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Predicting soil neonicotinoid content in agricultural landscapes using indirect indicators

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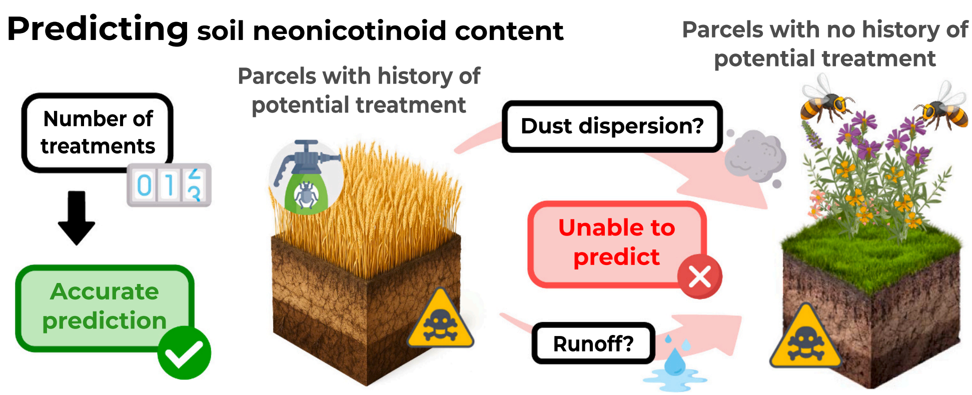
HIGHLIGHTS

- Neonicotinoids were detected in 78 % of sites, mainly imidacloprid and clothianidin.
- Residues found in untreated sites, suggesting contamination from surrounding areas.
- Number of treatments was a better proxy than time since last treatment.
- Dust dispersion and runoff showed weak performance as proxies of contamination.

GRAPHICAL ABSTRACT



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ABSTRACT

Neonicotinoid insecticides are a major driver of pollinator decline. Due to their persistence and mobility in soil, they can contaminate non-target vegetation through runoff or dust, reaching pollinator resources. However, predicting soil contamination is challenging, especially where pesticide use data is lacking. This study assessed the potential of using proxies such as cropping history and landscape structure to predict neonicotinoid content in soils. We analyzed seven neonicotinoids in 86 sites in agricultural landscapes of Belgium. Neonicotinoids were detected in 78 % of sites, with imidacloprid and clothianidin being the most frequently detected (up to 16.3 μg kg^{-1}). In 69 % of sites, contamination occurred without recent recorded treatment and for 33 %, contamination occurred with no recorded treatment history. Risk exposure through soil contact was often high, particularly for clothianidin (44 % of hazard quotient values >1) and imidacloprid (19 %). In potentially treated sites, the total number of treatments was a better predictor of contamination than time since last potential application. Landscape structure poorly predicted contamination, whether when reflecting subsurface water flow or dust dispersion in sites without recorded treatment history. Our research shows that predicting soil contamination at large scales can be approached using treatment history but may be difficult without accounting

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for contamination in non-arable sites. Further research is needed to inform agri-environmental policies with realistic contamination patterns.

1. Introduction

Pollinator health is gaining increased attention as there is growing evidence for important pollinator population declines across the world [54,7]. These declines could have significant consequences, such as causing deficits in crop production, as for example an estimated 5–8 % of global crop yields would be lost without the pollination services provided by bees [29]. Among the several causal mechanisms that are responsible for this decline, pesticide exposure has been considered to be one of the main contributing factors, because of their toxic effects on pollinators [22]. Studies have shown that pesticide residues are often detected in more agricultural soils than those where they were initially applied [26,52,8], degrading the overall habitat quality of farmland landscapes for pollinators [64].

Among pesticide molecules, the class of neonicotinoids, such as clothianidin, imidacloprid, or thiamethoxam, pose a high risk for pollinators and represented in 2014 a share of 25 % of the global insecticide market [53,6]. These molecules are primarily applied as seed coatings to enhance crop yield and performance. However, a major concern is that the vast majority of the active ingredient from the coating (approximately 80–98 %) remains in the soil [1,21]. Neonicotinoids can be absorbed by plant roots and later found in pollen and nectar through translocation and metabolism, or simply stay in nesting habitats of ground-nesting pollinators [69]. As a result, flowering areas are often contaminated, exposing pollinators to significant risks [21,72]. In agricultural landscapes, flowering areas such as wildflower strips and crop margins serve as key foraging habitats for pollinators [33], but they may become sources of exposure if their soils are contaminated with neonicotinoids. With field-realistic soil content (from 0.5 to 30 µg kg^{-1}), particularly under chronic exposure, these insecticides can impact cognitive abilities, larval development, and reproduction [32], as well as increase pollinator susceptibility to diseases [16]. Consequently, the main neonicotinoid molecules (imidacloprid, clothianidin, thiamethoxam, and thiacloprid) were definitely banned in the EU from agricultural use in January 2023 [15,4]. Despite this regulatory progress, new neonicotinoids molecules are being authorized in the EU and may represent a risk for pollinators because of their toxicity and their persistence [59,60].

In agricultural soils, studies consistently highlight neonicotinoids widespread presence [40,45,52]. For instance, they were detected in 97 % of soil samples coming from Swiss cultivated fields, including organically managed fields [26]. This widespread contamination can be explained by the high persistence of these molecules, with some of them remaining detectable in soils years after application [40], but also by their capacity to disperse beyond the site of application. Neonicotinoid contamination can occur both temporally, from residuals of previously treated soils [19,23,8], and spatially, through pesticide drift from adjacent fields [26,52], affecting in parallel nearby flowering crops and field margins [10,38,43]. One of the reason that allows neonicotinoid drifting is that they are highly soluble, which increases their mobility in soils and enables their transport through runoff of surface and subsurface waters [14,67,68]. Soil dust released during the sowing of coated seeds can also participate to neonicotinoid drift and contaminate nearby soils [38,41,65].

Predicting neonicotinoid presence in soil is essential for evaluating habitat quality and ecological risk at the landscape scale. However, since pesticide usage data is often unavailable, such predictions must rely on indirect proxies [13,58]. These proxies, based on the current literature, relate to two main contamination pathways: (1) temporal, via residuals persisting in the soil, which could be predicted using field cropping history as proxy; and (2) spatial, via dust dispersion or subsurface water fluxes, which could be predicted from the composition and the structure of the surrounding landscape. For instance, Chen et al. [13] demonstrated that surrounding cropland coverage can predict soil neonicotinoid residues at larger spatial resolutions, but their analysis did not account for finer landscape structure or the cropping history of individual sites, factors that could strongly influence residue levels. To address this issue and improve neonicotinoid soil content prediction, this study aimed to: (1) quantify the distribution and soil content of seven neonicotinoids in soils across different habitat types to identify where contamination is most likely to occur; and (2) evaluate the ability of two proxy variables (cropping history and surrounding landscape composition) to predict neonicotinoid occurrence and content in soils. This study ultimately aimed to deliver applied predictive tools to guide management decisions that could enhance landscape habitat quality for pollinators.

2. Materials and methods

2.1. Study area

We collected soil samples from 15 study zones in intensive cropland areas of Wallonia, southern Belgium (Fig. 1), a region where 44 % of the land is dedicated to agriculture [63]. Study zones were first selected randomly then filtered, based on several criteria (excluding zones closer than 500 m to urbanized areas, limiting forest surface area, only considering accessible zones; See Supplementary material for more details – Appendix S1). Selected study zones had a mean annual precipitation of 800–1000 mm and a mean annual temperature between 9.5 and 11°C [62]. Mean altitude of sampled study zones was 158 m above sea level.

2.2. Site selection & sampling design

Sites were chosen to represent the agricultural and soil diversity of Wallonia, by looking at the different landscape variables in our study zones (cropping history, soil texture and drainage, topography, proximity to other crops). We did not sample any organic crop parcel, as they only represented 1.7 % of our study area. Each site was sampled once over a three-year period (2023–2025), between late February and early April. We surveyed 86 different sites. In some sites, we collected two (35 % of all sites) to three (37 %) soil samples 10 m apart within the same visit to evaluate the average heterogeneity of neonicotinoid soil content (Fig. 1). In total, we collected 180 samples. Each soil sample was a composite of six subsamples taken from a 1 m radius circle, that were mixed and homogenized in the field. Samples were stored at −18°C until analysis. Samples were collected for 79 % of the time on loamy soils, 12 % on rocky soils and 9 % on other types of soil texture. Average soil pH of our samples was 6.46 ± 0.71, and average organic carbon content was 13.51 ± 6.60 g.kg^{-1}.

2.3. Chemical analysis

We sieved soil samples to 2 mm before extraction. We then mixed 5 g of soil with 5 mL of water. After shaking then waiting 30 min, we mixed the solution with 10 mL of acetonitrile acidified with 2 % formic acid, shook again and waited another 30 min. We then added QuEChERS salts (containing 4 g $MgSO_4$, 1 g NaCl, 0.5 g sodium citrate dibasic sesquihydrate, 1 g sodium citrate tribasic dihydrate) from Agilent (USA) to the solution. After homogenization and centrifugation 15 min at 4800 rcf at 4°C, we took the supernatant and filtered it to 2 µm on a PTFE filter. This filtered solution was then analyzed by liquid chromatography (Nexera

X2™Shimadzu, USA) coupled with a Time-Of-Flight Mass Spectrometer (Q-TOF) (X500R ABSciex, Singapore). The column used was a Waters ACQUITY UPLC™ HSS T3, 100 mm × 2.1 mm internal diameter., 1.8 µm particle size kept at 40°C with a mobile phase gradient (0.3 mL min^{-1}) consisting of: MilliQ water/ methanol (90/10) and 2 mM of ammonium formate acidified with 0.1 % of formic acid in lane A and methanol with 0.1 % of formic acid in lane B. The gradient goes from 100 % aqueous phase to 100 % organic phase in 4 min, stay 4.5 min in 100 % organic phase and goes back to 100 % aqueous phase in 1 min and stay in aqueous phase during 6 min for system conditioning between injections. Analysis was done in multi reaction monitoring mode (MRM) in electrospray positive ionization mode (ESI+). We measured concentrations of seven neonicotinoid molecules: acetamiprid, clothianidin, flupyradifurone, imidacloprid, sulfoxaflor, thiacloprid, and thiamethoxam. Detection limits were considered as half of the quantification limit. Samples that were above detection limits (DL) but below quantification limits (QL) were considered as having concentrations equal to 3/4 of the corresponding molecule QL (See Supplementary material – Appendix S2). We assumed no detection (below DL) was equivalent to 0.00 µg kg^{-1}. Variation of measure heterogeneity was assessed at three different concentrations using control samples (See Supplementary material – Appendix S2). All data used in these analyses are available in the Supplementary material.

2.4. Risk assessment for pollinators

For each site, we quantified neonicotinoid risk to pollinators by calculating the total neonicotinoid toxicity equivalent (TEQ) and the soil contact hazard quotient (HQ). TEQ values were calculated based on *Apis mellifera* oral acute LD_{50}, using imidacloprid as a reference [34]. Distribution of TEQ values for all sites is given in Supplementary material – Appendix S3. Hazard quotients were calculated based on Willis Chan et al. [69], and reflects the ratio between the estimated neonicotinoid dose an adult female *A. mellifera* receives from soil contact and the corresponding toxicity threshold. An HQ > 1 indicates that the exposure exceeds the level expected to cause adverse effects.

2.5. Landscape data

Landscape data (crop and soil information, landscape structure) was analyzed using R 4.5.2 [50], QGIS 3.32 [48], and ArcGIS Pro 3.2.0 [17]. Data sources and processing methods are described in the following sections.

2.5.1. Site soil information and cropping history

Crop rotation information was extracted from the administrative Walloon land parcel identification system (SIGEC) from 2015 to 2024. We simplified the crop classification system from 243 to 54 categories (Supplementary material – Appendix S4). Due to interannual variations in parcel boundaries, we used the geometrical parameter called pole of inaccessibility to track each parcel across years, which corresponds to the furthest point from the edges of the parcel [20] and intersected this point with the crop data for each year. Due to the limits of this method, cropping history of site locations were tracked across years using the site GPS position.

In the context of this study, because neonicotinoid usage data per farmer is not publicly available, we use the term "potentially treated crop" to refer as a crop that was legally permitted to be cultivated using neonicotinoids at a specific period, according to Belgian neonicotinoids authorization history [46]. To account for potential misclassification errors (i.e. crop parcels incorrectly classified as treated), we conducted a probability sensitivity analysis considering different misclassification scenarios of rates of 10, 20, 30, and 40 %. As information on intermediate crops was unavailable, we assumed each site had a single crop per year. "Sites with pollinator-attractive resources" is used in this study to refer to sites with grasslands, fallows and attractive flowering crops (specifically rapeseed and legume crops). Soil variables (i.e., soil texture and soil drainage) were extracted from the digital soil map of Wallonia (CNSW) after simplification of the categories (Supplementary material – Appendix S5).

2.5.2. Watershed definition

For each site, we computed the delimitation of the corresponding watershed area, i.e., the contributing area from where water fluxes

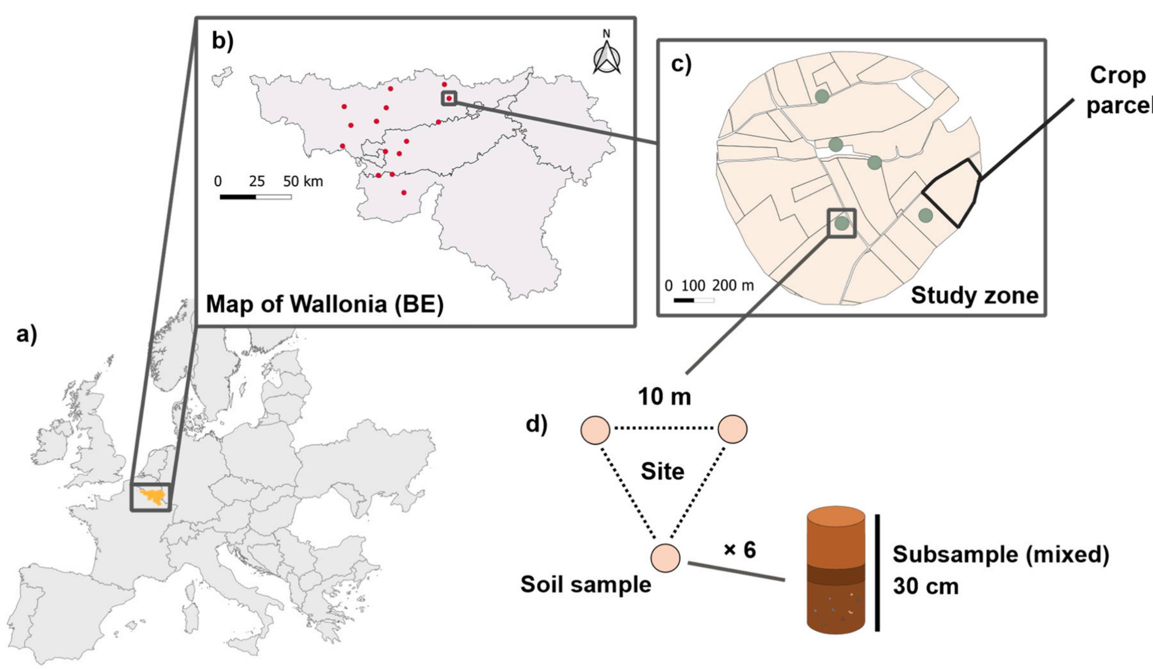

Fig. 1. Sampling design: location of the 15 study zones in Wallonia, Belgium. The background map represents the different agricultural regions of Wallonia (a-b), example of a study zone and sampling sites (c), with each site including samples of six soil subsamples (d). To evaluate the average heterogeneity of neonicotinoid soil content, sites were sampled one to three times.

could reach the site location. We used the *flow direction* tool followed by the *watershed* tool [28,49] from ArcGIS Pro software on a 10 m resolution digital elevation model [56]. We then measured the sum of area (land coverage) of potentially treated crops intersecting with the computed watershed zone over the course of one, two or three years prior to the sampling year of each location. We did not look further in time to avoid interfering with unrelated background noise (See detailed method, supplementary material – Appendix S6).

2.5.3. Proximity buffers

Around each site location we computed crop areas intersecting within a 200 m radius buffer, as dust dispersion has been proven to reach up to at least 90 m and probably even further [30]. Similarly, as for the watershed area, we measured the sum of the intersecting areas within those buffers with potentially treated crops, one, two or three years prior to the sampling year of each location.

2.6. Statistical analysis

All statistical tests were performed using R 4.4.1 [50]. Results were considered statistically significant at $p < 0.05$. P-values were adjusted to the multiplicity of tests made using a simple Bonferroni correction. Pearson' r correlation coefficient and Welch *t*-test significancy were assessed using the *stats* package. Models were only tested with clothianidin and imidacloprid due to a statistically insufficient number of samples above detection limit for the other molecules. To account for the spatial heterogeneity of agricultural landscapes, we added in all models a random effect considering the variability between study zones. All models were tested for over-/underdispersion using the *DHARMa* package [24]. All model results are given in Supplementary material (Appendix S7). Soil heterogeneity was evaluated by using the relative standard deviation (RSD), calculated by multiplying the mean of sample neonicotinoid content collected at a same site by the standard deviation.

2.6.1. Cropping history

Statistical tests performed to assess the impact of cropping history (time since last potential treatment and number of potential treatments) were computed using linear mixed-effect models made using the *lmer* function from the *lmerTest* package [31]. R^2 values shown in the figures are conditional R^2 from models, accounting for both fixed and random effects computed with the *r.squaredGLMM* function of the *MuMIn* package [5]. The model used for assessing the impact of cropping history with time since last treatment on soil neonicotinoid content was:

$$C = \mu + \beta \times \log(\theta + 1) + U_z + \varepsilon$$

Where C is the soil content of either clothianidin or imidacloprid of a site, β the effect of time since potential treatment, θ the number of years since the last cultivation of a potentially treated crop in the history of the site parcel, U_z a random effect linked to the study zone, and ε the residual term following $N(0, \sigma^2)$. Sites with no sugar beets (or sugar beets/vegetable for imidacloprid) in their cropping history were not included in this model since time since last potential treatment does not apply.

Similarly, the model for assessing the impact of cropping history with the number of potential treatments on soil neonicotinoid content was established as:

$$C = \mu + \beta \times \theta + U_z + \varepsilon_s$$

Where C is the soil content of either clothianidin or imidacloprid, β the effect linked to the number of potential treatments, θ the number of occurrences in the history of the parcel where the sample composite was collected, U_z a random effect linked to the study zone, and ε the residual term following $N(0, \sigma^2)$.

2.6.2. Landscape composition

The effects of crop surroundings were tested using presence/absence data of neonicotinoid with a generalized linear mixed model with a logit link made with the *glmmTMB* function of the *glmmTMB* package [11]. The tested variables were scaled (subtracted by the mean and divided by the standard deviation) before computation. Conditional R^2 values were computed with the *r2* function of the *performance* package [39]. The model used for assessing the impact of neonicotinoids transport (dust dispersion / subsurface water fluxes) on the probability of absence/-presence of neonicotinoids in soil was:

$$\log\left(\frac{p}{1-p}\right) = \mu + \beta_1 \times \textit{cumulated area}_{runoff} + \beta_2 \times \textit{cumulated area}_{dust\ dispersion} + U_z$$

Where p is the probability for a site to have detectable concentrations of either clothianidin or imidacloprid, *cumulated area*$_{runoff}$ is the sum of the surface area inside of the watershed delimited zone, across one, two or three years depending on the model and β_1 is its related effect. *cumulated area*$_{dust\ dispersion}$ is the sum of the surface area inside of the dust dispersion delimited zone (200 m buffer), across one, two or three years depending on the model, and β_2 its related effect. U_z a random effect linked to the study zone.

3. Results and discussion

3.1. Assessment of neonicotinoid content in agricultural soils

3.1.1. Concentrations and ranges

Among all sites, 67 out of 86 sites (78 %) were measured above DL for at least one of the seven analyzed neonicotinoid molecules. Frequency of detection, mean soil content and value ranges for each molecule are given in Supplementary material – Appendix S8. Clothianidin and imidacloprid were the most frequently detected, in 60 and 59 % of sites, respectively. Among the 101 samples above QL (56 % of all samples), clothianidin content ranged from 0.40 to 16.3 μg kg^{-1} , and imidacloprid content from 0.40 to 6.40 μg kg^{-1} (Fig. 2). When accounting for molecule-specific toxicity, the mean TEQ averaged 1.92 ± 2.79 μg kg^{-1}, ranging up to 21.79 μg kg^{-1}. Soil contents of both clothianidin and imidacloprid were highly correlated together (Pearson's r: 0.74; $p < 0.001$). Other neonicotinoids (thiacloprid, acetamiprid, thiamethoxam, and flupyradifurone) were detected in fewer than 9 % of sites, with soil content ranging up to 0.42 μg kg^{-1} , and sulfoxaflor was never detected. Our results were consistent with findings from Humann-Guilleminot et al. [26] and Riedo et al. [52], both of whom reported that clothianidin and imidacloprid were the most frequently found molecules in cultivated soils, but also those found with the highest content. Conversely, thiacloprid and thiamethoxam were even less frequently detected in our study (3 %) than in theirs (10–28 %).

This marked contrast in detection frequencies can largely be attributed to differences between molecules in soil persistence. Clothianidin and imidacloprid, have longer soil half-lives (120–280 days on average; [34,37]) than the remaining compounds (approximately 3–40 days). The only exception is flupyradifurone, which also exhibits a relatively long soil half-life (approximately 130 days), but was rarely detected. But additionally, differences in detection frequencies can also reflect historical use patterns and changes in regulatory status. Clothianidin and imidacloprid have been widely used on sugar beet and vegetable crops until their authorization was permanently withdrawn in 2022, shortly before our sampling campaign began [46]. At the time of sampling, only acetamiprid, flupyradifurone, and sulfoxaflor remained authorized within the EU [18,61], and yet all were detected at relatively low frequencies, suggesting either more rapid degradation in soil or in the case of flupyradifurone, probably limited use due to a recent introduction. Notably, clothianidin is also a degradation product of thiamethoxam [36], although we cannot quantify in this study the proportion of clothianidin that is derived from the degradation of thiamethoxam.

Clothianidin, imidacloprid and flupyradifurone showed RSD values

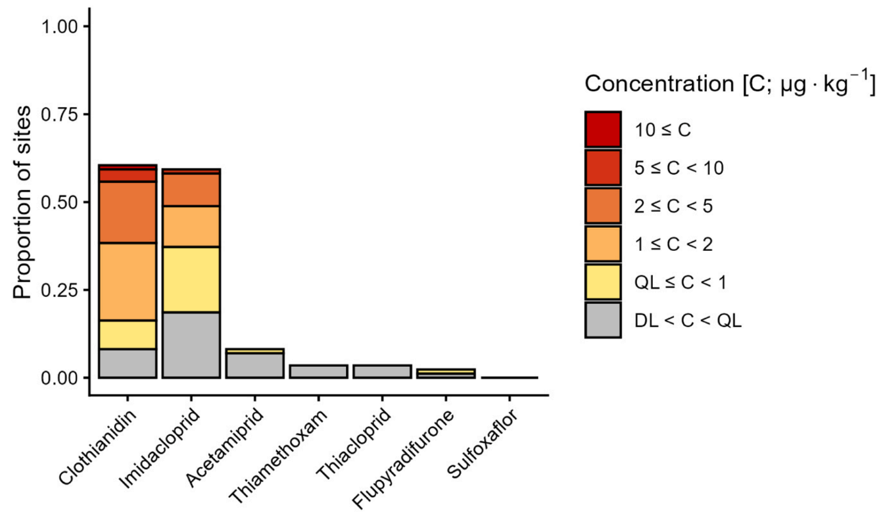

Fig. 2. Proportion of sites above detection limit for each neonicotinoid molecule. The color represents the measured concentration range of the collected samples. In total, 86 sites were analyzed. C = concentration; QL = quantification limit; DL = detection limit. QL (μg kg^{-1}): Clothianidin = 0.4; Imidacloprid = 0.4; Acetamiprid = 0.2; Thiamethoxam = 0.5, Thiacloprid = 0.5; Flupyradifurone = 0.4; Sulfoxaflor = 4. Detection limits and heterogeneity of sampling measurement are provided in Supplementary material – Appendix S2.

(mean RSD: 31.7 % for clothianidin, 33.7 % for imidacloprid, and 12.0 % for flupyradifurone) similar of what can be found in the literature for pesticide heterogeneity in soils [51]. The remaining molecules showed higher soil heterogeneity (mean RSD: 130–173 %). However, RSD estimations of flupyradifurone, acetamiprid, thiamethoxam, and thiacloprid should be considered carefully, due to the high amount of data below DL resulting in too few replicate data. This heterogeneity can be explained by variations in seeding density across farmers [27], but also by variations in local soil structure and composition [3]. Our findings suggest that the feasibility of predicting soil neonicotinoid content with a spatial resolution below 10 × 10 m is highly dependent on the molecule. This should be carefully considered in future fine-scale prediction efforts to reduce errors.

3.1.2. Characteristics of contaminated sites

Clothianidin and imidacloprid were the most detected molecules and were also banned at the time of sampling. We will thus mainly focus on these two molecules to allow for robust statistical analysis and to reduce the risk of confounding effects from recent seed treatments in soil samples. We found that 59 sites (69 % of all sites; 94 % of sites above DL) had no potentially treated crops in the year immediately preceding sampling (i.e., no sugar beets regarding clothianidin; no sugar beets and no vegetables regarding imidacloprid). In these sites in particular, soil contents ranged from 0.30 to 16.3 μg kg^{-1} for clothianidin (mean: 1.70 ± 2.16 μg kg^{-1}) and from 0.30 to 6.40 μg kg^{-1} for imidacloprid (mean: 0.99 ± 1.28 μg kg^{-1}). TEQ values averaged 1.84 ± 2.77 μg kg^{-1}. Two sites exhibited detectable content up to seven years after the last recorded use of a potentially treated crop. This order of persistence does not align with the average half-life estimates reported in the literature [35]. However, given the high variability in estimates coming from the literature (reaching up to 1400 days for clothianidin and 1300 days for imidacloprid; [21]), our observations are more consistent with the upper

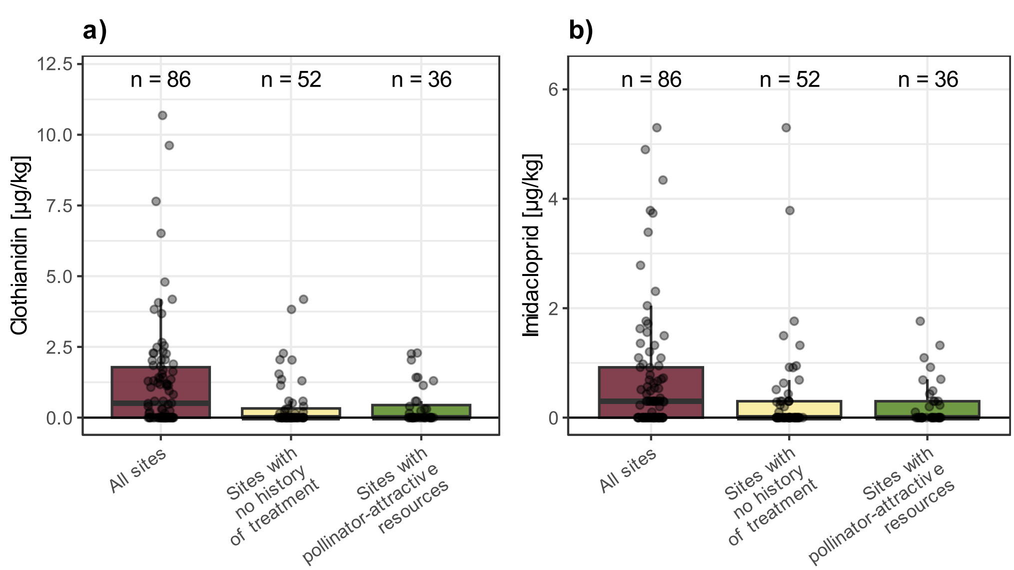

Fig. 3. Content of clothianidin (a) and imidacloprid (b) of site soils depending on the type of ecological habitat. “All sites” refer to all sites sampled in this study. “Sites with no history of treatment” refer to those with no history of cultivation of a potentially treated crop (see section 2.4.1). “Sites with pollinator-attractive resources” refer to grasslands, fallows and attractive flowering crops (rapeseed, legume crops). The sum of all category’s sites does not equal the total number of sites sampled in our study because categories overlap.

end of the reported range.

Some sites exhibited contamination despite having no recorded history of potentially treated crops between 2015 and 2024. These sites represented 33 % of all sampled sites (n = 28; Fig. 3). Soil content in these sites ranged from 0.15 to 4.18 μg kg^{-1} for clothianidin (mean: 0.70 ± 1.12 μg kg^{-1}) and from 0.10 to 3.79 μg kg^{-1} for imidacloprid (mean: 0.53 ± 0.80 μg kg^{-1}). TEQ values averaged 0.66 ± 1.28 μg kg^{-1}. These sites specifically did not share a specific crop rotation pattern. They show, along with precedent examples of the literature [10,43] that cropping history only is insufficient to predict neonicotinoid soil contamination, underscoring the need to identify alternative proxies to predict their occurrence.

When looking across all sites, risk exposure through soil contact was often high for pollinators, particularly for clothianidin: 44 % of sites had HQ values above 1 (up to 9.97), 19 % for imidacloprid (up to 4.43), and an absence of exceedance for thiamethoxam. Sites with no recorded history of potentially treated crops exceeded the hazard limit in only 8 % of cases for clothianidin and 5 % for imidacloprid. These levels are particularly concerning for ground-nesting pollinators, for which direct contact with contaminated soil poses a serious survival risk.

We observed contamination where pollinator-attractive resources were growing in 22 sites (59 % of all sites with attractive resources). In these sites, mean concentrations were 0.65 ± 0.79 μg kg^{-1} for clothianidin (up to 2.28 μg kg^{-1}) and 0.40 ± 0.49 μg kg^{-1} for imidacloprid (up to 1.76 μg kg^{-1}). Pollinator-attractive resources can be flowering habitats like fallows, grasslands, legumes crops, or rapeseed [42,44]. It is difficult to estimate how much content will contaminate the floral resources foraged by pollinators because plant uptake of neonicotinoids varies significantly depending on the molecule, the floral species, local environmental conditions and application methods [43,72]. Clothianidin, for example, may persist longer in annual plants, whereas imidacloprid could exhibit greater persistence in perennial species [12,9]. Consequently, sites with different plant communities and/or local conditions offer varying levels of contaminated resources, complicating the assessment of the overall risk through oral exposure for pollinators. Therefore, if soils supporting pollinator-attractive resources and those with no history of treatment are contaminated, it becomes essential to develop predictive methods at the landscape scale to effectively guide pollinator conservation strategies. This issue is particularly critical when former arable land is converted into wildflower strips as a conservation measure, since residual soil contamination may persist and potentially turn these habitats into "trap resources" for pollinators [2,66].

3.2. *Evaluating landscape proxies for predicting neonicotinoid soil contamination*

In this section, we investigate whether landscape-level proxies can predict soil neonicotinoid content in individual sites. This method is necessary because neonicotinoid usage data is often not available, or as it is the case in Belgium, only accessible at a regional scale, which is

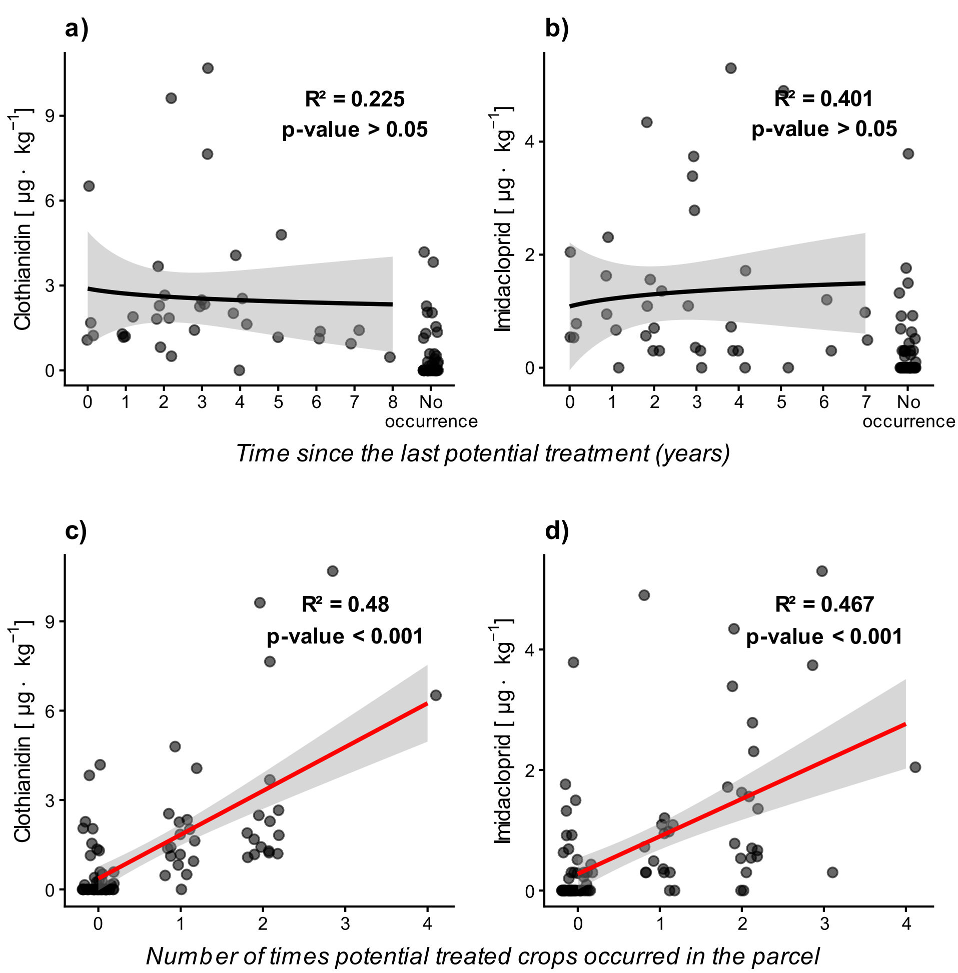

Fig. 4. Soil content of clothianidin and imidacloprid according to the time in years passed since the site has been potentially cultivated with neonicotinoids (a-b), and according to the number of times a site has been potentially cultivated with neonicotinoids (c-d). Effects of (a) and (b) were modeled using a logarithmic relationship. A red line indicates a significant correlation between content and the corresponding parameter, a black line a non-significant relationship. The grey area represents a 95 % confidence interval. Potentially treated crops with clothianidin refer to sugar beets, and with imidacloprid refer to sugar beets and vegetables.

insufficient for accurately predicting soil residues [13].

3.2.1. Cropping history

We used field cropping history, focusing on potentially treated crops (according to cropping history and Belgian legislation) and we first found no significant correlation between the number of years since the last cultivation of potentially treated crops and the soil content of clothianidin and imidacloprid ($p > 0.05$ for both molecule; Fig. 4a and b; see models, Supplementary material – Appendix S7). The number of potential treatments in the prior site history proved to be a better proxy for soil neonicotinoid content, with clothianidin and imidacloprid content showing a significant positive association with it (Fig. 4c and d). Accumulation patterns increasing with the number of treatment have been previously reported in the literature, with soil content typically plateauing after 3–5 years of repeated applications [55,71]. We did not observe such a plateau in our dataset, likely due to the limited number of sites that had received three or more potential treatments. Time since last treatment and number of treatments were highly correlated (Pearson's $r = -0.63$, $p < 0.001$). Results were robust when compared to scenarios simulating misclassification of potentially treated crops as non-treated (no significant changes between scenarios). Such misclassifications were likely minimal, as neonicotinoid-treated seeds represent the majority of sugar beet seeds in Belgium [25], and potentially contaminated crops were majorly sugar beets in our study (86 % of all potentially treated occurrences). Additionally, 97 % of sites cultivated within the past seven years with a crop potentially treated with clothianidin were contaminated, as well as 88 % for imidacloprid, reinforcing the idea that using potential treatment as a proxy is a feasible approach. Using such simplification for landscape scale predictions is likely to be accurate; however, its accuracy may vary depending on regional agricultural practices. Overall, for sites with a cropping history of treatment, the frequency of precedent potential treatments seems to be the best predictor for assessing soil neonicotinoid content.

3.2.2. Landscape composition

In this section, we examine whether landscape composition and structure can serve as proxies for neonicotinoid presence in soils in sites with no cropping history of treatment. Our results showed that some sites, despite having no record of potentially treated crop cultivation, still exhibited detectable content of neonicotinoids in the soil. Such contamination of apparently untreated sites is not uncommon and has been reported in previous studies [10,38,43]. Based on the current literature, we assume these contaminations could be explained by two main pathways: the spatial transport of neonicotinoids via dust dispersion through the air [41] or via runoff through surface and subsurface water fluxes [68,70]. To explore the relevance of these two pathways, we tested whether the land coverage of nearby potentially treated crops, either in close proximity to the sampling point (to account for dust deposition) or within the watershed boundary (for runoff), could be used as a proxy to predict the presence/absence of neonicotinoids in soils. However, we found no significancy regarding both routes of contamination ($p > 0.05$ for both routes and for clothianidin and imidacloprid; see models, supplementary material Appendix S7). More interestingly, most of these contaminations could not be explained by dust dispersion nor runoff, at least when looking at the area one year preceding the sampling (70 % of sites had no potentially treated crops in their surroundings; Fig. 5). Similarly to cropping history, results were robust when compared to scenarios simulating misclassification of potentially treated as non-treated crops (no significant changes). This suggests that local landscape structure seems to be an inefficient tool to reliably predict neonicotinoid drifting contamination.

Given the possibility that neonicotinoid drift may have occurred earlier in time, we also tested to extend the temporal window in which we were accounting for the land coverage of potentially treated crops in the surrounding landscape, two and three years prior to sampling. However, no significant correlations were observed ($p > 0.05$ for both time windows; see models in Supplementary Material – Appendix S7). Finding a predicting parameter that accounts for time windows longer than 3 years prior becomes increasingly challenging, as expanding the temporal scope introduces a lot of additional background noise into the data. Additionally, some of our models had low statistical power, which may explain the lack of detected effects of the surrounding landscape.

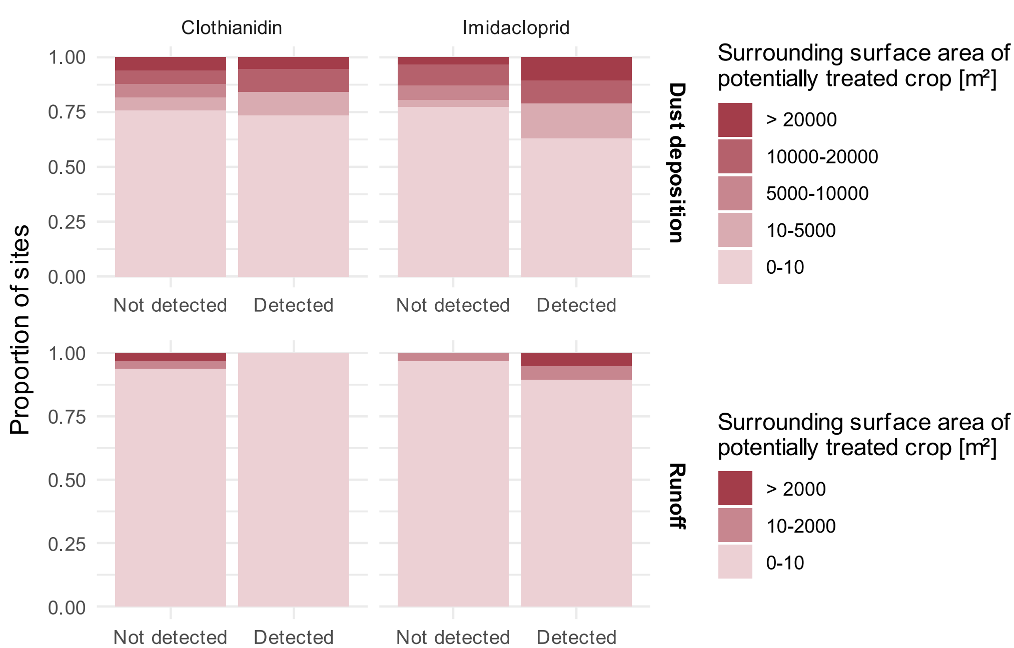

Fig. 5. Proportion of different ranges in quantity of land coverage surrounding the site location in contaminated versus uncontaminated samples for imidacloprid and clothianidin one year prior to sampling. The first row refers to the surrounding area in a 200 m buffer (hypothesis of transport through dust deposition). The second row refers to the delimitation of a watershed zone (hypothesis of transport through runoff). Potentially treated crops with clothianidin refer to sugar beets, and with imidacloprid refer to sugar beets and vegetables. To account for computation errors, we considered surroundings areas of less than 10 m² in the same range as null.

It is worth mentioning that surroundings of sites contaminated with imidacloprid exhibited a higher frequency of potentially treated crops three years prior to sampling (Supplementary material, Appendix S9), although the difference compared to uncontaminated samples was not statistically significant. Imidacloprid may exhibit greater spatial mobility across the landscape due to its higher water solubility relative to clothianidin (610 mg L^{-1} for imidacloprid compared to 327 mg L^{-1} for clothianidin), potentially facilitating broader dispersion through runoff.

Cropping history and spatial transport could act as parallel contamination mechanisms contributing to soil neonicotinoid levels. However, the methods used in this study cannot distinguish drift resulting from dust dispersion from that caused by runoff. This means that cultivated sites might have been contaminated via neonicotinoid drift, which could have partially offset the soil concentrations that we tried to correlate with local treatment history. Future prediction efforts should therefore consider incorporating both mechanisms simultaneously. These efforts might be improved by incorporating data on seeding calendars and local wind conditions together to more accurately capture drift dynamics.

Other sources of contamination, not considered in this study, may also play a role in explaining the presence of neonicotinoids in soils. For example, previous studies have highlighted the possibility of neonicotinoid-contaminated manure acting as a diffuse input into agricultural soils [57]. Similarly, neonicotinoids applied as foliar sprays (which were not included in the scope of this study) have been shown to enter the atmosphere and contaminate surrounding areas via precipitations [47]. Future attempts to predict neonicotinoid presence in soils should consider a broader range of contamination sources and pathways, particularly in relation to the mode of application.

Only the number of potential treatments of sites allowed prediction of soil neonicotinoid content in cases with a history of application. However, these results only apply to field- and parcel-scale conditions for now and should be tested before being extrapolated to larger administrative or regional scale. For sites with no such history, no reliable landscape predictors of contamination were identified. Due to the complexity of sources and pathways, the dominant processes driving contamination in these cases remain unclear. As a result, predicting soil content at large scales may not be feasible using indirect landscape proxies.

4. Conclusion

Our results show that neonicotinoids are widespread in agricultural soils and majorly present in sites with a history of potential treatments. Clothianidin and imidacloprid were the most detected molecules. Neonicotinoids residuals can be detected in sites where they were not directly applied, even in sites with pollinator resources, suggesting that these pesticides drift spatially. These findings highlight the need to develop methods for predicting soil contamination at broader spatial scales. We tested the ability of cropping history, local landscape structure and surrounding crop coverage to account respectively for soil residual contamination, contamination through runoff and through dust dispersion. In sites with a potential history of treatments, the number of applications provided a meaningful estimate of neonicotinoid content. In sites with no history of treatments, we found no reliable predictors of contamination. The processes driving the spatial drift of neonicotinoids at these sites remain unclear; however, the methods used in this study may not have allowed us to isolate the different pathways. Given the diversity of sources and contamination pathways, an accessible and low-cost proxy for predicting soil neonicotinoid content at large scales may not be achievable, emphasizing the need for more precise and integrative approaches. Further research is essential to ensure that agri-environmental policies, particularly those promoting nesting habitats, or melliferous habitats such as flowering strips, are guided by realistic contamination patterns, to avoid inadvertently creating ecological traps for pollinators.

Environmental implications

Neonicotinoids are systemic insecticides increasingly acknowledged as a major environmental threat to pollinators, consistently degrading habitat quality in agricultural landscapes. Due to their chemical properties and seed-coating application, most of the active ingredients often remain in the soil, where they persist for years and potentially disperse beyond treated fields, contaminating nearby pollinators foraging and nesting habitats. Developing large-scale, cost-efficient predictive tools is therefore essential to minimize chronic contamination of pollinator resources. Our research suggests that while such tools are feasible for assessing crop-soil contamination based on cropping history, predicting contamination in adjacent untreated habitats remains challenging.

CRediT authorship contribution statement

Anne-Laure Jacquemart: Project administration, Funding acquisition, Conceptualization. **Pierre Defourny:** Writing – review & editing, Project administration, Funding acquisition, Conceptualization. **Julien Radoux:** Writing – review & editing, Conceptualization. **Yannick Agnan:** Writing – review & editing, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Maxime Buron:** Writing – original draft, Visualization, Software, Methodology, Investigation, Formal analysis, Data curation. **Émile Foguenne:** Investigation, Formal analysis. **Alodie Blondel:** Writing – review & editing, Methodology, Investigation. **Thomas Marchetti:** Methodology, Investigation. **Léna Jeannerod:** Writing – review & editing, Methodology.

Declaration of Generative AI and AI-assisted technologies in the writing process

During the preparation of this work the author used OpenAI language model ChatGPT in order to improve readability, flow, and overall English quality. After using this tool/service, the author reviewed and edited the content as needed and takes full responsibility for the content of the publication.

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Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Anne-Laure Jacquemart reports financial support was provided by Walloon-Brussel federation. If there are other authors, they 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. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.140794.

Data availability

All scripts used in this paper are publicly available online at: https://github.com/VecTax/Buron_neonicotinoids. Data used in this study is available as an excel file in the Supplementary material.

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