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Primary and secondary pesticide drift profiles from a peach orchard

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HIGHLIGHTS

- Concentrations of airborne pesticides were measured both during and up to 6 h after ground pesticide application in orchard.
- Detailed and prolonged vertical drift profiles are presented, with detailed measurements of key meteorological parameters.
- The effect of volatility on drift was studied by simultaneously applying two pesticides that differ in their volatility.
- Pesticide concentrations decreased with time and showed clear dependency on wind conditions.
- Overall pesticide load drifted from the orchard during primary and secondary drift were found to be comparable.

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ABSTRACT

Atmospheric drift is considered a major loss path of pesticide from target areas, but there is still a large gap of knowledge regarding this complex phenomenon. Pesticide drift may occur during application (Primary drift) and after it (Secondary drift). The present study focuses on primary and secondary drift from ground applications in peach orchard (tree height of 3 m), under Mediterranean climate. Detailed and prolonged vertical drift profiles at close proximity to orchard are presented, together with detailed measurements of key meteorological parameters. The effect of volatility on drift was also studied by simultaneously applying two pesticides that differ in their volatility.

Drifting airborne pesticides were detected both during and after applications at sampling distances of 7 and 20 m away from orchard edge. Concentrations ranged between hundreds ng m^{-3} to a few μg m^{-3} and showed clear decrease with time and with upwind conditions. Almost no decline in concentrations with height was observed up to thrice canopy height (i.e., 10 m). These homogeneous profiles indicate strong mixing near orchard and are in line with the unstable atmospheric conditions that prevailed during measurements. While air concentrations during pesticide application were higher than after it, overall pesticide load drifted from the orchard during primary and secondary drift are comparable.

To the best of our knowledge this is the first work to show such large vertical dispersion and long duration of secondary drift following ground application in orchards. The obtained information indicates that secondary drift should not be neglected in exposure and environmental impact estimations.

1. Introduction

Pesticides play an essential part in modern agriculture, but causing an increasing concern regarding undesired exposure to these chemicals. In rural communities inhalation exposure may occur following pesticides drift from treated areas (Meli et al., 2003; Rull et al., 2009). Additional public exposure to pesticides drift evolves from the fact that airborne pesticides may reach distances of even few kilometers from point of application (LeNoir et al., 1999; Schummer et al., 2009; Zivan et al., 2016) and that the interface between agricultural and urban areas is continuously growing due to urbanization.

Pesticide drift may occur during application, primary drift (PD), and after it, secondary drift (SD). While PD mostly consists of droplets, SD mostly occurs due to evaporation and hence is expected to be almost entirely in the form of vapor (Van den Berg et al., 1999). Nevertheless, it should be noticed that ground agriculture applications in orchards often take several hours. During

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such duration, PD will include also pesticide vapor that had evaporated from already treated trees (Zivan et al., 2016).

Previous measurements of PD have mainly focused on droplet deposition outside treated areas (De Schampheleire et al., 2008; de Snoo and van der Poll, 1999; Piementel and Levitan, 1986). However, common PD measurements, based on deposition collectors (Carlsen et al., 2006a; Hewitt et al., 2001), impaction collectors (Gil et al., 2008, 2007), or even remote optical techniques (Kira et al., 2015; Stoughton et al., 1997), underestimate the drift of pesticides present in gaseous phase or as fine aerosols (Cross et al., 2003). The later account for only a small portion of applied pesticide, but can be transported over much longer distances and is more relevant to inhalation exposure assessments (Ferron et al., 1988).

Sampling fine aerosols and gaseous phase is often based on active air sampling through sorbing media (Coscolla et al., 2010; EPA, 1999a; Gouin et al., 2008; Harnly et al., 2005). Few studies have conducted such measurements of airborne pesticides following crops applications (Garron et al., 2012, 2009; Nuyttens et al., 2010; Siebers et al., 2003), while information concerning orchard applications is scarce. One such example is Miller et al. (2000) who measured, using aspirated filters, PD of Malathion applied on 11 m tall Pecan trees. Pesticide was detected, at distances of 33, 66, 132, and 198 m. Although part of the drift was transported upward sampling was only at a height of 2 m (Miller et al., 2000).

Most SD studies have focused on crops (Garron et al., 2009; Houbraken et al., 2016; Leistra et al., 2006; Siebers et al., 2003) or exposed soil (Prueger et al., 2005; Rice et al., 2002). Some SD studies are based on passive long term ambient measurement (Kosikowska and Biziuk, 2010), mainly for exposure assessments (Lee et al., 2002), while others use passive dosimeters at the field to measure the effect of direct evaporation (Carlsen et al., 2006b).

In semi-arid Mediterranean climate, most pesticide applications occur during the hot and dry spring and summer months (i.e., April–August). The lack of rain and the common usage of drip irrigation in these areas (due to water scarcity) minimize wash-off from treated surfaces. Both are expected to enhance evaporation and secondary drift in these highly cultivated areas (FOCUS, 2008).

The present work aims to better understand primary and, more importantly, secondary pesticide drift evolving from treated orchards under typical Mediterranean climate and application regulation.

The pesticide drift profile reported in this study were obtained from two different campaigns conducted under several environmental conditions (September 2013 and September–October 2014). Both campaigns were conducted at the Matityahu research station (Israel Ministry of Agriculture, 33°3′49″N/35°27′15″E). Each campaign included three applications, referred to as events 1–3 and 4–6. Two different pesticides, Spiroxamine and Myclobutanil, were applied and sampled. Airborne pesticides concentrations were measured over several periods, allowing to distinguish temporal patterns, and at several heights and distances from treated orchard to explore a possible spatial pattern.

2. Methods

The two pesticide applied in this work are Spiroxamine ($Vp = 1.7\ 10^{-2}\ Pa$ @ 20 C°; $K_H = 1.07\ 10^{-7}$atm m^{3}mol^{-1} (Schummer et al., 2010)) and Myclobutanil ($Vp = 2.0\ 10^{-4}\ Pa$ @ 20 C° (Tsiropoulos et al., 2006); $K_H = 4.28\ 10^{-9}$ atm m^{3}mol^{-1} (Schummer et al., 2010)). Analytical grade pesticides were acquired from Sigma-Aldrich at purity of 98.8% and 99.3%, respectively. For orchard applications, the commercial pesticide analogs, Impulse™ (500 g l^{-1} Spiroxamine) and Sheriff-Super™ (48 g l^{-1} Myclobutanil), were acquired from local vendors.

The apple orchard studied consists of 15 tree rows, each 150 m long, orientated north to south and 5 m apart (Fig. 1). Trees height (h) was ~ 3 m and their Leaf Area Index (during 2013 Campaign) was estimated as 2.5 m^2 m^{-2} based on radiation measurements using AccuPAR LAI Ceptometer Model LP-80. Air samplers and meteorological mast were placed east of the orchard edge within an adjacent fallow field, given western winds are common in the area during daytime (Fig. 1). Only the 12 rows closest to the samplers were sprayed.

Several meteorological parameters were measured during pesticide sampling. The meteorological data included wind speed, wind direction, relative humidity, real temperature and solar radiation. In 2013, a meteorological mast supporting 6 Young ultrasonic anemometers (model 81000) at 0.5 h, 0.75 h, 1 h, 2 h, 3 h, and 5 h, was positioned on the eastern edge of the orchard (Fig. 1) for collecting average profiles typical to the micrometeorology in the orchard. It has been shown that averaged profiles collected at the downstream edge of an orchard are similar to profiles obtained within the orchard (Moltchanov et al., 2011). In 2014 (due to technical limitations), only a single meteorological station (Young ultrasonic anemometer model 05103) was positioned at a height of 2.5 m and at a distance of 7 m.

The commercial pesticides were applied per local application regulations using a fan assisted boom sprayer equipped with ASJ-jet HCI8001 nozzles operating at a pressure of 8 bar with an air speed of 30 m s^{-1}.The sprayer was mounted on a tractor driving at 3 km h^{-1}. An amount of 42 mg m^{-2} and 4 mg m^{-2} for Spiroxamine and Myclobutanil, respectively, were applied simultaneously on a total area of 12000 m^2. An exception was in the first event of the 2013 campaign where only Spiroxamine was applied.

Air samples were collected onto Polyurethane foam (PUF) plugs (length 3 cm, diameter 5.6 cm) placed in fabricated glass holder attached to an industrial blower (GAST R-4110-2) via rotameters to control flow rates. The glass holders were placed at different heights using a Clark-Mast® telescopic pneumatic mast. Sampling locations and heights differed between campaigns: In 2013 (events 1–3) only one mast was deployed at 7 m distance from the orchard eastern edge with four sampling heights: 1.5, 3, 4.5, and 6 m above ground. In 2014 (events 4–6), two masts were deployed: first mast at 7 m from the eastern orchard edge with five sampling heights of 2, 4, 6, 8, and 10 m; second mast was placed at 20 m distance using the same sampling heights as the first mast, excluding 2 m (Fig. 1b).

Sampling intervals were divided into the following categories: PD – primary drift during application period, SD-1 – Secondary drift during first two hours after application, SD-2 – Secondary drift during third and fourth hours after application. In a few of the events (events 1, 4 and 5) SD-3 was also measured (fifth and sixth hours after application), while in events 5–6, air concentrations were also monitored during the consecutive nighttime (22:00 to 06:00 local time). All applications started between 6:00 and 15:00 (see Table S1 for detailed data). In 2013 two sprayers were used simultaneously to reduce the application time. In 2013 Applications required about 20 min and in 2014 40 min.

PUF samples were kept at −4 °C until extracted. Extraction was conducted using an Accelerated Solvent Extractor (Dionex, ASE-150) system. Extraction was done with hexane: diethyl ether (90:10 by volume) as solvent (EPA, 1999b), at 100 °C and 1500 *psi*. Extraction program included two statics cycles of 20 min each with a flush volume of 50% cell volume, using 66 ml stainless steel cells. Extract volume was then reduced to 1 ml using TurboVap-II system and centrifuged for 10 min @4000 rpm to remove any solid residue. Recovery efficiencies for Spiroxamine (93%; n = 3) and