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Environmental pesticide distribution in horticultural and floricultural periurban production units

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ABSTRACT

The environmental pesticide distribution on non-target systems (soil, drift and agricultural plastics) during the application step at small periurban production units, was studied in open field and greenhouses, for different crops (tomato, lettuce, broccoli, strawberry and flowers) using different pesticides (endosulfan, procymidone, chlorothalonil, chlorpyrifos and deltamethrin). In all cases, soil was the most exposed non-target system. For greenhouses, a general pesticide distribution was found of approximately 2/3 for crop, 1/4 for soil and 1/20 for plastic, of the total amount applied. In horticultural open fields, although the distribution was very dependent on the crop size and type, soil was also the most exposed non-target subsystem. Pesticide drift seems not to be significant in these production units, whilst pesticide accumulation on agricultural plastics reached up to 45% of the total applied, for polyethylene mulching in strawberry fields.

1. Introduction

The use of pesticides in modern agriculture has contributed to a sustained expansion of this activity in the last decades, mainly due to an increase in crop yields (Dias Ávila et al., 2010). Nevertheless, some negative impacts related to their application have been recognized, especially for the organochlorine pesticides, as consequence of their environmental persistence (Lv et al., 2010). In terms of their ideal performance, pesticides should reach the target organism, and having achieved its intended effect, should be decomposed rapidly into harmless compounds. Unfortunately, real pesticides perform differently under real application conditions, and the fate of these molecules directly affects not only the crops, but also non-target systems, like applicators (Flores et al., 2011), consumers (Harris et al., 2001), soil (González et al., 2010), non-target animals (Walker et al., 2010) and water sources (Mugni et al., 2011).

With the aim of improving the quality of environmental exposure information related to pesticides, and developing better predictive scenarios, experimental studies of pesticide environmental distribution are needed (Rice et al., 2007). This subject is particularly important in small scale periurban horticultural (Ramos et al., 2010) and floricultural (Flores et al., 2011) production units such as those surrounding Buenos Aires city. In these particular cases, potential environmental problems can be magnified by poor working conditions, lack of education, low technology, manpower dependence, and their geographical location close to the urban borders. In this context, our group has been studying labourer's exposure to pesticides (Hughes et al., 2008; Ramos et al., 2010; Flores et al., 2011), finding that other non-target subsystems like soil, plastic covers, and neighbouring fields may be significantly exposed. Thus, a quantitative estimation of non-target exposure levels is relevant in order to evaluate the fate of the phytosanitary products in these particular subsystems.

Unfortunately, non-target pesticide distribution in small scale production units, with manual application, has been scarcely investigated. Studies have been done in relation to the product drift outside the crop field. Snelder et al. (2008) reported that under manual application conditions in rice crops, drift shows a rapid decrease at over 0.5 m from the treated field edges. The same trend was observed by García-Santos et al. (2011) for a potato crop using a knapsack sprayer. In relation to soil, it is clear that the amount of pesticide that directly reaches this matrix during the application has profound effects on its biological state; additionally these substances can be transported to water resources, spreading the contamination. Although detailed studies analysing the fate of pesticides in different soils can be found in literature (Flores et al.,

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2009; González et al., 2010), to our best knowledge no systematic study of the initial amount of pesticide that is directly deposited on soil during the application process was performed at small scale production units.

Plastic sheeting is another important matrix affected by pesticides; this material is used for greenhouse construction or mulching purposes (Nerín et al., 1999). In this sense, most research has been directed to the study of the absorption phenomena of some organochlorine and organophosphate pesticides, mainly in low-density polyethylene (LDPE), (Nerín et al., 1996), and on assessing the recycling suitability of the LDPE used in mulching practices (Nerín et al., 1999).

This report presents an estimation of the distribution of endosulfan, procymidone, chlorothalonil, chlorpyrifos and deltamethrin on different non-target subsystems such as soil, plastic and neighbouring fields, during manual pesticide application in open and covered horticultural and floricultural production units with different crops.

2. Experimental methods

2.1. Study sites and conditions

Field trials were carried out as follows, using the producers' own equipment, (usually manually-operated backpacks); the spray-nozzle most used was a JD-12P.

2.1.1. Horticultural (H) and Floricultural (F) greenhouses

Horticultural greenhouses: experiments with tomato and lettuce were made at the Estación Experimental INTA San Pedro (San Pedro, Prov. de Buenos Aires, Argentina), during winter 2010 (15th July), using manual knapsacks, operating at flow rates between 0.76 and 1.11 L min^{-1}.

2.1.1.1. Experiences H1–H6. The tomato greenhouse was 52 m long and 8 m wide, with plants in 10 m × 1.5 m plots with 0.6 m separation; the distance between plots and walls was 1.7vm to rear and 1.3 m to sides. The lettuce greenhouse was 12.2 m long and 7.9 m wide, planted in 10 m × 1.5 m plots with 1 m separation; distance between plot and greenhouse walls was 1.5 m to back and 1.1 m to sides. In both instances inside weather conditions were: 10–15 °C, 20–43% relative humidity, 1012 hPa. Products applied were: endosulfan (H1–H4), procymidone (H2–H5), and chlorothalonil (H3–H6). All greenhouses have a lateral window along one side that was kept open during the pesticide application.

Floricultural greenhouses: measurements were made in three different commercial production sites in Moreno district (Prov. de Buenos Aires, Argentina). A manual knapsack was used in all cases, with flow rates between 0.31 and 1.0 L min^{-1}. Pots were always placed with minimum spacing, practically touching each other.

2.1.1.2. Experience F1. Chlorothalonil was applied during summer 2010 (December 10th), in a greenhouse 12 m long and 22 m wide. Seedling pots were laid out in columns of 2.1 m width separated by 0.6 m, and 0.1 m from the plastic wall. Plants were geranium (*Pelargonium hortorum*), fuchsia (*Fuchsia* hybrid), balsam (*Impatiens walleriana*). Indoor conditions were: 35 °C, 1005 hPa, 31% relative humidity, no wind.

2.1.1.3. Experience F2. Chlorpyrifos was applied during summer 2011 (February 11th), in a greenhouse 38 m long and 18 m wide. Geranium (*Pelargonium hortorum*) pots were laid out in an 18 m × 1 m plot, with 0.4 m aisles on three sides, and 0.1 m from the plastic wall. Indoor conditions were: 29 °C, 1011 hPa, 43% relative humidity, no wind.

2.1.1.4. Experience F3. Chlorpyrifos was applied during autumn 2011 (April 28th) in a greenhouse 18 m long and 6 m wide; with pots in 1.2 m plots separated by 0.4 m, and a lateral and rear separation from the plastic walls of 0.1 m and 0.4 m respectively; seedlings were gazania (*Gazania* hybrid), petunia (*Petunia* hybrid), indian tobacco (*Lobelia inflata*) and maiden pink (*Dianthus deltoides*). Indoor conditions were: 25 °C, 1012 hPa, 55% relative humidity, no wind.

2.1.2. Horticultural open fields

In all cases deltamethrin was applied using a manual knapsack, except in H15, where a pressurized hose with central pump was used. The flow rate of these applications was between: 0.94 and 1.33 L min^{-1}.

2.1.2.1. Experience H7–H10. Broccoli (H7–H9) and cauliflower (H10) of similar size and distribution, cultivated in a commercial plantation in Moreno district, during autumn 2008 (March–June). Plants were grown in rows 60–80 m long (0.85 m separation, 0.30 m between plants). Weather conditions were: 20–30 °C, relative humidity: 40–50%, 1005–1015 hPa, wind in intermittent gusts under 6 km h^{-1}. All plants were ready for harvest.

2.1.2.2. Experiences H11 to H13. Strawberries, grown in a commercial plantation in Moreno district. Experiments were done during winter 2008 (June–August). Plants were grown using black polyethylene mulching in ridges 90 m long, 1 m wide with 0.20 m furrows between rows. Seedlings were planted in holes in a zig-zag pattern at 0.20 m distance. Weather conditions were: 13–20 °C, relative humidity 26–40%, 1012–1016 hPa, wind in intermittent gusts under 12 km h^{-1}. Plants were between 4 and 6 months old.

2.1.2.3. Experiences H14 and H15. Lettuce, grown in a commercial plantation in Escobar district (Buenos Aires, Argentina), during summer 2011 (15th July). Plants were 15 d from transplant (5–7 leaves), 8×10^4 plants ha^{-1}, in rows 1.4 m wide. Weather conditions were: 22–24 °C, relative humidity 45–54%, 1016 mm Hg, wind irregular gusts up to 14 km h^{-1}.

2.2. Reagents, materials and chromatographic conditions

The pesticides applied were the following commercial formulations: 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 CropScience Argentina); procymidone (3-(3,5-dichlorophenyl)-1,5-dimethyl-3-azabicyclo[3.1.0]hexane-2,4-dione, CASRN [32809-16-8]), Sumilex® (CS, 50% w/v) (Summit Agro Argentina); endosulfan ((6,7,8,9,10,10-hexachloro-1,5,5a,6,9,9α-hexahydro-6,9-methano-2,4,3-benzodioxathiepine-3-oxide), CASRN [115-29-7]), Thionex® (EC, 35% w/v, Magan); chlorothalonil (2,4,5,6-tetrachloroisophthalonitrile, CASRN [1897-45-6]), Daconil® (EC, 72% w/v, Syngenta); chlorpyrifos (*0,0*-diethyl-*0*-(3,5,6-trichloro-2-pyridinyl)-phosphorothioate, CASRN [2921-88-2]), Lorsban® (EC, 48% w/v, Dow AgroSciences). Reference materials were prepared by recrystallization (>95% pure by GC-FID), and confirmed by ^1H and ^{13}C NMR. Primary solutions (100-600 ppm w/w) were prepared in cyclohexane, and diluted as needed. Cyclohexane (Aberkon p.a. grade) was used for all solutions and extracts, distilled prior to use and chromatographically checked as suitable for use under GC-ECD 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; ECD auxiliary flow 30 mL min^{-1}.

2.3. Sampling method and field procedure

Environmental pesticide distribution was studied by sampling these products in different subsystems: crop, soil, drift and plastic, in greenhouses and open fields after pesticide application. Cotton cloth was used as collector material placed on representative locations for wrapping whole plants; in special cases plants were cut off at ground level and rinsed whole with solvent.

2.3.1. Greenhouse experiences

For sampling pesticides from greenhouse plastics, 20 cm square cloth samplers were used (lined on one side by a polyethylene film to avoid external contamination) placed on the walls at three different heights (low: L, medium: M, high: H, Fig. 1) and roof, discriminating between the crop roof (RC) and the aisle roof (RA), as shown in Fig. 1. For measuring tomato plant exposure, because its shape prevented wrapping, the pesticide was extracted directly from the plant; the same procedure was used for small flower seedlings. Bigger plants and lettuces were individually wrapped using a piece of cloth. Pesticide falling on soil was sampled with strips of cloth covering the width of the exposed surface. Drift was sampled on the downwind side of the greenhouse, with 20 cm square samplers as previously reported (Ramos et al., 2010).

2.3.2. Open field experiences

For broccoli 75 cm × 20 cm samplers (6 replicates) were used for soil (from the top of a row, over the furrow to the next row), 80 cm × 75 cm for wrapping plants (6 replicates) and 122 cm × 20 cm samplers (6 replicates) for measuring drift, placed vertically on the downwind edges of the field. In strawberry fields, 25 cm × 30 cm samplers (6 replicates) were used for the furrow (exposed soil), 130 cm × 20 cm strips covered the plastic mulch (6 replicates), 70 cm × 50 cm samplers (6 replicates, each wrapping two units) were employed for plants, and 122 cm × 20 cm strips (7 replicates) placed vertically on the downwind edges of the field for measuring drift. For lettuce field studies, samplers were: 100 cm × 20 cm (6 replicates) for soil, 25 cm × 25 cm (6 replicates) for crop, and 20 cm × 20 cm (20 replicates) for measuring drift; these samplers were fixed to the ground at 0.5, 1.0, 2.5 and 7 m from the downwind edges of the field (see Supplementary material).

2.4. Extraction and analysis

Each of the cloth samplers was extracted separately in the laboratory with cyclohexane (20 min, rotary shaker with solvent volume depending on the section size e.g. 100 mL for 20 cm square sections) not later than 8 h after the field trial. Whole plants were placed in appropriate containers and shaken manually with cyclohexane (e.g. tomato plants with 3 L of solvent). All extracts were analyzed by GC-ECD, under the conditions previously described.

2.5. Method validation

Experiments were performed in order to investigate if deltamethrin, chlorothalonil, procymidone, chlorpyrifos and endosulfan suffered decomposition or were otherwise lost on the cotton cloth used for sampling. No loss was observed for storage periods under 24 h.

Pesticides linear range was studied finding that deltamethrin, chlorothalonil, procymidone, chlorpyrifos and endosulfan responses were linear between the following ranges 0.11–1.6 mg L^{-1} ($R^2 > 0.97$), 0.04–0.64 mg L^{-1} ($R^2 > 0.98$), 0.04–0.57 mg L^{-1} ($R^2 >$

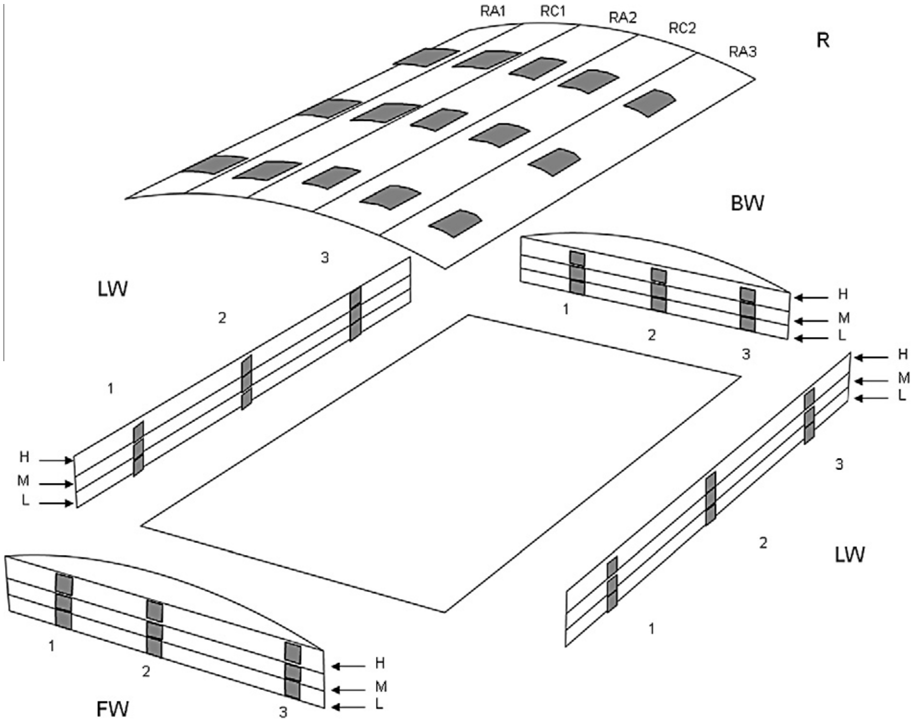

Fig. 1. Greenhouse sampling scheme and section definition.

Table 1
Pesticide distribution in horticultural and floricultural greenhouses subsystems.

Section	Pesticide percentage distribution in greenhouses (%)								
	Horticultural (tomato)			Horticultural (lettuce)			Floricultural		
	H1[a]	H2[b]	H3[c]	H4[d]	H5[e]	H6[f]	F1[g]	F2[h]	F3[i]
Plastic	4.0	3.9	3.1	0.06	0.16	0[j]	0.70	19.5	1.5
Soil	27.9	39.7	17.1	12.3	24.4	55.9	15.2	51.9	2.3
Crop	68.1	56.5	79.8	87.8	75.9	44.1	84.1	28.6	96.2
Drift	0[j]	0[j]	0.05	NM[k]					

[a–i] Total pesticide mass found were as follows: [a]1610.0 mg of endosulfan, [b]739.5 mg of procymidone, [c]3554.8 mg of chlorothalonil, [d]3716.4 mg of endosulfan, [e]1284.7 mg of procymidone, [f]397.2 mg of chlorothalonil, [g]8051.9 mg of chlorothalonil, [h]1343.8 mg of chlorpyrifos, [i]7193.5 mg of chlorpyrifos.

[j] Means that pesticide was non-detected using the previously described methodology.

[k] NM: not measured.

0.98), 0.06–1.07 mg L^{-1} ($R^2 > 0.968$), and 0.03–0.49 mg L^{-1} ($R^2 > 0.994$) respectively. The lowest points of each calibration curve were considered as the limit of quantitation. The precision was studied by injection of a complete calibration curve for deltamethrin, chlorothalonil, procymidone, chlorpyrifos and endosulfan by duplicate on six consecutive days and calculating the standard deviation of the slope of the calibration curves. A variation of 26% was found for deltamethrin, 25% for chlorothalonil, 10% for procymidone, 19.3% chlorpyrifos and 11.4% for endosulfan.

For all cases except tomato plants and the small flower seedlings, the calibration curves were made by spiking 10 cm square cotton cloth samples. For the tomato plants, blank runs were made by injection of extracts of this matrix obtained by rinsing plants not exposed to the pesticide. In this case pesticide recovery was estimated founding values between 83% and 115%, when samples were processed not later than 8 h after the field trial.

3. Results

3.1. Pesticide distribution in greenhouses and open fields units after product application

Table 1 shows the pesticide distribution after the product application in horticultural and floricultural greenhouses, and horticultural open fields, expressed as a percentage of the total product applied. This parameter permits the comparison of different situations in which diverse pesticides were applied on various crops, in different concentrations and volumes. The subsystems studied were: crops, plastic covers, soil and drift. The experiments were done applying deltamethrin, endosulfan, procymidone, chlorpyrifos and chlorothalonil, with manual knapsacks, in independent trials on different production units, under real working conditions with different workers. The horticultural crops studied were tomato, lettuce, broccoli and strawberries, while in the floricultural cases, potted plant seedlings were of a wider variety.

It is interesting to note that the greenhouse drift in the tomato case is practically negligible, being non-detectable in two experiments (H1, H2, Table 1) and finding only 0.05% of the total chlorothalonil applied in the third case (H3, Table 1), which was equivalent to 1.7 mg of this product.

In relation to the other subsystems, the crop was the most exposed system in seven of the nine measurements (H1–H5, F1, F3), ranging from 56.5% to 96.2% of the total applied pesticide. Only in one case for lettuce (H6, Table 1) and one other for floriculture (F2, Table 1), was soil the most exposed system.

Greenhouse plastic sheeting was the least exposed subsystem, with relative amounts varying from 3.1% to 4.0% for tomato, from non-detected to 0.16% for lettuce and from 0.70% to 19.5% for floricultural greenhouses. In this last case (F2, Table 1), the relative percentage of chlorpyrifos found on plastic was twelve times the mean pesticide amount found for the rest of the experiments. This

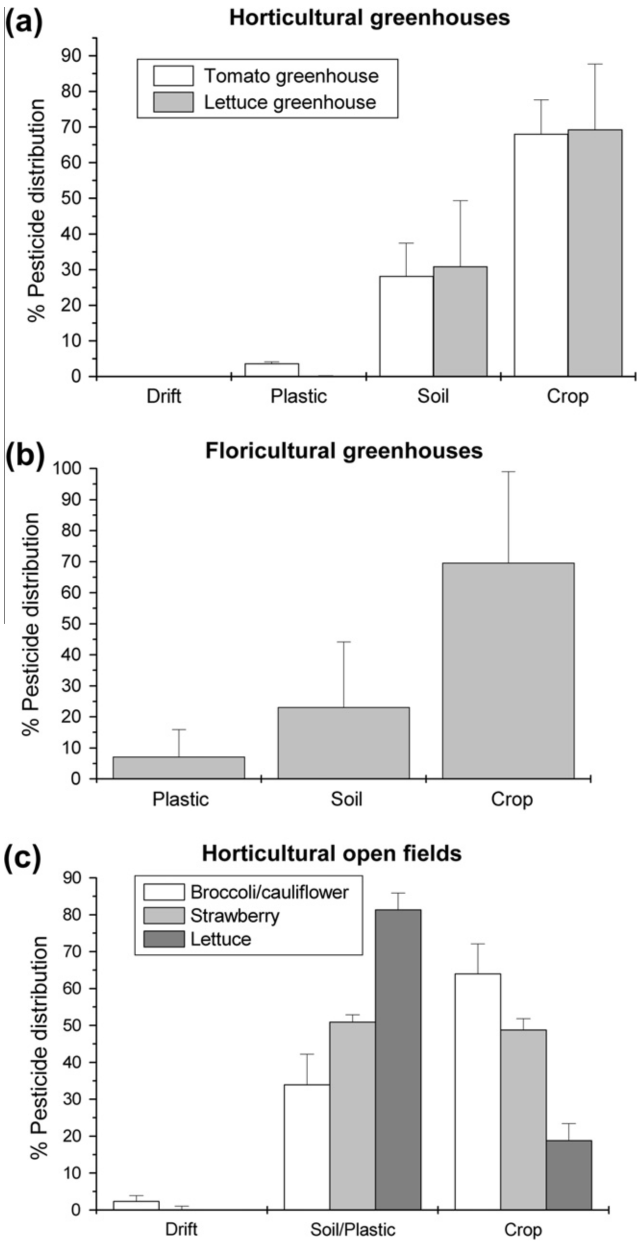

Fig. 2. Pesticide relative distribution in horticultural and floricultural greenhouses and open fields.

Table 2
Pesticide distribution in horticultural open fields.

Section	Pesticide percentage distribution in horticultural open fields (%)								
	Broccoli/cauliflower				Strawberry			Lettuce	
	H7[a]	H8[b]	H9[c]	H10[d]	H11[e]	H12[f]	H13[g]	H14[h]	H15[i]
Crop	50.0	67.8	67.5	73.5	56.0	45.6	44.7	14.2	23.3
Soil	48.2	27.2	30.9	25.5	4.5	5.4	2.9	85.8	76.7
Drift	1.8	5.0	1.5	1.0	NM[k]			0.034	0.16
Plastic	–[j]				38.8	48.9	52.2	–[j]	

[a–i] Total deltamethrin mass found were as follows: [a]509 mg, broccoli, [b]1615 mg, broccoli, [c]1925 mg, broccoli, [d]688, cauliflower, [e]6764 mg, [f]973.9 mg, [g]610 mg, [h]28.9 mg, [i]25.7 mg.

[j] No plastic was used in these cases.

[k] NM: not measured.

could be explained by the worker's incorrect application procedure, which would agree with the fact that half of the total fell on the soil instead of on the crop as occurred in the other cases. Fig. 2a and b shows the mean pesticide relative distribution on plastic, soil, crop and drift subsystems, for horticultural and floricultural greenhouses. Endosulfan, procymidone and chlorothalonil relative percentage distribution were averaged for horticultural results, and chlorothalonil and chlorpyrifos for floricultural cases, indicating on each bar the standard deviation of each average. For tomato and lettuce crops, a mean of 68–69% of the pesticide was found on the plants, and 28–30% on soil. While in lettuce greenhouse the amount of pesticide found on plastic was negligible, in the tomato and floricultural cases the amount was significant (mean values of 3.7% and 7.2 % respectively). It is interesting to emphasize the similar pesticide distribution between crop, soil, plastic and drift in greenhouses, independent of the crop type (Fig. 2a and b).

Table 2 shows the pesticide distribution between crop, soil and drift, for broccoli, strawberry and lettuce open field plantations. As plastic mulching was used for strawberry, the pesticide content of these plastic covers was also measured. All pesticide applications were done using manual knapsacks, with the exception of H15 where a pressurized line was employed. It is interesting to note, that in the broccoli and lettuce applications, the drift to surrounding land was quite low, ranging from 0.034% to 0.16% for lettuce (H14, H15, Table 2) and from 1.0% to 5.0% for broccoli and cauliflower (H7–H10, Table 2). These percentages were equivalent to 9.8–41.1 µg of deltamethrin for H14–H15, and 28.9–80.7 mg of the same product for H7–H10. For lettuce plots, drift outside the crop boundaries was measured with both spray systems (knapsack and pressurized line), studying the deltamethrin amount found as function of distance from the crop edges. Fig. 3A shows the pesticide mass (expressed in µg) when a pressurized line was used, and in Fig. 3B the same information is given for the manual knapsack application (for crop scheme and sampler locations see Supplementary material).

In relation to the other subsystems, results in Table 2 indicate that crop was the most affected matrix for broccoli and strawberry, but not for lettuce, at least at the reported growth stage. It is interesting to remark that if soil and plastic pesticide percentages are considered together for strawberry, which could happen if plastic mulching is not employed, the ground turns out to be a highly exposed matrix, with relative percentages of 43.3–55.1% (equivalent to 335.1–2907.2 mg of deltamethrin). For broccoli the relative soil exposure range was 27.2–48.2% (equivalent to 245.0–439.2 mg of deltamethrin, and 76.7–85.8% for lettuce (equivalent to 19.7–24.8 mg of the same pesticide). Fig. 2c shows the mean pesticide percentages and their standard deviations for broccoli, strawberry and lettuce in open field productions: for the first two, soil and crop showed almost the same relative pesticide exposure, whereas in lettuce fields most of the phytosanitary product was found on the soil.

3.2. Pesticide distribution in floricultural and horticultural plastic covers

Considering the exposure of plastics to pesticides, both in greenhouses (Table 1, H1–H6, F1–F3) and open fields (Table 2,

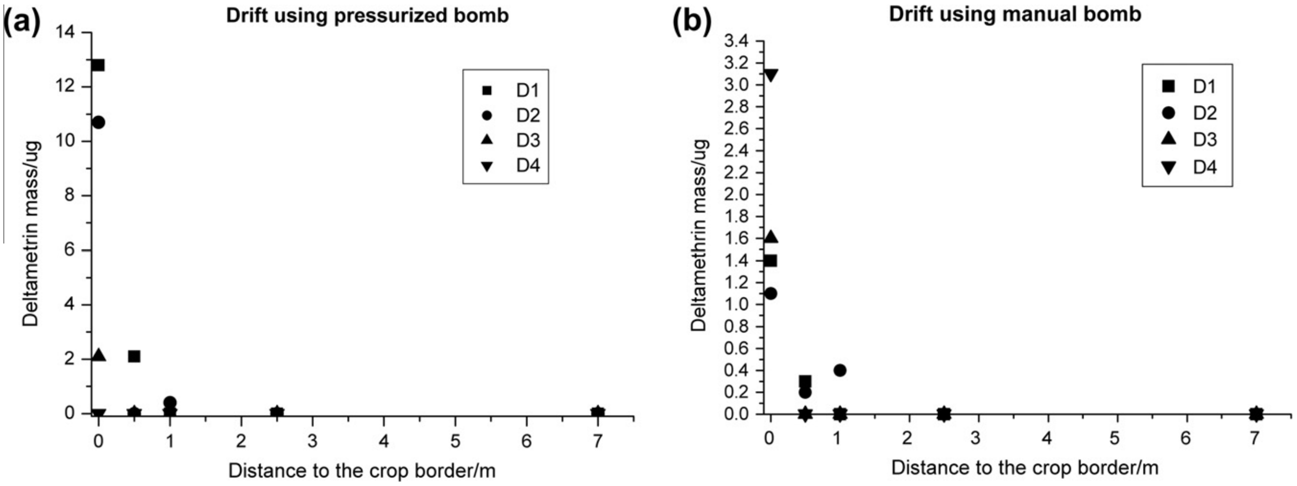

Fig. 3. Pesticide drift as distance function to crop border during deltamethrin application on lettuce.

Table 3
Pesticide distribution in horticultural and floricultural greenhouse plastic.

Section codes[a]	Pesticide percentage distribution in greenhouse plastic (%)								
	Horticultural (tomato)			Horticultural (lettuce)			Floricultural		
	H1[b]	H2[c]	H3[d]	H4[e]	H5[f]	H6[g]	F1[h]	F2[i]	F3[j]
LWL	3.4	2.0	10.7	9.7	0	0	25.9	97.9	66.8
LWM	5.1	3.9	9.4	–[k]			14.1	0.94	1.6
LWH	42.9	8.5	6.0	–			0.86	0.28	1.0
FWL	0[j]	0	0	0	0	0	10.3	–	–
FWM	0	0	0	0	0	0	4.1	–	–
FWH	0	0	0	0	0	0	1.5	–	–
BWL	8.4	12.2	4.9	NM[m]			0.24	0.85	0.18
BWM	9.1	34.8	19.8	NM			0	0.02	0.14
BWH	1.5	8.8	28.0	NM			0	0.03	0.17
RA	2.4	20.4	5.8	30.9	35.6	0	0	0	0
RC	18.7	9.7	52.2	59.4	64.4	0	0	0.39	30.1

[a] Code sections are as follows: lateral wall lower: LWL, lateral wall medium: LWM, Lateral wall high: LWH, front wall low: FWL, front wall medium: FWM, front wall high: FWH, back wall low: BWL, back wall medium: BWM, back wall high: BWH, roof aisle: RA, roof crop: RC.

[b–j] Total pesticide on plastic were as follows: [b]64.6 mg of endosulfan, [c]28.8 mg of procymidone, [d]109.1 mg of chlorothalonil, [e]2.2 mg of endosulfan, [f]2.0 mg of procymidone, [g]chlorothalonil below the detection limit, [h]56.9 mg of chlorothalonil, [i]262.8 mg of chlorpyrifos, [j]108.6 mg of chlorpyrifos.

[k] Means that pesticide was non-detected using the previously described methodology.

[l] The hyphen means that this section did not exist in the particular greenhouse.

[m] NM: not measured.

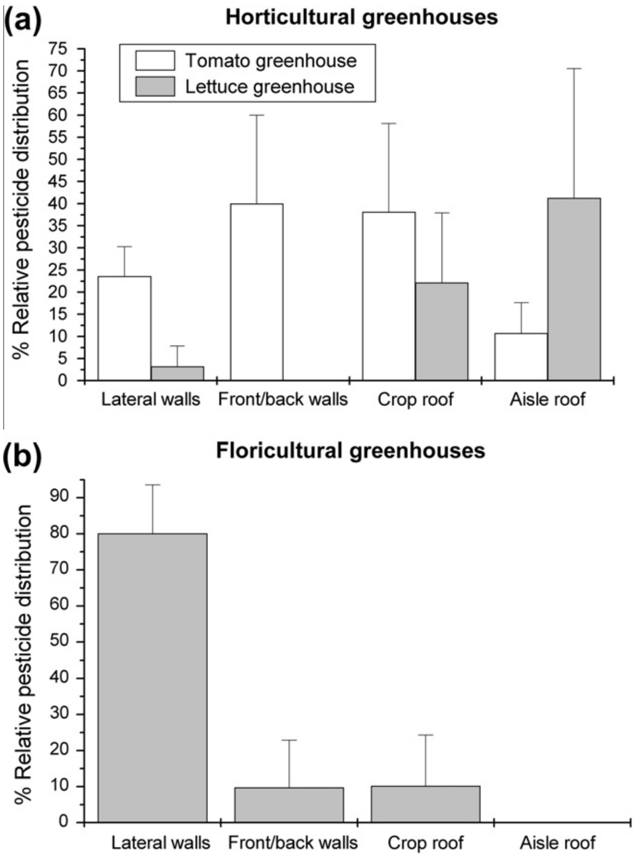

Fig. 4. Pesticide relative distribution on the greenhouse plastic.

H11–H13), it is clear that these surfaces could be affected by significant amounts of phytosanitary products. The polyethylene film used for mulching in strawberry crops was the most contaminated surface (H11–H13, Table 2), with relative deltamethrin percentage distribution values from 38.8% to 52.2% of the total applied product (equivalent to 318.3–2622.3 mg of pesticide), indicating that practically half of the total pesticide applied was directly deposited on the plastic.

A different situation was found for the plastic used in greenhouse construction. In horticultural experiences the pesticide deposited on the polymer surface after the application to a tomato crop, ranged from 3.1% to 4.0% of the total applied product (equivalent to a 28.9–109.1 mg range, footnotes Table 3); for the lettuce case, the amount of pesticide on the plastic was significantly lower, varying from not detected to 2.2 mg of product (footnotes, Table 3). In the case of floricultural greenhouse plastic the relative pesticide percentages fluctuated between 0.70% and 19.5%, which was equivalent to a 56.9–262.8 mg mass range (footnotes, Table 3).

Table 3 shows the pesticide percentage distribution in plastic greenhouses taking into account four main sectors: lateral walls, front/back walls, crop roof and aisle roof. Lateral, front and back walls were sampled at three heights: low, medium and high (Table 3). Fig. 4 presents the relative pesticide distribution of the aforementioned greenhouse sections, expressed as mean percentage of H1–H6 experiences for horticultural and F1–F3 for floricultural measurements, plus their respective standard deviations. While in the tomato case there is a more homogeneous distribution of pesticide on the four sections (Fig. 4a), in lettuce most of the pesticide was concentrated on the plastic roof. In floricultural greenhouses a completely different situation was found with the main amount of pesticide was found on the lateral walls (Fig. 4b).

4. Discussion

4.1. Discussion of the pesticide distribution in greenhouses and open fields

The results show that the relative pesticide distribution profile is quite similar for greenhouses, independently of the activity (horticulture: Fig. 2a, floriculture: Fig. 2b), although different from open field horticulture (Fig. 2c). In the case of horticulture greenhouses, either for tomato or lettuce, practically 65% of the applied pesticide reached the crop, 30% was directly deposited on the soil, less than 5% was deposited on the greenhouse plastic and the drift outside the plantation can be considered negligible. Although tomato and lettuce plants have very different heights (see Section 2), pesticide distribution patterns for both crops were similar, suggesting that plant height has a minor impact on pesticide distribution in the aforementioned subsystems inside the greenhouses.

A similar trend was found for the floricultural greenhouses, around 70% of the applied pesticide was found on the crop, 20–25% on the soil and 5–10% on the greenhouse plastic. So, in general for greenhouses, a pesticide distribution pattern after application of roughly 2/3 for crop, 1/4 for soil and 1/20 for plastic films was observed.

A different situation was found for open field crops (broccoli, strawberry, lettuce, Table 2, Fig. 2c). Although for broccoli a distribution trend similar to greenhouses (65–70% for broccoli, 35–40 for soil, less than 5% for drift) can be observed (Fig. 2C, white bars), for strawberry and lettuce the amounts of pesticide found on the crop were smaller (50–55% and 20–25% respectively), with the rest of the product falling on the ground. This fact could be explained by two factors: crop density and plant size. While broccoli were big plants, growing relatively close, covering almost all the ground, strawberry and lettuce were smaller and more separated, exposing more ground. In the last case, lettuce plants were very small (2 weeks old), thus increasing soil exposure to pesticides.

For open field lettuce crops, the total pesticide drift (as μg of deltamethrin) at different distances from the crop edge was measured for pressurized hose (Fig. 3a) and manual knapsacks applications (Fig. 3b). It is interesting to note that the absolute drift values (H14, H15, Table 2), although not considerable, were more important when using a pressurized hose than when applying with a manual knapsack. When the deltamethrin mass was measured using cotton samplers on soil at fixed distances from the crop border (0.5, 1.0, 2.5, 7.0 m, Supplementary material), the pesticide amounts found rapidly dropped to non-detectable values for distances greater than 1 m; the same pattern was observed for pressurized hose (Fig. 3A) and manual knapsacks (Fig. 3B).

4.2. Discussion of the pesticide distribution in the floricultural and horticultural greenhouse plastic

The pesticide relative percentages found on the tomato (3.1–4.0%, Table 1), lettuce (0–0.16%, Table 1) and floriculture plastics (0.70–19.5%, Table1), were different, as well as the relative distribution between the roof and walls. In the tomato case, pesticide seemed to be more homogenously distributed over the different sections (Fig. 4a), while for lettuce, exposure was more concentrated on plastic roof (Fig. 4a). In the case of the floriculture greenhouses, most of the pesticide was found on lateral walls (80.1 ± 13.5%, Fig. 4b) finding the rest on the front/back walls (9.7 ± 13.1%) and the crop roof (10.2 ± 14.1%).

The dissimilarities of these results could be explained taking into account the different crop distribution and the diverse greenhouse dimensions. For example, with tomatoes and lettuces there was a 60 cm wide aisle between the crop border and the wall, while in the floricultural greenhouses, plants were located as close as 10 cm to the plastic, which could explain why lateral walls were more exposed in these experiments.

5. Conclusions

During pesticide application in small horticultural and floricultural production units there was a considerable amount of product that landed on non-target systems, with soil/ground the most affected. For greenhouses, a general pesticide distribution of 2/3 for crop, 1/4 for soil and 1/20 for plastic could be proposed. In horticultural open fields the pesticide distribution was very dependent on the crop size and type, but soil was again the most exposed non-target system collecting 30–80% of the total pesticide. In horticultural production units with manual pesticide application, product drift was not significant, decaying to non-detectable amounts a few meters from the field border.

Considering agricultural plastics, polyethylene used for mulching was the most exposed, receiving practically half of the applied pesticide. For horticultural and floricultural greenhouse plastics, the relative pesticide percentages were significantly lower than for strawberry mulching, but the absolute pesticide mass found on these plastic films was significantly greater than the product that drifted outside the greenhouse.

It should be emphasized that these amounts correspond to the pesticide deposited in a single application operation, and these are usually repeated on a weekly schedule in the warm seasons.

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Appendix A. Supplementary material

Supplementary data associated with this article can be found, in the online version, at doi:10.1016/j.chemosphere.2011.12.074.

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