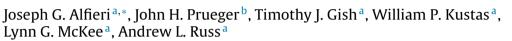
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The effective evaluation height for flux-gradient relationships and its application to herbicide fluxes



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ABSTRACT

Volatilization represents a significant loss pathway for many pesticides, herbicides and other agrochemicals. One common method for measuring the volatilization of agrochemicals is the flux-gradient method. Using this method, the chemical flux is estimated as the product of the vertical concentration gradient and a turbulent-transfer coefficient (eddy diffusivity). For computational simplicity, the evaluation height needed to calculate the eddy diffusivity is typically approximated as either the geometric or logarithmic mean. Both of these estimation methods are based on simplifying assumptions and can be a significant source of error, particularly when the separation distance between the measurement heights is large. Using data collected over an eight-year period at the USDA-ARS OPE3 experimental watershed, this study compared fluxes of metolachlor, a commonly-used herbicide, computed using the approximated evaluation heights with those calculated using the exact evaluation height. While it was found that the primary factor influencing the accuracy of the flux estimates using the approximate evaluation heights was atmospheric stability, errors in the estimate of the evaluation height can result in significant (>10%) errors in the flux-gradient technique.

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1. Introduction

Modern agricultural practices, including the use of agrochemicals such as herbicides and other pesticides, have greatly enhanced both the availability and quality of agricultural products during the last century. Looking forward, as the world's population continues to grow, agrochemicals will remain critical to meeting the increasing demand for food, fiber, and other agricultural commodities (Wheeler, 2002; Carvalho, 2006; Yates et al., 2011). At the same time, these chemicals have been associated with negative impacts on human health (Parron et al., 2011; Shirangi et al., 2011; Vinson et al., 2011), agricultural and native ecosystems (Schafer et al., 2007; Geiger et al., 2010), and the quality of water, air and soil resources (Harman et al., 2004; Hunt et al., 2006; Chopra et al., 2011; Rice et al., 2016). In order to minimize the adverse effects of

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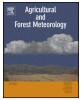
http://dx.doi.org/10.1016/j.agrformet.2016.10.010 0168-1923/Published by Elsevier B.V. agrochemical use, it is essential that their fate is accurately monitored after application.

Once applied, pesticides and other agrochemicals can either degrade *in-situ* or be transported from the field following any of three potential pathways: surface runoff, leaching into the ground water, and volatilizing into the atmosphere. The mode of transport depends on a number of factors including the method of application, field management practices, chemical characteristics of the compound, soil properties, and environmental conditions (Gish et al., 2009). A number of recent studies, however, indicate that the primary pathway for pesticide and herbicide loss is volatilization into the atmosphere (Prueger et al., 2005; Yates, 2006; Gish et al., 2009, 2011; Reichman et al., 2011). As pointed out by Bedos et al. (2002) and Yates (2009) among others, volatilization losses typically range between 5% and 25% of the amount applied, but they can be as great as 90% of the applied amount depending on the chemical characteristics of the compound applied and local environmental conditions.

One of the most prevalent and long-standing methods for determining pesticide fluxes is the flux-gradient approach (Glotfelty et al., 1984; Majewski et al., 1990; Majewski, 1999); the approach







is also commonly used to measure a number of other trace gases including ammonia, nitrous oxides, and methane (Laubach and Kelliher, 2004; Phillips et al., 2007; Harvey et al., 2008; Denmead, 2008; Xiao et al., 2014). The approach takes advantage of paired measurements of the chemical concentration at two different heights in conjunction with similarity theory to determine the flux; as the name suggests, similarity theory states that dimensionless vertical profiles of different scalar quantities, for example water or agrochemicals, can be described by the same universal function of height and atmospheric stability. More specifically, the chemical flux is calculated as the product of the mean concentration gradient and a proportionality parameter, often referred to as the eddy diffusivity, which accounts for the turbulent transport of the compound. As shown below, the accuracy of this approach is directly related to the accuracy of the estimates of the evaluation height, the height used to calculate the stability correction and eddy diffusivity.

For computational simplicity, the evaluation height is often estimated using the geometric or logarithmic mean assuming neutral atmospheric conditions. While it is only one of several potential sources of error associated with the flux-gradient approach [see Laubach and Kelliher (2004) and Mukherjee et al. (2014) for a further discussion of these] the error introduced by these approximations can propagate through to the flux estimates. Moreover, since, the magnitude of the error varies depending on the relative heights of the measurements and atmospheric stability, applications of the flux-gradient approach that require a large separation distance between the concentration measurements, such as measurements of pesticide volatilization, are particularly susceptible to errors due to inaccurate estimates of the evaluation height.

In an effort to characterize the uncertainty associated with the evaluation height and atmospheric stability, this study focuses on measurements of the herbicide metolachlor (2-chloro-N-(2ethyl-6-methyl-phenyl)-N-(1-methoxypropan-2-yl) acetamide) collected at the Optimizing Production for Economic and Environmental Enhancement (OPE3) experimental watershed near Beltsville, MD. Specifically, the impacts of both the stability correction and the method used to estimate the evaluation height on the metolachlor flux was evaluated by comparing flux calculated using the simplifying assumptions with the flux calculated using a stability correction and the exact evaluation height determined as discussed below. The following section provides further details regarding the theoretical background for determining the evaluation height. The third section provides an overview of the field site and measurements used herein while the fourth and fifth sections discuss the results and conclusions respectively.

2. Theoretical background

The flux-gradient approach, as it is applied to estimating herbicide fluxes, is typically expressed in terms of *K*-Theory to describe a flux analogously to Fick's law of molecular diffusion (Lumley and Panofsky, 1964; Stull, 1988). According to *K*-theory, the vertical transport of a conservative scalar quantity, such as water vapor, carbon dioxide, or herbicide vapor, within the surface boundary layer is driven solely by turbulent processes and is proportional to the concentration gradient of the scalar quantity. Using this conceptual framework, an herbicide flux can be expressed as

$$F_{\rm S} = -K_{\rm S} \,\partial S/\partial z \tag{1}$$

where F_S is the herbicide flux (ng m² s⁻¹), K_S is the eddy diffusivity (m² s⁻¹), and $\partial S/\partial z$ represents the vertical concentration gradient (ng m⁻⁴).

Unlike molecular diffusivity, K_S is not an intrinsic property; rather, the magnitude of K_S depends on both turbulent intensity and atmospheric stability. Within the surface boundary layer, the portion of the atmosphere nearest the land surface where the vertical transport of heat, moisture, and other scalar quantities is considered invariant with height, turbulence can be either suppressed or enhanced by the buoyant effects of thermally-stratified air (Stull, 1988; Garratt, 1994; Ayra, 2001). Under stable atmospheric conditions, the cooler overlying air inhibits vertical movement of the air. In the context of *K*-Theory, this is reflected in lower values of K_S compared to neutral conditions when there is negligible thermal stratification. During unstable conditions, the air near the surface is warmer, and therefore less dense, than the overlying air; the increased buoyancy enhances vertical movement of the air. To account for the enhanced vertical motion, K_S increases.

The vertical gradient of a scalar quantity, such as an herbicide, can be derived from the well-known logarithmic profile (Stull, 1988; Ayra, 2001):

$$S = \frac{F_S}{u_*k} \left[\ln\left(\frac{z}{z_0}\right) - \psi_z + \psi_{z_0} \right] + S_0 \tag{2}$$

where *S* is the herbicide concentration $(ng m^{-3})$ at height *z* (m), u_* is the friction velocity $(m s^{-1})$, *k* is the von Karman constant (dimensionless), z_0 is the surface roughness length (m), ψ_z and ψ_{z_0} (dimensionless) are the stability corrections at heights *z* and z_0 , respectively, and S_0 is the herbicide concentration $(ng m^{-3})$ at z_0 . The stability corrections are defined as

$$\psi = \begin{cases} \frac{+\gamma z}{L} & \frac{z}{L} \ge 0\\ 2\ln\left(\frac{1+\sqrt{1-\frac{-\gamma z}{L}}}{2}\right) & \frac{z}{L} < 0 \end{cases}$$
(3)

where γ and γ are empirically-derived constants [defined herein as -5 and 16, respectively, following Dyer and Hicks (1970) and *L* is the Obukhov Length (m).

From Eqs. (2) and (3), it follows that the vertical gradient in *S* can be expressed as

$$\frac{\partial S}{\partial z} = \begin{cases} \frac{F_S}{u_*k} \left[\frac{1}{z} + \frac{\gamma}{L} \right] & \frac{z}{L} > 0\\ \frac{F_S}{u_*kz} & \frac{z}{L} = 0\\ \frac{F_S}{u_*kz\sqrt{1 - \frac{\gamma}{L}}} & \frac{z}{L} < 0 \end{cases}$$
(4)

In practice, $\partial S/\partial z$ is approximated via a finite difference such that F_S is calculated according to

$$F_S = K_S \frac{\Delta S}{\Delta z} = K_S \frac{S_2 - S_1}{z_2 - z_1} \tag{5}$$

where S_1 is the herbicide concentration at height z_1 and S_2 is the herbicide concentration at height z_2 . The eddy diffusivity is calculated according to $K_s = k\hat{2}u_*/\phi_S$ where $\hat{2}$ is the evaluation height (m) and φ_S is the dimensionless profile function for *S*. In this study, it is calculated by assuming similarity in the turbulent transport of heat, moisture, and the herbicide using the Hicks and Dyer profile functions (1971; see also Dyer, 1974):

$$\phi_{S} = \begin{cases} 1 + \frac{+\gamma \hat{Z}}{L} & \hat{Z} \ge 0\\ \frac{1}{\sqrt{1 - \frac{-\gamma \hat{Z}}{L}}} & \hat{Z} \ge 0 \end{cases}$$
(6)

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