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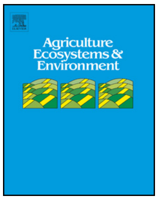

Snowmelt transport of neonicotinoid insecticides to Canadian Prairie wetlands

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ABSTRACT

During the growing season, neonicotinoid insecticides are frequently transported to surface water systems after rainfall events. However, detectable levels of neonicotinoids have also been found in wetlands during early spring, after ice-off but before crop seeding, representing an unexpected long-term exposure risk for aquatic organisms. This suggests long-term persistence, though origins and transport mechanisms remain unknown. We sampled 16 agricultural fields in the Canadian Prairies to investigate whether snow meltwater, particulate matter, top- (15 cm) or bottom-layer (15 cm) snow were potential sources of spring neonicotinoid contamination to receiving wetlands. Agricultural fields were selected based on the previous year's crop: eight canola fields (clothianidin-treated seed) and eight oat fields (untreated). We further sampled the wetlands draining those same oat and canola fields from ice-off to seeding to assess changes in neonicotinoid concentrations over time. Top-layer snow was below the limit of quantification for both canola and oat fields. Neonicotinoid concentrations (sum of clothianidin and thiamethoxam) were highest in meltwater (canola, mean: 267 ± 72.2 ng L^{-1}; max: 633), but also detected in bottom-layer snow (oat, mean: 36.1 ± 9.18 ng L^{-1}; max: 92.9), and soil particulate matter (canola, mean: 10.2 ± 1.82 μg/kg; max: 17.2). Concentrations in meltwater showed a stronger relationship ($R^2 = 0.35$) with initial concentrations in wetland water than any other source type. Temporary wetland hydrology is largely fed by meltwater thus spring total neonicotinoid concentrations were higher in temporary wetlands than seasonal/semi-permanent wetlands ($P = 0.003$). Only clothianidin was detected in soil particulate matter samples, including from oat fields not treated the year before, confirming this compound can persist over multiple years under local field conditions. The results of this study suggest that under normal agricultural practices, wetlands in colder climates are likely to be contaminated even before seeding occurs through persistence of neonicotinoids in soil and transport by snowmelt and particulate to surface water during spring runoff.

1. Introduction

Canada is home to an estimated 127 million ha of wetlands, accounting for 25% of the world's wetland area (Government of Canada, 1991). In the Prairie region, wetlands are often situated in productive farmlands where agricultural activities (e.g., drainage, agrochemical use) affect almost all wetlands directly or indirectly through mechanisms including increased siltation and destruction of wetland plants by herbicides (Kantrud et al., 1989; Bartzen et al., 2010). Agricultural wetlands are sometimes viewed as non-productive acreage (Wrubleski and Ross, 2011), but these surface water systems provide a suite of critical ecosystem services: water filtration, flood attenuation, and habitat and food resources for wetland-dependent organisms (e.g., birds, amphibians; Taft and Haig, 2005; Zhang et al., 2007). Production of diverse aquatic prey resources is especially critical during early spring staging and breeding periods for migratory insectivorous birds, waterfowl and shorebirds (Swanson et al., 1985; Davis and Smith, 2001; Mengelkoch et al., 2004; Baschuk et al., 2012).

More recent agrochemical threats to wetlands are the neonicotinoids, which are the fastest growing class of insecticides in modern crop protection (Jeschke et al., 2010). Neonicotinoids

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(e.g., acetamiprid, clothianidin, imidacloprid, and thiamethoxam) are frequently used as seed treatments on major agricultural crops across North America and Europe, including canola, cereals (e.g., wheat), corn and soybeans (Elbert et al., 2008). Clothianidin, imidacloprid, and thiamethoxam combined are registered for 295 crop uses in 120 countries (Jeschke et al., 2010). Although most seed treatments pre-date the advent of neonicotinoids (Buttress and Dennis, 1947), the large-scale production and mono-cropping common in current agricultural practices has led to over 95% of canola seeds being treated with some type of neonicotinoid active ingredient in Canada (Main et al., 2014), and 79–100% of corn in the United States (Douglas and Tooker, 2015). Pesticide use in Canada is considered confidential and seed-applied pesticide products were not accounted for by the National Agricultural Statistics Service major use survey in the United States, but neonicotinoid seed treatments continue to grow in popularity and spatial extent (USGS, 2012; Main et al., 2014; Douglas and Tooker, 2015). Insecticide applications typically extend throughout the Great Plains region – including the Canadian Prairies – which often directly overlaps with high density wetland environments.

Neonicotinoid seed treatments are designed to protect the individual plant while greatly reducing the amount of insecticide used in subsequent spray or soil drenching applications, thus theoretically lessening impacts on the environment (Elbert et al., 2008; Jeschke et al., 2010). However, typically 5% (max 20%) of the active ingredient may be absorbed by the target crop (Sur and Stork, 2003), while the remaining active ingredient may remain in soils (Goulson, 2013). Estimates of clothianidin and thiamethoxam persistence in soils are variable with clothianidin persisting in fields (half-life: DT_{50}) from 277 to 1386 days (DeCant and Barrett, 2010), and thiamethoxam persisting in fields for 7–109 days (Goulson, 2013). Neonicotinoids also accumulate in soils over time (Bonmatin et al., 2005; Jones et al., 2014) and are expected to persist longer in colder regions at mid to higher latitudes due to lower temperatures and lower sunlight intensity (Bonmatin et al., 2015). Indeed, a previous Saskatchewan field study demonstrated little to no measurable dissipation over 775 days (PMRA, 2004). Additionally, both insecticides are highly water soluble (clothianidin = 327 mg/L; thiamethoxam = 4100 mg/L) with high potential for transport into water bodies (HSDB, 2012). Neonicotinoids have been detected across a range of agricultural surface water systems in North America including rivers, streams and wetlands with peak neonicotinoid concentrations typically detected after seeding or during the growing season following rainfall events (Starner and Goh, 2012; Anderson et al., 2013; Hladik et al., 2014; Main et al., 2014; Smalling et al., 2015). However, traditional studies reporting water concentrations have rarely assessed whether contamination persists outside the growing season.

In the Canadian Prairie region, 80–85% of water stored in wetlands comes from the shallow snowpack which is highly susceptible to wind erosion or redistribution (Gray, 1970). Prairie soils can also freeze to depths of 1 m or more (Van der Kamp et al., 2003). As the infiltration capacity of frozen soils is limited (Granger et al., 1984), a large amount of surface meltwater can be generated in a short amount of time during spring snowmelt which flows into depressions forming small wetlands (Hayashi et al., 2003). During the melt process, soil particles may be picked up and transported with the runoff (Gray, 1970). Our previous studies found that after ice-off (i.e., opening of surface water) and before seeding has occurred, between 36% (2012) and 91% (2013) of Prairie wetlands contained at least one neonicotinoid insecticide (Main et al., 2014) at total concentrations up to six times greater than those which may induce chronic effects to aquatic insects (Morrissey et al., 2015). Consequently, snowmelt transport of neonicotinoids to wetlands, during a critical spring period, may have important implications for wetland biota.

Although we previously reported pre-seeding neonicotinoid concentrations in Prairie wetlands, the origin of those spring detections remained unknown. Therefore, our objectives were to (1) identify the major source of neonicotinoids to wetlands in spring; and (2) examine what factors affect change in spring wetland neonicotinoid concentrations over time. We hypothesized that pre-seeding wetland water contamination originated either from snow contact with agricultural fields or soil particulate matter containing residual neonicotinoid active ingredients which may be scoured and transported to wetlands during seasonal snowmelt runoff.

2. Methods

We conducted fieldwork during April and May of 2014 at 16 agricultural fields (65 ha) near Alvena, Saskatchewan (52.5167°N, 106.0167°W; Fig. 1). Saskatchewan is characterized by warm, dry summers (e.g., daily average, July: 18.5 °C) and cold, dry winters (e.g., daily average, January: −15.5 °C); average precipitation is approximately 276.7 mm of rainfall and 91.3 cm of snow (Environment Canada, 2010). Our study fields were situated in the Black soil zone (Order: Chernozemic) which is typically associated with shorter growing periods and lower temperatures, but increased moisture leading to a wider variety of potential crops in production (average soil organic matter = 4.5 to 5.5%; SCWG, 1998). Typical crop rotation in this area of Saskatchewan is an alternating canola/cereal rotation where the majority of canola is treated with clothianidin or thiamethoxam products while a small but growing fraction of cereals are treated. We controlled for previous crop type (i.e., crop planted in spring 2013) by selecting an even distribution of fields previously seeded to either canola or oat crops. All oat fields were planted with un-treated seed in the year prior (2013), whereas canola fields were previously planted using clothianidin-treated seeds (Prosper®, Bayer CropScience) at standard application rates. It should be noted that in years prior to our study (2011 and 2012), 20% of landowner canola fields were planted with thiamethoxam-treated seeds (Helix Xtra®, Syngenta). Study wetlands ($n = 16$; one per agricultural field) spanned a range of classes (defined by Stewart and Kantrud, 1971) including: temporary ($n = 6$), seasonal ($n = 7$) and semi-permanent ($n = 3$). All wetlands were <1 ha in size, ranged in initial depth from 20 cm to over 1 m and were randomly chosen based on consistent timing of availability after ice-off.

2.1. Snow and meltwater collection

Our snow sampling technique was modified from previously published methods (McConnell et al., 1998; Hageman et al., 2006). Top- and bottom-layer snow samples were collected using a stainless steel spade that was rinsed thoroughly between samples with deionized water. Top-layer samples were extracted from a ~15 cm × 15 cm × 30 cm core at three random points surrounding the study wetlands (~3 m from the wetland edge). The top ~5 cm were removed to lessen the possibility of atmospheric deposition or wind-scoured soil that may contaminate the perceived “clean layer”. An additional three bottom-layer snow samples were collected at the same locations from the ~15 cm of snow that directly interacts with the soil. All snow samples were transferred to individual polyethylene bags, transported to the lab in the dark in coolers, and immediately placed in a freezer at −20 °C until analysis. Prior to analysis, snow samples were placed in a large stainless steel basin, covered and allowed to melt overnight at room temperature (McConnell et al., 1998). Composite top- and bottom-layer samples were created from the three sample points and poured into chemically-cleaned (acetone: hexane) 1 L amber