



# Analysis of pesticides in surface water, stemflow, and throughfall in an agricultural area in South Georgia, USA



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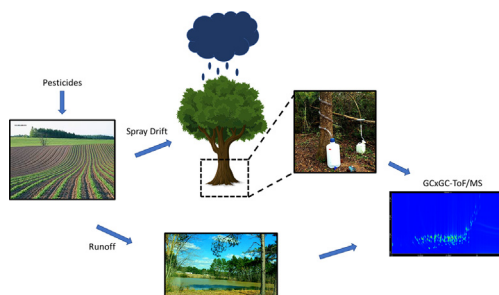
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## HIGHLIGHTS

- Pesticide concentrations in surface water, stemflow and throughfall were analyzed.
- Metolachlor and tebuconazole were the most frequently detected pesticides.
- Higher concentrations were observed in throughfall compared to stemflow samples.
- Understanding migration of pesticides will help estimate risk to non-target species.

## GRAPHICAL ABSTRACT



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## ABSTRACT

To study spray drift contributions to non-targeted habitats, pesticide concentrations in stemflow (water flowing down the trunk of a tree during a rain event), throughfall (water from tree canopy only), and surface water in an agriculturally impacted wetland area near Tifton, Georgia, USA were measured (2015–2016). Agricultural fields and sampling locations were on the University of Georgia's Gibbs Research Farm, Tifton, GA. Samples were screened for more than 160 pesticides, and cumulatively, 32 different pesticides were detected across matrices. Data indicate that herbicides and fungicides were present in all types of environmental samples analyzed while insecticides were only detected in surface water samples. The highest pesticide concentration observed was 10.50 µg/L of metolachlor in an August 2015 surface water sample. Metolachlor, tebuconazole, and fipronil were the most frequently detected herbicide, fungicide, and insecticide, respectively, regardless of sample origin. The most frequently detected pesticide in surface water and stemflow samples was metolachlor (0.09–10.5 µg/L), however, the most commonly detected pesticide in throughfall samples was biphenyl (0.02–0.07 µg/L). These data help determine the importance of indirect chemical exposures to non-targeted habitats by assessing inputs from stemflow and throughfall into surface waters.

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## 1. Introduction

In 2007, approximately 5.2 billion pounds of pesticides were used worldwide; of that amount, 1.1 billion pounds were applied on U.S. soils (Grube et al., 2011). Along with detect concentrations of pesticides in surface waters, pesticides have also been found globally in rainwater (Hüsken and Levsen, 1997; Ahmed et al., 1998; McConnell et al., 1998; Coupe et al., 2000; Dubus et al., 2000; Goel et al., 2005; Sauret et al., 2009; Zhang et al., 2011; Potter et al., 2014; Rice et al., 2016; Potter and Coffin, 2017). Many researchers have monitored pesticides in rainfall, where concentrations have reached 22.9 µg/L of methyl parathion near agricultural sites (Coupe et al., 2000). In rainwater, herbicides were frequently found to be more prevalent, based on detection frequency, (46–61%) and at higher concentrations than insecticides and fungicides (Trevisan et al., 1993; Goel et al., 2005).

Spray drift is the process whereby, during application, an amount of pesticide does not reach the intended application zone due to wind transportation (Briand et al., 2002; Vischetti et al., 2008). Pesticide spray drift often contributes to contamination of nearby surface waters and other non-agriculturally targeted areas. Many parameters can affect spray drift such as climatic conditions and land topography, as well as spray characteristics (i.e. droplet diameter, viscosity of the formulation, and spraying technique) (Briand et al., 2002; Vischetti et al., 2008). Due to variations in climatic conditions, pesticide spray drift may contaminate areas 10–100 m from the intended application site (de Snoo and de Wit, 1998). Even with the presence of a 20-m buffer zone surrounding agricultural fields, spray drift can significantly pollute the surrounding area (Cunha et al., 2012).

Throughout the world, 40% of applied pesticides are herbicides, 33% are insecticides, while 10% are fungicides and 17% are classified as others. Many of these compounds can be readily volatilized after application due to their physical properties (Stokstad and Grullón, 2013). Volatilization coupled with spray drift likely causes many agriculturally-applied pesticides to be transported to the atmosphere, resulting in quantifiable amounts in rainwater far from application sites (Hüsken and Levsen, 1997; Ahmed et al., 1998; Coupe et al., 2000). Gish et al. (2011) determined that volatilization of herbicides occurred immediately after application and measurable concentrations were detected more than 5 days later. It has been estimated that, on average, 23–65% of applied metolachlor can volatilize under certain climatic conditions, within a few days of application (Gish et al., 2011). For other current-use pesticides, up to 90% of the total mass applied can volatilize during and following agricultural application (LeNoir et al., 1999).

Due to volatilization, long-range atmospheric transport of pesticides can occur; compounds can traverse many miles and have been found in remote regions such as the Arctic (Cotham and Bidleman, 1991; Rice and Chernyak, 1997). Conveyance of pesticides to the atmosphere can be caused by spray drift, volatilization, runoff, and wind erosion of soil (Briand et al., 2002). Both runoff and volatilization, as previously discussed, have been investigated for sources of pesticide loss. It was found that runoff accounts for less than 3% loss, while volatilization accounts for approximately 2–25% total loss (Gish et al., 2011). Atmospheric transport has also been shown to deposit pesticides to nearby pristine areas and it is well-documented that pesticides applied in the agriculturally intensive Central Valley of California have been transported and deposited to the Sierra Nevada mountain range (McConnell et al., 1998; LeNoir et al., 1999). Of note: tebuthiuron, a herbicide, was detected at concentrations four times higher in a Brazilian riparian forest compared to other areas, possibly due to air pollution concentrating the pesticide in the tree canopy and then releasing into the soil during rain events (Bicalho et al., 2010).

Pesticide residues may enter the ecosystem through precipitation, leaching, spray drift, discharge of wastewater and other routes (Griffini et al., 1997; Konstantinou et al., 2006; Rice et al., 2016). During a rain event, contaminating pesticides are often washed off trees and surrounding foliage which results in stemflow (precipitation that flows down along the tree trunk) and throughfall (rain that passes through the tree canopy). These routes of environmental release have been well-studied and higher concentrations of pesticides are often detected in stemflow and throughfall samples when compared to open-field rainfall (Bernhardt and Ruck, 2004; Rice et al., 2016). Further, stemflow encounters more arboreal surface area for a longer period of time than throughfall, resulting in higher pesticide concentrations in stemflow than in throughfall (Rice et al., 2016). Pesticides studied by Bernhardt and Ruck (2004) were detected in stemflow samples for a longer period of time than rainfall samples due to the trees acting as filters and concentrating contaminants from the atmosphere. Large pulses of pesticides have also been shown to flow into rivers or surface waters after spray application that coincides with a rain event (Thurman et al., 1991; Griffini et al., 1997; Konstantinou et al., 2006; Rice et al., 2016). Understanding how concentrations of pesticides enter aquatic habitats is paramount in quantifying the cumulative environmental exposure and risk associated with spray drift.

In a recent survey that analyzed pesticide concentrations in streams throughout the U.S., the occurrence rate of detecting one or more pesticides in agricultural, urban or mixed use watershed matrices was over 90% (Gilliom et al., 2006). The most frequently detected herbicides in these waters were atrazine, glyphosate, aminomethylphosphonic acid (AMPA; a metabolite of glyphosate) and metolachlor (Gilliom et al., 2006; Battaglin et al., 2016). In agreement, Smalling et al. (2012) detected 24 pesticides in pond water samples collected from several states throughout the U.S. The most frequently detected pesticides were, again, herbicides, mainly atrazine, glyphosate and AMPA. Battaglin et al. (2016), however, reported that fungicides were more frequently detected in water samples collected throughout the U.S. compared to insecticides and herbicides, likely due to their more frequent, intensive application schedules even with lower application rates. Overall, concentrations of pesticides in surface waters tend to follow seasonal trends with higher detectable concentrations observed during periods of heavy use (such as in spring and summer due to preparation for planting) and lower concentrations in winter and after crop harvesting (Konstantinou et al., 2006).

To date, few studies have attempted to quantify pesticides in stemflow and throughfall and determine how these concentrations impact surface water concentrations (Trevisan et al., 1993; Bernhardt and Ruck, 2004; Zhang et al., 2011; Rice et al., 2016). The overall objective of this study was to identify and quantify pesticides in surface water, stemflow and throughfall, adjacent to agricultural fields, over the course of a year (February 2015 through January 2016). We also compared stemflow to throughfall concentrations after rain events at three paired sites. This research will ultimately inform regulatory aspects of pesticide application such as environmental residence time in rainwater, surface waters/rivers, and surrounding foliage as a result of spray drift and its impact on non-target organisms inhabiting the areas.

## 2. Materials and methods

### 2.1. Chemicals

In total, we screened for 160 pesticides by combining those commercially available in the gas chromatography (GC) multi-residue pesticide kit from Restek ( $n > 150$ ); Bellefonte, PA) and supplementing additional pesticides that were obtained from the

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