True eddy accumulation - Part 1: Solutions to the problem of non-vanishing mean vertical wind velocity

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Abstract. The true eddy accumulation method (TEA) provides direct measurements of ecosystem-level turbulent fluxes for a wide range of atmospheric constituents. TEA utilizes conditional sampling to overcome the requirement for a fast sensor response demanded by the state-of-the-art eddy covariance method (EC).

The TEA method is formulated under the assumption of ideal conditions with a zero mean vertical wind velocity during the averaging interval. However, this idealization is rarely met under field conditions. Additionally, unlike in EC, this assumption can not be imposed in post processing due to the real-time nature of sampling and the absence of high-frequency measurements of the scalar. Consequently, fluxes measured with the TEA method are biased with a non-turbulent advective term that scales with the scalar mean concentration.

Here, we explore the magnitude of this biased advective term and potential ways to minimize or remove it. We propose a new formulation to calculate TEA fluxes that minimizes the bias term. The new formulation shows that the magnitude of the error is constrained to $\bar{w}/\overline{|w|}$ when the stationarity criterion is fulfilled. Here, w is the vertical wind velocity, and the overbar denotes time averaging. The error is shown to be dependent on the asymmetry of atmospheric transport, represented by the coefficient α_c. Two methods of estimating the coefficient α_c are proposed, a probabilistic treatment of turbulent transport, and a method utilizing the assumption of scalar similarity. We show how other formulas for calculating the TEA flux are linked to the new formulation and explore the different corrections in a numerical simulation.

The new formulation avoids the direct dependence of the bias term on the scalar background concentration. This result increases the confidence in applying the TEA method to measuring fluxes of atmospheric constituents. This is particularly relevant to scalars with a large background concentration and a small flux. This paper is part one of a two-part series on true eddy accumulation.

1 Introduction

Micrometeorological methods provide non-invasive, in situ, integrated, and continuous point measurement for ecosystem fluxes on a scale ideal for ecosystem study (Baldocchi et al., 1988; Baldocchi, 2014). Among micrometeorological methods, eddy covariance (EC) has become the de-facto method for measuring ecosystem fluxes for the past 40 years. The EC method is the most direct micrometeorological method. It is also relatively easy to set up and operate. These features have led to the wide

use and adoption of the EC method at hundreds of sites worldwide, including several regional and global flux measurements networks such as ICOS and FLUXNET (Hicks and Baldocchi, 2020).

The EC method depends on the fast measurement of vertical wind velocity and the scalar concentration (such as an atmospheric constituent). The requirement for fast measurement frequency (10 to 20 Hz) limits the application of the method to a handful of atmospheric constituents where fast gas analyzers are available. Alternative methods that work for slow gas analyzers include: (i) signal downsampling methods (Lenschow et al., 1994) such as disjunct eddy accumulation (Rinne et al., 2000a; Turnipseed et al., 2009) and disjunct eddy covariance (Rinne and Ammann, 2012), and (ii) indirect methods such as flux gradient methods (e.g, Rinne et al. (2000b)) which depend on the Monin-Obukhov similarity theory (Monin and Obukhov, 1959) and the relaxed eddy accumulation (REA) which assumes flux-variance similarity (Businger and Oncley, 1990). The true eddy accumulation method (TEA) (Desjardins, 1977) is the most direct and mathematically equivalent alternative to eddy covariance among accumulation methods. Unlike EC, the TEA method requires the concentration measurements to be carried out once every averaging interval (30 minutes) Businger and Oncley (1990). The TEA method is formulated under ideal conditions assuming a zero mean vertical wind velocity during the averaging interval. This assumption is almost never met under field conditions and it is not possible to enforce in post-processing due to lacking high-frequency information of the scalar concentration. As a result, the non-vanishing vertical mean velocity will contribute to a systematic error in the flux. Nonzero mean vertical wind velocity is a source of error for all eddy accumulation methods, including TEA (Hicks and McMillen, 1984), relaxed eddy accumulation (REA) (Pattey et al., 1993; Businger and Oncley, 1990; Bowling et al., 1998), and disjunct eddy accumulation (DEA) (Turnipseed et al., 2009). The reported bias on the flux due to nonzero $\bar{w}$ varied with different studies and accumulation methods. For TEA, Hicks and McMillen (1984) recommended that $\bar{w}$ should not exceed $0.0005\,\sigma_w$ if accumulated mass is measured and $0.02\,\sigma_w$ when concentrations are measured directly. Turnipseed et al. (2009) reported that a mean vertical wind bias of $\pm\,0.25\,\sigma_w$ lead to $\pm\,15\%$ mean systematic bias in the flux using the disjunct eddy accumulation method. Values reported for the REA method showed that a systematic bias of approximately 5% of the flux due to a $\bar{w}$ of $0.20\,\sigma_w$ (Pattey et al., 1993), which agrees with the recommendations of Businger and Oncley (1990). The magnitude of the residual mean vertical velocity depends on the meteorological and topographic features of the measurement site and is larger in complex sites (Rannik et al., 2020).

In this paper, we revise the theory of the true eddy accumulation method and obtain a generalized equation that isolates the error due to nonzero vertical wind velocity. The new equation shows that the error in the flux is a function of the atmospheric transport represented by the transport asymmetry coefficient, α_c. We study the value and the interpretation of this coefficient in the framework of quadrant analysis and define its boundary conditions. Next, we show analytical and empirical ways to obtain the transport asymmetry coefficient and explore the implications of these estimates on the flux in a numerical simulation. Finally, we show how existing formulations for calculating the TEA flux are special cases of the new equation.

2 Theory

2.1 Eddy covariance

The net ecosystem exchange (NEE) of a scalar c (such as an atmospheric constituent), $\overline{N_c}$ is the total vertical flux $\overline{wc}$ across the measurement plane at a height h and the change of storage below that height (Gu et al., 2012).

$$60 \quad \overline{N_c} = \overline{wc}\big|_h + \int_0^h \frac{\partial c}{\partial t} \mathrm{d}z \tag{1}$$

Where w is the vertical wind velocity ($\mathrm{m\,s^{-1}}$), c is the molar density ($\mathrm{mol\,m^{-3}}$) of the scalar of interest (such as CO_2). The previous equation can be reached either from a holistic mass balance approach or by averaging the continuity equation for the scalar, c and integrating from the surface to measurement height, h. In both cases, horizontal advection is ignored as a virtue of the assumption of horizontal homogeneity and molecular diffusion is ignored due to its small magnitude (Gu et al., 2012). For a full discussion on the equations of surface flux, see, for example (Finnigan et al., 2003; Foken et al., 2012a).

The storage term measurements and value are beyond the scope of this study, therefore we ignore it. Consequently, the total vertical flux is represented by the first term on the right hand side of Eq. (1) which can be further decomposed into turbulent and mean advective parts.

$$\overline{wc} = \overline{w'c'} + \bar{w}\,\bar{c} \tag{2}$$

The overlines denote ensemble averages that obey Reynolds averaging rules. Primes represent departures from the mean. The ensemble averages are estimated experimentally by time averages, thus for a stationary time series drawn from an ensemble, the turbulent flux for the averaging period, Δt can be written as

$$F_{\Delta t} = \frac{1}{\Delta t} \int_0^{\Delta t} w'(t) c'(t) \, \mathrm{d}t = \overline{w'c'} \tag{3}$$

where $w(t)$ and $c(t)$ are realizations of the vertical wind velocity and the scalar quantity such as CO_2 concentration, respectively.

2.2 True Eddy Accumulation

The true eddy accumulation method circumvents the need to record the fluctuations of scalar concentration at a frequency sufficient to represent the individual flux transport eddies. Instead, it is sufficient to measure the mean product $\overline{wc}$ for updraft and downdraft once for each averaging interval, Δt (e.g. 30 minutes).

The product of w and c is realized by physically collecting air samples with a flow rate proportional to the vertical wind velocity, w. The method is formulated assuming ideal conditions where the mean vertical wind velocity during the averaging

period is assumed to be zero. When $\bar{w} = 0$, the second term on the right hand side of Eq. (2) will be zero and the turbulent flux $\overline{w'c'}$ will equal the total ecosystem flux $\overline{wc}$. By separating $\overline{wc}$ depending on the direction of the vertical wind velocity we can write

$$85 \quad \overline{wc} = \frac{1}{\Delta t} \int_{0}^{\Delta t} (\delta^{+} cw + \delta^{-} cw)\, \mathrm{d}t \tag{4}$$

$$\text{where } \begin{cases} w > 0 & \delta+ = 1; \ \delta- = 0 \\ w < 0 & \delta+ = 0; \ \delta- = 1 \end{cases} \tag{5}$$

Hence, by sampling air with a flow rate proportional to the magnitude of vertical wind velocity and accumulating it according to its direction in updraft and downdraft reservoirs, one can measure the quantity $\overline{wc}$ and consequently the flux without having to measure the high-frequency fluctuations of the scalar, c (Desjardins, 1977; Hicks and McMillen, 1984).

90   Sampling air proportional to the magnitude of vertical wind velocity requires a scaling parameter, A that ensures the proportionality of the flow rate to the magnitude of vertical wind velocity. The scaling parameter is the product of the pump calibration coefficients and other coefficients used to adjust the system's dynamic range. For a short interval of time $\mathrm{d}t$, a sample of the volume $V_{sample} = A|w|\,\mathrm{d}t$ will be collected in the system. The accumulated sample volume in each of the two reservoirs during a long enough averaging period Δt (30 to 60 minutes) will be

$$95 \quad V_{total} = \frac{1}{\Delta t} \int_{0}^{\Delta t} A|w|\,\mathrm{d}t \tag{6}$$

By the end of the averaging period, Δt, the flux will be equal to the difference in the scalar accumulated mass between updraft and downdraft reservoirs.

If it is desired to formulate the flux in terms of the accumulated scalar concentration $(\mathrm{mol\,m^{-3}})$ instead of the accumulated mass, the average scalar density of accumulated samples in each of the reservoirs will equal the accumulated mass of the scalar

100   divided by the accumulated volume

$$C_{acc}^{\uparrow\downarrow} = \frac{m}{V} = \frac{\overline{c\,|w^{\uparrow\downarrow}|}}{\overline{|w^{\uparrow\downarrow}|}} \tag{7}$$

Where C_{acc} is the accumulated scalar density and the arrows indicate the reservoir. The measured concentration in Eq. 7 is the weighted mean of the scalar concentration and the magnitude of the vertical wind velocity.

When $\overline{w}$ is assumed to be zero, $\overline{|w^{\uparrow}|} = \overline{|w^{\downarrow}|} = \overline{|w|}/2$, and we can write the flux in terms of concentrations of accumulated

105   samples, similar to Hicks and McMillen (1984)

$$F_{\text{TEA}} = \frac{\overline{|w|}}{2}(C_{acc}^{\uparrow} - C_{acc}^{\downarrow}) \tag{8}$$

Where $\overline{|w|}$ is the mean of the magnitude of the vertical wind velocity.

2.3 The problem of nonzero mean vertical wind

Although the total ecosystem flux is defined to be $\overline{wc}$ in Eq. (2), it is not possible to directly use the measured w and c to calculate the total flux. The reason is the difficulty of obtaining an accurate measurement of w. Any non-turbulent offset (bias) in the mean vertical velocity will lead to a flux biased with $\bar{w}\bar{c}$. Several reasons contribute to a biased mean vertical wind velocity including: topography in particular in complex sites, non-alignment of the anemometer to local topography, biases in anemometers, flow perturbations, and meteorological reasons induced by local circulation or topographical drainage (Lee et al., 2005; Paw U et al., 2000; Heinesch et al., 2007). Therefore, the measured biased advective term needs to be discarded and the true physical term, known as "Webb term" or Webb-Pearman-Leuning (WPL) term, needs to be estimated by other means (Webb et al., 1980; Fuehrer and Friehe, 2002). The original formulation of the TEA method assumes a zero mean vertical wind velocity during the flux averaging interval thus assumes the total ecosystem flux to be equal to the turbulent flux, $\overline{w'c'}$. However, this assumption is rarely valid under field conditions due to the reasons outlined earlier and the measured TEA flux will be a biased total vertical flux, $\overline{wc}$. If the turbulent flux is to be measured using the TEA method, the biased term $\bar{w}\bar{c}$ needs to be removed.

Previous efforts have been focused on minimizing $\bar{w}$ to reduce the bias in the TEA flux. However, since the wind information can not be changed after sampling, any treatments for the wind velocity measurements are final when the air samples have been collected. Thus, there is no way to guarantee a zero mean vertical velocity. A common approach to nullifying mean vertical wind velocity in EC measurements is to rotate the wind coordinates in post-processing to force $\bar{w}$ to zero for each averaging interval, this method - commonly referred to as double rotation - is not feasible in eddy accumulation methods. The planar fit method (Wilczak et al., 2001) and its variants, such as the sector-wise planar fit (Foken et al., 2004), are better suited for the online application in the TEA method (Siebicke and Emad, 2019).

The planar fit method aligns the sonic coordinates to the long-term streamline coordinates by aligning the wind vector to the plane that minimizes the sum of squares of the vertical wind velocity means for a long period of time (weeks to months). This approach, while minimizes the vertical wind velocity means of the individual averaging intervals, does not force them to be zero. Considerable spread of $\bar{w}$ values around zero can still be observed after applying the planar fit method. (Sun, 2007; Rannik et al., 2020).

2.4 TEA equation under nonzero $\bar{w}$ conditions

The goal here is to enable measuring the turbulent flux $\overline{w'c'}$ from TEA measurements when $\bar{w} \neq 0$. The key to extending the TEA equation to conditions of nonzero $\bar{w}$ is to obtain an estimate of the scalar mean $\bar{c}$ from TEA measurements, and

consequently remove the biased advective term $\bar{w}\bar{c}$. We achieve this by using the weighted mean of c and $|w|$ as an estimate for c after correcting for the correlation between them.

The weighted mean of the scalar, c and the vertical wind velocity magnitude, $|w|$ can be obtained from TEA measurements Eq. (7). By decomposing $\overline{c|w|}$ into mean and fluctuating parts

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$$\overline{c|w|} = \overline{c'|w|'} + \bar{c}\,\overline{|w|} \tag{9}$$

The value of $\bar{c}$ can be found to be

$$\bar{c} = \frac{\overline{c|w|}}{\overline{|w|}} - \frac{\overline{c'|w|'}}{\overline{|w|}} \tag{10}$$

Substituting $\bar{c}$ in Eq. (2), we can write the flux as

$$\overline{w'c'} = \overline{wc} - \frac{\bar{w}}{\overline{|w|}}\,\overline{|w|}\bar{c} + \frac{\bar{w}}{\overline{|w|}}\,\overline{|w|'c'} \tag{11}$$

145
We can obtain all the terms in Eq. (11) from our measurements except for the covariance term $\overline{|w|'c'}$. We define the "transport asymmetry coefficient" for the scalar c, (α_c) as the ratio of the covariance between the wind magnitude and the scalar to the covariance between the wind and the scalar.

$$\alpha_c \equiv \frac{\overline{c'|w|'}}{\overline{c'w'}} \tag{12}$$

We notice α_c is conveniently independent of the scalar standard deviation. It can be written as

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$$\alpha_c = \frac{\rho_{c|w|}\sigma_{|w|}}{\rho_{cw}\sigma_w} \tag{13}$$

where $\rho_{c|w|}$, ρ_{cw} are the correlation coefficients between c and $|w|$, and c and w, respectively. $\sigma_{|w|}$ and σ_w are the standard deviations of $|w|$ and w, respectively. After substitution, we write the turbulent flux as

$$\overline{w'c'} = \overline{wc} - \frac{\bar{w}}{\overline{|w|}}\,\overline{|w|}\bar{c} + \frac{\bar{w}}{\overline{|w|}}\alpha_c\overline{w'c'} \tag{14}$$

Finally, we rearrange Eq. (14) and obtain the generalized TEA flux equation that gives a turbulent TEA flux when the mean
155 vertical wind velocity is nonzero

$$\overline{w'c'} = \frac{\overline{wc}\,\overline{|w|} - \overline{|w|}\bar{c}\,\bar{w}}{\overline{|w|} - \alpha_c\bar{w}} \tag{15}$$

2.4.1 Calculating the corrected TEA flux

The new general equation for TEA (Eq. 15) extends the validity of the method to conditions where the mean vertical wind velocity is nonzero. We show here how the turbulent TEA flux can be calculated from the measured physical quantities.

160   The weighted mean over an averaging period Δt can be written as

$$\frac{\overline{c|w|}}{\overline{|w|}} = \overline{c|w\uparrow|}\,\frac{\overline{|w\uparrow|\Delta t^{\uparrow}}}{\overline{|w|}\Delta t} + \overline{c|w\downarrow|}\,\frac{\overline{|w\downarrow|\Delta t^{\downarrow}}}{\overline{|w|}\Delta t} \tag{16}$$

which in terms of the quantities we are measuring, translates to

$$\overline{c|w|} = \overline{|w|}\left(\frac{C_{acc}^{\uparrow}V^{\uparrow} + C_{acc}^{\downarrow}V^{\downarrow}}{V_{\text{total}}}\right) \tag{17}$$

Similarly

$$165 \quad \overline{cw} = \overline{|w|}\left(\frac{C_{acc}^{\uparrow}V^{\uparrow} - C_{acc}^{\downarrow}V^{\downarrow}}{V_{\text{total}}}\right) \tag{18}$$

After substitution and simplification, we obtain the TEA flux in terms of the measured quantities

$$F_{\text{TEA}} = \frac{C_{acc}^{\uparrow}V^{\uparrow}\left(\overline{|w|}-\bar{w}\right) - C_{acc}^{\downarrow}V^{\downarrow}\left(\overline{|w|}+\bar{w}\right)}{\overline{|w|}-\alpha_c\bar{w}} \times \frac{\overline{|w|}}{V_{total}} \tag{19}$$

Where F_{TEA} is the kinematic flux density ($\mathrm{mol\,m\,s^{-1}}$). $C_{acc}^{\uparrow}$ and $C_{acc}^{\downarrow}$ are the mean concentrations ($\mathrm{mol\,m^{-3}}$) of the scalar c in updraft and downdraft reservoirs at the end of the averaging period, Δt. $V^{\uparrow}$ and $V^{\downarrow}$ are the accumulated sample volumes

170   ($\mathrm{m^3}$) in updraft and downdraft reservoirs during the averaging period. $\overline{|w|}$ is the mean of the absolute vertical wind velocity ($\mathrm{m\,s^{-1}}$) during the averaging period.

2.5 Values of the transport asymmetry coefficient α_c

2.5.1 Quadrant analysis of α_c

The value of α_c can be analyzed using the framework of quadrant analysis. Quadrant analysis is commonly used to inspect

175   the contributions from different quadrants in the (w', c') plane by sorting the instantaneous values into four categories (S_1 .. S_4) according to the sign of the two fluctuating components (e.g., (Katul et al., 1997; Raupach, 1981; Katsouvas et al., 2007)). Where S_i is the fraction of the flux transported by contributions in quadrant i. Following the definition of Thomas and Foken (2007), the pairs S_2 and S_4 are ejections and sweeps for downward directed net flux (negative ρ_{wc}) and S_1 and S_3 for upward directed net flux (positive ρ_{wc}).

The total flux is the sum of the contributions from the four quadrants, we similarly find that the covariance term $\overline{|w'|c'}$ can be written as

$$\overline{|w'|c'} = S_1 + S_4 - S_2 - S_3 \tag{20}$$

It follows that α can be written in terms of quadrants as

$$\alpha = \frac{S_1 + S_4 - (S_2 + S_3)}{S_1 + S_2 + S_3 + S_4} \tag{21}$$

It should be noted that when $\bar{w} \neq 0$, $\overline{|w'|c'}$ is an approximation for $\overline{|w|'c'}$, the latter can be found to be

$$\overline{|w|'c'} = \overline{|w' + \bar{w}|'c'} \tag{22}$$

The contributions of $\overline{|w|'c'}$ can be accommodated in this analysis by partitioning the contributions into six categories, the four quadrants and two additional bands for the contributions when w' falls between 0 and $\bar{w}$. However, the contribution from the additional bands is small and has little impact on the interpretation of this analysis.

Considering that the quadrants $S1$ and $S4$ represent the contribution of updrafts to the flux. Similarly, $S2$ and $S3$ represent the contribution of downdrafts to the flux. We define

$$\text{flux}^{\uparrow} = S1 + S4 \tag{23}$$

$$\text{flux}^{\downarrow} = S2 + S3 \tag{24}$$

Here, the arrows indicate the direction of the wind and not the sign of the flux. e.g., $\text{flux}^{\uparrow}$ is the portion of the flux transported with updrafts which can be either positive or negative flux. If follows that α_c can be written as

$$\alpha_c = \frac{\text{flux}^{\uparrow} - \text{flux}^{\downarrow}}{\text{flux}^{\uparrow} + \text{flux}^{\downarrow}} \tag{25}$$

The previous equation indicates that if the flux transported with updrafts, $\text{flux}^{\uparrow}$ have the same sign as the flux transported with downdrafts, $\text{flux}^{\downarrow}$, then the value of $|\alpha_c|$ will be smaller than one. If $|\alpha_c| > 1$ then $\text{flux}^{\uparrow}$ and $\text{flux}^{\downarrow}$ have opposing signs which indicates that the wind and the scalar are correlated for updrafts and anti-correlated for downdrafts or vice versa, indicating non-stationary conditions.

2.5.2 Analytical value of α_c

An analytical expression for the value of α can be obtained from the knowledge of the joint probability distribution of the vertical wind velocity and the scalar. If the wind and the scalar are assumed to follow a Gaussian joint probability density function, we find the analytical value of α_c in terms of the moments of the joint probability density function to be

$$\alpha_c = \mathrm{erf}\left(\frac{\bar{w}}{\sqrt{2}\,\sigma_w}\right) \tag{26}$$

Where erf is the error function. σ_w is the wind standard deviation.

We can use the analytical value of α_c and further substitute the expected value of $|w|$ with the mean of the folded normal distribution (Leone et al., 1961) to obtain an analytical expression for the expectation of the flux error due to a nonzero vertical wind velocity using $\bar{w}$ and σ_w. By substituting into the last term of Eq. (14), $\bar{w}/\overline{|w|}\alpha_c$

$$F_{\mathrm{err}} = \overline{w}\sqrt{\pi}\left(\sqrt{2}\sigma_w \mathrm{e}^{-\frac{(\overline{w})^2}{2\sigma_w^2}} + \mathrm{erf}\left(\frac{\overline{w}}{\sqrt{2}\,\sigma_w}\right)\overline{w}\sqrt{\pi}\right)^{-1}\alpha_c \tag{27}$$

Here, F_{err} is the error in the TEA flux when failing to account for the correlation between the scalar and the magnitude of vertical wind velocity.

2.5.3 Scalar similarity to estimate α_c

The assumption of scalar similarity provides a potential empirical way to estimate the value of α_c, i.e., by calculating the value of α_c from another scalar where high-frequency measurements are available, e.g. sonic temperature. The assumption of scalar similarity is supported by experimental evidence that has shown that different scalars behave similarly due to a similar transfer mechanism (Ohtaki, 1985; Wesely, 1988). However, the assumption of scalar similarity can not be always guaranteed and should be used with caution. Nonetheless, we believe it is a useful assumption to approximate the value of α_c given that the value of α_c is determined by the distribution of turbulent transport in different quadrants which is expected to have the same effect on different scalars under good mixing conditions.

3 Methods

3.1 Numerical simulations

We set up a numerical simulation to test the magnitude of the error due to nonzero $\bar{w}$ on the flux and investigate the values of the coefficient α_c. For this simulation, we used 10 Hz measurements obtained from a field experiment measuring vertical wind velocity and scalar concentration using an infrared gas analyzer (IRGA) and a sonic anemometer. We used data from a period of 12 days from 15 June 2020 to 26 June 2020. The data were collected at an ideal flat agricultural site in Braunschweig, Germany. A full description of the site and the instrumentation are provided in the accompanying paper (Emad and Siebicke, 2022b). We added a random $\bar{w}$ offset in the range (-0.25 to $0.25\,\mathrm{m\,s^{-1}}$) to each averaging interval but limited $\bar{w}$ to smaller than $2\sigma_w$. We obtained 3 repetitions and calculated the flux according to different formulas. In total, there were about 1400 30-minute averaging intervals. The methods compared were: i) the flux calculated using the concentrations formula of Hicks and McMillen (1984) shown in Eq. (8), ii) the equation for DEA including the non equal volume correction of Turnipseed et al.

(2009), and iii) the new generalized equation proposed in the current study (Eq. 14) utilizing α_θ values calculated from sonic temperature and the analytical value of α_c.

We applied minimal quality checks on the resulting fluxes before the comparison. Tests for stationarity following Foken et al. (2005) removed 22% of the averaging intervals. We limited the values of $|\alpha_\theta|$ to less than 1, which removed an additional 4% of the averaging intervals. Furthermore, when the sonic temperature was used for calculating α_θ, periods with low turbulence intensity, $|\rho_{w\theta}| < 0.2$ where excluded. The excluded averaging intervals occurred almost exclusively during night-time conditions.

4   Results and Discussion

We first discuss the newly proposed TEA equation, then compare it to different TEA formulations. Then, we discuss the interpretation of the transport asymmetry coefficient α and different ways of estimating it.

4.1   Nonzero mean vertical wind velocity

The newly proposed TEA equation (Eq. (14)) successfully constrained the biased advective term $\bar{w}\bar{c}$. The new equation employs information about the scalar transport to allow the estimation of $\bar{c}$ from available TEA measurements and, consequently get an estimate of the biased advective term. Besides the correction of the nonzero $\overline{w}$ bias, the estimation of the scalar mean, $\bar{c}$ is essential for the WPL correction and the calculation of storage fluxes.

The terms of Eq. 14 account for different contributions to the flux. The first term on the right-hand side is equivalent to calculating the flux as the difference in accumulated mass between updraft and downdraft. When $\bar{w} = 0$, the equation is reduced to this term only. The second term accounts for the bias introduced by the biased advective term $\bar{w}\bar{c}$ by using the weighted mean of the scalar and the magnitude of wind as an estimate for $\bar{c}$. We show that when $\bar{w} \neq 0$, the first two terms are equivalent to using the concentration formula of Hicks and McMillen (1984) shown in Eq. (8), with the unequal volume correction of Turnipseed et al. (2009) that accounts for the small difference between the weighted mean $\bar{c}_{\mathrm{W}}$ and average of concentrations $(C_{acc}^{\uparrow} + C_{acc}^{\downarrow})/2$. Refer to appendix A for details about this equality. The new third term $\overline{c'|w|'}/\overline{|w|}$ corrects for the correlation between the scalar and the magnitude of the wind. Ignoring the third term will result in a flux biased with the ratio $\bar{w}/\overline{|w|}\alpha_c$.

The new TEA equation reveals an important insight. When using the new equation to calculate the flux, the error in the flux when $\overline{w} \neq 0$ is independent of the scalar concentration and is governed by the characteristics of the turbulent transport. This strengthens the confidence in using the TEA method for measuring atmospheric constituents with high background concentration and small flux (low deposition velocity).

Using the new TEA formula with an estimated value for α_c was effective in reducing the uncertainty and the systematic error in the calculated fluxes (Fig. 1). To quantify the magnitude of the systematic bias and uncertainty resulting from nonzero $\bar{w}$ on the fluxes, we used the simulation results to obtain the slope and the coefficient of determination, R^2 from a linear fit of
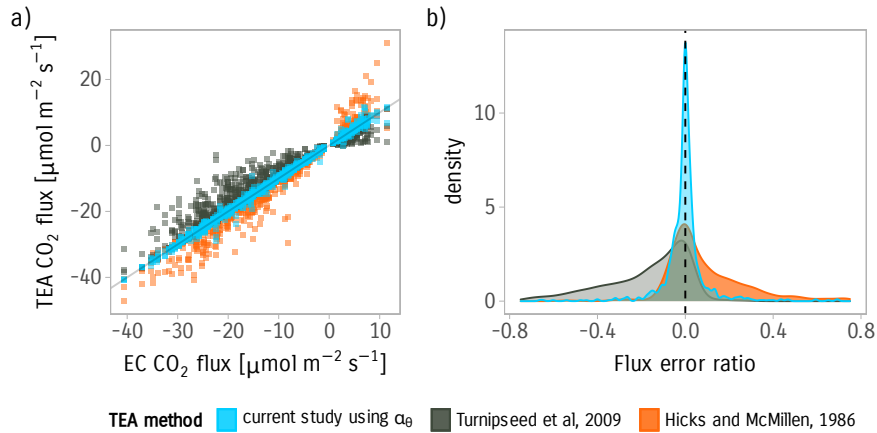
Figure 1. a) Comparison of different equations of TEA flux calculation against a reference EC flux. Data were obtained from high frequency measurements over 12 days from 15 June 2020 to 26 June 2020 and included an added random $\bar{w}$ offset in the range (-0.25 to 0.25 $\mathrm{m\,s^{-1}}$)). Colors represent different formulas. b) Kernel density estimates of the flux error ratio using the three different formulas.

the calculated fluxes against the reference EC flux. The simulation results show an increased bias and uncertainty in the fluxes when $\bar{w} \neq 0$ and a significant improvement when using an estimate of α for correction (Fig. 1).

265   We found that using the accumulated mass difference to calculate the TEA flux (first term in Eq. 14) produced the largest errors. Values of $\bar{w}$ as small as $0.01\,\sigma_w$ was sufficient to produce more than 10% mean bias in the flux magnitude for CO_2 in our dataset.

     The use of the concentration TEA equation of Hicks and McMillen (1984) was a considerable improvement over using the mass difference but still overestimated the TEA flux. The slope of the linear fit was 1.12 ($R^2 = 0.94$). Using the DEA

270 equation of Turnipseed et al. (2009) which includes an additional term to correct for the effect of unequal volume on the flux, led to underestimating the TEA flux yielding a slope of 0.84 ($R^2 = 0.93$). The correction of nonzero $\bar{w}$ using α_θ utilizing the assumption of scalar similarity significantly reduced the bias and the uncertainty and gave slope of 1.005 ($R^2 = 0.995$). The use of the analytical value of α_c using Eq. 26 similarly reduced the bias but with a smaller reduction in uncertainty, yielding a slope of 0.991 ($R^2 = 0.97$). The higher uncertainty when using the analytical value of α_c is likely due to the deviation from

275 the assumed Gaussian probability distribution.

     These results indicate that the proposed corrections using an estimate of α_c are very effective in minimizing or removing the bias from TEA flux when $\bar{w} \neq 0$ even when using the analytical value of α_c.

4.2   Value and interpretation of the transport asymmetry coefficient α

The value of α_c defined in Eq. (12) indicates the disparity of the flux transport between updrafts and downdrafts. Values of α_c

280 larger than 1 indicate that updraft flux ($S_1 + S_4$) and downdraft flux ($S_2 + S_3$) have opposing signs. This pattern indicates that
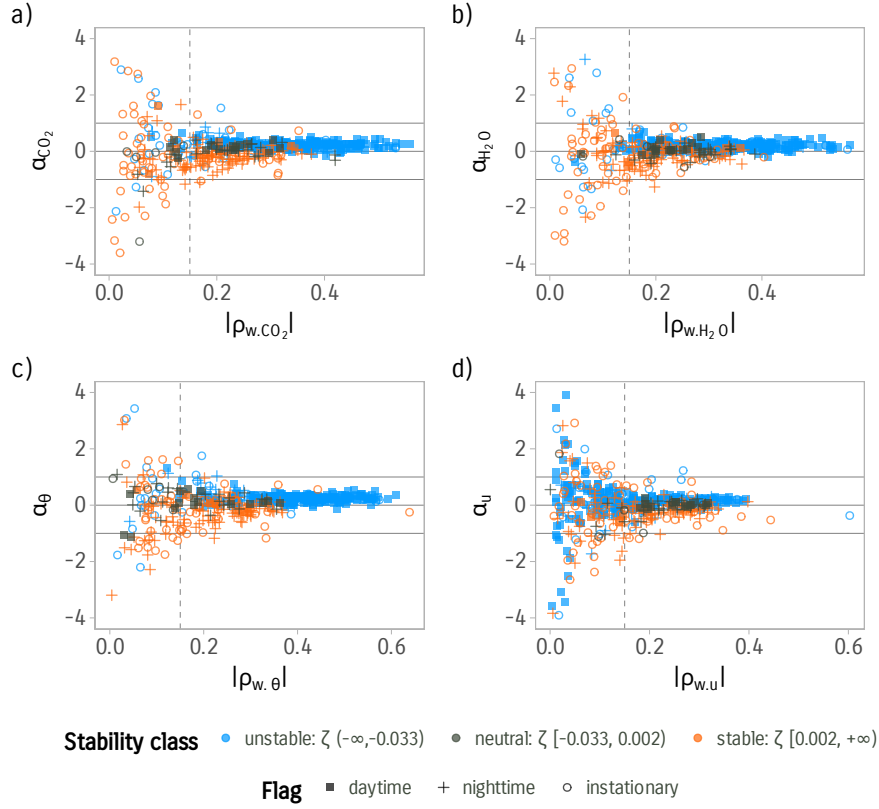
Figure 2. Observed values of the transport asymmetry coefficient α vs. the correlation coefficient for four variables calculated from hight-frequency measurements. a) CO_2, b) H_2O, c) sonic temperature, θ, and d) wind velocity component, u. Colors distinguish different stability classes. Point shape differentiates daytime, nighttime, and instationary conditions following Foken and Wichura (1996). Vertical dashed line is set to $x = 0.15$ and the horizontal lines are set to $y = -1$ and $y = 1$.

the wind and the scalar are correlated for updraft flux and anti-correlated for the downdraft flux (or the other way around). This pattern violates the stationarity conditions.

Therefore, we conclude that for stationary flows the systematic error in the TEA flux is smaller than $\pm \bar{w}/\overline{|w|}$. Observed values of α for H_2O, CO_2, θ, and the wind component, u are shown in Fig. 2. The data confirm that values of $|\alpha|$ are

285 consistently below 1 for the four scalars when the stationarity criterion is met. However, when the correlation between w and the scalar is low during conditions associated with low developed turbulence, spurious correlations might lead to values of $|\alpha|$ larger than one.

We found that the value of α for CO_2 correlates moderately with the skewness of the measured scalar ($r = 0.61$, data not shown). The observed mean of α for CO_2 and sonic temperature calculated from high-frequency measurements for periods

290 with negligible $\bar{w}$ was approximately 0.2 for unstable and good turbulent mixing conditions ($|\rho_{wc}| > 0.25$) with a standard error, $SE = 0.01$. For stable stratification ($\zeta > 0$), the mean of α was approximately equal to -0.18 but with a higher spread

around the mean, $SE = 0.09$. These values indicate that updrafts have a larger contribution to the flux under unstable stratification and smaller contribution during stable stratification. The results generally agree with values found from studies using conditional sampling (Greenhut and Khalsa, 1982) and LES simulations (Wyngaard and Moeng, 1992) which found that updrafts contribution to the flux is 2 to 3 times larger than downdrafts under unstable conditions due to the contribution of convective thermals.

The sign of α indicates whether updrafts or downdrafts have a larger contribution to the flux. Inspecting Eq. 25, we find that a positive α indicate that the magnitude of updrafts contribution to the flux is larger than the magnitude of the downdrafts contribution, $|\text{flux}^{\uparrow}| > |\text{flux}^{\downarrow}|$ while the opposite is true for a negative α.

The analytical value of α from Eq. (26) was effective in minimizing the systematic bias as confirmed by the simulation results. However, the assumption of a Gaussian distribution, although used in the literature, e.g., (Wyngaard and Moeng, 1992), is not adequate. While the wind might be normally distributed for most stability classes (Chu et al., 1996), the scalar can depart significantly from normality (Berg and Stull, 2004). Other distributions might be more suited for approximating the joint probability distribution (Frenkiel and Klebanoff, 1973). For example, Katsouvas et al. (2007), using experimental data, showed that a third-order Gram–Charlier distribution was necessary and sufficient in most of the cases for describing the quadrant time and flux contributions. It is worth considering this distribution to find a better analytical formula to calculate the expectation of α.
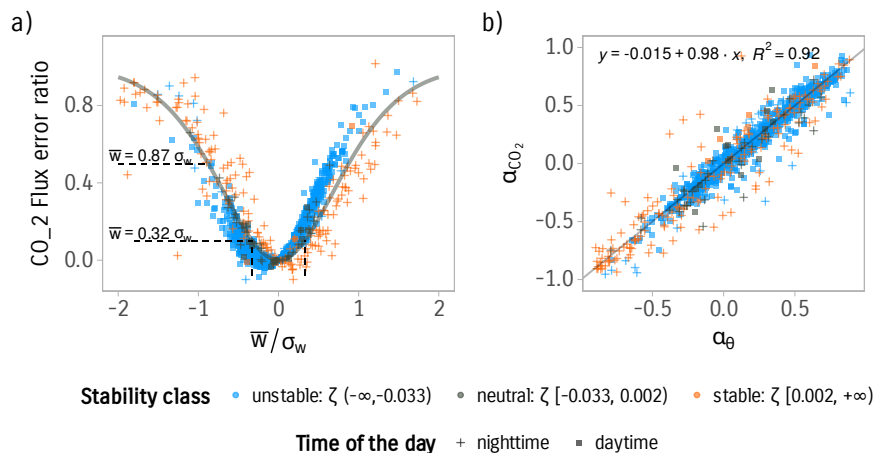
Figure 3. a) The error ratio in the CO_2 flux calculated using the TEA method due to nonzero mean vertical wind. The solid gray line represents the analytical values of the error in the flux (if a joint Gaussian probability distribution is assumed). The points are the observed error calculated from high-frequency measurements colored according to stability classes. The point shape distinguishes day-time and night-time data. b) Relation of α_{CO_2} calculated for CO_2 and α_{θ} calculated from sonic temperature and a 1-to-1 line for reference.

The hypothesis of scalar similarity was proposed as another source for estimating the values of α. The similarity was confirmed empirically by investigating the values of α_{θ} and α_c from high-frequency measurements (Fig. 3). A linear fit with

310 a slope of 0.98 and R^2 of 0.92 was obtained during steady-state and well-developed turbulence conditions. During such conditions, α_θ can substitute α_c to calculate the flux correction ratio. However, the correction becomes large and unreliable in periods where σ_w and ρ_{cw} are small, associated with small fluxes during night-time and stable conditions. Additionally, temperature is considered a poor proxy during near-neutral conditions due to its contribution to buoyancy (McBean, 1973; Hicks et al., 1980). We noticed that the variance in α values is higher under weakly developed turbulence and experimentally

315 determined the threshold for the optimum use of α_θ for the correction as $|\rho_{cw}| = 0.2$. Below this threshold, values of α_θ larger than 1 are observed, making the correction unreliable. This threshold can be seen as an indicator for the violation of assumptions of homogeneity and stationarity or other problematic conditions. Similar uses for the correlation coefficient are common in the literature e.g. (Foken and Wichura, 1996).

Another use of the formulation using α is to find a $\bar{w}$ threshold, above which, the TEA flux measurement becomes unreliable.

320 For example, if we define that the bias in the flux should not exceed 10% of the flux, we can find experimentally that the error in the flux due to nonzero $\bar{w}$ becomes larger than 10% when $\bar{w}$ exceeds $0.21\sigma_w$ for periods with good turbulent mixing conditions ($|\rho_{w,\mathrm{CO_2}}| > 0.2$). This threshold is close to the analytical value of $0.323\ \sigma_w$ obtained from the Gaussian joint probability distribution. To push this threshold further, α_θ calculated from sonic temperature can be used during good turbulent mixing conditions ($|\rho_{w,\mathrm{CO_2}}| > 0.2$). Simulations indicate that the average relative confidence interval for the predicted value of α_θ

325 from $\alpha_{\mathrm{CO_2}}$ is 0.17% of the fit value. In summary for this example, to keep the error in the flux below 10%, α_θ can be safely used to correct for biased $\bar{w}$ as long as $\bar{w} < 0.7\sigma_w$. This limit is considered forgiving and easy to achieve with online coordinates rotation and further rather simple online treatments. The only times where this limit is expected to be reached is when σ_w is very small (e.g., during night-time conditions) where other problems such as low turbulent mixing and violations of the assumptions of the EC and TEA methods are expected to occur. These periods largely overlap with periods considered

330 of low quality and are usually excluded from the analysis (Foken et al., 2012b).

To summarize, we find that the error in the TEA flux is constrained to $\bar{w}/\overline{|w|}$ for $|a| < 1$, which was shown here to be true for stationary conditions, which are at the focus of turbulent flux measurements. If a correction is desired to minimize this error, two options were presented to estimate α_c: first, an analytical solution, and second, an estimate employing scalar similarity. Finally, with the use of α_c, the typically observed systematic flux bias due to nonzero mean vertical wind velocity could be

335 effectively characterized and minimized.

5   Conclusions

In this paper, we revised the theory of the true eddy accumulation method and extended its applicability to measure turbulent fluxes under non-ideal conditions where the mean vertical wind velocity during the averaging interval is not zero. The new generalized equation allows estimating the scalar mean during the flux averaging interval and defining conditions where the

340 error in the flux is significant.

The new formulation allowed to constrain the relative systematic error in the TEA flux to the ratio $\bar{w}/\overline{|w|}$ under stationarity conditions. This systematic error was reduced to be a function of the disparity of atmospheric transport instead of having it

scale with the scalar background concentration. This development significantly reduces the systematic bias in TEA fluxes under non-ideal conditions allowing the TEA method to be used indifferently with various atmospheric constituents.

345   The coefficient α_c, defined to quantify the atmospheric transport asymmetry, has proved to be very useful in estimating and removing the error in measured TEA fluxes. We showed two methods for estimating α_c to reduce the flux systematic bias: i) an estimate of α_c based on the assumption of flux variance similarity, and ii) an analytical expression based on the assumption of a Gaussian joint probability distribution of the scalar concentration and vertical wind velocity. Both of these estimation methods were shown to be effective in minimizing the systematic error in the flux when compared to conventional TEA formulas.

350   In conclusion, the results presented in this paper showed that it is possible to achieve minimum bias in the TEA flux under most atmospheric conditions as well as identify those conditions which are less favorable. We believe that these results increase the confidence in using the TEA method for different atmospheric constituents and under a variety of atmospheric conditions.

Code and data availability. Data and scripts needed for producing the results and the figures presented in this paper are provided at (Emad and Siebicke, 2022a).

355 **Appendix A: Hicks and McMillen formulation**

We show here how the TEA flux formula of Hicks and McMillen (1984), originally formulated under the assumption of $\bar{w} = 0$ is equivalent to using $(C_{acc}^\uparrow + C_{acc}^\downarrow)/2$ as an estimate for $\bar{c}$ in the second term on the right hand side of Eq. (2).

We write the conditional expectation of $\bar{w}$ as

$$\overline{w} = \overline{\left(\overline{w|\mathrm{sign}(w)} \right)} = \overline{|w^\uparrow|}\, \mathrm{P}(w^\uparrow) - \overline{|w^\downarrow|}\, \mathrm{P}(w^\downarrow) \tag{A1}$$

360   where $\mathrm{sign}(w)$ is the sign of vertical wind velocity. $\mathrm{P}(w^\uparrow)$ and $\mathrm{P}(w^\downarrow)$ are the observed probabilities of the sign of w, which equals the ratio of the time the wind is positive or negative to the total integration interval time.

$$\overline{w} = \overline{|w^\uparrow|}\frac{\Delta t^\uparrow}{\Delta t} - \overline{|w^\downarrow|}\frac{\Delta t^\downarrow}{\Delta t} \tag{A2}$$

and similarly

$$\overline{|w|} = \overline{|w^\uparrow|}\frac{\Delta t^\uparrow}{\Delta t} + \overline{|w^\downarrow|}\frac{\Delta t^\downarrow}{\Delta t} \tag{A3}$$

365   by substituting $\overline{|w|}/2$ with

$$\frac{\overline{|w|}}{2} = \overline{|w^\uparrow|}\frac{\Delta t^\uparrow}{\Delta t} - \frac{\bar{w}}{2} = \overline{|w^\downarrow|}\frac{\Delta t^\downarrow}{\Delta t} + \frac{\bar{w}}{2} \tag{A4}$$

we obtain

$$C_{acc}^{\uparrow}(\overline{w^{\uparrow}}\,\frac{\Delta t^{\uparrow}}{\Delta t} - \frac{\bar{w}}{2}) - C_{acc}^{\downarrow}(\overline{|w^{\downarrow}|}\,\frac{\Delta t^{\downarrow}}{\Delta t} + \frac{\bar{w}}{2}) \tag{A5}$$

After rearrangement and simplification we get to

370 $$F_{\text{EA}} = \overline{cw} - \bar{w}\left(\frac{C_{acc}^{+} + C_{acc}^{-}}{2}\right) \tag{A6}$$

When Eq. (A6) is compared with Eq. (2), it is clear that the term $\frac{C_{acc}^{+}+C_{acc}^{-}}{2}$ is used as an estimate for $\bar{c}$.

Appendix B: List of symbols

Table B1. Symbols and subscripts with units

Symbols

c	$\mathrm{mol\,m^{-3}}$	Molar density of a scalar
w	$\mathrm{m\,s^{-1}}$	Vertical wind velocity
Δt	s	Flux averaging interval
A	–	TEA sampling scaling factor
V	$\mathrm{m^3}$	Volume
C	$\mathrm{mol\,m^{-3}}$	Mean concentration of accumulated samples
α_c	–	Transport asymmetry coefficient for scalar c
ρ	–	Correlation coefficient
S_i	–	Flux contribution from quadrant i

Subscripts

acc	Accumulated samples
$\uparrow$	Updraft buffer volume
$\downarrow$	Downdraft buffer volume
c	Atmospheric constituent

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