to scalar mixing ratios. Scalar concentrations in the atmosphere can be expressed in various ways. Among those are densities, ρ, (mass per unit volume), the mole fraction, χ, (moles scalar per mole humid air) and mixing ratio, r, (moles scalar per mole dry air). However, practically all available fast response sensors for CO_2 and water vapour sense absolute numbers of molecules in a given volume and not mixing ratios.

The way WPL had derived a vertical mean flow has been critically examined and controversially discussed (see review by Fuehrer and Friehe, 2002). But no finding or consideration disproved their principal concept of what happens in the atmospheric boundary layer, when heat and water vapour fluxes occur together with a scalar flux. Thus consideration of so-called density effects has become a standard procedure of turbulent flux data post-processing (Baldocchi, 2003) not only for open-path eddy correlation (EC) systems but also for closed-path EC systems, on which we focus here.

Currently there are two methods considering density fluctuations in closed-path EC measurements. Aubinet et al. (2000), who were the first to summarise recommendations for a common flux calculation procedure within the European flux network, EUROFLUX, noted that their infra red gas analyser (IRGA, LI-6262, LICOR; Lincoln, NE, USA), corrected the calculated CO_2 mole fractions for water vapour fluctuations automatically, and consequently, no further WPL consideration had to be taken into account. Their approach will be further discussed and a small inaccuracy will be detailed in the Appendix. This instrument dependent feature is equivalent to converting the raw data or mole fractions to mixing ratios—point-by-point—at the raw data level, provided that fast temperature, pressure and water vapour concentration readings are available. The alternative method uses covariances and averages together with an extended flux equation to consider the WPL theory.

WPL described basically the processes in the atmosphere, which are in most cases accurately monitored by open-path EC sensors. However, closed-path sensors measure the scalar concentration in a cuvette after an air sample has been transported there from the atmosphere through filters and tubes. Here it is generally accepted that temperature fluctuations are effectively

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attenuated through rapid heat exchange between the EC system walls and the air sample and can thus be neglected (Leuning and Moncrieff, 1990; Massman, 1991; Leuning and Judd, 1996; Rannik et al., 1997). Hence, the only remaining effects of the WPL theory that are relevant for the calculation on scalar fluxes with closed-path sensors are associated to the water vapour flux through the atmospheric boundary-layer that is causing water vapour fluctuations in the sample. Although closed-path sensors were not even mentioned in the original WPL publication, their equation is still used for closed-path sensors (Leuning and Moncrieff, 1990). However, applying the original WPL equations to closed-path sensors in such manner, clearly considers the instruments as *perfect* sensors, that is, assuming that the measured concentrations represent the scalar concentrations in the atmosphere. This has been proven incorrect in practice.

A little closer to the problem we address here, was the work of Massman (2004) who investigated the effects of signal attenuations due to low-pass filtering, that is, the amplitude damping. He concluded that attenuation of water vapour fluctuations due to active physical low-pass filtering by the closed-path EC system, that is, interaction with the tubing system, must not be corrected for when considering water vapour fluctuations with the WPL theory, because this would overcorrect the effects. In order to complete this analysis, we add to Massman's analysis of *amplitude* effects the analysis of *phase* effects of the low-pass filtering on scalar concentrations that are measured in closed-path EC systems.

Phase effects can lead to an additional delay in travelling time, τ, of the scalar and water vapour compared to the dry air component. This means that gases might be decoupled and thus the WPL effects for the scalar in the sample cell of the IRGA are changed compared to the atmosphere. Here we discuss the consequences for the estimated CO_2 fluxes that arise from different τ for CO_2 and water vapour in closed-path EC systems.

2. Materials and methods

2.1. Theory

Using Reynolds decomposition, it follows for the vertical turbulent flux, F_m, in ideal situations with homogeneous and stationary conditions and no chemical reactions of a scalar in the atmosphere that

$$F_m = \overline{\rho_C' w'} + \overline{w}\ \overline{\rho_C}, \tag{1}$$

here expressed as a mass flux per unit area and time. Index C stands for the scalar and w is the vertical wind velocity. Overbars denote temporal averages, primes fluctuations. Note that $\overline{w}$ can differ from 0 m s^{-1}; thus the second term does not necessarily vanish. This effect is considered by applying the WPL theory adapted to the closed-path EC system. $\overline{w}$ can be calculated with the WPL theory, but other formulations exist (Fuehrer and Friehe, 2002). WPL quantified a turbulent density flux that is caused by density fluctuations and which is balanced by a mean flow, that is, the second right-hand side (rhs) term of eq. (1). Their original formulation for the scalar flux is:

$$F_m = \overline{w'\rho_C'} + \mu\left(\frac{\overline{\rho_C}}{\overline{\rho_d}}\right)\overline{w'\rho_v'} + (1+\mu\sigma)\frac{\overline{\rho_C}}{\overline{T}}\overline{w'T'} \quad \text{(WPL24)}$$

(Webb et al., 1980), with indices 'v' and 'd' for water vapour and the dry air component, respectively. Symbols T, σ and μ represent air temperature, the ratio of molecular masses of dry air and vapour, $\frac{m_d}{m_v}$, and the water vapour mixing ratio on a mass basis, $\frac{\overline{\rho_v}}{\overline{\rho_d}}$.

WPL used the term *expansion* to express the cause for scalar density fluctuations in the atmosphere that cannot be attributed to a scalar flux through the surface. These density fluctuations have however two reasons: (i) temperature fluctuations in the atmospheric boundary-layer cause expansion/contraction and (ii) water vapour fluctuations cause dilution/concentration of the scalar. Consequently, the two additional rhs terms in WPLs' eq. (24) (WPL 24) can be interpreted as a dilution/concentration term that is related to the water vapour flux and an expansion/contraction term that is related to the sensible heat flux.

As opposed to the atmosphere, it is not obvious how to deal with density effects in a closed-path sensor, because ρ is usually highly distorted by the processes in the tubing system compared to the atmosphere. In this case the term *dilution* explains the effects better. Other than density effects, the dilution effect is still conserved in closed-path sensors, at least to a certain extent. Beyond its effect on scalar density in the atmosphere, dilution decreases the mole fraction, χ_C, of the scalar. No effect of dilution can be seen if the reference is dry air (Webb et al., 1980), that is, when using mixing ratios, r_C.

Now we examine how these atmospheric processes are related to the processes in a closed-path EC sensor. As mentioned by Aubinet et al. (2000), the raw signals of the IRGA are transformed from voltage signals into mole fractions on the basis of calibration with reference gases. Transforming eq. (1) from using densities and mass flux densities to using mole fractions and molar flux densities, F_C (mol m^{-2} s^{-1}), can be calculated as

$$F_C^{\mathrm{WPL}} = \frac{\overline{\chi_C' w'} + \overline{\chi_C}\ \overline{w}}{\overline{v_{\mathrm{mol}}}}, \tag{2}$$

where v_{mol} is the volume of one mole in the sampled air in the sensor, $v_{\mathrm{mol}} = \frac{\Re T}{p}$, with the universal gas constant $\Re$, that is, 8.3144 J K^{-1} mol^{-1}.

We used (WPL 24) and set the covariance term between $\overline{T'w'}$ equal to 0 K ms^{-1}, assuming quantitative damping of atmospheric temperature fluctuations ($T' = 0$ K) in the closed-path EC sensor (Leuning and Moncrieff, 1990). Thus the expansion term of the original WPL equation becomes zero mol m^{-2} s^{-1} and the molar flux per unit time and area is

$$F_C^{\mathrm{WPL}} = \frac{\overline{\chi_C' w'} + \frac{M_d}{M_C}\frac{\overline{\rho_C}}{\overline{\rho_d}}\overline{\chi_v' w'}}{\overline{v_{\mathrm{mol}}}} = \frac{\overline{\chi_C' w'} + \overline{r_C}\,\overline{\chi_v' w'}}{\overline{v_{\mathrm{mol}}}}, \tag{3a}$$

where the scalar densities in the covariance terms of the original equation have again been converted to mole fractions. M_d and M_C are the molecular masses and $\overline{\rho_d}$ and $\overline{\rho_C}$ the averaged densities of dry air and the scalar, respectively. The WPL dilution term is reduced to the simple product of the scalar mole mixing ratio, $r_C = \frac{\chi_C}{\chi_d}$, and the covariance of the water vapour mole fraction and w. Equation (3a) is the classical approach to apply the WPL theory to closed-path sensors. From (3a) it becomes clear that a comparably small scalar flux at comparably high scalar background concentration will be most sensitive to the WPL theory. Because the water vapour flux is in most cases either zero or positive, upward scalar fluxes will increase, whereas the absolute magnitude of downward fluxes will decrease when applying the WPL theory.

Our criticism of the above approach is that the explicit use of the original WPL theory closed-path sensors treats them as perfect sensors, that is, the dilution processes in the atmosphere are precisely represented in the measurement. Phase effects cause decoupling of scalars in an air stream, like it is, for example, used in gas chromatography to separate gases in a mixture from each other. Many investigations have shown that CO_2 and water vapour behave differently in different designs of closed-path EC systems (Laubach and Teichmann, 1996; Aubinet et al., 2000; Ibrom et al., 2007). Lag times for water vapour, τ_v, were always larger than that for CO_2, τ_c, which can be determined empirically as the lag times with which the w time series had to be shifted against the scalar to yield maximum absolute covariances (Pilegaard et al., 2001; Clement, 2004).

If $\overline{\chi_v' w'}$ and $\overline{\chi_C' w'}$ are maximal in the atmosphere, the water vapour dilution effect on $\overline{\chi_C' w'}$ is also maximal. Decoupling and different amplitude attenuation of CO_2 and water vapour in the EC system however desynchronise the dilution of χ_C by water vapour in the sensor and thus reduces its effect on $\overline{\chi_C' w'}$, whereas the second rhs term in eq. (3a) still represents the full effect in the atmosphere, if frequency corrections were applied to $\overline{\chi_v' w'}$. Thus using eq. (3a) for an imperfect sensor would overcorrect the flux. In order to quantify these effects we need, however, to know the correct flux. For this we suggest

$$F_C^{\mathrm{WPLcp}} = \frac{\overline{\chi_C' w'} + \overline{r_C}\left(\overline{\chi_v' w'}\right)_{\tau_C}}{\overline{v_{\mathrm{mol}}}}. \tag{3b}$$

This new version differs only from (3a) in the fact that is uses measured $\overline{\chi_v' w'}$, which means with this respect that it has *not* been frequency corrected, following the suggestion of Massman (2004) but, as we note here, additionally also calculated at the same lag time as for maximum $\overline{\chi_C' w'}$, τ_c.

As WPL note, the scalar turbulent surface flux can also be expressed sufficiently accurately in terms of covariances of mixing ratios with w. Transforming their original eq. (20) ($F_C = \overline{\rho_d} \overline{s_C' w'}$ in kg m^{-2} s^{-1}) from using mass mixing ratios, s_C, to using molar mixing ratios, it follows that

$$F_C = \frac{\overline{p} - \overline{e}}{\overline{p}\ \overline{v_{\mathrm{mol}}}} \overline{r_C' w'} \tag{4}$$

expressed in mol m^{-2} s^{-1}(Ibrom et al., 2007). p is the atmospheric pressure, e the partial pressure of water vapour. Comparison of (4) and (3b) allows us to check the consistency of the specified WPL theory for closed-path EC systems, that is, we test the hypothesis that these expressions are quantitatively identical.

We exemplify the effects of applying the WPL theory to a real closed-path EC system with measured turbulence time series and demonstrate their impact on the calculated scalar covariances and annual net flux budgets.

2.2. Sites and methods

In the following section we use measurements from the Beech forest site near Sorø, Zealand, Denmark (Pilegaard et al., 2001; Pilegaard et al., 2003). The closed-path EC system samples the air at ca. 18 m above the tree tops and carries it through a 48 m long tube at 22 l min^{-1} to a hut. In the hut, part of the air sample is taken at 8 l min^{-1} from the main stream via a secondary circuit, filtered and pressed through the IRGA (LI-6262, LI-COR, Lincoln, NE, USA) (see also Aubinet et al., 2000).

The low-pass filtering effects on turbulence time series have been analysed using degraded air temperature time series as described earlier (Ibrom et al., 2007) using

$$T_{lp_n} = T_n A_{lp} + \left(1 - A_{lp}\right) T_{lp_{n-1}} \tag{5}$$

with index lp as low-pass filtered signal and n the number of the current observation. For very small values of the dimensionless low-pass-filter constant (A_{lp}) it is important to initialise the filter with the last value of a previously filtered time series.

The low-pass-filter constant is related to the cut-off frequency of the filter (f_{co}) as noted in eq. (6) with the sampling frequency, f_s.

$$A_{lp} = 1 - e^{-2\pi \frac{f_{co}}{f_s}}. \tag{6}$$

The post-processing software *rcpm* (Morgenstern, 2000; Ibrom, 2001) was used to calculate the lag times of filtered and unfiltered time series and as well for the calculation of covariances of lagged scalars, here either the mole fractions or the mixing ratios of CO_2 and water vapour, with w.

In total five different flux estimates were calculated to describe the effects of different ways to apply the WPL theory, either incorrectly, 1–3, or correctly, 4 and 5:

(1) The scalar flux calculated from mole fractions and w without considering the WPL theory at all, $F_C^0 = \frac{\overline{\chi_C' w'}}{\overline{v_{\mathrm{mol}}}}$.

(2) The scalar flux calculated with the classical WPL theory for open-path sensors, F_C^{WPL}, that is, using incorrectly (3a) with $\overline{\chi_v' w'}$ that is corrected for high frequency loss and calculated with w that is lagged by τ_v. This approach ignores that the dilution effect on the CO_2 concentration in the closed-path sensor is actually caused by a low-pass filtered water vapour signal, with different amplitude damping and phase shift than for CO_2.

(3) The scalar flux calculated using the classical WPL theory but ignoring decoupling of water vapour and CO_2 in the sensor (F_C^{WPLM}) using (3a) with $\overline{\chi_v' w'}$ that is not corrected for high frequency loss but calculated with w that is lagged by τ_v.

(4) The two estimates of the correct CO_2 flux using either using either (3b), F_C^{WPLcp}.

(5) Equation (4), F_C.

The flux estimates were corrected for high frequency loss as described by Ibrom et al. (2007), that is, considering the high frequency loss correction for $\overline{r_C' w'}$. Using power spectra for r_C instead of, for example, χ_C, when deriving the spectral transfer function for CO_2, H_C, has the advantage that neither the CO_2 power spectra nor the estimated H_C are contaminated by the water vapour dilution effect. Because (3b) closely approximates (4), as outlined in the previous section, the spectrally corrected flux is equal to $F_l \frac{\overline{p-e}}{\overline{p}\overline{v_{mol}}} \overline{r_C' w'}$ and $F_l \frac{\overline{\chi_C' w'} + \overline{r_C}\left(\overline{\chi_v' w'}\right)_{\tau_C}}{\overline{v_{mol}}}$ for (4) and (3b), respectively, using F_l as the low-pass filtering correction factor (see Ibrom et al., 2007). Applying the spectral correction factor to F_C gives the correct result in both cases—applying it however only to $\overline{\chi_C' w'}$ in eq. (3b) instead of the whole term $[\overline{\chi_C' w'} + \overline{r_C}(\overline{\chi_v' w'})_{\tau_C}]$ would be intuitive but incorrect. The high-frequency loss correction increased the flux estimates on average by ca. 4%.

From these five different flux estimates the annual net flux budgets were calculated using only data, when the EC system worked properly and friction velocity exceeded 0.25 m s^{-1}. Gaps in the data were filled with a sequence of steps.

(1) If meteorology data existed, the daytime fluxes where filled with a look-up table that consisted of a 10 × 10 matrix of F_C averages which were calculated for each available combination of T and photosynthetically active radiation classes. The nighttime fluxes were filled with a look-up table of 20 T classes. To minimize effects of outliers, the classes were defined as equally sized bins between the 1 and 99% percentiles of the frequency distribution of the class variable. The lower and upper percents of observations were subsumed into the first and the last classes, respectively. Only those elements of the look-up tables were used, for which two or more valid flux observations existed. The look-up tables were calculated with moving 24 d data windows centred around the day where fluxes were filled.

(2) Small remaining gaps (≤1 h) were linearly interpolated.

(3) The still remaining gaps were filled with running half-hourly ensemble averages [mean diurnal variation in Falge et al. (2001)], increasing the averaging interval from 3 to 6, 12, 24 and 48 d, incrementally.

The net biotic CO_2 flux was calculated by adding the rate of CO_2 storage change underneath the sensor, S_C, to the turbulent flux. S_C was estimated from CO_2 concentrations above the canopy, which is a close approximation to the real storage change term, after filtering data for turbulent conditions with the friction velocity.

3. Results

3.1. Decoupling of CO_2 and H_2O concentrations in a closed-path EC system

Ibrom et al. (2007) have shown that the low-pass filtering of water vapour in the Sorø EC system is much stronger than for CO_2, and, additionally, that this spectral effect is not constant but varies strongly with the relative humidity. Here we show the effects on lag-times between the atmospheric scalars that have been calculated by cross-correlation analysis with r_C and w time series. We expect the time lag to consist of two components: the average traveltime of dry air in the EC system and, additionally, the time lag due to phase effects from low-pass filtering of the scalar by the tubing system of the EC system. The cross-correlation analysis detects only the sum of the two components – here we suggest an approach on how to distinguish between the two of the components.

The dependency of the lag time values from phase shift effects alone can be demonstrated with the help of T time series that are degraded with a recursive digital filter using different f_{co} (Fig. 1). These time series have been measured with the same instrument, at the same time and in the same volume as the w

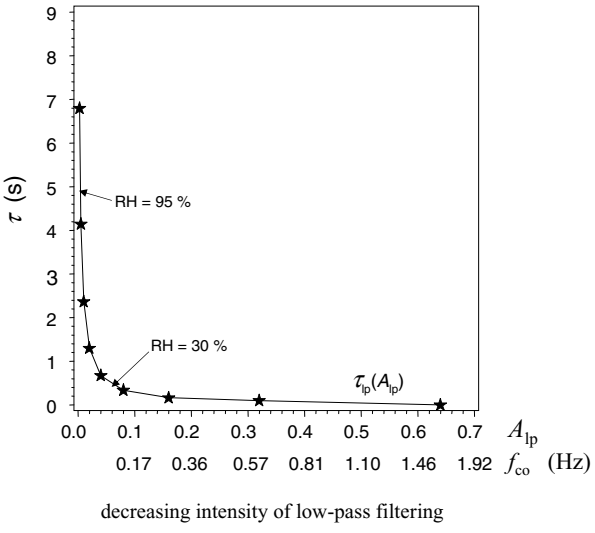

Fig. 1. Dependency of the time lag (τ_{lp}), caused by low-pass filtering as determined by covariance maximisation of degraded temperature time series with the vertical wind speed, on the low-pass filter constant, A_{lp}, and cut-off frequencies, f_{co}. Averaged time lags for water vapour and CO_2 and their difference were 8.7, 6.1 and 2.6 s, respectively. These values were calculated from 22 time series during a summer day (06:00–18:00, 2 July 2005). f_{co} for CO_2 is 0.35 Hz. Individual values of f_{co} for H_2O vary with relative humidity, RH, as shown by Ibrom et al. (2007).

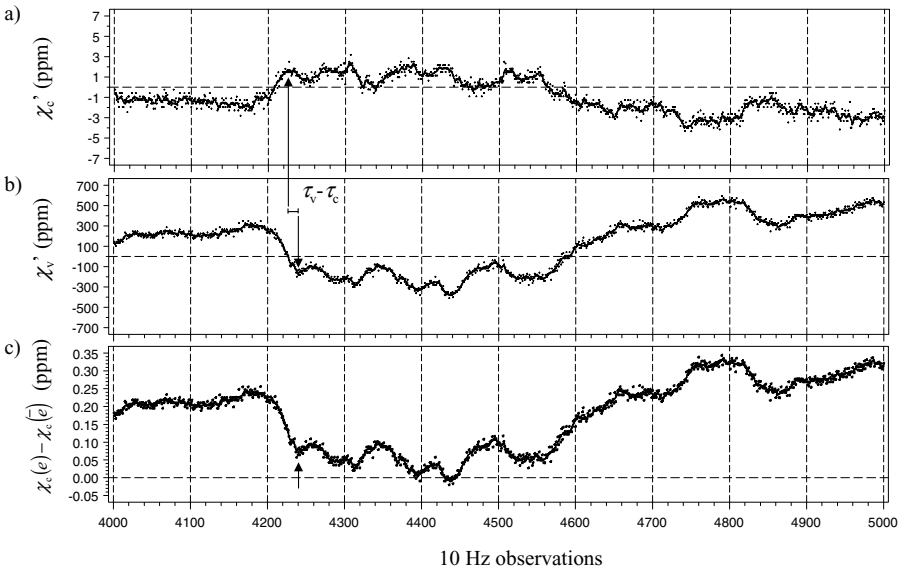

Fig. 2. Turbulence time series of (a) CO_2 and (b) water vapour mole fractions and (c) the difference between the mole fractions that were either calculated with the averaged or with the instant water vapour pressure (e). The time series was measured during a summer day (24.06.2005 14:30–15:00).

time series. Ibrom et al. (2007) found a more or less constant f_{co} value of 0.35 Hz for CO_2. One can read from Fig. 1 that the time lag due to low-pass filtering with $f_{co} = 0.35$ Hz is smaller than 0.2 s. Consequently the measured τ_C is a close approximation to the averaged traveltime of air in the EC system. The difference $\overline{\tau_v} - \overline{\tau_C}$ is on average 2.6 s and is caused by the stronger low-pass filtering of water vapour as compared to CO_2. The order of magnitude of this difference is equal to the observed range of τ_v as affected relative humidity (see arrows in Fig. 1). For our considerations it is important to note that the water vapour is on average decoupled from CO_2 by ca. 2 to 3 s. This value varies between 0.5 s at very low relative humidity and 7 s at high relative humidity.

Now we examine the effect of decoupling on χ_c at the raw data level. Figure 2 shows a case of moderate decoupling between CO_2 and water vapour at the example of a measured turbulence time series. The latent heat flux (λE) is 164 W m^{-2}, thus dilution effects on $\overline{\chi_C' w'}$ can be expected. The upper two panels show 100 seconds of χ_C' and χ_v' time series measured with the same instrument, at the same time. The visually obtained lag time difference is indicated by a bar and two arrows. The calculated values are 6.1 and 7.6 s for τ_C and τ_v, respectively. The resulting difference is 1.5 s, which is a little smaller than the difference in Fig. 2. The lower panel shows the difference between $\chi_C'(e)$ that was calculated with the actual 10 Hz water vapour pressure and $\chi_C'(\bar{e})$ that was calculated from the fast IRGA CO_2 readings but using a constant average vapour pressure value, thereby extinguishing the water vapour dilution effects on χ_C'. One can see that the difference ranges between 0 and 0.3 ppm, which corresponds to ca. 5% of the χ_C' range. The covariance of $\chi_C(e) - \chi_C(\bar{e})$ with w is equal to the water vapour dilution effect on $\overline{\chi_C' w'}$. It is obvious that the difference $\chi_C(e) - \chi_C(\bar{e})$ is perfectly synchronised with χ_v' and thus covaries with w in the same manner. Thus if we had lagged the w time series by τ_v this covariance would have been larger. In the atmosphere at the measurement point there are no time lags between χ_C' and χ_v' and, thus, the dilution effect would be even stronger in the atmosphere than in the EC system. With the data we can try to quantify the effect this has on the covariances and fluxes.

Using (4) and (3a) and calculating the difference $F_C - F_C^{WPL}$ yields a measure of the overcorrection by the traditional approach. It amounts for the 30 min sample time series to -1.2% of F_C. When using uncorrected $\overline{\chi_v' w'}$ in the dilution term of (3a), as suggested by Massman (2004), the estimate, F_C^{WPLM}, is still 0.4% smaller than F_C. Using (3b) includes also the effect of decoupling between CO_2 and H_2O in the EC system and yields a very close approximation to F_C; F_C^{WPLcp} is only 0.05% smaller than F_C. Hence, the sole decoupling effect contributes in our sample case to 1/3, the amplitude effect about two-third of the overestimation, if the original WPL theory would be applied for closed-path sensors.

3.2. *Effects of the WPL theory on measured covariances*

Figure 3 shows comparisons of four different CO_2 flux estimates from χ_C and w taking various aspects of the WPL theory for closed-path sensors into account with the alternative estimate using point-by-point conversion, F_C. All half-hourly data of the year 2005 were used when the IRGA worked properly. But even using quality checked flux data, a difficulty arose from an effect

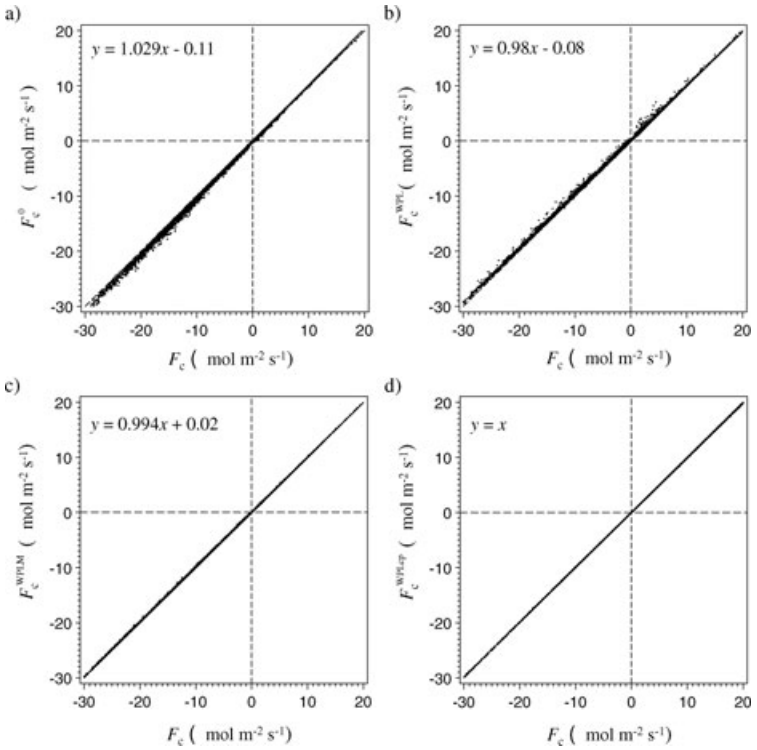

Fig. 3. Comparison of an uncorrected CO_2 flux estimate using mole fractions (F_C^0, a), applying the WPL-theory by using the best estimate of $\overline{\chi_v' w'}$ in the atmosphere in the dilution term (F_C^{WPL}, b), using $\overline{\chi_v' w'}$ that were not high frequency corrected (F_C^{WPLM}, c), and using $\overline{\chi_v' w'}$ in the WPL dilution term like it was measured in the close-path EC-system, that is, at the lag time for CO_2 (F_C^{WPLcp}, d) with the estimate that was calculated from molar CO_2 mixing ratios, F_C, eq. (4).

of the flux calculation on the lag time. This introduced some noise in the comparison, which we avoided by selecting only those covariances, where the lag times did not differ when using either mixing ratios or mole fractions. Due to the directional effects of the WPL theory, regressions have also been analysed for unidirectional subdata sets, that is, for positive and negative fluxes, respectively (Table 1).

Ignoring WPL theory when using $\overline{\chi_C' w'}$ in the flux calculations increased its absolute value on average by 2.9% (Fig. 3a). If, however, only data with negative F_C were selected, the average increase was 3.3% (Table 1). Applying the WPL theory to these covariances as originally suggested, reduced the flux estimate relative to F_C on average by 2% (Fig. 3b)—the flux was overcorrected. The result for the entire data set is very similar to that we described at the example of one time series (Section 3.1). Two-third of the overcorrection could be attributed to the fact that $\overline{\chi_C' w'}$ had been corrected for amplitude damping and the rest was caused by not taking decoupling of water vapour and CO_2 in the EC system into account. The results show also that applying the modified approach on covariances (3b) is equivalent to converting the concentrations to mixing ratios prior to the covariance calculations (4). F_C^{WPLcp} is practically identical with F_C, for time series where the lag times did not depend on whether mole mixing ratios or mole fractions were used (Fig. 3d). We show here for the first time that the two different concepts yield almost the same result, given the appropriate application of the WPL theory in closed-path EC systems, thereby also confirming the principle of the WPL theory. The comparisons in Table 1 show that effects of dilution (F_C^0 versus F_C) or of decoupling (F_C^{WPL} versus F_C) are only evident, if $F_C < 0\ \mu$mol m^{-2} s^{-1}, that is, they are asymmetric with respect to the CO_2 flux direction.

Table 1. Statistics of linear regression and correlation analysis of CO_2 fluxes (F_C) and water vapour fluxes as latent heat fluxes (λE). The fluxes from CO_2 mixing ratios are used as independent variables. For definitions of symbols, see Section 2.2. Only time series were used, where the lag times did not differ between using either mixing ratios or mole fractions in the calculations.

	Slope	Intercept	R^2
F_C^0 versus F_C	1.029	−0.109	0.999
F_C^0 versus F_C only positive	1.001	−0.020	0.999
F_C^0 versus F_C only negative	1.033	−0.083	0.998
F_C^{WPL} versus F_C	0.981	0.082	0.999
F_C^{WPL} versus F_C only positive	1.001	0.017	0.997
F_C^{WPL} versus F_C only negative	0.980	0.087	0.999
F_C^{WPLM} versus F_C	0.994	0.022	1.000
F_C^{WPLM} versus F_C only positive	0.999	0.003	1.000
F_C^{WPLM} versus F_C only negative	0.003	0.024	1.000
F_C^{WPLcp} versus F_C	1.000	−0.004	1.000
F_C^{WPLcp} versus F_C only positive	0.999	−0.001	1.000
F_C^{WPLcp} versus F_C only negative	1.000	−0.004	1.000
λE^0 versus λE	0.989	0.035	1.000
λE^{WPL} versus λE	0.999	0.001	1.000

The effects on the water vapour flux estimation were smaller than for CO_2 fluxes, but still measurable (Table 1). Neglecting

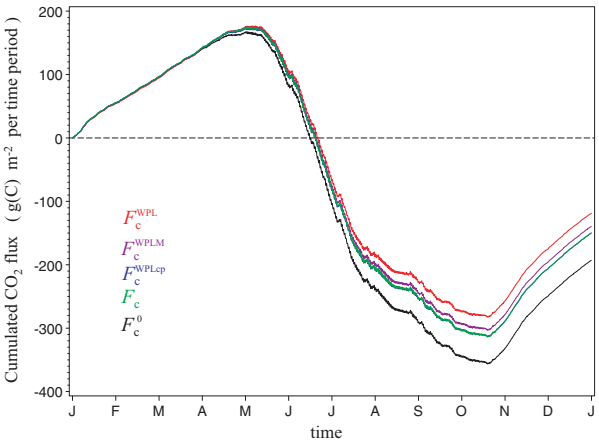

Fig. 4. Cumulative gap-filled CO_2 fluxes of the year 2005 calculated from the five different CO_2 flux estimates (definitions, see Fig. 3). Estimates F_C^{WPLcp} and F_C were so close to one another that they can not be distinguished. Annual sums are given in Table 2.

WPL theory reduced water vapour flux estimates by 1.1%; ignoring decoupling of H_2O and CO_2 in the EC system had no effect on water vapour fluxes at all.

Figure 4 displays the effects of the way the WPL theory is applied in the CO_2 flux calculation on the annual net CO_2 fluxes. It can be seen that differences between the cumulated flux estimates start to increase in late spring until the beginning of the winter period, when they stayed constant. Table 2 lists estimates of the annual net fluxes as calculated from the different approaches. Using the conventional formulation to consider WPL theory in the flux calculation from covariances of mole fractions and w, leads to the underestimation of the annual net CO_2 uptake by 21% or 31 g m^{-2} yr^{-1}; ignoring the WPL theory would overestimate the flux by 29%. The decoupling accounts for 1/3 of the overcorrection, that is, 7% or 11 g m^{-2} yr^{-1} of the most realistic annual net flux estimates (F_c and F_C^{WPLcp}). The flux estimate using the WPL theory that is adapted to the imperfections of closed-path EC systems (3b) comes very close to the estimate that was calculated from mixing ratios (4).

4. Discussion

The comparison of flux estimates from different calculation methods confirmed the theory (see Section 2.1) in all cases, which will be explained briefly. First of all, although much smaller than in open-path EC systems, the WPL theory has to be considered, when calculating fluxes from mole fractions in closed-path EC systems, otherwise the absolute flux magnitudes of downward directed fluxes will be overestimated and upward directed fluxes will be underestimated.

Applying the WPL theory reduced the absolute CO_2 flux estimates, because the CO_2 fluxes where downward directed, whereas the water vapour fluxes were upward directed. The opposite effect could not be shown (Table 1), because in situations with net upward F_C, that is, during night and during the whole winter, the water vapour fluxes were then generally low. The coincidence between the direction of the CO_2 flux and the magnitude of the water vapour flux introduced an asymmetry, that is, effects of WPL theory on the CO_2 uptake—no effect on CO_2 release, which increased the effect on the annual CO_2 net uptake strongly. By this, the WPL effect on flux estimates becomes much larger on a daily or annual basis than expected from the effects on single covariances.

As expected, ignoring the decoupling of water vapour and CO_2 in the EC system when applying the WPL theory leads to an overcorrection of the fluxes ($|F_C^{WPL}| < |F_C^{WPLM}| < |F_C^{WPLcp}| \approx |F_C|$). The reason is that the effect of water vapour dilution on the covariance of the CO_2 mole fraction with w is smaller in the closed-path EC system than in the atmosphere, because water vapour fluctuations are attenuated and shifted against CO_2 in the EC system and thus less correlated with w. This was proved at the raw data level (Fig. 2), and the consequences on the flux estimates were demonstrated with the covariances (Fig. 3) and the annual budgets (Fig. 4). Because this effect has not yet been included in existing recommendations on how to consider the WPL theory in calculating fluxes from closed-path sensors they have to be revised and, consequently, the recalculation of carbon fluxes to be considered using, for example, (4) and raw data series of CO_2 and H_2O mole mixing ratios. For this it is necessary to measure and record the internal physical conditions in the samples cell at the same temporal resolution like the water vapour and the scalar concentration. A similar result can be obtained using (3b), which requires the calculation of $\overline{\chi_v' w'}$ at exactly the same time lag as for the calculation of $\overline{\chi_C' w'}$. As with the temperature fluctuations, which are effectively filtered, the dilution term of the WPL theory should contain the same $\overline{\chi_v' w'}$ that imposes the dilution effect in the sensor, that is, $\overline{\chi_v' w'_{\tau_c}}$. A bias between both methods arises from different lag times, either if mixing ratios or mole fractions are used for covariance calculations.

Table 2. Different estimates of annual net biotic CO_2 fluxes for the year 2005 (definitions of symbols, see Section 2.2).

$F_C^0 + S_C$	$F_C^{WPL} + S_C$	$F_C^{WPLM} + S_C$	$F_C^{WPLcp} + S_C$	$F_C + S_C$
		g (C) m^{-2} yr^{-1} (%)		
−192.8 (128.5)	−118.8 (79.2)	−139.3 (92.9)	−149.6 (99.7)	−150.0 (100)

Contrary to Massman (2004), we recommend using mixing ratios instead of mole fractions directly at the raw data level when calculating the fluxes from closed-path EC measurements, because this considers the WPL theory and excludes effects of decoupling on closed-path EC systems at the same time. Massman (2004) argued that the lack of fast recordings of pressure and temperature in the sample cells of the IRGA might lead to errors in the calculations of the mixing ratios in this case. However, because these parameters are also needed to calculate mole fractions of CO_2 from the raw voltage recordings of the sensor, the effects are the same, irrespectively of whether one calculates mole fractions or mixing ratios. We investigated the effect by comparing covariances calculated with half-hourly averages of internal pressure and temperature with those calculated with 1 min averages and did not find any systematic effects. The reason is that fluctuations of internal pressure and temperatures do not correlate with w.

Although the tight relationships in Fig. 3a and b imply the possibility of a simple correction without recalculation from raw data, we do not recommend this. It has to be noted, that these strict relationships do only exist, when τ_c was not affected by using either r_C or χ_C in the cross-correlation analysis with w. Because the effects depend on the magnitude of low-pass filtering in the EC system, we rather recommend investigating, whether the effects are similarly strong in a certain design and application of a closed-path EC system and need to be corrected anyway. A first parameter that shows, whether the effects will be relevant for a closed-path EC system, is the difference between τ_v and τ_C. The larger the difference, the larger will be the effect of decoupling on the WPL terms.

As has been shown by Clement (2004) and in this study (cf. Fig. 1) using the results from a previous study (Ibrom et al., 2007), these differences in lag times depend strongly on relative humidity. At high relative humidity, low-pass filtering of the water vapour signal in closed-path EC systems is much stronger due to absorption and desorption of H_2O molecules to or from the surfaces of the EC system. These effects can be seen in every EC system in which relative humidity is not kept at very low values. The lag time for CO_2 has been reported much more often than for H_2O (see, e.g. Aubinet et al., 2000). Clement (2004) used two different time windows, in which the lag times were searched for. While the lag time for CO_2 ranged between 6 and 8 s, the lag time for water vapour could reach up to 25 seconds in his EC system. In this study it is shown that the differences in lag times range from 0.5 to 7 s(Fig. 1). The technical designs of the two EC systems differ substantially. Different tube lengths are used and the Sorø system uses a secondary sampling circuit, whereas the EC system at the Griffin site does not (see Aubinet et al., 2000; Pilegaard et al., 2003). The values reported here may, therefore, well be typical for a number of closed-path EC systems operating over forests. The effects on measurements above smoother surfaces might well be stronger, because the contribution of higher frequencies to the flux is comparably larger than in applications over forests. The high frequency domain is mostly affected by the decoupling of water vapour and CO_2 in the EC system.

The results showed also that application of the WPL theory had only a small effect on the estimation of water vapour fluxes—the fluxes were slightly increased after correction. Although the effects are small, it has to be noted that they are systematic and thus should be considered. Decoupling between water vapour and CO_2 in the EC system had, however, only negligible effect. The reason for this is that the CO_2 concentration in the sample is under ambient conditions much smaller than the water vapour concentration, and thus has no measurable effect on neither values of the water vapour mole fractions nor of mixing ratios. Consequently, the conventional method applying the WPL theory yields accurate results for water vapour and latent heat flux estimates.

The results of this study apply in principle not only for CO_2 flux measurements but for every closed-path EC system, for example, measuring N_2O, CH_4, O_3 or NO_x fluxes, in which the gas concentration is being diluted by water vapour fluctuations. To take the WPL theory into account, pressure, air temperatures and water vapour concentrations need to be estimated for the same sample cell where the other gases are measured. The results of this study show clearly that using water vapour fluxes in the atmosphere is not sufficient when considering the WPL theory, because the dilution effects in the closed-path EC system would be overestimated to some degree. This becomes in particular a problem, if the closed-path system does not offer the feature to measure water vapour fluctuations in the sample cell as it is the case with some ozone and NO_2 closed-path EC analysers (Massman et al., 1995; Pilegaard et al., 1998; Gerosa et al., 2005). A solution for such systems is drying the air sample physically before the air sample enters the sample cell of the EC system (Edwards et al., 2003; Pattey et al., 2006). If amplitude attenuation and phase shift of water vapour in the EC system are known, eq. (3b) will give an accurate estimation of the WPL effects. In those systems in which the low-pass filtering effects on water vapour concentrations are clearly dominated by processes in the tubing system, a second closed-path system can be attached to the same tubing system and might be used to estimate the water vapour fluctuations in an analyser that does not measure water vapour concentrations. In any case it is recommended to measure and record the internal physical conditions in the sample cell at the same temporal resolution like the water vapour and the scalar concentration. The above-described effects of decoupling are restricted to closed-path systems. Open-path systems will only be affected from errors in the water vapour and sensible heat flux estimation.

5. Conclusions

In an analysis of the water vapour dilution effects on scalar concentrations in closed-path sensors we showed that the WPL the-

ory (Webb et al., 1980) can not directly be applied to these EC systems, because water vapour and CO_2 molecules are decoupled in the tubing system. We recommend using mole mixing ratios rather than mole fractions to calculate covariances that are neither biased by decoupling nor by WPL terms. Using this flux estimate as a reference, we showed considerable systematic effects of different ways of considering WPL theory. Neglect of decoupling and amplitude damping led to overcorrection of the annual net CO_2 fluxes of in our case of 21% of which 7% was caused by the decoupling effect. The traditional WPL theory can be adapted to low-pass filtering effects in closed-path sensors by using the covariance of water vapour and the vertical wind velocity that is calculated at the same lag time as for CO_2 rather than that for water vapour. Given this small modification, we could prove here for the first time that applying the WPL theory to covariances from mole fractions yields practically the same results as calculating fluxes from raw data that has been converted point-by-point to mole mixing ratios. We specified correct ways to apply spectral corrections to CO_2 fluxes using either point-by-point converted raw data or the specified WPL correction for closed-path systems.

The recommendations described here for closed-path sensors do not apply for open-path sensors, because these measure the processes directly in the atmosphere, that is, the original expression by Webb et al. (1980) describes the effects of density fluctuations appropriately accurately.

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Appendix

In Aubinet et al. (2000) it was mentioned that the WPL theory does not need to be applied to their closed-path EC measurements, because their IRGA (LI-6262) automatically corrected for water vapour dilution. We add here that this automatic correction has to be activated by a certain setting (FCT 76 set to 2, see LI-COR Manual). If done so, the LI-6262 will calculate mole fractions in the sample cell, which will be related to the moles of humid air in the reference cell. If the reference gas is scrubbed for water vapour or purged with nitrogen, which is a very common set up, the LI-6262 will output mixing ratios for CO_2 instead of mole fractions. This particular setting does not have any effect on the vapour concentrations, that is, these are still mole fractions (LI-COR, personal communication 2006) and it has also no effect on the fast voltage output of the sensor.

In their eq. (18) Aubinet et al. (2000) expressed the CO_2 flux similar to

$$F_C = \frac{\bar{p}}{\Re \overline{T}_{son}} \overline{r'_C w'}. \tag{A1}$$

We replaced here mole fractions by mole mixing ratios, according to the IRGA output as mentioned above. Equation (A1) differs from eq. (4) by the factor with which the covariance is multiplied. Because T_{son} is a close approximation to the virtual temperature, the factor $\frac{\bar{p}}{\Re \overline{T}_{son}}$ is almost identical with the number of moles of humid air per m^3. The appropriate measure would be the number of moles of dry air per m^3. The resulting error in the flux calculation is however only small. It scales with vapour pressure with ca. –1% per 1500 Pa and is other than the decoupling effect not asymmetric with regard to night or daytime. In case of water vapour fluxes (A1) gives correct results, because the LI-6262 outputs mole fractions irrespectively of any of the possible configurations.

The new IRGA LI-7000 does not include the dilution correction anymore. For this and similar closed-path analysers the application of the WPL theory requires either the transformation of mole fractions to mole mixing ratios by the user, or the use of eq. (3b). For the point-by-point conversion of CO_2 mole fractions to mole mixing ratios the pressure and temperatures in the sample cell are needed at a high sampling frequency. Thus it is strongly recommended to keep these supplementary data together with the scalar concentrations as raw data. They can also be used for future methodical analyses and for data quality assessment.

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