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Polyethylene film incorporation into the horticultural soil of small periurban production units in Argentina

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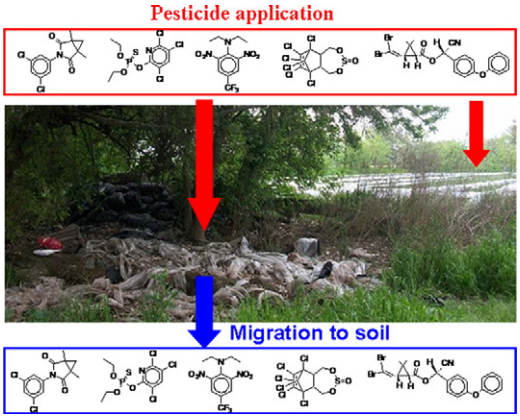
HIGHLIGHTS

- PE plastic residue was found in horticultural soils (equals to 10% of the area).
- The predominant fragment size was (28 $cm^2 \pm 13$ cm^2).
- Pesticides can migrate to the inside of the PE film.
- Further bidirectional migrations between soil and plastic film can occur.

GRAPHICAL ABSTRACT



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ABSTRACT

Horticulture makes intensive use of soil and extensive use of polyethylene (PE) sheeting and pesticides, producing an environment where the dynamics between soil and plastics can affect pesticide fate. We have determined that the presence of plastic residues in the horticultural soil of small production units equals 10% of the soil area, being meso and macro-sections the predominant fragment sizes. All soil samples were taken from different plots located in Cuartel V, Moreno district, in the suburbs of Buenos Aires city, Argentina. Laboratory experiments were conducted to see the relations among pesticide, soil and PE film. Endosulfan recovery from LDPE films (25 μm and 100 μm) was studied, observing evidence that indicated migration to the inside of the plastic matrix. To further analyze the dynamics of pesticide migration to soil and atmosphere, experiments using chlorpyrifos, procymidone and trifluralin were performed in soil–plastic–atmosphere microenvironments, showing that up to 24 h significant amounts of pesticides moved away from the PE film. To determine whether PE residues could act as potential pesticide collector in soil, column elution experiments were done using chlorpyrifos, procymidone and trifluralin. Results showed an important pesticide accumulation in the mulch film (584 μg–2284 μg pesticide/g plastic) compared to soil (13 μg–32 μg pesticide/g soil). Finally, chemical and photochemical degradation of deltamethrin adsorbed in PE film was studied, finding a protective effect on hydrolysis but no protective effect on photodegradation.

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We believe that a deeper understanding of the dynamics among soil, plastic and pesticides in horticultural productive systems may contribute to alert for the implications of PE use for plastic sheeting.

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1. Introduction

Horticultural productivity has partially increased due to plastic sheeting (Espí et al., 2012) and pesticide application (Hillocks, 2012). PE sheeting is used in greenhouses, walk-in tunnels, low tunnel covers and mulching applications. More than 80% of the agricultural film sold worldwide is made of low density polyethylene (LDPE), mainly used in greenhouse 150 μm–200 μm films containing ethyl vinyl acetate (EVA) as a copolymer. The mean lifetime of greenhouse sheeting is 6–8 months. Mulch film is the second most common agricultural film application, usually as black monolayer LDPE 12 μm–80 μm film, with a mean lifetime of 2 to 4 months. China, Japan and South Korea represent about 80% of the worldwide mulched surface, employing 700,000 t LDPE/year for this purpose (Espí et al., 2012). Plastic residues of various sizes have been reported in the ocean and freshwaters (McCormick et al., 2014; Rochman et al., 2004); plastic particles are also common in soil, but are infrequently measured (Rilling, 2002).

Intensive pesticide use is another factor affecting horticultural productivity (Cooper and Dobson, 2007; Nakajima and Ortega, 2014). Studies show the negative environmental aspects related to the application of pesticides, including their presence on non-target systems such as water (Licciardello et al., 2011; Masiá et al., 2015a,b; Oliver et al., 2012), soil (Jacobsen and HjelmsØ, 2014) and non-target organisms (Damalas and Eleftherohorinos, 2011).

In horticulture, the concurrent combination of intensive pesticide use, plastic sheeting and soil exploitation sets an environmental scenario that requires further attention. Fragments of plastic sheeting may represent a novel vector of pesticides into soil, considering the significant amount of pesticides that reach these plastic covers during the application process (Querejeta et al., 2012).

It has been reported that plastic film can absorb pesticides in aqueous solutions (Nerín et al., 1996) and that these molecules can be desorbed from plastic surfaces to organic solvents (Nerín and Battle, 1999). The development of PE membrane passive pesticide samplers (Khairy et al., 2014) is based on this phenomenon. Pesticides migrate through the polyethylene pores in the non-crystalline areas of the film (Huckins et al., 1993). It has also been shown that volatile molecules like methylene bromide, propargyl bromide or chloropicrin can permeate polyethylene films in the gaseous phase (Papiernik et al., 2001). The migration of small organic stabilizers from plastic films has also been reported (Haider and Karlsson, 2001).

In previous pesticide degradation studies in horticultural soils (Querejeta et al., 2014), we noticed the ubiquitous presence of plastic residues. Taking into account that pesticide mobility and degradation in soil may be affected by the presence of plastic fragments, we aimed at quantifying the presence of plastic residues in horticultural soil and studying the interactions among plastic films, pesticides and soil.

2. Materials and methods

2.1. Chemicals and solvents

Chemicals and solvents were of the best analytical grade. To prepare each reference material, technical grade pesticides were purified by recrystallization (>95% pure by GC-FID). The identity and purity of the active principles were confirmed by ^{1}H- and ^{13}C-NMR. A primary solution of 300–1000 ppm w/w was prepared in acetone or cyclohexane, and the working solutions were obtained by dilution as needed. Acetone and cyclohexane (Aberkon p.a. grade) used for all solutions and extracts were previously distilled and chromatographically checked as suitable for GC-ECD use.

2.2. Pesticides

Endosulfan, chlorpyrifos, procymidone, trifluralin and deltamethrin were chosen for this study due to their extensive use in horticultural and floricultural production units in Argentina (Berenstetin et al., 2014; Flores et al., 2011; Hughes et al., 2008; Querejeta et al., 2012; Querejeta et al., 2014; Ramos et al., 2010). Additionally, endosulfan was selected for swabbing and immersion experiments due to its high stability. Chlorpyrifos, procymidone and trifluralin were used for migration trials in microenvironments and columns, as three different examples of hydrophobic pesticides with different volatility. Finally, deltamethrin was adopted for chemical and photochemical degradation studies because of its fast reaction kinetics under the described experimental conditions.

Commercial products used in the laboratory experiments were as follows:

- Endosulfan (6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9a-hexahydro-6,9-methano-2,4,3-benzodioxathiepine-3-oxide), CASRN [115-29-7]): Thionex® (EC, 35% w/v, Magan).
- Procymidone (3-(3,5-dichlorophenyl)-1,5-dimethyl-3-azabicyclo [3.1.0]hexane-2,4-dione, CASRN [32809-16-8]) liquid: Sumilex® (CS, 50% w/v) (Summit Agro Argentina); and solid: Sumilex® (WP, 50% w/w) (S. Ando Argentina).
- Chlorpyrifos (*0,0*-diethyl-*0*-(3,5,6-trichloro-2-pyridinyl)-phosphorothioate, CASRN [2921-88-2]), Lorsban® (EC, 48% w/v, Dow AgroSciences)
- Trifluralin (α,α,α trifluoro-2,6-dinitro-N, N-dipropyl-p-toluidine), CASRN [1582-09-8], Trigermin® (CS, 48% w/v, Cheminova)
- Deltamethrin ((*S*)-α-cyano-3-phenoxybenzyl-(1*R*,3*R*)-3-(2,2-dibromovinyl)-2,2-dimethylcyclopropanecarboxylate, CASRN [52918-63-5]): Decis Forte® (EC, 10% w/v) (Bayer CropScienceArgentina).

2.3. Plastics used in the assays

Two types of plastic PE film were used: LDPE black 25 μm mulch film and LDPE crystal 100 μm film with EVA (used for tunnels and greenhouses). The specific kinds of plastic used are defined in each experiment.

2.4. Chromatographic conditions

All chromatographic analysis were performed on a Perkin-Elmer (Norwalk CT, USA) AutoSystem XL Gas Chromatograph with Autosampler automatic injector, equipped with an electron capture detector (ECD), and a fused silica capillary column (PE-5, 5% diphenylpolysiloxane — 95% dimethylpolysiloxane stationary phase, 30 m length, 0.25 mm i.d. and 0.25 μm film thickness). The GC-ECD operating conditions were injector temperature: 280 °C; ECD temperature: 375 °C; oven temperature: 190 °C for 1.5 min, 45 °C min^{-1} to 300 °C then 10 °C min^{-1} to 320 °C and hold 2 min; injection volume 1 μL, splitless; carrier gas: N_2, 30 psi; and ECD auxiliary flow 30 mL min^{-1}.

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