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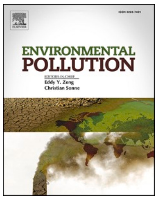

Pesticides in doormat and floor dust from homes close to treated fields: Spatio-temporal variance and determinants of occurrence and concentrations☆

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A B S T R A C T

Indoor dust has been postulated as an important matrix for residential pesticide exposure. However, there is a lack of information on presence, concentrations and determinants of multiple pesticides in dust in residential homes close to treated fields. Our objective was to characterize the spatial and temporal variance of pesticides in house dust, study the use of doormats and floors as proxies for pesticides in indoor dust and identify determinants of occurrence and concentrations. Homes within 250 m from selected bulb fields were invited to participate. Homes within 20 km from these fields but not having agricultural fields within 500 m were selected as controls. House dust was vacuumed in all homes from floors (VFD) and from newly placed clean doormats (DDM). Sampling was done during two periods, when pesticides are used and not-used. For determination of 46 prioritized pesticides, a multi-residue extraction method was used. Most statistical analyses are focused on the 12 and 14 pesticides that were detected in >40% of DDM and VFD samples, respectively. Mixed models were used to evaluate relationships between possible determinants and pesticides occurrence and concentrations in DDM and VFD. 17 pesticides were detected in more than 50% of the homes in both matrixes. Concentrations differed by about a factor five between use and non-use periods among homes within 250 m of fields and between these homes and controls. For 7 pesticides there was a moderate to strong correlation (Spearman rho 0.30–0.75) between concentrations in DDM and VFD. Distance to agricultural fields and air concentrations were among the most relevant predictors for occurrence and levels of a given pesticide in DDM. Concentrations in dust are overall higher during application periods and closer to fields (<250 m) than further away. The omnipresence of pesticides in dust lead to residents being exposed all year round.

1. Introduction

1.1. Pesticides in house dust

Pesticides play an important role in the agricultural production. An average amount of four million tons of pesticides are sprayed every year worldwide, with about 12% being applied in Europe (FAO, 2019). As pesticides may be dispersed outside the intended areas of application (Bueno et al., 2017), this may lead to exposure of the surrounding population (Zivan et al., 2016). Several studies have reported pesticides in house dust (e.g. Audy et al., 2018; Lee et al., 2018; Dong et al., 2019) and, although concentrations in the environment typically reflect annual usage, many of the active ingredients can still be detected after one year in house dust (Smith et al., 2017). Some studies even show that

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pesticides that were banned or restricted for many years still can be found in the indoor environment (Rudel et al., 2003).

1.2. Dust as exposure route to pesticides

The body of evidence regarding routes of human (residential) exposure to pesticides via house dust increased in recent years (Deziel et al., 2017). Dust has moved more into the focus as a possibly relevant contributor to human pesticide exposure (Golla et al., 2012; Bennett et al., 2019). This is due to the following reasons: firstly, dust ingestion, inhalation and contact with house dust have been shown to be primary routes of exposure for residents (Melymuk et al., 2020), especially for small children (Whitemore et al., 1994; Roberts & Dickey, 1995); secondly, people usually spend most of their time indoors at home (Brasche & Bischof, 2005), making this environment a prime source for exposure to contaminated dust (Dalvie et al., 2014). Thirdly, several studies have found associations between residential pesticide exposure and a wide range of health effects (e.g. Sabarwal et al., 2018; Rappazzo et al., 2019; Raherison et al., 2019), although only very few studies, like the one from Wickerham et al. (2012) single out dust exposure from residential exposure.

1.3. How do pesticides end up in house dust?

Pesticides can accumulate in indoor dust via different routes. Drift of pesticides usually occurs over short distances (0–250 m), with higher concentrations in both air and ground deposits closer to agricultural fields (0–50 m) (Garron et al., 2009; Zande et al., 2017) and declining exponentially with distance from the applied field (Carlsen et al., 2006). Pesticides can however be bound to particles and can travel longer distances and penetrate into homes further away and settle as house dust (Coronado et al., 2011). Additionally, the gas-phase fraction of pesticides can be bound to indoor dust particles (Wei et al., 2019). Pesticides can also reach the house dust by the take-home route (Hyland & Laribi, 2017), where contaminated soil is dragged into the residence by contaminated clothing and shoes or by pets (López-Gálvez et al., 2019).

1.4. Dust from doormat and vacuuming – current and historical pesticide use

Although pesticides can end up in indoor house dust via several routes, most previous studies assessing pesticide concentrations in dust have used solely vacuumed floor dust (e.g. Colt et al., 2008, Salis et al., 2017; Ten Brinke,) or wipe dust sampling (e.g. Schultz et al., 2019; Mercier et al., 2011) to investigate the occurrence. These methods reflect both current and historical pesticide use (Béranger et al., 2019), since it is not known for how long the collected dust has been present. Therefore they fail to capture pesticides exclusively used in the study period. A solution to this, is using a bespoke clean doormat for the study period. By doing so, the sample taken from the doormat reflects solely currently used pesticides (Plascak et al., 2019). Only a limited number of studies have compared different dust matrixes (Lu et al., 2000; Moschet et al., 2018; Rostkowski et al., 2019; Dubocq et al., 2021) and few have looked into determinants of occurrence and concentrations of pesticides in indoor dust (Gunier et al., 2011, Deziel et al., 2019).

1.5. Aim of our study

Here we present the results of indoor dust measurements of 46 pesticides across two different dust matrixes, vacuumed floor dust (VFD) and dust from a bespoke study doormat (dust from doormat, DDM). Five aims were a-priori defined: i) Study patterns for different pesticides occurrence in both dust matrixes; ii) Study temporal differences, by comparing concentrations between a period when pesticides are applied and a period when they are not applied; iii) Study spatial differences, by comparing concentrations in homes located close to fields with homes located further away; iv) Study the relation between concentrations of pesticides in VFD and DDM and increase our knowledge on the take-home exposure route; and v) Identify determinants of occurrence and concentration of pesticides in indoor dust samples (VFD and DDM), as an effort to further improve future pesticide exposure models.

2. Materials and methods

2.1. Study design

This research is part of the Dutch OBO study (OBO, 2019). The study took place from May 2016 to December 2017 in homes in the vicinity of bulb fields, a cultivation representative of down-ward boom spraying. “Location” was defined as an area consisting of homes surrounding a selected field, on which information about the applied pesticides was available. All sampling sites were located in the Netherlands, in the North-Holland and South-Holland provinces. Dust samples were taken during pesticide *use* and *non-use* periods in 9 different locations. In the use period, pesticides in dust were sampled per location for one week, with a spray event on the selected field as starting timepoint. In the non-use period (i.e. period when pesticides are not used), sampling was carried out also for one week.

2.2. Selection of pesticides for targeted analysis

Pesticides were selected based on registration, usage in tulip and lily cultivation and availability of a single analytical method. A more detailed description of the selection process can be found in Kruijne et al., (2019). In summary, a total of 46 pesticides were selected for analysis, comprising 29 pesticides that are frequently sprayed in bulb fields, 3 pesticides used in bulb disinfection, 6 breakdown products of some of these pesticides and 8 pesticides that were found in a previous study in soil and plant material from flower bulbs (OBO, 2019).

The selected pesticides represent a vast range of different physicochemical properties as well as the three product types. These include 11 herbicides: asulam, chloridazon, chlorpropham, dimethenamid-p, linuron, metamitron, metamitron-desamino, pendimethalin, s-metolachlor, sulcotrione and terbuthylazine; 12 insecticides: acetamiprid, cyhalothrin-lambda, deltamethrin, flonicamid, fosthiazate, imidacloprid, oxamyl, primicarb, pymetrozine, spirotetramat, spirotetramat-enol and thiacloprid; and 23 fungicides: azoxystrobin, boscalid, cyprodinil, difenoconazole, dimethomorph, fludioxonil, fluopicolide, fluopyram, fluopyram-benzamide, flutolanil, kresoxim-methyl, mepanipyrim, prochloraz, propamocarb, prothioconazole, prothioconazole-desthio, pyraclostrobin, tebuconazole, thiophanate-methyl, carbendazim, toclofos-methyl, trifloxystrobin, trifloxystrobin-acid. The list of analysed pesticides and relevant physical-chemical properties can be found in supplementary material A. Excluded from selection were chlorothalonil, diquat, esfenvaleraat, folpet, glyfosaat, iprodione and mancozeb. All these pesticides required an analytical method different from the selected one. Detailed information on these selection can be found in Figueiredo et al., (2021a).

2.3. Recruitment

With the aim of getting a good spatial distribution of houses around sprayed fields with at least one of those fields being treated with a pesticide listed for analyses, we initialized a recruitment process (Fig. 1). First, farmers of bulb fields were contacted to participate in the study and provide information on their fields and then, in case of acceptance, the residents living in the vicinity of those fields were approached. Here, recruitment and selection are briefly described. More details can be found in Figueiredo et al., (2021a), including the power calculation for minimum samples needed (study size).

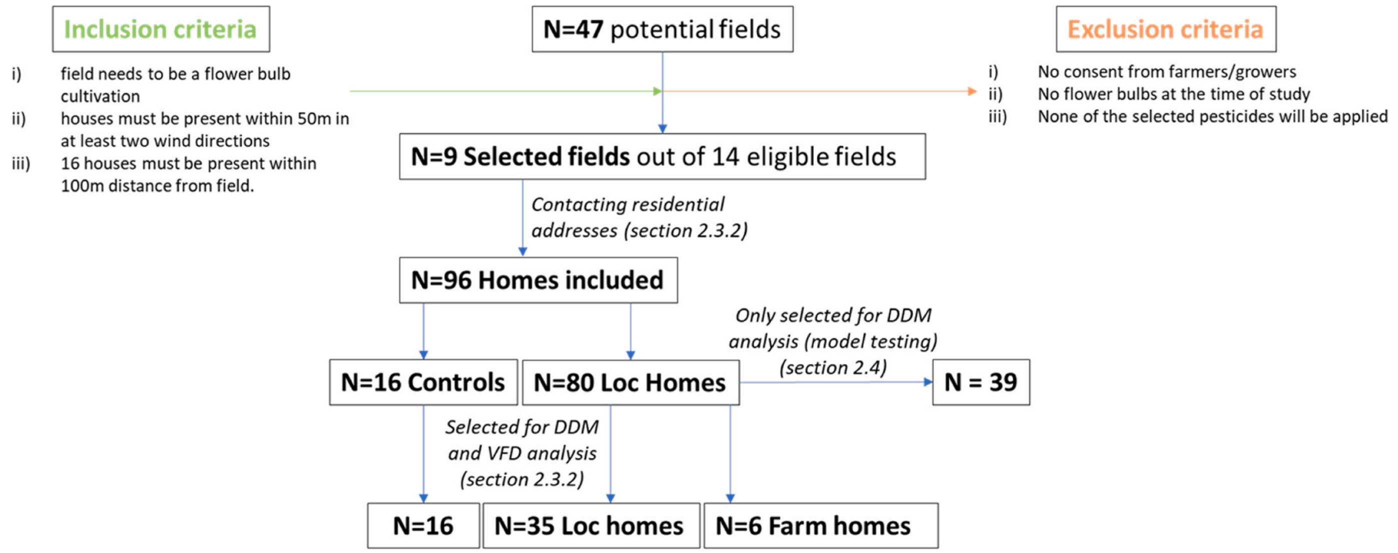

Fig. 1. Flowchart of recruitment process for both fields and homes.

2.3.1. *Fields*

A search for eligible fields resulted in 47 potential fields. This selection was based on 1) field needs to be a flower bulb cultivation and 2) houses must be present within 50 m in at least two wind directions, and at least 16 houses must be present within 100 m distance. Thirty-three locations did not participate because farmers did not give consent ($N = 26$), because there were no flower bulbs present at the time of the study or none of the selected pesticides were going to be applied ($N = 7$). From the 14 eligible fields 9 were randomly selected for the study.

2.3.2. *Homes*

After inclusion of a field in the study, all residential addresses within 250 m of the perimeter of the field, here called “Location” (Loc) Homes, were selected using the Dutch cadastral data “Basisregistraties adressen en gebouwen (BAG)”. Potential control homes, further called “Controls”, were also selected using BAG to identify homes in medium to low urbanized areas (i.e. <1500 addresses/km2), situated within 20 km from a target field, and not having any agricultural fields within 500 m of the home.

Invitation letters and a brochure were sent to all identified addresses and interested invitees were interviewed by phone using a structured interview script.

In total, 1778 residential addresses and 482 addresses at control locations received an invitation to the study. Eighty potential Loc Homes responded, corresponding to a response rate of 4.5% (range 2.1%–33.3% by location). Additionally, 16 control homes were included (response rate: 3.3%). Not all homes partook in all measurement campaigns as three homes missed one of the two seven-day measurement campaigns due to holidays and residents from four homes ended their participation before the end of the study.

Due to budgetary reasons dust samples (both DDM and VFD) were only analysed for 41 homes that were selected out of the 80 homes initially included in the study. In short, the aim was to have a good spatial distribution of Loc Homes (i.e. different distances from the field were equally represented). For this, we selected some homes located very close to the fields (<50 m) ($N = 16$), some more further away (50 m–150 m) ($N = 14$) and some located between 150 m and 250 m ($N = 11$). These buffers are based on previous research done on pesticide concentrations at different distances downwind (Siebers et al., 2003; Figueiredo et al., 2021b) and ensured that homes were located both up and down-wind of the application (all cardinal directions). All controls were included in the sample analyses.

Some of the participants were also growers. These homes ($N = 6$), defined here as “Farm” homes, were treated as a separate group in all analyses, since it is known from previous studies that these homes are more prone to pesticide accumulation and take-home exposures (Curl et al., 2002; Curwin et al., 2005).

2.4. *Additional homes – modelling testing purposes*

Of the homes initially not selected ($N = 39$) we later analysed the dust doormat sample from the *use* period for each home, using identical protocols and laboratory as the initial analyses. We here use this separate dataset solely for model testing purposes (test dataset). We chose DDM over VFD for additional analyses as DDM has a determined sampling time and surface, increasing comparability and avoids the influence of long-term pesticide accumulation (Harnly et al., 2009).

2.5. *Sample collection*

2.5.1. *Vacuumed floor dust (VFD)*

In all participating homes, VFD was collected from the living room by a research assistant. In the *use* period, it was collected 7 days after a spray event. The recruited farmers informed us a-priori on which day and time they would spray. This defines the spray event. Spray events were not the first spraying occurring in the selected fields. In the *non-use* period, VFD was collected at the time of doormat retrieval. For this, a sample sock (Allied Filter Fabrics, Hornsby, Australia) was attached to the hose of a vacuum cleaner. Initially, the research assistant vacuumed 2 m^2 of carpet or 4 m^2 of smooth floor for 2 min. After analysing the first samples ($N = 18$) this was increased to 4 m^2 of carpet or 6–8 m^2 of smooth floor, depending on available free floor space, to increase the amount of dust collected. Sampling duration was increased to 5 min. Sampling duration and sampled area were recorded for each home and the results were standardised per gram of collected dust. In a sensitivity analysis, we saw that the increase in sampling time and surface area vacuumed did not significantly affect pesticide concentration per gram dust and therefore, for the final analysis, results were pooled. The sample amount varied from 0.02 to 28 g, with a median value of 0.37 g. Samples were stored at $-18\ ^{\circ}C$ until analysis.

2.5.2. *Dust doormat (DDM)*

In each participating home, a clean doormat (100% polypropylene) was cut to applicable size and placed indoors at the main entrance by a participant. In the *use* period, the participant placed the doormat on the day of the spray event. In the *non-use* period it was placed during a month where no sprayings occurred. The doormat was collected by the research assistant within 5 days after the end of the measurement campaign and transported in a clean box to the laboratory. We recorded the size of the doormat and start and end date of collection for each home and standardised the result per gram of collected dust. In the laboratory, we used a sample sock (Allied Filter Fabrics, Hornsby, Australia) to vacuum clean all dust material from the doormat. Samples were stored at −18 °C until analysis The amount of dust material retrieved from the doormat varied from 0.55 to 196 g, with a median of 6.0 g.

2.6. Analysis method for determination of pesticides in dust samples

For determination of pesticides in the dust samples, a multi-residue extraction method was used. This is based on salt-induced phase partitioning technique (QuEChERS) (Lehotay, 2007; Perestrelo et al., 2019) and Liquid Chromatography-tandem Mass Spectrometry (LC-MS/MS). This way, all 46 selected pesticides and relevant metabolites could be measured simultaneously. We chose not to do fractioning or sieving of the dust sample given that we are interested in total exposure, several other studies also used this approach (Cao et al., 2012).

In brief, the entire dust sample was extracted with water and acetonitrile/1% acetic acid by mechanical shaking. Salts were added to induce phase partitioning. The organic phase containing the pesticides was used for LC-MS/MS analysis. Dust is a complex, variable and heterogenous matrix, resulting in variable and often strong matrix effects (ion suppression) in the LC-MS/MS analysis. Therefore quantification was based on the standard addition method. To this end, for each sample extract, two aliquots were taken. To one aliquot the mix standard of 46 pesticides was added. After 2-fold dilution with water, the extracts with and without standard addition were analysed by LC-MS/MS. For details on sample preparation,LC-MS/MS conditions and quality assurance see supplementary material B.

In-house validation and on-going analytical quality control were done according to EU guidance document SANTE/11945/2015 (currently SANTE/12682/2019). In most cases (83%), recoveries were between 70 and 120%. The precision (RSD) were below 20% at the 50 μg/kg level, and around 20% at lower levels. The limit of quantification (LOQ) was 1 μg/kg for most pesticides (N = 33), 3–50 μg/kg for 12 pesticides. The limit of detection (LOD) was estimated in case the LOQ was higher than 1 μg/kg, and in these cases ranged from 1 to 20 μg/kg. See the supplementary material B for details.

2.7. Treatment of left-censored data

For left-censored data (<LOD) imputation was performed when at least 40% of the measured samples had levels above the LOD. Concentrations between LOD and LOQ, although semi-quantitative, were used as such since these are likely more accurate than imputed values (Succop et al., 2004). For each pesticide, imputation was performed using the method proposed by Lubin et al. (2004). Here, unbiased estimates are obtained by imputing the values below LOD based on the maximum likelihood estimation, while accounting for the distribution and correlation of all pesticide data. Here, imputation was performed including 100 iterations.

2.8. Sampling period and number of analysed samples

Both types of dust samples were grouped according to the period of sampling: during the period the pesticide was used, normally between March and August, or outside the period the pesticide was used, October to December. Periods of application of each pesticide were defined based on reported spraying schedules. Samples were therefore grouped by *use* and *non-use* for each pesticide separately. The reported spraying periods of each pesticide can be found in supplementary material C.

In total 292 dust samples were analysed, with 125 being DDM samples and 128 being VFD samples. From the DDM samples there were 14 from Farm Homes (N = 7 in both periods), 79 from Loc Homes (N = 48 *use* period and N = 31 *non-use* period) and 32 from Controls (N = 16 in both periods). From the VFD samples there were 14 from Farm Homes (N = 7 in both periods), 82 from Loc Homes (N = 48 *use* period and N = 34 *non-use* period) and 32 from Controls (N = 16 in both periods). As indicated before, additional samples (n = 39; 3 from Farm Homes and 36 from Loc Homes) were analysed for validation of the pesticide occurrence model in DDM.

2.9. Questionnaires and variables used for modelling purposes

Per home, a questionnaire on home characteristics was collected as well as lifestyle information and demographics for all participants within a home. Detailed information on the filling of questionnaires and list of all questions asked can be found in Figueiredo et al., (2021a). These questions pertained to *a priori* identified variables that might be related to occurrence and variance in concentrations of pesticides in indoor dust. In short, this information consists of variables that i) are related to house characteristics and can affect the dynamics of pesticides in indoor dust, such as having a smooth vs carpeted flooring, forced vs natural ventilation, sealed against draught or having visible leakages, amongst others; ii) are related to house dynamics, such as leaving shoes outside or inside, number of inhabitants, number of pets and type of pets, use of pesticides, amongst others.

In addition to the above, we also use meteorological variables, such as humidity, precipitation, wind speed and direction. Distance from home to closest agricultural field is used as a spatial variable. See Figueiredo et al. for details on collection of both meteorological and spatial variables (section 2.7, Figueiredo et al., 2021b).

Finally, as an additional variable, we also predicted concentrations in dust (Dustpred) based on the deterministic equation by (Weschler and Nazaroff, 2010). Here, air concentrations sampled via active air samplers parallel to the dust collection are used as input. Detailed methods and results regarding air measurements can be found in Figueiredo et al., (2021b). The full list of variables and type (i.e. discrete or continuous) as well as information on the equation used to calculate Dustpred can be consulted in Supplementary material D. All variables were included as independent variables in the undermentioned modelling steps.

2.10. Statistical analysis

All data analyses were performed using R, version 4.0.0 (R Core Team 2017). The pesticide concentration data was log10 transformed to meet the assumptions of inferential statistics.

2.10.1. Samples categorization and focus of analysis

Not all of the 46 targeted pesticides were applied during the course of the study, therefore, for data analysis and interpretation purposes, pesticides were categorized into three groups: i) pesticides that were reported as being applied in the selected field and/or on fields located in the vicinity of the included homes (<250 m) during the course of the study; ii) pesticides that were not reported as being applied but were used for bulb disinfection purposes; and iii) pesticides that were neither reported as being applied or used for bulb disinfection. The results are presented separately for these three groups given that bulb disinfection is not bound to a fixed period of usage and might be used inside facilities that are not located close to agricultural fields.

The field participating in our study was often not the only field applying pesticides in the proximity of the participating Loc Homes. Information regarding spraying applications and applied mixtures was a posteriori collected or estimated (based on expert decision) for all fields within 250 m of location homes. Information on the different spraying applications is reported in OBO 2019. Data on pesticides used for bulb disinfection in 2017 was retrieved from local farmers and data available from Ten Brinke,), an agricultural advisory company.

We first summarized the detection frequency for all 46 targeted pesticides. In subsequent statistical analysis, however, the focus was solely on pesticides that were quantified in at least 40% of the measured samples. This comprises some of the pesticides applied in bulb fields (N = 9 for DDM and N = 10 for VFD) and almost all pesticides used in bulb disinfection (N = 3 for DDM and N = 4 for VFD). No quantitative assessment can be performed for the remaining pesticides given that more than 60% data is missing (Jakobsen et al., 2017).

2.10.2. Spatial and temporal differences in concentrations

In order to study spatial differences we compared concentrations in Loc Homes vs Controls. For temporal differences we compared concentrations in the *use* period vs *non-use* period. Concentrations were plotted for easy visualization of the aforementioned comparison. Student's *t*-Test were used to determine whether the means of different groups (i.e. samples taken during the *use* and *non-use* period and Loc Homes vs Controls) were equal to each other. For this comparisons data was analysed from 41 Loc Homes and from 16 Controls, both during use and non-use period, respectively.

2.10.3. Correlations between the two matrixes

Spearman's rank correlation coefficient was used to study the relationship between pesticide concentrations in the two types of dust samples (DDM and VDF). Here, Spearman correlation coefficients for Loc Homes and Controls were calculated separately, instead of grouping all samples together (i.e. Loc Homes + Controls). We chose to look at correlations per group given that concentrations in Loc Homes were generally much higher than in Controls in both periods. This would drive the correlations if all samples were taken together and would likely hide any pattern between DDM and VFD that solely occurs in Loc Homes or Controls. All analyses were performed with concentrations in nanograms per collected amount of dust (ng/g). As a sensitivity analysis, correlations were additionally calculated using concentrations in nanograms per surface area (ng/m^2).

2.10.4. Identifying possible determinants of occurrence and concentrations of different pesticides in indoor dust

To assess possible determinants of occurrence and concentrations of pesticides in indoor dust, mixed models were built using the *lme4* package for R. Two correlated random effects (intercept and slope) were estimated for each level of the HouseID factor (i.e. in *lme4* Period | HouseID). Analyses were carried out for both the occurrence (binary – logistic regression) and log-transformed concentrations of pesticides (continuous - linear regression) in dust.

A multivariate logistic regression model was built using a backward stepwise algorithm for variable selection in combination with the *glmer* function to identify the best model to predict occurrence of pesticides in indoor dust. Here, percentage of values above LOD was used as dependent variable.

To predict concentrations in dust the *lmer* function as implemented in R was used. Here, each pesticide concentrations was used as dependent variable. The obtained models were tested using the independent dataset of 39 DDM samples.

Finally, for the logistic model, the AUC (area under the curve) was calculated as performance measurement using the *pROC* library for R. For the linear regression model, we calculated the R2 and RMSE.

3. Results

Table 1 shows the percentage of samples above LOD for the three groups: Farm Homes, Loc Homes and Controls. The results are ordered by decreasing half-life in soil and clustered by type of dust sample (DDM, VFD) and by period (*use* and *non-use*). All pesticides were detected at least once in both types of dust with the exceptions of cyhalothrin-Lambda, terbuthylazine and sulcotrione, detected only in VFD.

3.1. Detection of pesticides in DDM and VFD

3.1.1. DDM – pesticides detection frequency

Regarding pesticides applied in bulb fields (group I, Table 1), these were, on average, detected in 64% and 54% of the samples collected in Farm Homes during the *use* and *non-use* periods, respectively. For Loc Homes, these were detected in 32% and 23% of the samples collected during the *use* and *non-use* periods, respectively. For Controls, these were detected in 12% and 8% of the samples collected during the *use* and *non-use* periods, respectively. Regarding pesticides used in bulb disinfection (group II, Table 1), these were, on average, detected in 94% and 92% of the samples collected in Farm Homes during the *use* and *non-use* periods, respectively. For Loc Homes, these were detected in 49% and 58% of the samples collected during the *use* and *non-use* periods, respectively. For Controls, these were detected in 45% and 30% of the samples collected during the *use* and *non-use* periods, respectively. For pesticides that were not used in either of the above-mentioned situations (Group III, Table 1) detection was very low (overall average of 6%).

3.1.2. VFD – pesticides detection frequency

Regarding pesticides applied in bulb fields (Group I, Table 1), these were, on average, detected in 59% and 50% of the samples collected in Farm homes during the *use* and *non-use* periods, respectively. For Loc Homes, these were detected in 32% and 21% of the samples collected during the *use* and *non-use* periods, respectively. For Controls, these were detected in 11% and 13% of the samples collected during the *use* and *non-use* periods, respectively. Regarding pesticides used in bulb disinfection (Group II, Table 1), these were, on average, detected in 75% and 88% of the samples collected in Farm homes during the *use* and *non-use* periods, respectively. For Loc Homes, these were detected in 70% and 67% of the samples collected during the *use* and *non-use* periods, respectively. For Controls, these were detected in 63% and 59% of the samples collected during the *use* and *non-use* periods, respectively. For pesticides that were not used in either of the above-mentioned situations (Group III, Table 1) detection was very low (overall average of 14%).

3.2. Concentrations in DDM and VFD

Imputation of values below LOD was performed for pesticides with more than 40% of the samples having levels above the LOD, resulting in 12 pesticides with imputed values for DDM and 14 for VFD. Most of these pesticides had at least 50% of measured samples above LOD, with exception of fluopyram and prothioconazole-desthio.

In Fig. 2 we present the results of the comparison between group means for these pesticides for DDM, by comparing Loc Homes vs Controls, during *use* and *non-use periods*. Fig. 3 shows the same comparison for pesticides in VFD. All Student's *t*-Test results from the spatial and temporal comparisons can be found in supplementary material E.

3.2.1. DDM – concentrations in space (Loc Homes vs controls) and time (use vs non-use)

Differences between Loc Homes and Controls in the *use* period were statistically significant for 9 out of the 12 pesticides for DDM. For 8 of these 9 pesticides, Loc Homes had higher concentrations and imidacloprid was the only pesticide with higher concentrations in Controls compared with Loc Homes. In the *non-use* period, significant differences between Loc Homes and Controls were as pronounced, with 10 out of 12 pesticides having higher concentrations in Loc Homes.

For Loc Homes, we only observed significantly higher concentrations for 3 pesticides applied in bulb fields in the *use* when comparing with the *non-use* period (panel A, Fig. 2). For Controls, significant differences were only observed for tebuconazole, imidacloprid and carbendazim, with the last two belonging to the bulb disinfection group (panel B, Fig. 2).

Table 1
Percentage of detectable pesticide concentrations by exposure group, type of dust sample and use period.

	Farm Homes				Loc Homes				Controls			
	Use period		*Non-use* period		*Use* period		*Non-use* period		*Use* period		*Non-use* period	
Active ingredient (pesticide group)	VFD	DDM	VFD	DDM	VFD	DDM	VFD	DDM	VFD	DDM	VFD	DDM
Group I - Reported as being **applied in bulb fields** during the study period												
boscalid (F)	88	100	100	100	92	98	88	84	43	88	63	63
azoxystrobin (F)	88	88	83	83	66	65	59	52	50	19	50	25
fluopyram (F)	88	88	67	100	54	56	38	61	14	13	13	13
flutolanil (F)	88	100	83	83	34	42	26	39	14	13	0	0
pendimethalin (H)	75	100	67	100	68	96	41	71	29	19	31	13
mepanipyrim (F)	50	50	0	17	6	19	0	6	0	0	0	0
linuron (H)	75	100	17	100	36	52	9	19	0	6	0	0
tebuconazole (F)	75	100	83	83	84	81	79	61	64	75	69	44
prothioconazole-desthio (F)	63	63	83	50	58	67	41	45	21	19	19	13
chloridazon (H)	88	75	83	83	24	10	6	6	7	0	0	0
pyraclostrobin (F) [a]	100	100	100	100	92	96	79	84	21	75	38	50
metamitron-desamino (H)	50	63	67	33	34	21	9	3	0	0	13	0
tolclofos-methyl (F)	88	100	83	83	24	17	18	29	0	0	6	0
chlorpropham (H)	38	50	0	0	26	48	18	13	14	19	6	0
cyhalotrin-lambda (I)	13	0	0	0	2	0	0	0	0	0	0	0
pymetrozine (I)	13	38	33	17	14	17	3	19	0	0	0	0
S-metolachlor (H)	100	100	50	83	46	69	6	13	14	6	0	0
deltamethrin (I) [a]	13	0	17	0	0	4	12	6	7	6	13	0
prochloraz (F) [a]	100	100	100	100	82	90	79	94	14	38	63	50
metamitron (H)	63	75	67	83	38	23	12	10	0	0	0	0
asulam (H)	75	88	50	67	30	31	3	19	7	0	0	0
pirimicarb (I)	13	38	50	17	12	8	9	6	7	0	6	0
fluopyram-benzamide (F)	13	25	17	33	2	0	0	0	0	0	0	0
thiacloprid (I)	50	38	67	33	30	2	21	3	0	6	0	6
dimethenamid-P (H)	50	38	33	17	14	6	0	3	0	0	0	0
oxamyl (I)	38	25	33	17	10	6	9	0	0	6	0	0
flonicamid (I)	63	75	50	83	56	19	24	16	21	0	19	6
acetamiprid (I)	75	75	50	67	18	6	9	0	14	0	13	0
trifloxystrobin (F)	50	50	17	67	12	8	6	10	7	6	0	6
trifloxystrobin-acid (F)	13	38	0	0	0	0	0	0	0	0	0	0
kresoxim-methyl (F)	50	63	33	50	16	2	21	6	0	0	13	0
prothioconazole (F) [a]	50	38	50	50	6	15	0	10	0	0	0	0
spirotetramat (I)	63	63	33	33	4	4	0	3	0	0	13	0
spirotetramat-enol (I)	50	50	33	17	2	2	3	3	0	0	6	0
Group II - Not applied in bulb fields but reported as **used in bulb disinfection** in 2017												
imidacloprid (I)	63	100	83	83	82	25	65	35	93	75	88	44
propamocarb (F)	75	75	67	83	62	29	41	29	57	6	44	25
thiophanate-methyl (F)	75	100	100	100	46	71	71	84	36	19	56	13
carbendazim (F)	88	100	100	100	90	73	91	84	64	81	50	38
Group III - **Not applied** in bulb fields and **not reported** as used in bulb disinfection												
fluopicolide (F)	13	0	33	0	10	2	9	3	7	0	6	0
difenoconazole (F)	25	0	33	0	28	6	18	13	7	6	13	6
cyprodinil (F)	13	13	0	0	20	17	18	13	0	13	13	0
dimethomorph (F)	13	13	33	17	28	19	21	10	7	6	13	19
fludioxonil (F)	38	13	17	0	50	33	50	16	14	25	31	6
terbuthylazine (H)	0	0	17	0	4	0	0	0	7	0	0	0
fosthiazate (I)	25	25	17	17	0	0	0	0	0	0	0	0
sulcotrione (H)	0	0	0	0	0	0	0	0	7	0	0	0

Farm Homes – Homes where farmers live; Loc Homes – Homes located within 250 meters from a selected field; Controls – Homes located more than 500 meters from any agricultural field but in the same region as the Loc Homes. Use – Period when pesticides are sprayed; Non-use – Period when pesticides are not sprayed. VFD – Vacuumed floor dust; DDM – Dust from doormat. F – Fungicide; H – Herbicide; I – Insecticide. Pesticides that are not used but are transformation products are in italic. All values are in percentage (%) of samples where a given pesticide was detected above the limit of detection. The colour scheme is used to highlight detection frequency, darkest colour = 100% detection and lightest colour = 0% detection. Colour scheme is divided into the following 8 detection (%) range intervals (from lighter to darker): 0 -> (0,11] -> (11,25] -> (25,50] -> (50,75] -> (75,90] -> (90,100) -> 100.[a] Can also be used for bulb disinfection (Ten Brinke 2017)

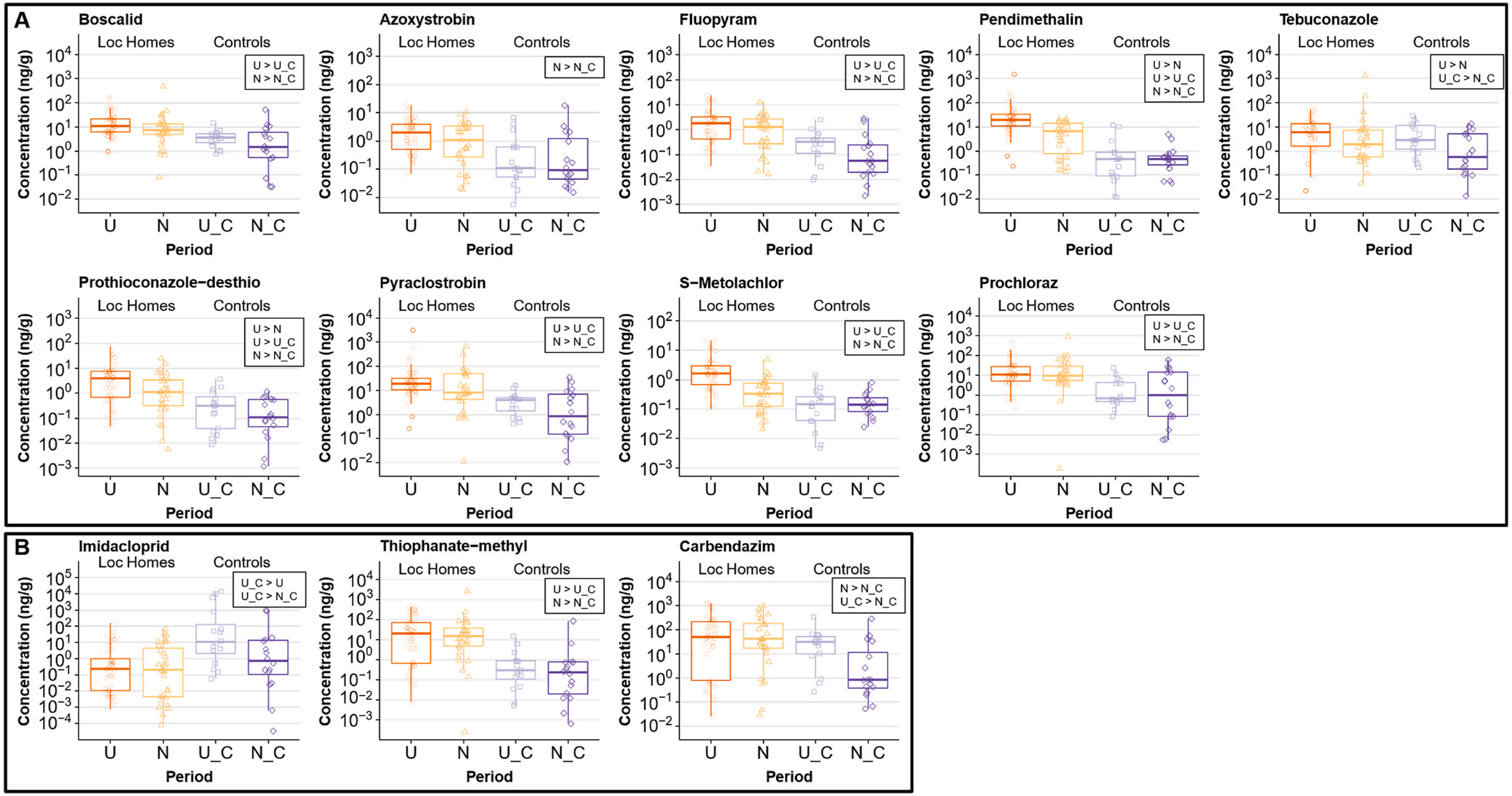

Fig. 2. Pesticide concentrations in house doormat grouped by Use (U) and Non-Use period (N) for locations and U_C and N_C for Controls. Panel (A) refers to pesticides applied in bulb fields and panel (B) to pesticides used in bulb disinfection. Summary statistics in boxplots (min, max, 1st and 3rd quartile and median). The box in the upper right corner of each graph is a comparison between the different groups and indicates which differences between group means is statistically significant at the 0.05 level.

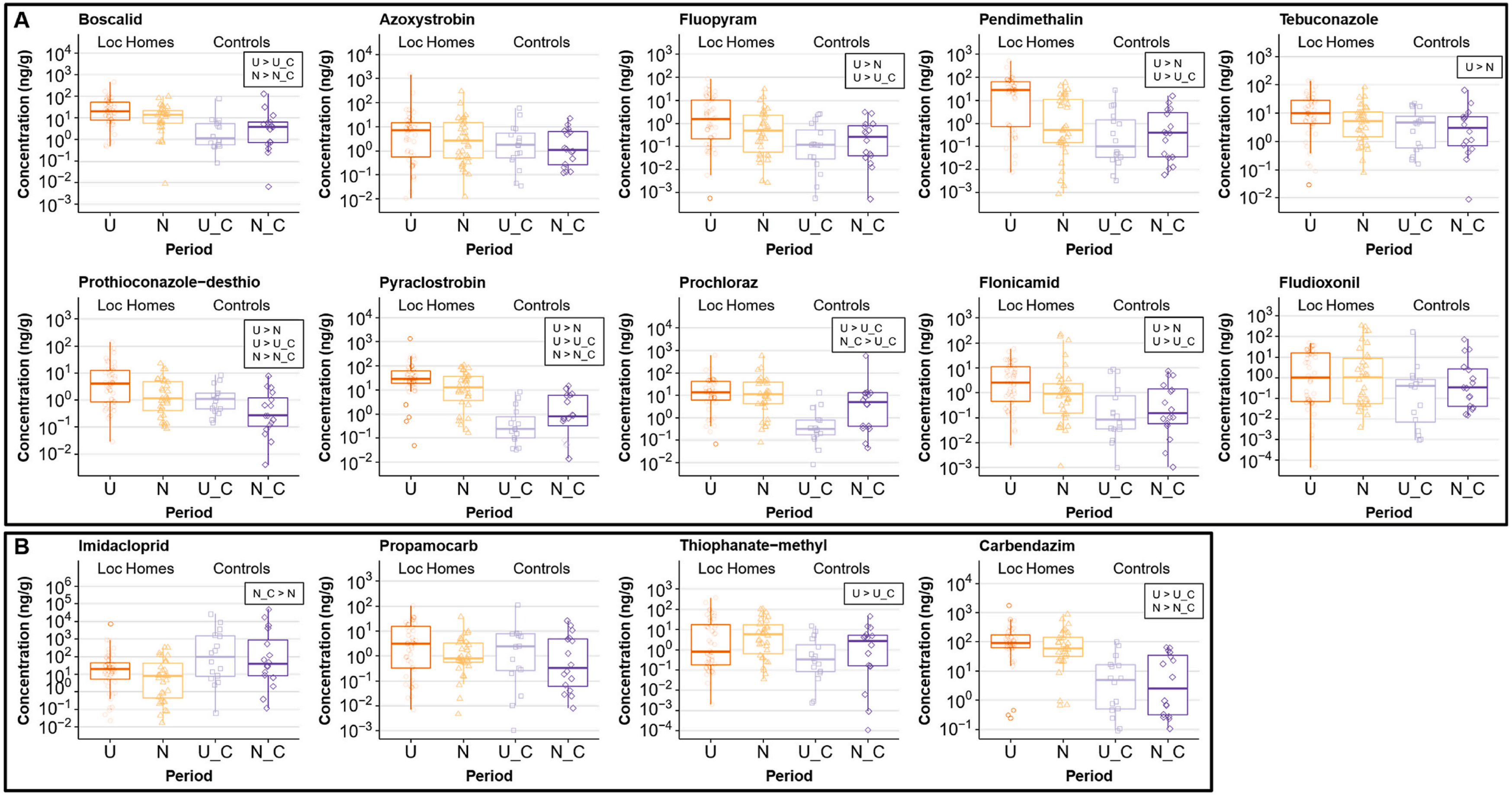

Fig. 3. Pesticide concentrations in vacuumed house dust grouped by Use (U) and Non-Use period (N) for locations and U_C and N_C for Controls. Panel (A) refers to pesticides applied in bulb fields and panel (B) to pesticides used in bulb disinfection. Summary statistics in boxplots (min, max, 1st and 3rd quartile and median). The box in the upper right corner of each graph is a comparison between the different groups and indicates which differences between group means is statistically significant at the 0.05 level. No box = no statistically significant difference between groups.

3.2.2. VFD – concentrations in space (Loc Homes vs controls) and time (use vs non-use)

Differences between Loc Homes and Controls in the *use* period were statistically significant for 9 out of the 14 pesticides for VFD. In the *non-use* period, significant differences between Loc Homes and Controls were less pronounced, with 6 out of 14 pesticides having higher concentrations in Loc Homes. Similar to DDM, imidacloprid was found in higher concentrations in Controls than in Loc Homes.

We observed for both VFD samples collected in Loc Homes higher concentrations in the *use* as compared to *non-use* period, for 6 pesticides applied in bulb fields (panel A, Fig. 3). For Controls, no clear differences were observed except for prochloraz, where concentrations were significantly higher in the *non-use* period as compared to the *use* period. Although reported as being sprayed during the measuring period, prochloraz can also be used in bulb disinfection.

3.3. Correlation between pesticides in DDM and VFD

Correlations between concentrations in DDM and VFD were calculated for each pesticide that had >40% detects in both dust matrixes. These were 11 pesticides out of the 14 initially imputed. Each correlation comprised 111 paired observations. All calculated Spearman correlation coefficients can be consulted in Supplementary material F.

The average correlation between concentrations in both types of dust is, for Loc Homes, 0.24 [−0.01, 0.55] and 0.27 [−0.06, 0.40], in the *use* and *non-use* period, respectively. For Controls, 0.23 [−0.18, 0.74] and 0.25 [−0.36, 0.60], in the *use* and *non-use* period, respectively.

In Loc Homes, correlations were absent (−0.01) to moderate (0.55) in the *use* period. Three pesticides showed statistically significant ($\alpha < 0.05$) correlations between both matrixes. Prothiconazole-destio and pyraclostrobin showed moderate correlation coefficients, 0.38 and 0.55, respectively. Whist imidacloprid showed a weak correlation coefficient of 0.14. There were no statistically significant correlations for Loc Homes in the *non-use* period.

In Controls, correlations were very weak (0.06) to moderately-strong (0.74) in the *use* period. Three pesticides showed statistically significant correlations between both matrixes. Fluopyram and tebuconazole showed moderate correlation coefficients, 0.57 and 0.55, respectively. Imidacloprid displays a moderately-strong correlation coefficient of 0.74. In the *non-use* period, two different pesticides showed strong statistically significant correlations, pendimethalin and prochloraz, both 0.60.

From the sensitivity analysis (see supplementary material G), where the correlation between concentrations in nanograms per surface area (ng/m^2) were calculated, no significant correlation between pesticides in Loc Homes was observed. Whilst, for Controls, concentrations of imidacloprid, pendimethalin and prochloraz were moderate to strongly correlated between both dust matrixes.

3.4. Pesticides occurrence in dust – multivariate mixed-effect logistic models

In the multivariate analysis, for DDM, and after selection via a stepwise approach, the resulting model encompasses 5 variables, namely half-life in soil, vapor pressure, average kg applied per year, distance to field and predicted dust concentration. Predicted performance (calculated AUC) of the DDM model was 75% using the independent DDM dataset.

In the multivariate analysis, for VFD, and after selection via a stepwise approach, the resulting model encompasses the same variables as the DDM multivariate model, except for distance to nearest agricultural field, which was only selected for the DDM model. The odds ratio for each predictor variable can be found in supplementary material H.

3.5. Pesticides concentration in dust – multivariate mixed-effect linear models

Multivariate models for pesticide concentration in dust varied significantly per pesticide for both DDM and VFD. However, for DDM, distance to field and predicted concentration in dust were the most selected variables between models (Table 2). Models were only built for imputed pesticides. Results from univariate linear models for each pesticide can be consulted in Supplementary material I.

Specifically, five pesticides, namely fluopyram, pendimethalin, prothioconazole-desthio, pyraclostrobin and s-metolachlor, have predicted concentration in dust and distance in common as predictive variables with similar beta coefficient (β) signs.

For three pesticides, namely azoxystrobin, prochloraz and imidacloprid, the presence of dogs in the household was a predictive variable, but with a positive β for imidacloprid. The self-reported use of snail or slug bait products (Pest vs Snails) showed to be an important predictor in models for prochloraz, thiophanate-methyl and carbendazim, all fungicides. Meaning that concentrations for these pesticides were higher in homes where residents reported using products against snails.

For VFD, five pesticides, namely pendimethalin, tebuconazole, pyraclostrobin, prochloraz and imidacloprid, have predicted concentration in dust (Dustpred) in common with similar positive effect estimates (positive β). For five pesticides, namely azoxystrobin, fluopyram, tebuconazole, flonicamid and fludioxonil, presence of dogs in the home is correlated with lower concentrations (negative β) in house dust. Distance to closest field showed to be an important variable in models for boscalid, pyraclostrobin, prochloraz and carbendazim, all with a similar negative β, meaning a decrease in indoor concentrations when living further away from the fields.

Reported pesticide smell was associated with increase (positive β) in boscalid and prothioconazole-desthio concentrations in VFD. Increase in evaporation from crops was associated with increase (positive β) in indoor concentrations for four different pesticides.

Finally, predicted performance of the DDM models is low (R^2 0.004–0.460), with the explained variance being higher for pesticides that were applied in bulb fields. For example, 0.46 for the pyraclostrobin, and 0.26 for the tebuconazole model. All β values, as well as calculated R^2 and RMSE for the DDM model can be consulted in Supplementary material J.

4. Discussion

4.1. Detection of pesticides in house dust

All 46 targeted pesticides were present in at least one dust sample, with most of them being detected in both VFD and DDM. This is in line with previous studies that also detected several pesticides in indoor dust (Blanchard et al., 2014; Bennett et al., 2019), not just sprayed pesticides but even others that are no longer allowed (Béranger et al., 2019).

When comparing with Béranger et al. where settled dust was analysed from homes located in different agricultural areas in France (Béranger et al., 2019), some pesticides were found with similar detection rates, like lambda-cyhalothrin, cyprodinil and s-metolachlor (1–15%), whereas others, like tebuconazole, chlorpropham and imidacloprid (used in bulb disinfection), were detected much more frequently in our study. A recent study done in China (Wang et al., 2019), had similar detection rates as our study for carbendazim and imidacloprid in indoor floor dust, both being detected in more than 70% of all samples.

It is difficult to ascertain why some pesticides used in the study period are detected at low rates in comparison with others. This is because many variables influence occurrence, such as application frequency, applied dosage (Degrendele et al., 2016), persistence in the environment (Richards et al., 2016), amongst others. For most pesticides we don't have enough information to infer on the reason(s) for low detection. We do see that lamba-cyhalothrin and deltamethrin, although

Table 2
Determinants of pesticide concentration in doormat (DDM) and vacuumed floor dust (VFD) samples.

Grouping	Variables	DDM Multivariate model [b] per pesticide												VFD Multivariate model [b] per pesticide												
		1	2	3	4	5	6	7	8	9	10	11	12	1	2	3	4	5	6	7	9	10	11	12	13	14
Presence of pets	Dog = Y		-							-	+				-	-		-							-	-
	Cat = Y										+															
	Rodent = Y													-												
House information	Living room size										+															
	Sealed against draught										-															
	Leakage								+																	
	Natural ventilation								-																	
	Roof = Flat																						-			
	Number of persons																	+								-
	Distance[a]			-	-		-	-	-					-						-	-			-		
Pesticide smell or usage	Pesticide Smell														+				+							
	Pest vs Snails = N									-		-	-													
	Pest vs Fungi = N																						-			
	Pest vs Fleas & Ticks = N																					-				
Climatic conditions	Wind Speed																				+			+		
	Humidity					-							-													
	Cloudness												+					-								
	Evaporation[a]	+			+														+	+						
	Precipitation Duration				+															+						
Other	Duspred[a]			+	+	+	+	+	+	+							+	+		+	+	+				

[a] This variables were log10 transformed.

[b] Beta coefficients sign (+ or -) presented solely for statistically significant variables with p-value < 0.05. Pesticides: (1) Boscalid, (2) Azoxystrobin, (3) Fluopyram, (4) Pendimethalin, (5) Tebuconazole, (6) Prothioconazole-desthio, (7) Pyraclostrobin, (8) S-Metolachlor, (9) Prochloraz, (10) Imidacloprid, (11) Thiophanate-methyl, (12) Carbendazim, (13) Flonicamid, (14) Fludioxonil.

persistent in the environment, are rarely detected in VFD and DDM. This is likely because i) they are both are pyrethroids, which usually have rather low application rates and ii) their LOD/LOQ are a higher compared to most other pesticides.

Closer to agricultural fields the detection frequency increased for pesticides in both DDM and VFD, as also reported in other studies (e.g. Lemley et al., 2002; Colt et al., 2004, Bennett et al., 2020). However, the above was not found for pesticides that were not applied in the bulb fields and not reported in bulb disinfection. For this group, detection frequency was, as could be expected, independent of the proximity to the fields.

Our results show a less pronounced contrast in detection frequency for pesticides that are used solely in bulb disinfection. This was expected, given that this group is not bound to a fixed period of usage and might be used inside facilities that are not located close to agricultural fields. This information is new and adds to the body of evidence regarding spatio-temporal exposure to pesticides, since exposure to this specific group is more continuous and not bound to a certain time-interval.

The presence of multiple pesticides in both types of dust in the non-use period makes evident that exposure to dust containing pesticides continues outside the actual spraying season. Also, high detection rates might be an indication of slower degradation times (i.e. higher half-life) in the indoor environment. This can be due to a combined indoor accumulation, recirculation and absence of photodegradation in shaded areas inside the household.

4.2. *Concentrations and spatio-temporal distribution*

Our results indicate that, overall, concentrations in indoor dust are higher in farm homes and in location homes closer to fields, and are higher in the period of pesticide usage. These findings match previous reports. Quirós-Alcalá et al. reported that pesticide concentrations were higher in indoor dust of farmer homes compared to non-famer-homes (Quirós-Alcalá et al., 2011). Also, a study done in farm, rural, and urban houses in the New York State (Obendorf et al., 2006) concluded the same, here, samples were taken in different seasons and pesticide concentrations were also higher in summer (i.e. during spraying time) in rural farm homes. Smith et al. also measured higher pesticide levels in indoor dust in the spraying season as compared to the non-spraying season (Smith et al., 2017).

However, the above conclusions do not apply to all pesticides. An interesting finding is that for imidacloprid, an insecticide used in bulb disinfection, concentrations were significantly higher in DDM and VFD of Controls, in the *use* and *non-use* period, respectively. We suspect that these levels are likely driven by either household use (as seen in a study by Deziel et al., 2017) or presence of bulb disinfection sites closer to Controls than Loc Homes. The first being more likely, given that, although the sale of products for agricultural use that contain this insecticide was prohibited in the EU starting from December 2018 (EU, 2018), imidacloprid can still be used against ticks/fleas and also as biocide (against ants, flies, etc) in households. A recent study also found similar imidacloprid concentrations in indoor dust (Shin et al., 2020).

In a recent study, azoxystrobin, pyraclostrobin and trifloxystrobin were also observed in indoor dust from 188 North Carolina homes with similar detection frequencies. However, average azoxystrobin concentrations in Loc Homes were factor 5 to 10 higher than the concentrations observed in that study (Cooper et al., 2020).

Another important finding was the high concentrations of carbendazim measured at both Loc Homes and Controls. These high concentrations are in the same order of magnitude as those found in household dust form homes in California (Shin et al., 2020). Although no longer approved for usage, this fungicide is a degradation product of thiophanate-methyl, which was still allowed for spraying in several different crops until late 2020 (EU, 2019). Thiophanate-methyl is a fungicide with known endocrine disruptive effects (Lu et al., 2004), and other potential adverse health effects (Götte et al., 2020). Exposure to thiophanate-methyl is likely more local given its rapid degradation (see dt50 in Table 1), whilst for carbendazim, a more persistent fungicide in bare soils (6–12 months) (Singh et al., 2016), exposure can be spread across larger areas due to medium and long-range transport and be long-lasting. Concerns regarding the possible long-term risk associated to carbendazim and thiophanate-methyl were also recently reported in a peer-review performed by the European Food Safety Authority (Arena et al., 2018). These concerns were strengthened by the finding of carbendazim in several handwipes (taken from participants in OBO) and strong correlation with urine samples from participants of the OBO study (Oerlemans et al., 2021).

4.3. Pesticides in VFD and DDM: how do they correlate?

To the best of our knowledge, this is the first study to measure, simultaneously, pesticides in dust from doormats and from vacuum floor dust samples. Overall, correlations were low to moderate between the two matrixes with some exceptions.

Statistically significant correlations were found between VFD and DDM for seven different pesticides, which is likely a reflection of the take-home route. We also noticed that these seven pesticides share one commonality: persistence in the soil. Both pyraclostrobin and prothioconazole-desthio are moderately persistent (Zhang et al., 2012 and EPA, 2007, respectively), whilst the remaining five are persistent (Cooper et al., 2020; Matadha et al., 2019; Lewis et al., 2016). Therefore, it seems likely that solely more persistent pesticides are taken home via clothes, skin, vehicles, pets and shoes (i.e. take-home route).

Finally, the poor correlation between VFD and DDM for several pesticides shows that results can be variable depending on the method used. Though, a recent study comparing pesticides in vacuumed outdoor and indoor dust also reported similar correlation ranges (overall moderate) (Simaremare et al., 2021). Correlations between the two matrixes can also be influenced by cleaning habits and other parameters (such as leaving windows open). As previously discussed, DDM captures only a snapshot of the spraying season (a single week in our case), while VFD captures an accumulation over a longer period, therefore also capturing pesticides susceptible to medium to long-range transport. So, it might be that for health assessment studies VFD becomes more relevant, while for exposure assessment DDM has advantages due to the more defined surface and time period of measurements.

4.4. Take-home route

It is evident, from our results, that the take-home route is relevant for Farm Homes, given that occurrence of almost all pesticides in Farm Homes doormats was higher than Loc Homes and Controls. This is also indicated in several publications (Bradman et al., 2009; Marwanis et al., 2019). By extension, the take-home route might also be relevant in homes located further away from agricultural areas, given that several pesticides were detected in the doormats from these homes in both spraying and non-spraying seasons. Moreover, it's not just an important factor in the spraying season but throughout all year, as also concluded by Gunier et al., (2016). Given that humans spend most of their lifetime indoors (Farrow et al., 1997; Baker et al., 2007), the take-home route might be a relevant source for higher indoor exposure to pesticides.

4.5. Determinants and prediction of different pesticides occurrence in dust

With the developed logistic regression model we were able to accurately predict occurrence of a given pesticide in dust in 75% of the samples. This is similar to a study done in the central valley of California, where the occurrence-model had an accuracy around this percentage (ROC C-statistics 70–74%) (Nuckols et al., 2008). The selected explanatory variables, being vapor pressure, half-life in soil, distance to agricultural fields and air concentrations (used to predict concentration in dust) are known parameters in deterministic models for pesticide concentrations and remain the most important determinants of pesticide occurrence in dust.

4.6. Determinants and prediction of different pesticides concentration in dust

Concentrations were more difficult to estimate than occurrence. The linear regression models explained only a small part of the variance in concentrations of different pesticides in dust. Similar results were described by Gunier et al. reporting predictive performance of their models between R^2 0.04–0.28 (Gunier et al., 2011).

Selected explanatory variables vary a lot between each model and it is important to stress that correlation is not necessarily causation. For example, we do not find a rationale for dogs being a factor influencing azoxystrobin and prochloraz concentrations in dust, whilst for imidacloprid it could make sense since it can be used against ticks/fleas on dogs. We also did not find a rational behind the influence in concentrations of three fungicides by reported use of snail or slug bait products. Overall, only the predicted concentration in dust based on air concentrations and distance appear more often as important determinants across all models for pesticide in DDM.

In summary, it is easier to predict which pesticides will be present in dust, but quantitative estimation remains challenging especially for pesticides not directly used in the vicinity of the homes. For quantitative estimates more information about the actual source strengths are needed, but difficult to obtain.

4.7. Strengths and limitations

To our knowledge, this is the first study employing a combination of DDM and VFD sampling techniques. This is one of the main strengths of our study since it allowed us to, first, study what drives pesticide concentrations in dust, second, to understand if patterns between homes and periods are the same for both types of dust, and finally, to better asses the take-home route and infer on possible predictors of exposure to pesticides in dust. Moreover, this is the first study to look at possible determinants of occurrence and concentrations for some current-used pesticides, such as fluopyram, s-metolachlor, flonicamid and thiophanate-methyl.

Our approach allowed us to analyze two different types of dust samples, which is the recommended procedure to ensure complete characterization of contaminants in indoor dust (Schultz et al., 2019). We succeeded in having a good spatial distribution of homes around bulb fields in different locations. Adding to this, collected samples were representative of both use and non-use periods. Lastly, the targeted pesticides are a good representation of varying physico-chemical properties, and the results show the capacity of our approach to detect very low concentrations in dust.

Our study also has some limitations. Not all collected samples were analysed, but we tried to ensure good spatial distribution and analyze samples from homes located in all four main cardinal directions. Dust samples were quite heterogeneous with various types of materials (e.g. pet hair, fibers, dirt) (see Figure B1, supplementary material B) adding to the total mass. These materials differ in physico-chemical properties which will affect pesticide sorption (different for each material) (Mattei et al., 2019). This limitation is however not restricted to our study but to every study that looks into home dust.

Some of the low detection rates might be due to collection of dust before a certain pesticide was applied. Although sampling of VFD and DDM was performed in the middle of the use period, it is still possible that certain pesticides were only sprayed after dust collection. We chose not to collect at the end of the use period to avoid loss of information due to the environmental degradation of the sprayed pesticides.

Although the sampling of VFD was done in the living room to ensure comparability between homes, it might not represent average concentration in indoor dust for each home. The presence of different sources and major activities associated with each type of room are the main drivers of dust composition, therefore it is highly probable that concentrations within the same home vary per room (Lioy et al., 2002). In future studies it is recommended to vacuum more rooms to also have an idea about intra-home variations in pesticide in dust. Especially in attics, chemical burden might be higher than in the rest of the home, as suggested by Cizdziel & Hodge (2000).

Finally, there is also possible variation in DDM due to the placement of the mat. The doormats might not capture everything during the measuring period, as they were covering the main entrance but not all entrances to the home.

5. Conclusion

We found pesticides in indoor dust of all homes included in our study. There is clear evidence that exposure to contaminated dust occurs for longer periods and is not solely bound to homes close to agricultural fields. There is also a clear spatial pattern for both the probability of detection and measured concentrations. Pesticide concentrations in dust from homes closer to fields was in general a factor five higher than controls, as well as factor five higher in the spraying season compared to the non-spraying season.

A statistical model to estimate occurrence of different pesticides in dust was developed. The main determinants were similar to the ones included in current deterministic models. As long as the input data is available, this model can be used in studies to predict what pesticides might be present in the homes.

Lastly, DDM might be a better proxy for pesticides in indoor dust for exposure assessment studies, given that it can be deployed for a certain period and capture exposure in a clearly defined time-frame, whilst VFD might be more appropriate for health assessment, given that it captures both past and current exposures, result of a continuous indoor accumulation and degradation of sprayed pesticides.

Credit author statement

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2022.119024.

Farm Homes – Homes where farmers live; Loc Homes – Homes located within 250 m from a selected field; Controls – Homes located more than 500 m from any agricultural field but in the same region as the Loc Homes.

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