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### Concentration and movement of neonicotinoids as particulate matter downwind during agricultural practices using air samplers in southwestern Ontario, Canada



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### Luis Gabriel Forero, Victor Limay-Rios, Yingen Xue, Arthur Schaafsma\*

Department of Plant Agriculture, University of Guelph, Ridgetown Campus, Ridgetown, Ontario NOP 2CO, Canada

#### HIGHLIGHTS

- TSP Neonic levels under field conditions averaged 6.2 times higher during planting than during tillage or wind events.
- Tillage and wind contributed to offsite residue transport at much lower levels than planting.
- Neonicotinoid particulate matter during planting of treated seed declined significantly over a distance of 250 m.
- The highest honeybee exposure occurs on field edges high enough to induce chronic effects.

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#### G R A P H I C A L A B S T R A C T



#### ABSTRACT

Atmospheric emissions of neonicotinoid seed treatment insecticides as particulate matter in field crops occur mainly for two reasons: 1) due to abraded dust of treated seed generated during planting using vacuum planters, and 2) as a result of disturbances (tillage or wind events) in the surface of parental soils which release wind erodible soil-bound residues. In the present study, concentration and movement of neonicotinoids as particulate matter were quantified under real conditions using passive and active air samplers. Average neonicotinoid concentrations in Total Suspended Particulate (TSP) using passive samplers were 0.48 ng/cm<sup>2</sup>, trace, trace (LOD 0.80 and 0.04 ng/cm<sup>2</sup> for clothianidin and thiamethoxam, respectively), and using active samplers 16.22, 1.91 and 0.61 ng/m<sup>3</sup> during planting, tillage and wind events, respectively. There was a difference between events on total neonicotinoid concentration collected in particulate matter using either passive or active sampling. Distance of sampling from the source field during planting of treated seed had an effect on total neonicotinoid air concentration. However, during tillage distance did not present an effect on measured concentrations. Using hypothetical scenarios, values of contact exposure for a honey bee were estimated to be in the range from 1.1% to 36.4% of the reference contact LD<sub>50</sub> value of clothianidin of 44 ng/bee.

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#### 1. Introduction

The use of neonicotinoid insecticides as seed treatments is common for field crops in southwestern Ontario. In 2013 almost

\* Corresponding author. *E-mail address:* aschaafs@uoguelph.ca (A. Schaafsma).

100% of maize and 60-80% of soybeans crop areas were planted using seed treated with clothianidin or thiamethoxam (Stewart and Baute, 2013). Vacuum type planters have been shown to dislodge, as a result of abrasion, insecticide seed coatings during planting, and exhaust them to the atmosphere (Greatti et al., 2003, 2006; Nikolakis et al., 2009; Nuvttens et al., 2013). These abraded particles or contaminated dust, are potentially toxic to exposed honevbees and other pollinators (Krupke et al., 2012; Sgolastra et al., 2012; Girolami et al., 2012; Pistorius et al., 2009, 2015; Marzaro et al., 2011; Pochi et al., 2012; Tapparo et al., 2012), with some cases of acute mortality by direct contact reported (EFSA, 2013b; Sgolastra et al., 2012; Health Canada, 2012, 2013; Pistorius et al., 2009; Girolami et al., 2012; Marzaro et al., 2011; Cutler et al., 2014; Sgolastra et al., 2017). Fugitive residues could also contaminate other environmental matrices such as surface water (Anderson et al., 2015; Bonmatin et al., 2015; Morrissey et al., 2015; Schaafsma et al., 2015; Starner and Goh, 2012) or nearby vegetation (Greatti et al., 2003, 2006; Krupke et al., 2012; Nikolakis et al., 2009). In addition to contaminated dust from treated seed, residues of neonicotinoids persisting from previous years' applications bound to airborne soil particles can contaminate off-target matrices such as surface water or foraging resources. This source, not considered previously, is at concentrations 5 orders of magnitude lower than 15 mg/g of neonicotinoids coming from fugitive residues escaping vacuum planters (Krupke et al., 2012; Limay-Rios et al., 2016).

The movement of other groups of pesticides in agricultural areas of Ontario has been studied (Hayward et al., 2010; Hoff and Grift, 1992; Meijer et al., 2003; Gouin et al., 2005; White et al., 2006) focusing mainly on organochlorines and other semi-volatile organic compounds that were applied to foliage. These compounds have a moderate vapour pressure and low water solubility, characteristics which make them widely distributed in the atmosphere (Hoff and Grift, 1992). Neonicotinoids represent a completely different scenario of low volatility because of their low vapour pressure, which explains why residues from seed treatments only occur with dust in the atmosphere.

Concentrations of neonicotinoid fugitive residue sampled under field conditions using vacuum type planters while planting neonicotinoid-treated seed have been measured previously. Tapparo et al. (2012), using active air samplers, collected the total suspended particulate (TSP) matter and dust particles less than or equal to 10  $\mu$ m in diameter (PM<sub>10</sub>) at various distances from planter exhaust having different modifications. They found average clothianidin concentrations in the range from 800 to 21,700 ng/m<sup>3</sup> and 200–700 ng/m<sup>3</sup> for TSP and PM<sub>10</sub>, respectively. In Italy, Pochi et al. (2012) using active air samplers located at different distances from the edge of the field measured neonicotinoid air concentration during planting, reported clothianidin concentrations in TSP of 546, 136 and 227 ng/m<sup>3</sup> at 5, 10 and 20 m, respectively. Using similar planter configurations and active air sampling, Biocca et al. (2014) assessed the dust drift from planting of treated seed, and found TSP mean concentrations for clothianidin ranging from 225 to 247 ng/m<sup>3</sup>.

Using passive air samplers, various authors estimated the deposition of neonicotinoid residues on the ground (Biocca et al., 2011; Pochi et al., 2015; Greatti et al., 2003, 2006; Tremolada et al., 2010; Heimbach et al., 2014; Xue et al., 2015). Xue et al. (2015) using vertical sticky traps, reported a mean neonicotinoid rate coming from planting of treated seed of 29 mg/ha with the assumption of a field of 100 m long  $\times$  150 m wide, and dust restricted to a 3-m high boundary layer. There are few data available on neonicotinoid concentration in dust plumes during planting of treated seed (Krupke et al., 2017) or during tillage by passive air samplers, and even fewer using active samplers. Both

sources of data are important when determining exposure to nontarget organisms. Furthermore, there are no published data about concentrations of neonicotinoid pesticide in those particles or possible distances that those particles can travel under field conditions in a maize ecosystem.

The main factors that could affect dust dispersion are seed quality, type of planter and air outlet, wind speed and soil conditions (Pistorius et al., 2009, 2015). It is important to understand the dynamics of movement that neonicotinoid residues originating from planting of treated seed can follow after being released to the atmosphere under realistic field conditions. This study measures the concentrations of neonicotinoid insecticides in the boundary layer originating from planting of treated maize and soybean seeds using vacuum style planters in commercial fields, with the objective of estimating how far biologically important concentrations of residues travel. These will be the first data of this kind reported for maize and soybean production in the Great Lakes region of North America.

#### 2. Materials and methods

#### 2.1. Experimental fields

This research was part of a larger study of pollinator exposure to neonicotinoid insecticides used as seed treatment in vacuum-style planters, in a commercial maize ecosystem (Limay-Rios et al., 2016; Schaafsma et al., 2015, 2016; Xue et al., 2015). The study enlisted 10 farmer co-operators who contributed a total of 31 fields where neonicotinoid-treated seed was planted during the period between 2013 and 2015. Another 8 fields randomly chosen, were used to collect samples of wind borne particulate matter during tillage and visible wind erosion events during 2014 and 2015. Fields were located in five counties (Essex, Chatham-Kent, Lambton, Middlesex, and Elgin) in southwestern Ontario, Canada. More detailed field information is given in Supplemental information Table S2.

#### 2.2. Sample collection

Particulate matter leaving agricultural fields was collected during three different field scenarios in years 2013, 2014 and 2015 using active and passive air samplers. The first scenario was during vacuum-planting of maize or soybean neonicotinoid treated seed managed under common agricultural practices for Southwestern Ontario, data collected during planting using passive sampling for year 2013 were already reported in Xue et al. (2015). The second was during tillage of fields which had a previous history of being planted with neonicotinoid treated seed. The third scenario was during wind events in non-tilled maize fields around one month after planting, and a tilled soybean field around 4 days after planting. Samplers were deployed during the total time of planting or tillage for the field sampled. Sampling devices were located at the leeward edge of the subject field (referred to as 0 m from source), and the far leeward edge of the downwind neighboring field (distances were variable, between 10 and 694 m, Table S1) always facing directly into the wind. For some locations because neighboring fields presented some conflicting operations, data for the neighboring field edge were not collected. Samplers were engaged five minutes before any activity occurred in the field, and removed 10 min after the event finished to maximize the collection period related to a field operation event. Details about planter, seed treatment and doses used for each fields are summarized in Table S2. Mean meteorological conditions were measured during each test using a pocket weather meter (Kestrel 4000), and are given in Table S3.

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