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Fate and transport of agriculturally applied fungicidal compounds, azoxystrobin and propiconazole

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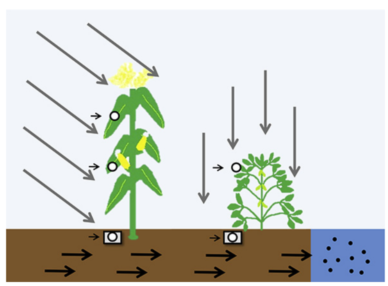
HIGHLIGHTS

- Aerial application of fungicides resulted in detectable drift up to 75 m.
- Both aerial and ground application types resulted in canopy penetration to the soil level.
- Azoxystrobin and propiconazole both persisted in the field up to 301 d following application.
- The primary mode of transport for applied fungicides was agricultural runoff.
- Rain events significantly affected the level of fungicide transport.

GRAPHICAL ABSTRACT



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ABSTRACT

Fungicidal active ingredients azoxystrobin and propiconazole, individually and in combination, have been marketed worldwide in a range of fungicide treatment products for preventative and curative purposes, respectively. Their presence in streams located throughout the midwestern and southeastern United States warrant the need for research into the potential routes of transport of these fungicides in an agricultural field setting. Potential canopy penetration and drift effects of these fungicides during aerial and ground applications were studied in the current project. Canopy penetration was observed for both application types, however drift was associated only with the aerial application of these fungicides. Azoxystrobin and propiconazole persisted in the soil up to 301 d, with peak concentrations occurring approximately 30 d after application. The predominant mode of transport for these compounds was agricultural runoff water, with the majority of the fungicidal active ingredients leaving the target area during the first rain event following application. The timing of application in relation to the first rain event significantly affected the amount of loss that occurred, implying application practices should follow manufacturer recommended guidelines.

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1. Introduction

The use of fungicides on corn and soybean crops has markedly increased in the United States due to increased demand for corn production and concern of the global rise in soybean rust emergence over the last decade (Battaglin et al., 2010). In 2004, cases of soybean rust pathogens, *Phakopsora pachyrhizi* (Asian species) and *Phakopsora meibomiae* (New World species), were first documented in the United States. Due to the potential impact of these pathogens

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on crop yield, the U.S. Environmental Protection Agency (USEPA) approved previously restricted fungicides for Section 18 Emergency Quarantine Exemption in 2006. Included in these exemptions were the strobilurin and azole fungicides (Ochoa-Acuna et al., 2009). This exemption led to a rise in the use of fungicides on United States' crops from 2% in 2002 to an approximately 30% in 2009 (Belden et al., 2010). Active fungicidal ingredients azoxystrobin and propiconazole, individually and in combination, have been marketed worldwide in a range of fungicide treatment products for preventative and curative purposes, respectively (Battaglin et al., 2010).

Azoxystrobin and propiconazole are applied at late growth stages to corn and soybean requiring the use of aerial and ground application techniques, respectively, indicating a potential route of transport to nearby aquatic environments via drift as well as runoff. Application recommendations from product labels typically suggest thorough crop coverage, a reduction in crop overlap, avoidance of use if rain is forecasted, and avoidance of conditions favorable for application drift at the discretion of the applicator. However, there are still concerns for the potential of fungicide drift due to the use of aerial and ground applications, a push for maximum leaf coverage, and sufficient canopy penetration. Parameters contributing to pesticide drift during aerial and ground application events include wind speed, application height, application speed, boom length, nozzle type, angle of the nozzle, and droplet size (SDTF, 1997a; b). Droplet size is considered the most important of these variables affecting pesticide drift (Lamondia et al., 2006; SDTF, 1997a; b) as droplet sizes smaller than 150 microns descend more gradually and are therefore more susceptible to wind effects. Droplet sizes greater than 300 microns descend more quickly and are thus less affected by wind variables. Due to the inherent variability associated with aerial and ground pesticide application techniques, drift may be a contributing factor determining the presence of azoxystrobin and propiconazole detected in streams and lakes.

In addition to drift, canopy penetration of aerial and ground applied fungicides may result in azoxystrobin and propiconazole reaching the soil surface, providing a potential pathway to non-target aquatic systems through agricultural runoff. A wide range of DT_{50}s (a measure of the time it takes for 50% of the parent compound to dissipate from a given area) have been reported for azoxystrobin and propiconazole with observations ranging from 121 to 262 d (EFSA, 2010; Jorgensen et al., 2012) and 9–462 d (Bromilow et al., 1999; EC, 2003; Thorstensen and Lode, 2001), respectively. With variable field dissipation rates, these fungicides may persist in the environment, increasing their likelihood for transport off-site via agricultural runoff. Studies investigating the vertical movement of azoxystrobin and propiconazole through agricultural soils have suggested little mobility of these fungicides likely due to an affinity of both compounds to bind to soil particles (EC, 2003; EFSA, 2010; USEPA, 1997), however soil particles may act as sorbent carriers for these fungicides during rain events, transporting azoxystrobin and propiconazole into non-target aquatic environments. This phenomenon has already been noted for propiconazole (Wu et al., 2003). Currently, there are few large-scale studies that have been conducted to determine the major transport pathways for azoxystrobin and propiconazole in an agricultural landscape (Jorgensen et al., 2012; Rodrigues et al., 2013). Understanding the movement of these fungicides is a key factor in minimizing their presence in midwestern United States streams.

The primary objectives of the current research project were to: (1) utilize Empore™ disks at multiple plant heights to determine the occurrence of drift and canopy penetration during aerial and ground fungicide applications; (2) monitor fungicide dissipation rates to determine the DT_{50} in agricultural field soils; and, (3) determine the transport of applied azoxystrobin and propiconazole via agricultural runoff following significant rain events (>1.27 cm).

2. Materials & methods

2.1. Large-scale agricultural field experiment

2.1.1. Study site and application

The study area consisted of four adjacent agricultural fields located in Moultrie County, Illinois, United States (Fig. 1). Fields 1 (31.07 ha) and 2 (32.34 ha) were planted in continuous corn for 2013 and 2014, while fields 3 (32.34 ha) and 4 (32.47 ha) were planted on a yearly corn/soybean rotation, with soybean planted in 2013 and corn planted in 2014. Planting was targeted for corn in early April and soybeans in mid-May and no till practices were utilized in all fields. Fields 1 and 4 were treated with a fungicide formulation (13.5% azoxystrobin, 11.7% propiconazole) on July 25, 2013 (field 1), August 8, 2013 (field 4), and July 11, 2014 (fields 1 and 4) and fields 2 and 3 were untreated to serve as experimental controls for the study. The treated field (field 4) planted in soybeans in 2013 used a ground application with a rate of 0.768 L per ha (93.8 g propiconazole and 108.5 g axozystrobin per ha) with 17.96 L water per ha and treated fields planted in corn (field 1 in 2013 and fields 1 and 4 in 2014) used an aerial application with rates of approximately 93.8 g propiconazole and 108.5 g axozystrobin per ha.

2.1.2. Sample collections

Empore™ solid phase extraction disks (C18, 47 mm diameter, polytetrafluoroethylene fibers with bonded silica, Supelco, St. Louis, MO, USA) were placed in 20 different locations in each field 48 h prior to application (Fig. 1a) to assess potential canopy penetration and drift effects of the fungicides. At each location in the fields planted with corn, Empore™ disks were placed at three separate heights, one at the top of the plant, one at mid-height (approximately 45 cm from the soil), and one on the surface of the soil. Empore™ disks were placed at two heights at each location in soybean fields, one at the top of the plant and one at the soil level. Sampling design and number of locations were chosen to maximize field coverage and capture the spatial variability associated with azoxystrobin and propiconazole concentrations at each plant height. Following application, Empore™ disks were collected and placed into acetone washed glass scintillation vials and stored at 4 °C, in the dark, prior to analysis with a 14 d maximum holding time (Slaina et al., 1979).

Surface soil samples were collected at 10 different locations in each field (Fig. 1b, $n = 40$). Samples were collected prior to application to determine background concentrations and were collected 48 h post-application and approximately every two weeks for the first month with a single collection in subsequent months until background concentrations were observed. Surface soils (0–6 cm depth) were collected using stainless steel trowels, the plant debris removed, and the soil placed into acetone washed 0.25 L clear glass mason jars. Samples were stored at 4 °C, in the dark, with a maximum hold time of 30 d (USEPA, 1997, 2006).

In addition to monitoring soil, runoff water and sediment were also collected. Three polyvinyl chloride (PVC) runoff troughs (4.57 m (l) x 0.15 m (h)) were installed within each field after planting (Fig. 1b, $n = 18$) with an additional four troughs installed on the edges of the fields ($n = 4$) in order to collect runoff water and sediment samples. Troughs were buried so that the top of each trough was level with the surface of the soil. Runoff water and sediment were collected from each trough following a significant rain (>1.27 cm) event prior to application to determine background fungicide concentrations. Water and sediment were subsequently collected within 24 h of a significant rain event following fungicide application for the 2013 and 2014 field seasons. Water and sediment were mixed within the trough before collection to maximize

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