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Surface runoff and subsurface tile drain losses of neonicotinoids and companion herbicides at edge-of-field[☆]

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ABSTRACT

With their application as seed coatings, the use of neonicotinoid insecticides increased dramatically during the last decade. They are now frequently detected in aquatic ecosystems at concentrations susceptible to harm aquatic invertebrates at individual and population levels. This study intent was to document surface runoff and subsurface tile drain losses of two common neonicotinoids (thiamethoxam and clothianidin) compared to those of companion herbicides (atrazine, glyphosate, *S*-metolachlor and mesotrione) at the edge of a 22.5-ha field under a corn-soybean rotation. A total of 14 surface runoff and tile drain discharge events were sampled over two years. Events and annual unit mass losses were computed using flow-weighted concentrations and total surface runoff and tile drain flow volumes. Detection frequencies close to 100% in edge-of-field surface runoff and tile drain water samples were observed for thiamethoxam and clothianidin even though only thiamethoxam had been applied in the first year. In 2014, thiamethoxam median concentrations in surface runoff and tile drain samples were respectively 0.46 and 0.16 μg/L, while respective maximum concentrations of 2.20 and 0.44 μg/L were measured in surface runoff and tile drain samples during the first post-seeding storm event. For clothianidin, median concentrations in surface runoff and tile drain samples were 0.02 and 0.01, μg/L, and respective maximum concentrations were 0.07 μg/L and 0.05 μg/L. Surface runoff and tile drain discharge were key transport mechanisms with similar contributions of 53 and 47% of measured mass losses, respectively. Even if thiamethoxam was applied at a relatively low rate and had a low mass exportation value (0.3%), the relative toxicity was one to two orders of magnitude higher than those of the other chemicals applied in 2014 and 2015. Companion herbicides, except glyphosate in tile drains, exceeded their water quality guideline during one sampling campaign after application but rapidly resumed below these limits.

1. Introduction

Insect and weed chemical controls together with disease suppression are important components of integrated pest management strategies in commercial corn and soybean cropping systems.

Drastic changes have recently occurred in pest insects management strategies. Use of broadcast organophosphate and carbamate insecticides are declining while neonicotinoids are increasing since the introduction of imidacloprid in the 1990s and later with the use of the second generation neonicotinoid insecticides, thiamethoxam and clothianidin (Main et al., 2014; MDDELCC, 2016). The latter is also a degradation product of thiamethoxam (Nauen et al., 2003). The use of neonicotinoids increased dramatically during the last 10 years with their application as seed coatings. According to Baker and Stone (2014), about 4, 3 and 14 g/ha of imidacloprid, thiamethoxam and clothianidin, respectively, were used in 2013 in selected watersheds of the Midwestern US corn belt. Douglas and Tooker (2015) have synthesized publicly available data in the U.S. and estimated that 34–44% of soybeans and 79–100% of corn hectares were treated in 2011. In 2012, it was estimated that applications of neonicotinoid covered about 11

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million hectares with over than 216,000 kg of active ingredients in the Prairie cropland of Canada (Main et al., 2014). In Quebec (Canada), it is estimated that almost 100% of corn and 50% of soybeans seeds planted are coated with neonicotinoids which correspond to about 500,000 ha (MDDELCC, 2015). These usage rates coupled with long persistence and high water solubility have led to frequent detection of neonicotinoids in aquatic environments. For instance, in intensive corn and soybean producing areas, Giroux (2015) reported a mean detection frequency of thiamethoxam and clothianidin varying from 93% to 98% from 2012 to 2014 within four Quebec watersheds. In 2013 in southwestern Ontario, water samples collected in 5 corn-producing counties showed detection frequencies of 98.7% and 100% for thiamethoxam and clothianidin, respectively (Schaafsma et al., 2015). Hladik et al. (2014) reported mean detection frequencies at 9 sampling locations across Iowa in 2013 of 23, 47 and 75% for imidacloprid, thiamethoxam and clothianidin, respectively. In the Prairie Pothole region, Main et al. (2014) also reported neonicotinoid detection in 91% of the 136 wetlands sampled in spring 2013.

Neonicotinoids, which act on insects by irreversibly binding to the nicotine acetylcholine receptors, have received increasing attention in the recent years due to their potential impacts on non-target organisms such as pollinators, predatory insects, earthworms and aquatic species (TFSP, 2015). Overall, it is recognized that neonicotinoids have the potential to cause negative effects at the individual and population levels of aquatic invertebrates at low concentrations (Pisa et al., 2014). In laboratory studies, negative relationships have been found between macroinvertebrate abundance and imidacloprid concentrations (Alexander et al., 2007; Mohr et al., 2012; Roessink et al., 2013; Stoughton et al., 2008). Effects on macroinvertebrates were observed at concentrations as low as 0,024 μg/L (Roessink et al., 2013). In a statistical analysis between macroinvertebrate abundance and imidacloprid concentration Van Dijk et al. (2013) showed a negative relationship. Indoor stream mesocosm studies had also demonstrated negative effect on the emergence of macroinvertebrates (Mohr et al., 2012) and that repeated pulses of imidacloprid also lead to massive drift of benthic macroinvertebrates (Beketov and Liess, 2008; Berghahn et al., 2012).

Concurrently, chemical weeds control has continued to evolve in the last few decades primarily due to the introduction of new molecules, the development of herbicides-resistant crops and the apparition of herbicides-resistant weeds (Appleby, 2005). Chemicals introduced in the 1950s (e.g. atrazine) and the 1970s (e.g. glyphosate and metolachlor) continue to be widely used in North-America while newer products, including mesotrione which became available in the late 1990s, are increasing in popularity (Giroux, 2015).

Aquatic ecosystems may be contaminated by pesticides, including neonicotinoids, through various routes such as atmospheric deposition, surface runoff, tile drain discharge and groundwater seepage losses (Anderson et al., 2013; Bonmatin et al., 2015; Smalling et al., 2015). As reported by Hladik et al. (2014), monitoring data for neonicotinoids transport to surface water are limited and more research on geographic occurrence, concentrations and transport mechanisms is needed. The intent of this study was to document surface runoff and tile drain losses of two commonly used neonicotinoids (thiamethoxam and clothianidin) and companion herbicides (atrazine, glyphosate, *S*-metolachlor and mesotrione) at the edge of a 22.5 ha field under corn and soybeans rotation. A particular attention was given to tile drain lines as they may contribute to a significant proportion of pollutant mass load exports in a cold and humid climate. To our knowledge, this study is the first conducted in the St. Lawrence lowlands providing flow-weighed event mean concentrations at the edge-of-field as well as surface and subsurface partitioning of neonicotinoid losses.

2. Materials and methods

2.1. Site description

The experimental site was established on a 22.5-ha field under corn and soybean rotation located in Saint-Samuel, Quebec, Canada (4602′14″N - 7212′20″W) (Fig. 1). Environment Canada station No. 7022160 located at 30 km from the site shows a historical annual temperature average of 6.4 °C, varying from −10.2 °C in January to 20.9 °C in July, on average. The mean growing season length from 1979 to 2008 is 204 days based on a temperature above 5.5 °C. The average annual precipitation is 1114 mm out of which 242 cm fall as snow, and 598 mm fall as rain during the growing season (Agrométéo Québec, 2015). No supplemental irrigation was used on the experimental site.

The field is composed of three soil series, namely the Des Sault, Courval and Lévrard loams, issued from deep marine depositions to superficial lagoon layers. According to the Canadian System of Soil Classification, the three soil series belong to the orthic humic gleysol (Humaquept) sub-group of the gleysolic order. All three soils are characterized as imperfect to poor drained (Rompré et al., 1984). The average slope of the field is 0.25% and is artificially drained through a network of nine shallow ditches and thirteen tile drain lines flowing into two collectors.

Hay was produced for four years without use of insecticides before it was burned with glyphosate in June 2013 and seeded with soybean after conventional tillage (CT) with a deep moldboard plow in half of the field and directly drilled through crop residues in the no-till (NT) second half. Successive crops were also grown at a proportion of 50% CT and 50% NT. In 2014, the first monitored year, corn was planted on May 20th with thiamethoxam coated seeds corresponding to 116 g/ha of active ingredients (a.i.). Chemical weeds control was achieved with commercial mixtures of atrazine, glyphosate, *S*-metolachlor and mesotrione at respective total concentrations of 285, 1717, 1050, and 105 g a.i./ha. The applications of the herbicide mixtures were made on June 9th, 2014 in the CT section and on June 17th in the NT section. Corn silage was harvested on October 1st, 2014. In 2015, soybean was partially planted on May 27th and completed on June 2nd with fungicides treated seeds (Baccilus Subtilis MBI600, Sedaxane, Fludioxonil, Metalaxyl-M and S-isomer). Weeds were controlled using glyphosate and chlorimuron ethyl at concentrations of 1334 and 9 g a.i./ha, respectively. No neonicotinoid treated soybean seeds were used. Soybean was harvested on October 25th, 2015. The two fungicides Baccillus Subtilis and sedaxane, and the herbicide chlorimuron ethyl were not monitored in this study.

2.2. Flow monitoring and sampling

Flow monitoring and water quality sampling was implemented between May 21st' 2014 and December 1st, 2015. Winter and spring snowmelt events could not be monitored due mainly to issues with back flooding and freezing of sampling equipment. Surface monitoring was performed at the outlet of four shallow ditches (Fig. 1) using for each outlet a 2H flume (Plasti-Fab, Tualatin, OR, USA) equipped with an ISCO 720 Flow Module mounted in a stilling well to control a programmed ISCO 6712 Autosampler (Teledyne ISCO, Lincoln, NE, USA) to collect flow-weighted water samples. Surface runoff volumes triggering the collection of sub-samples were periodically adjusted to account for differences in runoff coefficient during the growing and the non-growing seasons. Flow-weighted samples, collected at the four surface runoff monitoring

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