

An analysis and implications of alternative methods of deriving the density (WPL) terms for eddy covariance flux measurements

W. J. Massman · J.-P. Tuovinen

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Abstract We explore some of the underlying assumptions used to derive the density or WPL terms (Webb et al. (1980) *Quart J Roy Meteorol Soc* 106:85–100) required for estimating the surface exchange fluxes by eddy covariance. As part of this effort we recast the origin of the density terms as an assumption regarding the density fluctuations rather than as a (dry air) flux assumption. This new approach, which is similar to the expansion/compression approach of Liu (Boundary-Layer Meteorol 115:151–168, 2005), eliminates the dry-air mean advective vertical velocity from the development of the WPL terms and allows us to directly compare Liu's assumptions for deriving the WPL terms with the analogous assumptions appropriate to the original expression of the WPL terms. We suggest, (i) that the main difference between these two approaches lies in the interpretation of the turbulent exchange flux, and (ii) that the original WPL formulation is the more appropriate approach. Given the importance of the WPL terms to accurate and reliable measurements of surface exchange fluxes, a careful analysis of their origins and their proper mathematical expression and interpretation is warranted.

Keywords Air parcel expansion/compression · Mass conservation equations · Trace gas fluxes

1 Introduction

Recently, Liu (2005) proposed an alternative derivation of the density terms necessary for estimating atmospheric trace gas exchange fluxes using eddy covariance that

W. J. Massman (✉)

USDA Forest Service, Rocky Mountain Research Station, 240 West Prospect, Fort Collins, CO 80526 USA

e-mail: wmassman@fs.fed.us

J.-P. Tuovinen

Finnish Meteorological Institute, Climate and Global Change Research, P.O. Box 503, FI-00101 Helsinki, Finland

differs from that originally proposed by Webb et al. (1980). This alternative approach, which is based on air parcel expansion/compression concepts, produced nighttime CO_2 exchange fluxes at a forested ecosystem that were about 4% less than the equivalent WPL-corrected fluxes and increased the daytime CO_2 exchange fluxes at the same site by about 20% (Liu 2005). Given that these density terms are absolutely critical to accurate and reliable measurements of eddy covariance fluxes, and therefore to issues involving atmospheric chemistry, the global network of eddy covariance systems (FLUXNET: Baldocchi et al. 2001), and carbon cycle research, it is essential to understand and resolve the differences between Liu's (2005) result and Webb et al.s (1980), henceforth denoted by WPL. This study examines these two approaches with the intent of determining which method is preferred. However, to be as clear as possible during this comparison it is beneficial to view the WPL terms from a perspective based on density fluctuations rather than employing assumptions on the dry air flux. In keeping with our desire for clarity, we will also express all relevant hypotheses and relationships in terms of density and density fluctuations, rather than rely on volume-based formulations (e.g., Liu 2005).

2 Original WPL approach to the density terms

The key elements of the approach taken by WPL are (a) the ideal gas law and Dalton's law of partial pressures, and (b) the assumption of zero total dry air flux. [We do not include Reynolds's averaging in this discussion because it is common to all approaches when deriving the density terms.] The original WPL derivation will not be repeated here, however, it can be found under various guises in WPL, Paw U et al. (2000), Fuehrer and Friehe (2002), Massman and Lee (2002), and Leuning (2004). WPL's basic results can be summarized in the following two equations:

$$\rho'_d = -\bar{\rho}_d(1 + \bar{\chi}_v) \left(\frac{T'}{\bar{T}} - \frac{p'}{\bar{p}} \right) - \mu_v \rho'_v, \quad (1)$$

$$\overline{w' \rho'_{t,S}} = \overline{w' \rho'_t} - \overline{\bar{w}_t w' \rho'_d}, \quad (2)$$

where (henceforth) any unsubscripted variable will refer to the ambient atmosphere, e.g., ρ refers to mass density [kg m^{-3}] of the ambient (moist) air, and a subscripted variable will refer to a specific component of the atmosphere, e.g., the subscript d refers to dry air, the v subscript refers to water vapour, the t subscript refers to a generic trace gas and as such can mean water vapour, CO_2 (which would be specified by a c subscript), etc.; Reynolds decomposition is denoted in the usual manner [i.e., $x = x' + \bar{x}$ when referring to the fluctuating (x') and mean ($\bar{x}$) of the quantity x]; T is temperature [K]; p is pressure [Pa]; μ_v is the ratio of the molecular mass of dry air ($m_d = 28.966 \text{ g mol}^{-1}$) to molecular mass of water vapour ($m_v = 18.016 \text{ g mol}^{-1}$); $\bar{\chi}_v = \mu_v \bar{\rho}_v / \bar{\rho}_d$ is the mean mole fraction (or mean molar mixing ratio) of water vapour relative to dry air [mol mol^{-1} -dry air]; $\bar{w}_t = \bar{\rho}_t / \bar{\rho}_d$ is the mean mass fraction (or mean mass mixing ratio) of trace gas relative to dry air [kg kg^{-1} -dry air]; $\overline{w' \rho'_{t,S}}$ is the source/sink-related turbulent (mass) exchange flux between the atmosphere and the underlying surface; $\overline{w' \rho'_t}$ is the covariance between the fluctuating vertical velocity, w' [m s^{-1}], measured with a sonic anemometer and the fluctuating mass density measured with a fast response scalar sensor; and the turbulent dry air flux, $-\overline{\bar{w}_t w' \rho'_d}$,

encapsulates the WPL or density terms associated with heat and water vapour fluxes. Note here that (a) we include the pressure term, $p'/\bar{p}$, which WPL did not, in keeping with the findings of Massman and Lee (2002) and Fuehrer and Friehe (2002) that this term may be important for some conditions, and (b) we are not discussing any issues involving spectral attenuation of measured fluctuations or associated covariances (e.g., Massman 2004).

3 An alternative perspective on the density terms

In this section we propose an alternative approach for obtaining Eq. (2) that does not require assuming that the total dry air flux is zero and using a dry air mean advective vertical velocity, i.e., $\bar{w} = -\bar{w}'\rho'_d/\bar{\rho}_d$.

We begin with the ideal gas law, $p = nRT$, where n is the molar density [mol m^{-3}] of ambient air and $R = 8.314 \text{ J mol}^{-1} \text{ K}^{-1}$ is the universal gas constant. Next, employing Reynolds averaging and solving for n' yields

$$n' = -\bar{n}(T'/\bar{T} - p'/\bar{p}). \quad (3)$$

This equation does not include the second-order term, proportional to $n'T'$, because it is assumed to be negligible when compared to the other terms. Next expanding n to $n_d + n_v$, isolating n'_d on the left-hand side, and dividing both sides of the resulting relationship by $\bar{n}_d$ yields

$$\frac{n'_d}{\bar{n}_d} = -(1 + \bar{\chi}_v) \left(\frac{T'}{\bar{T}} - \frac{p'}{\bar{p}} \right) - \frac{n'_v}{\bar{n}_d}. \quad (4)$$

Note that at this point of the derivation Eq. (4) is simply a restatement of (or is in essence identical to) Eq. (1) above. Therefore, Eq. (4) contains no more information than Eq. (1); but the derivation of Eq. (4) is simpler than that used in previous studies to arrive at Eq. (1).

Now we hypothesize that in situ (Eulerian) measurements of trace gas density fluctuations, n'_t or equivalently ρ'_t , are composed of two components, one related to atmospheric expansion/compression, $n'_{t,\text{exp}}$ or $\rho'_{t,\text{exp}}$, and another associated with the source/sink-related turbulent exchange fluxes, $n'_{t,S}$ or $\rho'_{t,S}$. Therefore, $n'_t = n'_{t,\text{exp}} + n'_{t,S}$. The original WPL approach can now be recast by assuming that

$$n'_{t,\text{exp}}/\bar{n}_t = n'_d/\bar{n}_d. \quad (5)$$

In terms of mass density this yields

$$\rho'_{t,\text{exp}} = -\bar{\rho}_t(1 + \bar{\chi}_v) \left(\frac{T'}{\bar{T}} - \frac{p'}{\bar{p}} \right) - \bar{\omega}_t \mu_v \rho'_v. \quad (6)$$

Consequently, $\rho'_{t,\text{exp}}$ can be identified as the WPL density term expressed in terms of mass density fluctuations, rather than in terms of a mean advective velocity, and the expansion/compression assumption can be written as $\rho'_{t,\text{exp}} = \bar{\omega}_t \rho'_d$. Subtracting the expansion/compression component $\rho'_{t,\text{exp}}$ from the measured density fluctuation, ρ'_t , yields the density fluctuation associated with the turbulent exchange flux, $\rho'_{t,S}$, given as

$$\rho'_{t,S} = \rho'_t - \bar{\omega}_t \rho'_d. \quad (7)$$

The above description suggests that the expansion/compression approach is consistent with the original formulation of the WPL terms. It does have the advantage of being relatively straightforward. It also provides an alternative to the original approach, which relies on a (potentially unnecessary) mean advective vertical velocity (Paw U et al. 2000; Massman and Lee 2002) and its antecedent assumption that $\overline{w\rho_d} = 0$, which has yet to be justified on a physically rigorous basis. On the other hand, the expansion/compression approach [i.e., Eq. (5)], although useful, does not provide a physically rigorous explanation of the origin of the density terms either. As a result, we assert that the expansion/compression approach neither confirms nor invalidates the assumption that the total dry air flux is zero. Rather it is simply a different way of looking at the physical processes involved in the origin of the density terms.

Liu (2005), using a volume-based version of the expansion/compression analogy, concluded that

$$\rho'_{t,\text{exp}} = -\frac{\bar{\rho}_t}{\bar{\rho}} \left[\bar{\rho}_d(1 + \bar{\chi}_v) \left(\frac{T'}{\bar{T}} - \frac{p'}{\bar{p}} \right) + (\mu_v - 1)\rho'_v \right], \quad (8)$$

which is clearly not the same as Eq. (6). However, Liu expressed the expansion/compression hypothesis in terms of moist air rather than dry air, i.e., he assumed

$$\frac{\rho'_{t,\text{exp}}}{\bar{\rho}_t} = \frac{\rho'}{\bar{\rho}}. \quad (9)$$

Here we note Liu's (2005) expression for $\rho'/\bar{\rho}$ for Eq. (9) can be obtained from a simple set of manipulations performed on the expression for n' , i.e., Eq. (3), which is the same starting point for Eq. (4). First (as before), isolate n'_d on the left-hand side of the expression by subtracting n'_v from both sides of the expression for n' , convert to mass density by multiplying both sides by m_d , add ρ'_v to both sides, which results in an expression for fluctuating moist air density, ρ' , on the left-hand side; next divide both sides by $\bar{\rho}$, which yields

$$\frac{\rho'}{\bar{\rho}} = -\frac{1}{\bar{\rho}} \left[\bar{\rho}_d(1 + \bar{\chi}_v) \left(\frac{T'}{\bar{T}} - \frac{p'}{\bar{p}} \right) + (\mu_v - 1)\rho'_v \right]. \quad (10)$$

Liu (2005) formally identifies the expansion/compression hypothesis using the concept of an air parcel and a volume-based approach obtained with $\rho'/\bar{\rho} = -V'/\bar{V}$, where V is the instantaneous volume of the air parcel undergoing expansion or compression. Comparing the basic assumption made by Liu (2005), as expressed with Eq. (9), with that given by Eq. (5) for WPL indicates a major difference between these two approaches. WPL expresses relative change in an air parcel undergoing expansion or compression in terms of molar density ($n'_d/\bar{n}_d$), whereas Liu (2005) expresses it in terms of mass density ($\rho'/\bar{\rho}$). For dry air there is no distinction between molar and mass density; however, for moist air there is because $n'/\bar{n} \neq \rho'/\bar{\rho}$ due to the presence of water vapour. Note that this last assertion follows directly from the equation of state for ambient moist atmosphere (e.g., Jacobson 1999, pp. 19–25), which can be used to show that the molecular mass of ambient moist air, m , can be expressed as $m = m_d(1 + \omega_v)/(1 + \mu_v\omega_v)$. This last relationship clearly indicates that any Reynolds decomposition relating n' and ρ' must also include terms associated with either $\omega_v = \bar{\omega}_v + \omega'_v$ or, equivalently, $m = \bar{m} + m'$. The consequence of this simple difference between Liu's (2005) approach and the original WPL is subtle, as discussed next, and relates to how one interprets the turbulent exchange flux.

Summarizing Liu (2005) for $\rho'_{t,S}$ yields $\rho'_{t,S} = \rho'_t - \bar{\omega}_{tm}\rho'_t$, where $\bar{\omega}_{tm} = \bar{\rho}_t/\bar{\rho}$ is the mass mixing ratio relative to moist air. However, now $\rho'_{t,S}$ must be interpreted carefully as it no longer refers to an individual trace gas, but rather to a mixture of the trace gas and water vapour. This assertion can be deduced from the term, $\bar{\omega}_{tm}\rho'_t$ by rewriting it as $\bar{\omega}_t(\rho'_d + \rho'_v)/(1 + \bar{\omega}_v)$. As long as $\bar{\omega}_v \neq 0$, then any mass flux corresponding to this term will be a mixture of the trace gas ($\bar{\omega}_t$) and water vapour ($\bar{\omega}_v$). This can be seen more explicitly by comparing Liu's expression for $w'\rho'_{t,S}$, given next, with the corresponding expression from WPL given by Eq. (2).

$$\overline{w'\rho'_{t,S}} = \overline{w'\rho'_t} - \frac{\bar{\omega}_t}{1 + \bar{\omega}_v}(\overline{w'\rho'_d} + \overline{w'\rho'_v}). \quad (11)$$

In the case of CO_2 , Liu (2005), by associating the flux term $\overline{w'\rho'_{t,S}}$ of Eq. (11) solely with CO_2 , fails to recognize the water vapour component and thereby introduces a systematic negative bias into the calculations for true CO_2 turbulent exchange flux. During the daytime, when $\overline{w'\rho'_{c,S}} < 0$ and $\overline{w'\rho'_v} > 0$, Eq. (11) results in a significant overestimation of the downward turbulent CO_2 exchange flux, while at night, when $\overline{w'\rho'_{c,S}} > 0$ and $\overline{w'\rho'_v} \approx 0$ the upward turbulent CO_2 exchange flux is slightly underestimated, precisely as Liu (2005) found.

In the event that the trace gas is water vapour, this misinterpretation results in an incorrect estimation of the turbulent water vapour exchange flux as can be seen by comparing the following two equations, where the first is the original WPL expression and the second is Liu's (2005) equivalent,

$$\overline{w'\rho'_{v,S}} = \overline{w'\rho'_v} - \bar{\omega}_v\overline{w'\rho'_d}, \quad (12)$$

$$\overline{w'\rho'_{v,S}} = (\overline{w'\rho'_v} - \bar{\omega}_v\overline{w'\rho'_d})/(1 + \bar{\omega}_v). \quad (13)$$

Except for the term $(1 + \bar{\omega}_v)$, these last two expressions are the same. Therefore, because $\bar{\omega}_v \ll 1$ is virtually always true, the result is a small underestimation of the turbulent water vapour exchange flux when using Liu's (2005) expression, again in agreement with Liu's (2005) results.

The next section reinforces our interpretation of $\overline{w'\rho'_{t,S}}$ by examining the equation of continuity for a trace gas and the relationship between the turbulent exchange flux and the trace gas source term.

4 Equation of continuity

By combining the equations of continuity for dry air and a trace gas, Paw U et al. (2000), Massman and Lee (2002), and Leuning (2004) essentially derive a 3-D version of the equation of continuity of the turbulent trace gas mass exchange flux. For this section we will specify the trace gas as CO_2 . Their results are summarized by

$$\begin{aligned} \frac{\partial \rho_c}{\partial t} - \bar{\omega}_c \frac{\partial \rho_d}{\partial t} + \nabla \bullet (\mathbf{v}'\rho'_c - \bar{\omega}_c \mathbf{v}'\rho'_d) \\ + (\mathbf{v}'\rho'_d + \mathbf{v}'\rho_d) \bullet \nabla \bar{\omega}_c = \left[1 + \bar{\omega}_c \left(\frac{m_{\text{O}_2}}{m_c} - 1 \right) \right] \bar{S}_c, \end{aligned} \quad (14)$$

where the boldface quantities are 3-D vectors, $\mathbf{V}$ is the mean wind velocity, $\mathbf{v}'$ is the velocity perturbation, ∇ is the gradient operator, t is time, m_{O_2} ($= 32.00 \text{ g mol}^{-1}$) is the molecular mass of O_2 , m_c ($= 44.01 \text{ g mol}^{-1}$) is the molecular mass of CO_2 , and $\bar{S}_c$ is the mean CO_2 source term [$\text{kg m}^{-2} \text{ s}^{-1}$]. Here we follow Massman and Lee (2002) by allowing for a source of dry air [$\bar{S}_d = (1 - m_{\text{O}_2}/m_c)\bar{S}_c$], which is modelled from the photosynthesis/respiration stoichiometric relationship between O_2 and CO_2 . Consequently, the source term, $\bar{S}_c - \bar{\omega}_c \bar{S}_d$, in Eq. (14) can be written as $[1 + \bar{\omega}_c(m_{\text{O}_2}/m_c - 1)]\bar{S}_c$. Equation (14) also expresses the storage (or time rate of change) term as a linear combination of two terms, rather than combine them into one term as is usually done, because we wish to emphasize that these conservation equations refer to mass conservation, which could be lost by expressing the storage term as $\bar{\rho}_d \partial \bar{\omega}_c / \partial t$. Note that this expression does not assume incompressibility ($\nabla \bullet \mathbf{V} = 0$) of the mean flow and that the traditional WPL terms are included in the dry air flux term, $-\bar{\omega}_c \nabla' \rho'_d$, which is the 3-D equivalent of the term used in previous sections.

In his appendix Liu (2005) used the same equation of continuity for CO_2 , but instead of the equation of mass conservation for dry air he used the equation of continuity for moist air. Following exactly the same mathematical manipulations as with Eq. (14), one arrives at

$$\frac{\partial \bar{\rho}_c}{\partial t} - \bar{\omega}_{\text{cm}} \frac{\partial \bar{\rho}}{\partial t} + \nabla \bullet (\nabla' \rho'_c - \bar{\omega}_{\text{cm}} \nabla' \rho') + (\nabla' \rho' + \nabla \bar{\rho}) \bullet \nabla \bar{\omega}_{\text{cm}} = \left[1 + \bar{\omega}_{\text{cm}} \left(\frac{m_{\text{O}_2}}{m_c} - 1 \right) \right] \bar{S}_c - \bar{\omega}_{\text{cm}} \bar{S}_v. \quad (15)$$

In the case of Eq. (14) the turbulent exchange flux is clearly CO_2 because the source term $[1 + \bar{\omega}_c(\frac{m_{\text{O}_2}}{m_c} - 1)]\bar{S}_c \approx \bar{S}_c$ so long as $\bar{\omega}_c \ll 1$. In the case of Eq. (15), however, the source term is a combination of CO_2 and water vapour, i.e., source term $\bar{S}_c - \bar{\omega}_{\text{cm}}(\bar{S}_d + \bar{S}_v) \approx \bar{S}_c - \bar{\omega}_{\text{cm}}\bar{S}_v = \bar{S}_c - [\bar{\omega}_c/(1 + \bar{\omega}_v)]\bar{S}_v$. For a hot dry surface, for which there is little evaporation and the ambient air is dry, then Eq. (15) is quite similar to Eq. (14) and the turbulent CO_2 exchange fluxes will be similar. Of course, this is the simplest and most obvious bounding scenario on Eq. (15). But consider the other bounding scenario, which could occur over a large body of water. In this situation the relative importance of the CO_2 and water vapour sources would be reversed. As a result the 3-D turbulent exchange flux, $\nabla' \rho'_c - \bar{\omega}_{\text{cm}} \nabla' \rho'$, is going to relate more strongly to water vapour flux than to CO_2 flux because the source term is dominated by the source term for water vapour, i.e., $-\bar{\omega}_{\text{cm}}\bar{S}_v > \bar{S}_c$. Consequently, the interpretation of turbulent exchange flux in Eq. (15) is as a combination of CO_2 and vapour fluxes, not as a pure CO_2 flux. These scenarios and the correct interpretation of the WPL terms and related fluxes clearly indicate that the preferred approach for formulating the turbulent trace gas exchange flux is in terms of Eq. (14), dry air density fluctuations, and the original WPL formulation because it refers solely to the trace gas flux and does not include a component that is related to the water vapour flux.

In the case of water vapour all that is necessary is to examine the source term of the equation of continuity for water vapour. In this case the source term is given as $\bar{S}_v - \bar{\omega}_{\text{vm}}(\bar{S}_d + \bar{S}_v) = (1 - \bar{\omega}_{\text{vm}})\bar{S}_v - \bar{\omega}_{\text{vm}}\bar{S}_d = \bar{S}_v/(1 + \bar{\omega}_v) - [\bar{\omega}_v/(1 + \bar{\omega}_v)][1 - m_{\text{O}_2}/m_c]\bar{S}_c$; where, in general, the strength of the dry air source term, $[\bar{\omega}_v/(1 + \bar{\omega}_v)][1 - m_{\text{O}_2}/m_c]\bar{S}_c$, is negligible compared with the water vapour source term, $\bar{S}_v/(1 + \bar{\omega}_v)$. Nonetheless, the term $(1 + \bar{\omega}_v)^{-1}$ is the same as that identified in Liu's (2005) expression for the water vapour flux, Eq. (13). Consequently, the turbulent exchange water vapour fluxes

are misrepresented by the numerical factor $(1 + \overline{\omega_v})^{-1}$ because they correspond to the source term $\overline{S_v}/(1 + \overline{\omega_v})$ rather than the unmodified source term, $\overline{S_v}$. Note that the storage (or time rate of change) terms and the advective terms in the conservation equations associated with the turbulent exchange fluxes for any trace gas, including water vapour, are also misrepresented in a manner similar to that we have just outlined above for the fluxes and the source terms.

5 Conclusions

This paper explores alternative methods of deriving the density or WPL terms required for estimating the turbulent exchange fluxes by eddy covariance. The expansion/compression approach proposes that the WPL terms result from the inherent turbulent expansion/compression of the atmosphere at the point of measurement (density condition), rather than as a result of a mean advective velocity (flux condition) as in the original formulation. Consequently, the expansion/compression approach cannot be used to either justify or invalidate WPL's original assumption that the total dry air flux is zero, because it is independent of the condition $\overline{w\rho_t} = 0$. Nonetheless, the expansion/compression model does produce the same mathematical expression for the density terms as originally derived by Webb et al. (1980). Thus the expansion/compression approach confirms the validity of formulating the WPL terms in terms of the dry air density fluctuations (as opposed to Liu's (2005) formulation), because to do otherwise is to misconstrue the meaning of the turbulent exchange flux.

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