



Transport of a neonicotinoid pesticide, thiamethoxam, from artificial seed coatings☆



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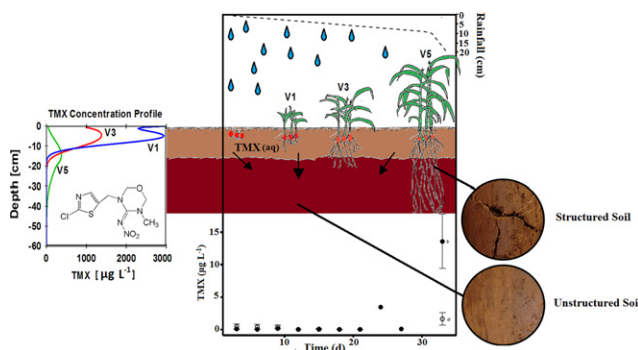
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HIGHLIGHTS

- Transport of neonicotinoid thiamethoxam (TMX) was quantified from corn seed coats.
- A column study with viable corn plants simulated field-realistic leaching.
- Evapo-concentration reduced and soil structure and plants enhanced TMX movement.
- TMX was transported from surface horizons in levels acutely toxic to aquatic life.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 29 August 2017

Received in revised form 2 November 2017

Accepted 3 November 2017

Available online 17 November 2017

Editor: Kevin V. Thomas

Keywords:

Soil structural features

Evapo-concentration

Evapotranspiration

Corn stage

Insecticide

ABSTRACT

Neonicotinoid insecticides coat the seeds of major crops worldwide; however, the high solubility of these compounds, combined with their toxicity to non-target organisms, makes it critical to decipher the processes by which they are transported through soils and into aquatic environments. Transport and distribution of a neonicotinoid (thiamethoxam, TMX) were investigated by growing TMX-coated corn seeds in coarse-textured and fine-textured soil columns (20 and 60 cm lengths). To understand the influence of living plants, corn plants were terminated in half of the columns (no plant treatment) and allowed to grow to the V5 growth stage (33 days of growth) in the other half (with plant treatment). TMX was analyzed in leachate 12 times over 33 days and in bulk soil after 8, 19, and 33 days of corn growth. All 20 cm columns leached TMX at levels exceeding the United States Environmental Protection Agency benchmark for aquatic invertebrates ($17.5 \mu\text{g L}^{-1}$). TMX migrated from seeds to adjacent bulk soil by the eighth day and reached deeper soil sections in later growth stages (e.g., 30–45 cm depth by Day 33). Fine-particle soils transported over two orders of magnitude more TMX than coarse-textured soils (e.g., $29.9 \mu\text{g}$ vs $0.17 \mu\text{g}$, respectively), which was attributed to elevated evapotranspiration (ET) rates in the sandy soil driving a higher net retention of the pesticide and to structural flow occurring in the fine-textured soil. Living plants increased TMX concentrations at depth (i.e., 30–60 cm) compared to the no plant treatment, suggesting that corn growth may drive preferential transport of TMX from coated seeds. Altogether, this study showed that neonicotinoid seed coatings can be mobilized through soil leachate in concentrations considered acutely toxic to aquatic life.

Published by Elsevier B.V.

☆ Notes: The authors declare no competing financial interest.

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1. Introduction

Each year >20,000 t of neonicotinoids are produced (Codling et al., 2016) and applied to 140 + crops worldwide (Elbert et al., 2008; Main et al., 2015), with an estimated global market value of ~\$2.6 billion (Goulson and Kleijn, 2013; Jeschke et al., 2011). Imidacloprid (IMD), thiamethoxam (TMX), and clothianidin (CLO) are the most common neonicotinoids, and are used extensively as seed coated insecticides. These three compounds have high selectivity for the insect nicotinic acetylcholine receptor, which makes them an effective pesticide against a broad spectrum of invertebrate pests while remaining relatively non-toxic to mammals (Jeschke and Nauen, 2007; Jeschke et al., 2011; Zalom et al., 2005). Despite their effectiveness as insecticides, neonicotinoids have been scrutinized for high toxicity to non-target invertebrate organisms and insectivorous birds (Douglas et al., 2015; Hallmann et al., 2014), including an association with significant declines in bee populations (Alix et al., 2009; Sanchez-Bayo and Goka, 2014; Sandrock et al., 2014; Whitehorn et al., 2012).

Globally, >60% of all neonicotinoids are applied via seed dressings (Goulson and Kleijn, 2013). Neonicotinoid seed coatings are advertised to provide continued protection from insect herbivory throughout the growing season, as the highly soluble compounds move into the root zone, enter the plant, and arm its aboveground tissue (Jeschke et al., 2011; Jones et al., 2014; Zalom et al., 2005). Because they are applied directly to the plant roots, seed coatings typically have lower amounts of active pesticides compared to spray applications (in terms of g ha⁻¹) and are therefore often considered to be more environmental friendly (Jeschke et al., 2011). However, only 2–20% of the pesticide is taken up by the target crop, with the remainder left in the soil environment where it may become transported into ground water and eventually incorporated into surface water systems (Sanchez-Bayo, 2014). Neonicotinoids can be applied as corn coatings at amounts up to 1.25 mg per seed. Using corn production (typical planting rate: 74,000 seeds ha⁻¹) as an example, an estimated 3300 t of neonicotinoids can be applied each year to the 36 million ha of maize in the United States (USDA, 2017). If only 20% of the neonicotinoids are taken up by the corn plants, then up to 2700 t of neonicotinoids may be mobilized in the surrounding environment. As a possible consequence, neonicotinoids are now detected in surface waters across North America (Hladik et al., 2014; Main et al., 2015; Schaafsma et al., 2015). In regions with intensive soybean and maize production, neonicotinoids have been detected in nearly all surface water bodies (Hladik et al., 2014; Schaafsma et al., 2015), with seed coatings identified as the most likely source. However, the linkage between seed coatings and the aquatic environment as not been directly assessed, and no work to date has identified or quantified the underlying mechanisms by which neonicotinoids are transported from agricultural fields.

Neonicotinoids have moderate to high leaching potential, with leaching patterns largely explained by the aqueous solubility of the compounds (Banerjee et al., 2008; Cox et al., 2001; Cox et al., 1997; Gupta et al., 2008; Katagi, 2013; Kurwadkar et al., 2014; Leiva et al., 2015; Oi, 1999). Leaching typically increases with soil water content; for example, the half-life of thiamethoxam (TMX) in soil columns decreased from 301 days to 46 days as soil water content increased from dry to near-saturated conditions, with degradation rates increasing with TMX concentration (Gupta et al., 2008). Because neonicotinoids are polar, highly soluble, and exhibit low affinity for soil mineral matrix, the partial equilibrium conditions provided by a rain storm may promote leaching via bulk flow or advection (Hu and Brusseau, 1996; Katagi, 2013; Kurwadkar et al., 2014; Oi, 1999).

Soil structural features, which arise as unconsolidated soil material arranges into a more stable hierarchy of aggregates, often form secondary pore networks that may affect solute transport. As soil water pressure increases during a rain event, flow can preferentially follow these structural pathways and bypass the soil matrix, where water is tightly held at more negative potentials (Jarvis, 2007). This non-equilibrium flow can

result in pesticides being rapidly mobilized through the soil profile (FOCUS, 2001; Jarvis, 2007; Molz, 2015). For example, mass balances performed on the herbicide bentazone have showed that up to 8% of the applied dose can be lost through soil structural pathways, resulting in concentrations as high as 200 µg L⁻¹ in tile drainage (FOCUS, 2001). Thus, it is important to understand if neonicotinoids can also be mobilized via flow through soil structural pathways.

The transport and distribution of pesticides in soil is also further complicated by the presence of plants. For instance, maize can apply suction forces well above 9.5 bars (Ionescu, 1969). For highly soluble compounds such as neonicotinoids, this plant-induced suction could translate to a vertical stratification in the soil profile, in which neonicotinoids can remain close to the soil surface under stable unsaturated conditions. On the other hand, roots may increase the size of soil structural pathways, which can cause increased preferential leaching (Bundt et al., 2000; Jørgensen et al., 2002). Such plant-related processes have not yet been well-studied in the context of pesticide transport.

The primary aim of this study was to quantify the transport of thiamethoxam (TMX) from coated corn seeds in fine-textured and coarse-textured soils, while also accounting for the roles of soil structure and viable plants. We hypothesize that: i) coarse-textured soil will transport more TMX than fine-textured soil with intact structural features, ii) structured soil will transport more than unstructured soil of the same texture, and iii) the presence of viable plants will result in less leaching of the pesticide from soil columns. Since seed coatings are the dominant form of neonicotinoid application for many crops, direct measurements of their movement from the seeds is an imperative step in assessing the overall environmental impact of this practice.

2. Materials and methods

2.1. Soil characterization

Soils were taken from pastures in New Kent and Whitehorse, VA. The New Kent soil was a Bojac series (Typic Hapludult) and coarse-textured (hereafter referred to as a “sand”). The Whitehorse soil was a Shottower series (Typic Paleudults) and a fine-textured, moderately structured soil (hereafter referred to as a “loam”).

Intact soil cores (stainless steel, 2.5 cm × 5 cm) taken from the two sites were analyzed for bulk density [M L⁻³] and saturated hydraulic conductivity (K_s) [L T⁻¹]. Samples were collected every 5 cm depth from the surface to 30 cm depth ($n = 6$ cores per depth increment). The 0–20 cm cores were considered to represent the A_p horizon and the 20–30 cm cores were considered to represent the B_t layer. K_s was determined using the falling head method with a UMS KSAT Benchtop Saturated Hydraulic Conductivity Instrument (UMS Inc.; Munich, Germany). K_s was determined per the manufacturer's recommendation as:

$$K_s = bL \left(\frac{A_{burette}}{A_{sample}} \right) \quad (1)$$

where $A_{burette}$ [L²] is the cross-sectional area of the water column, A_{sample} [L²] is the cross-sectional area of the sample, L is the length of the sample, and b is an exponent determined via curve-fitting between measured pressure head (h , starting at some initial pressure head h_0) and time:

$$h(t) = h_0 e^{-bt} \quad (2)$$

Loose soil samples from the A_p (0–20 cm) and B_t (20–60 cm) horizons were air dried, sieved to 2 mm, and analyzed for pH, cation exchange capacity (CEC) [Mol M⁻¹], total organic carbon (TOC) [M M⁻¹], and texture. Five replicates ($n = 5$) were used samples per test; additional details regarding the soil CEC, pH, TOC, and texture measurements are found in Supporting Information. TMX sorption coefficient (K_d) [L³ M⁻¹] and sorption coefficient normalized to soil organic carbon

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