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Pesticide exposure of wild bees and honey bees foraging from field border flowers in intensively managed agriculture areas

Laura T. Ward [a,*], Michelle L. Hladik [b], Aidee Guzman [a], Sara Winsemius [a,c], Ariana Bautista [a], Claire Kremen [a,d], Nicholas J. Mills [a]

[a] Department of Environmental Science, Policy, and Management, University of California, 130 Mulford Hall #3114, Berkeley, CA 94720-3114, USA
[b] U.S. Geological Survey, California Water Science Center, 6000 J St., Placer Hall, Sacramento, CA 95819, USA
[c] Department of Land, Air, and Water Resources, University of California, One Shields Ave, Davis, CA 95616-8627, USA
[d] Institute for Resources, Environment and Sustainability, Dept of Zoology, Biodiversity Research Centre, 429-2202 Main Mall, University of British Columbia, Vancouver, BC V6T 1Z4, Canada

HIGHLIGHTS

- Flowering field border plants contain pesticides from greater landscape.
- Different bee types foraging in the same region exhibit different exposure profiles.
- Pesticide residues (ng per bee) and bee size are positively correlated.
- Exposure to bifenthrin and pesticide mixtures could adversely affect bee health.
- Landscape level analysis is needed to understand pesticide exposure for bees.

GRAPHICAL ABSTRACT

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ABSTRACT

Bees are critical for food crop pollination, yet their populations are declining as agricultural practices intensify. Pollinator-attractive field border plantings (e.g. hedgerows and forb strips) can increase bee diversity and abundance in agricultural areas; however, recent studies suggest these plants may contain pesticides. Pesticide exposure for wild bees remains largely unknown; however, this information is needed to inform agricultural practices and pesticide regulations meant to protect bees. It is important to determine whether border plantings that attract and support pollinators may also deliver pesticides to them. In this study, we collected various samples for pesticide residue analysis, including: multiple species of wild bees, honey bees, flowers from four types of bee-attractive field border plants, and soil. Silicone bands were also utilized as passive aerial samplers of pesticide residues. The five pesticides detected most frequently across all samples were the insecticide bifenthrin, the herbicides thiobencarb, metolaclor, and propanil, and the fungicide fluopyram. We detected the greatest number of parent pesticides in bands (24), followed by soil (21). Pesticides were also detected in field border plant flowers (16), which do not receive direct pesticide applications, and included many products which were not applied to adjacent field crops. Pesticide concentrations were lower in bees than in flowers but higher in bees than in soils. Pesticide residue per bee (ng/bee) increased with increasing wild bee size, though pesticide concentration (ng/g) did not increase. While honey bees and wild bees contained a similar number and concentration of pesticides overall, pesticide mixtures varied by bee type, and included some

* Corresponding author.
E-mail address: lauraward@berkeley.edu (L.T. Ward).

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mixtures known to cause sublethal effects. The results from this study highlight the benefits of measuring more sample types to capture the total exposome of bees, including a greater range of bee species, as well as the need to consider exposure to pesticides at the landscape level.

1. Introduction

Wild bees and honey bees (*Apis mellifera*) are crucial in agroecosystems because pollination services are critical to human nutrition and global food security (Eilers et al., 2011; Klein et al., 2007). However, bees are declining worldwide due to land-use change, especially agricultural intensification, which has caused significant habitat loss and increased pesticide exposure to bees (Goulson et al., 2015; Kovács-Hostyánszki et al., 2017; Potts et al., 2010).

One practice to promote bee conservation in agricultural landscapes has been the restoration of bee-attractive field border plants, such as hedgerows and flower strips (Albrecht et al., 2020; M'Gonigle et al., 2015; Morandin and Kremen, 2013; Ponisio et al., 2016; Venturini et al., 2017). Field border plantings are considered a cost-effective conservation strategy (Morandin et al., 2016) that can provide continuous floral resources for bees in the otherwise forage limited monocultural landscapes (Blaauw and Isaacs, 2014) while also increasing crop pollination and crop yield (Boyle et al., 2020; Garibaldi et al., 2014). In fact, these border plantings are known to support higher bee species richness than crop fields (Sardiñas and Kremen, 2015), improve long-term population dynamics (Kremen et al., 2019; Ponisio et al., 2019), and decrease parasite presence (Cohen et al., 2021). Recent evidence, however, suggests that both cultivated, bee-attractive and uncultivated (i.e., weedy) border plants harbor pesticides that have been applied to crops (Botías et al., 2015; David et al., 2016; Long and Krupke, 2016) and may be a source of pesticide exposure for honey bees and wild bees. Significant pesticide exposure for bees from field border plants, including plants purposefully introduced to support pollinators, could counteract the intended conservation benefits of this land use practice.

Wild bees contribute to pollination services in many agricultural areas (Dainese et al., 2019; Pitts-Singer and James, 2008), yet most pesticide research with bees to date has largely focused on honey bees (Kopit and Pitts-Singer, 2018; Sgolastra et al., 2019). Bees are incredibly diverse with thousands of species and thus exposure and sensitivity to pesticides likely varies among species, especially for those with differing life histories (Kopit and Pitts-Singer, 2018). Though more efforts have been made to include a select number of other managed and wild bee species as models for pesticide risk assessment (e.g., Botías et al., 2017), this research is still limited (Sgolastra et al., 2019). Furthermore, few studies have looked at pesticide exposure for wild bees in the field (Hladik et al., 2016; Longing et al., 2020; Main et al., 2020); most have been laboratory or cage studies (Lundin et al., 2015) using a subset of species that can be reared for laboratory research (Kopit and Pitts-Singer, 2018; Sgolastra et al., 2019). Routes of exposure, particularly the variation in exposure, in field-realistic conditions remain largely unknown for the majority of wild bee species, yet this information is essential for understanding pesticide risk to wild bees to complement bee conservation efforts.

To better understand pesticide exposure for different groups of bees in an agricultural landscape, we examined pesticide residues from different sample types, including wild bees, honey bees, flowers, soil, and air (via passive samplers) at field sites with both perennial and annual, cultivated and uncultivated bee-attractive field border plantings. The objectives of this study were to determine if (1) the flowers of field border plants harbor pesticides, (2) the composition and/or concentration of pesticides differed among bees, flowers, soil, and air, and (3) pesticide exposure differed among flower types (i.e., planted herbaceous, weedy, and woody plants) and/or among wild bees of differing size and foraging ranges. This study is one of the first to examine pesticide exposure for a wide variety of bees visiting field border plants in an agricultural landscape. It is also novel in that it compares pesticides detected in wild bee and honey bee bodies to potential sources of pesticide exposure for bees, namely, flowers, soil, and air. Understanding the pesticide profile of honey bees and wild bees foraging from field border plants in agricultural areas compared to the potential sources of exposure can help to inform best management practices for pesticide use and farmland conservation practices.

2. Methods

2.1. Study field sites

Samples were collected at field sites on eight farms in an intensively managed agricultural landscape in Yolo and Colusa Counties, California. Field sites were selected such that they were no closer than 4.5 km apart; the maximum distance between field sites was 49.5 km. Each field site had a pollinator hedgerow, a forb strip, and weedy areas, all along the perimeter of an agricultural field. Hedgerows were generally mature (>10 years since establishment), 3–6 m in width and 350 m long, consisting largely of native perennial trees and shrubs (Morandin et al., 2016; Morandin and Kremen, 2013; Ponisio et al., 2016). Forb strips had been established (i.e. seeded) in the previous three to four years and consisted of primarily native annual herbaceous plants in rectangular plots (800 m^2) either directly adjacent to, or in one case, interdigitated with the hedgerows (Williams, 2016). Weedy areas included patches directly adjacent to hedgerows and forb strips, in addition to patches adjacent to crops, in irrigation ditches or in otherwise fallow land within the farm field. In general, insecticides and fungicides were not directly applied to hedgerows and forb plantings, though herbicides were sometimes used during establishment and follow-up maintenance (Long and Anderson, 2010). The herbicides glyphosate and clethodim were occasionally spot applied to weedy areas in the forb strips, to help the pollinator plants become established (Williams, 2016). While the agricultural region consists primarily of conventionally managed row crops, vineyards, and orchards, the crops are relatively diverse. Crops occupying 5% or more of the region within a radius of four kilometers of the field sites included: alfalfa, almonds, hay (non-alfalfa), oats, olives, rice, sunflower, tomatoes, walnut, and winter wheat (California Department of Pesticide Regulation, 2018).

2.2. Sampling design

Samples were collected from the eight field sites (described above) between mid-April and June 2016, with collection focusing on peak bloom times for two distinct sets of bee-visited flowering plants. Period one ranged from mid-April to mid-May and period two ranged from mid-May to June. In order to better understand the different routes of pesticide exposure for bees, different sample types were collected: wild bees, honey bees, flowers, soil, and silicone bands (as aerial passive samplers). Sampling methods are described below for each sample type. Not all samples could be collected from each of the eight field sites during the two sampling periods (Table S1).

2.2.1. Wild bees and honey bees

Both honey bees and wild bees visit border plants in agricultural fields; however, they have different behavioral and life history patterns that could influence exposure to pesticides (Kopit and Pitts-Singer, 2018; Sponsler et al., 2019). Thus, both honey bees and wild bees were collected from the eight field sites. Since bee size measured by intertegular span (IT-span) is correlated with foraging distance (Greenleaf et al., 2007), we divided the wild bees collected into three size categories corresponding to estimated foraging distances (Table S2). All bees were collected via sweep net, transferred individually to micro-centrifuge tubes, and placed on dry ice in the field, then stored at −20 °C in the laboratory prior to pesticide

analysis. Small brushes and gel (solidified mixture of gelatin (50 g), glycerin (150 ml), and distilled water (175 ml)) were used to remove the majority of pollen from the surface of each bee in an effort to analyze the pesticide load of the bee itself rather than pesticides in the pollen it carried. Though not all nectar ingested by bees is necessarily digested, we did not remove the contents of the crop as our objective was to analyze residues of the entire bee body.

Our target was a minimum sample weight of 1 g for pesticide analysis, and therefore sweep netting continued until the target was reached and consequently, the number of individuals in each wild bee sample varied (Table S2). It was not feasible to sample a fixed number of individuals; we sometimes needed >100 small bees to reach the minimum sample weight for chemical analysis and sampling that many large bees would have been impractical and potentially detrimental to local populations of large bodied species. Bees were typically collected when observed foraging on flowers. To obtain the minimum number of individuals required for pesticide analysis, bees were occasionally collected in flight in flower patches. For one small wild bee sample in the first sampling period, five of the bees included in the total composite sample of 28 bees were collected one month prior to the official sampling period; these were included to be able to achieve the minimum weight for the sample. At one field site, three bumble bees that had been observed to be entering and/or exiting an underground nest were collected. Since bees sometimes forage a great distance from their nests and thus may not have been foraging within the field site at all, these three bees were excluded from statistical analysis and are reported separately in the results.

While individual wild bees forage for both pollen and nectar, individual honey bees tend to have distinct castes of pollen or nectar foragers (Free, 1963). To avoid biasing our data, we attempted to collect an equal number of pollen and nectar foraging honey bees for each sample. During sweep netting, honey bees were collected and labeled as pollen foragers if they had large pollen loads in their corbicula. Honey bees were collected and labeled as nectar foragers if they had been observed foraging for nectar for at least 30 s. A total of 10 honey bees were collected per field site per time period for each composite sample. In all but one sample, there was a 50:50 ratio of pollen to nectar foragers; in one sample from period 1 there was a 60:40 ratio of pollen to nectar foragers due to limitations in field sampling.

2.2.2. Flowers

Pesticides applied to crops can be present in field border plants due to both aerial drift and uptake from the soil (Bonmatin et al., 2015; Botías et al., 2016; David et al., 2016; Krupke et al., 2012). Pesticide uptake and dissipation can vary by plant characteristic (e.g., growth, acidity, shape, leaf texture, and transpiration) and pesticides can be present in differing amounts in distinct parts of a plant (e.g., stem, leaves, petals, pollen, and nectar), leading to the expectation that pesticide detection and concentration could differ among herbaceous and woody plants (Bonmatin et al., 2015; Fantke and Juraske, 2013). Bees consume nectar from flowers and collect pollen to feed to their progeny. To estimate pesticide exposure for bees visiting field border flowering plants, flowers from four types of plants were collected from each field site during each sampling period, including two planted herbaceous, one weedy, and one woody plant (see Table 1 for species). Flower petals and sepals were removed to the extent possible with scissors and/or cork borers to isolate the pollen and nectar producing structures. Flowers from each plant type were placed into glass containers and transferred to coolers in the field before storage at −20 °C for pesticide analysis. To achieve a minimum sample weight of 0.3 g for pesticide analysis, the number of individual flowers per sample was inversely related to flower size. When possible, flowers were collected from different individual plants at each field site.

2.2.3. Soil

Pesticides applied to crops can leach through soils and be taken up by field border plants (Bonmatin et al., 2015; Botías et al., 2016, 2015; David et al., 2016). Also, many wild bees nest in soil (Kopit and Pitts-Singer, 2018). To understand exposure of field border plants and nesting bees, a composite soil sample (5 soil cores, 0–7 cm) was collected using a 2.5 cm diameter soil probe across each field site from locations where other bee and flower samples were collected. The five soil samples were collected once during each sampling period, combined, and stored in zip-top bags. Bags were placed in coolers in the field, then stored at −20 °C for pesticide analysis.

2.2.4. Silicone bands

As bees can encounter pesticides via aerial spray drift and dust (Krupke et al., 2012), silicone bands (100% silicone wristbands from 24HourWristbands.com, 1.27 cm with 6.7 cm inner diameter) were deployed as passive samplers to collect aerial deposits of pesticides (Swanson et al., 2018). One band was used per field site per sampling period (described above); the sampling period was around 1 month (Hladik and Ward, 2022). At the beginning of the sampling period, pre-cleaned bands (O'Connell et al., 2014) were removed from foil storage on site, cut into pieces, and each piece was stapled vertically to separate aboveground stakes at a height between 1.25 and 1.75 m. The stakes were spread across each field site, such that their locations corresponded with locations within fields where flower, bee, and soil samples were collected. At the end of each sampling period, bands were recovered, stored in foil, placed in a cooler, and then stored at between −20 °C for pesticide analysis. Bands from each field site were stored separately.

2.3. Analytical methods for pesticide detection

Bees, flowers, and soil were extracted via pressurized liquid extraction and solid phase extraction cleanup (Hladik et al., 2016; Hladik and McWayne, 2012; Main et al., 2020) and silicone bands were extracted via sonication with ethyl acetate (Swanson et al., 2018). Samples were analyzed for 168 pesticides and degradates using both gas and liquid chromatography-tandem mass spectrometry. The complete list of compounds, analytical limits of detection (LOD), and surrogate compound recoveries can be found elsewhere (Hladik and Ward, 2022). While a wide range of pesticides were included in the analysis, some exceptions include the herbicides glyphosate, 2,4-D, and triclopyr. Although these herbicides are widely used and can remove floral resources for bees, herbicides in general are considered less toxic to pollinators than insecticides or fungicides (Goulson et al., 2015).

2.4. Statistical analysis

We constructed separate generalized linear mixed models to examine how the number of pesticides detected (Poisson error) varied across samples, pesticide type (fungicide, herbicide, or insecticide), and their interaction. Three distinct models examined samples in terms of: (1) sample type (i.e., wild bees, honey bees, flowers, soil, and silicone bands), (2) wild bee size (i.e., small, medium, large), and (3) flower type (i.e., herb 1, herb

Table 1
Flower species sampled during each sampling period (P1 = period one and P2 = period two).

	Herb 1	Herb 2	Weedy	Woody
P1	Scorpion-weed (*Phacelia* spp. mostly *tanacetifolia*)	California poppy (*Eschscholzia californica*)	Mustard (Brassicaceae: *Brassica* spp., *Hirschfeldia* spp., *Raphanus* spp.)	California coffee berry (*Frangula californica)*
P2	Gumweed (*Grindelia* spp. mostly *camporum*)	California poppy (*Eschscholzia californica*)	Yellow star-thistle (*Centaurea solstitialis*)	Toyon (*Heteromeles arbutifolia)*

2, woody, and weedy; see Table 1). For all analyses of the number of pesticides, degradates and parent compounds were combined and counted as a single pesticide when their association with each other was indisputable. Specifically, the degradates DDE and DDD and parent compound DDT were combined and counted only once, and the degradate DCPMU and parent compound diuron were combined as one. We removed the degradate 3,4-DCA from these analyses because it is a degradate of both propanil and diuron and so it could not be assigned to a single parent compound. Models for sample type and flower type included the interaction of field site and time period as random effects; models for wild bee size included field site and time as separate random effects due the lack of balance in the dataset.

Next, we composed separate generalized linear mixed models to determine how the concentrations of the pesticides detected (ng/g; Gaussian error) differed among samples, pesticide type (fungicide, herbicide, or insecticide), and their interaction. Four distinct models examined samples in terms of: (1) sample type (i.e., wild bees, honey bees, flowers, and soil), (2 and 3) wild bee size (i.e., small, medium, large), and (4) flower type (i.e., herb 1, herb 2, woody, and weedy; see Table 1). In the first model, silicone bands were removed from the sample type model because pesticide concentration in silicone bands is more dependent on surface area (ng/band) not mass (ng/g). The second and third models (i.e., the two wild bee size models) were distinct in that we examined how pesticide concentration differed among wild bee sizes using pesticide residues per bee (ng/bee) and pesticide concentration per mass of bee (ng/g). Pesticide residues per bee is commonly used as a measure of pesticide toxicity for honey bees and is included here for comparison. For all pesticide concentration analyses, pesticide degradates and parent compounds were treated as distinct pesticides (i.e., they were not combined, as in the previous analysis), only detected compounds were analyzed, and concentration was transformed with natural logs to linearize the data. All pesticide concentration models included the interaction of field site and time period as random effects.

Since bifenthrin was the most commonly detected pesticide in our samples, post facto, as an indicator of potential routes of exposure for bifenthrin, linear mixed models with Gaussian errors were used to test whether the concentration of bifenthrin in wild bees was associated with sample type and either wild bee size or flower type. The same analysis was also conducted for honey bees. In these comparative analyses of pesticide concentrations, sample type was included as a fixed effect in the model and field site was included as a random effect. Concentrations were scaled so that silicone bands could be included as a sample type despite the different unit of measurement for concentration.

All generalized linear mixed models were run using the lme4 package in R version 3.6.1 (Bates et al., 2015; R Core Team, 2019). Type II Wald chi square tests were used to determine the significance of fixed effects in the models using the Anova function from the car package in R (Fox and Weisberg, 2019). Post hoc comparisons of marginal means among sample types within significant fixed effects were made using the emmeans function from the emmeans package in R (Lenth, 2020).

3. Results

3.1. Overview of pesticide detection in samples

Of the 148 samples analyzed for the potential occurrence of 168 pesticide residues, a total of 37 pesticides and degradates were detected, of which there were 8 insecticides, 13 herbicides, 12 fungicides, and 4 degradates (Hladik and Ward, 2022). There was a total of 17 pesticides and degradates detected in wild bees overall (16 in small bees, 17 in medium bees, and 12 in large bees), 10 in honey bees, 17 in flowers overall (12 in planted herb 1, 11 in planted herb 2, 12 in weedy, and 12 in woody), 25 in soil, and 24 in silicone bands. Percent detection of insecticides, herbicides, and fungicides within sample types are depicted in Fig. 1.

The three most frequently detected pesticides in all samples were the insecticide bifenthrin (44%) and the herbicides thiobencarb (42%) and metolachlor (37%). Pesticides that were detected in over 20% of all samples also included the herbicides pendimethanlin and propanil, and the fungicides boscalid, carbendazim and fluopyram. For wild bees, the insecticide bifenthrin and the herbicides hexazinone, metolachlor, propanil, and thiobencarb were detected in over 25% of samples. For honey bees the insecticide bifenthrin, the herbicide thiobencarb, and the fungicides carbendazim and fluopyram were detected in over 25% of samples.

Among the insecticides examined, neonicotinoids are known to be particularly harmful to bees though toxicity varies by active ingredient. Neonicotinoids in general were infrequently detected at our field sites; imidacloprid was the sole neonicotinoid detected (Fig. 1). Imidacloprid was detected in 20% of flower samples at concentrations of 8.0 to 28.7 ng/g. It was also detected in 13% of soil samples at a concentration of 0.3 to 0.9 ng/g (at only one field site during both sampling periods). All imidacloprid detections in flowers were in planted herbaceous or weedy plants sampled during period one (*Phacelia* spp., *Eschscholzia californica*, and *Brassicaceae*); imidacloprid was not detected in woody plant flowers in either sampling period. At the field site where imidacloprid was detected in the soil, it was also detected in weedy flowers, not in the planted herbaceous flowers.

The insecticide bifenthrin was detected at all field sites and was the most frequently detected pesticide in our samples in general. Bifenthrin was detected in 29% of wild bee samples, 31% of honey bee samples, 40% of flower samples, 63% of soil samples, and 100% of the silicone band samples. Of the flower samples, bifenthrin was notably detected in all of *Centaurea solstitialis* samples (weedy, period 2) and all but one sample of *Heteromeles arbutifolia* (woody, period 2). Bifenthrin is a pyrethroid insecticide and the contact lethal dose required to kill 50% of honey bees (LD_{50Hb}) is 14.6 ng/bee (EPA, 1985; Sanchez-Bayo and Goka, 2014); the oral LD_{50Hb} is 100 ng/bee (University of Hertfordshire, 2022). None of our bee samples contained bifenthrin in concentrations near the oral or contact LD_{50Hb}; however, 60% of honey bee samples and 55% of wild bee samples with bifenthrin detections exhibited a toxic unit (TU, concentration detected/ contact LD_{50}, von der Ohe and de Zwart, 2013) $\geq$ 0.07. The greatest TU for a honey bee was 0.166 and the greatest TU for a wild bee was 0.20 (large wild bee). The TU based on the oral LD_{50} would be <0.06 for all bees. The sample of three bumble bees collected above the underground nest at one field site had a TU of 0.377 for bifenthrin. TUs for all other pesticides in all other bee samples were $\leq$ 0.0004 and would not be expected to appreciably contribute to any toxicity.

Of the fungicides known to be harmful to bees either independently or synergistically with insecticides, three were detected in our samples: chlorothalonil, myclobutanil, and propiconazole (Fig. 1). While all three of these fungicides were detected at levels (<9.6 ng/bee) far below the honey bee LD_{50} levels (33,900 ng/bee or greater; (University of Hertfordshire, 2022)), sublethal effects are not well studied. Chlorothalonil was detected at all field sites and in 88% of silicone band samples, 19% of soil samples, 10% of wild bee samples, and 10% of flower samples (only *Phacelia* spp., and *Frangula californica*). Myclobutanil was detected at 63% of field sites but only in bee samples, including 19% of honey bee and 10% of wild bee samples. In two cases (one honey bee and one large bee sample), myclobutanil was present in the same sample as bifenthrin. Propiconazole was detected at all field sites and in all silicone band samples, 15% of wild bee samples, 2% of flower sample, and 25% of soil samples.

Results from all models are described below and summarized in Supplemental Table 3.

3.2. Number of pesticides

3.2.1. Sample type and pesticide type

A total of 33 pesticides were detected in our samples after combining parent pesticides and their degradates. The mean number of pesticides detected differed by pesticide type and sample type ($\chi^2_{\text{pesticide type}}$ = 66.86, df = 2, $P < 0.001$; $\chi^2_{\text{sample type}}$ = 422.15, df = 4, $P < 0.001$) with no significant interaction between these factors ($\chi^2_{\text{interaction}}$ = 13.97, df = 8, P = 0.083). Typically, less than two pesticides from each category

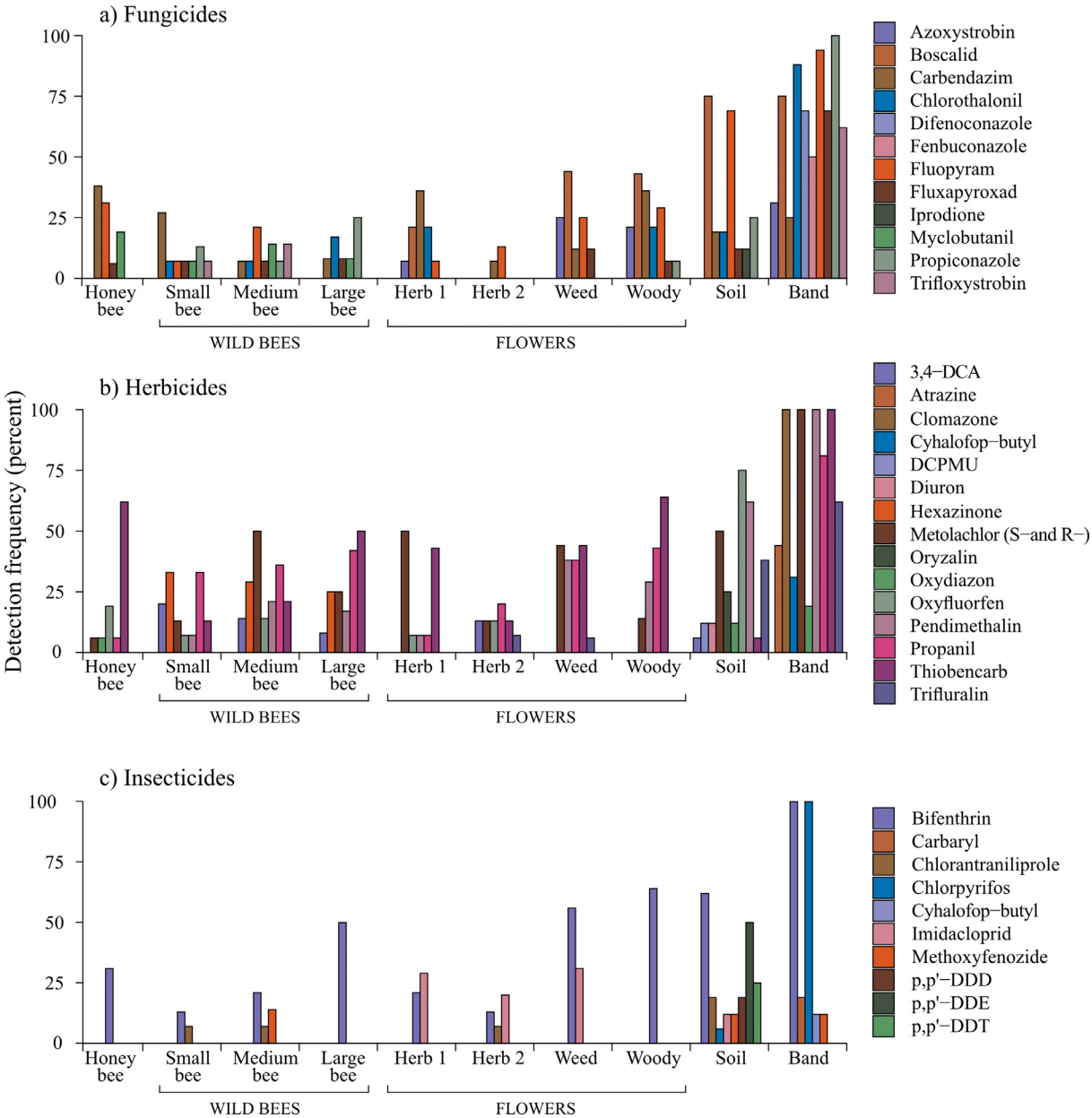


Fig. 1. Percent detection of (a) fungicides, (b) herbicides, and (c) insecticides and their degradates for all sample types.

were detected in each sample type; the one exception was the silicone bands, which detected from 6 to 8 herbicides and fungicides (Fig. 2A). A greater number of pesticides were detected in soil samples than in flower or bee samples, but the greatest number was detected in silicone band samples. The number of pesticides detected generally increased from insecticides to fungicides to herbicides (Fig. 2A).

3.2.2. Wild bee size

Among wild bees, the mean number of pesticides detected differed by pesticide type ($\chi^2_{\text{pesticide type}} = 25.54$, df = 2, $P < 0.001$), following a similar trend as for the overall dataset. The number of pesticides detected did not differ by wild bee size ($\chi^2_{\text{wild bee}} = 2.27$, df = 2, $P = 0.322$; Fig. 2B) and there was no interaction with pesticide type ($\chi^2_{\text{interaction}} = 1.77$, df = 4, $P = 0.778$).

3.2.3. Flower type

Among the different flowers examined, the mean number of pesticides detected differed by pesticide type and flower type ($\chi^2_{\text{pesticide type}} = 12.15$, df = 2, $P = 0.002$; $\chi^2_{\text{flower type}} = 17.78$, df = 3, $P = 0.001$, Fig. 2C), with no interaction ($\chi^2_{\text{interaction}} = 5.15$, df = 6, $P = 0.525$). Planted herbaceous plants in general, and specifically Herb 2 (*Eschscholzia californica*; see Table 1) had fewer distinct pesticides detected than other flower types.

3.3. Pesticide concentration

3.3.1. Sample type and pesticide type

Analyses of pesticide concentrations revealed a significant interaction between pesticide and sample type ($\chi^2_{\text{interaction}} = 21.91$, df = 6, $P = 0.001$), and thus each pesticide type was examined separately. The mean concentration of pesticides differed among sample types for all three pesticide types examined ($\chi^2_{\text{insecticides}} = 27.54$, $\chi^2_{\text{herbicides}} = 162.73$, $\chi^2_{\text{fungicides}} = 90.68$ with df = 3 and $P < 0.001$ for all). For all pesticide types, pesticide concentrations tended to be highest in flowers and lowest in soils (Fig. 3A). While pesticide concentrations detected in wild bees and honey bees did not differ, concentrations detected in wild bees were typically lower than in

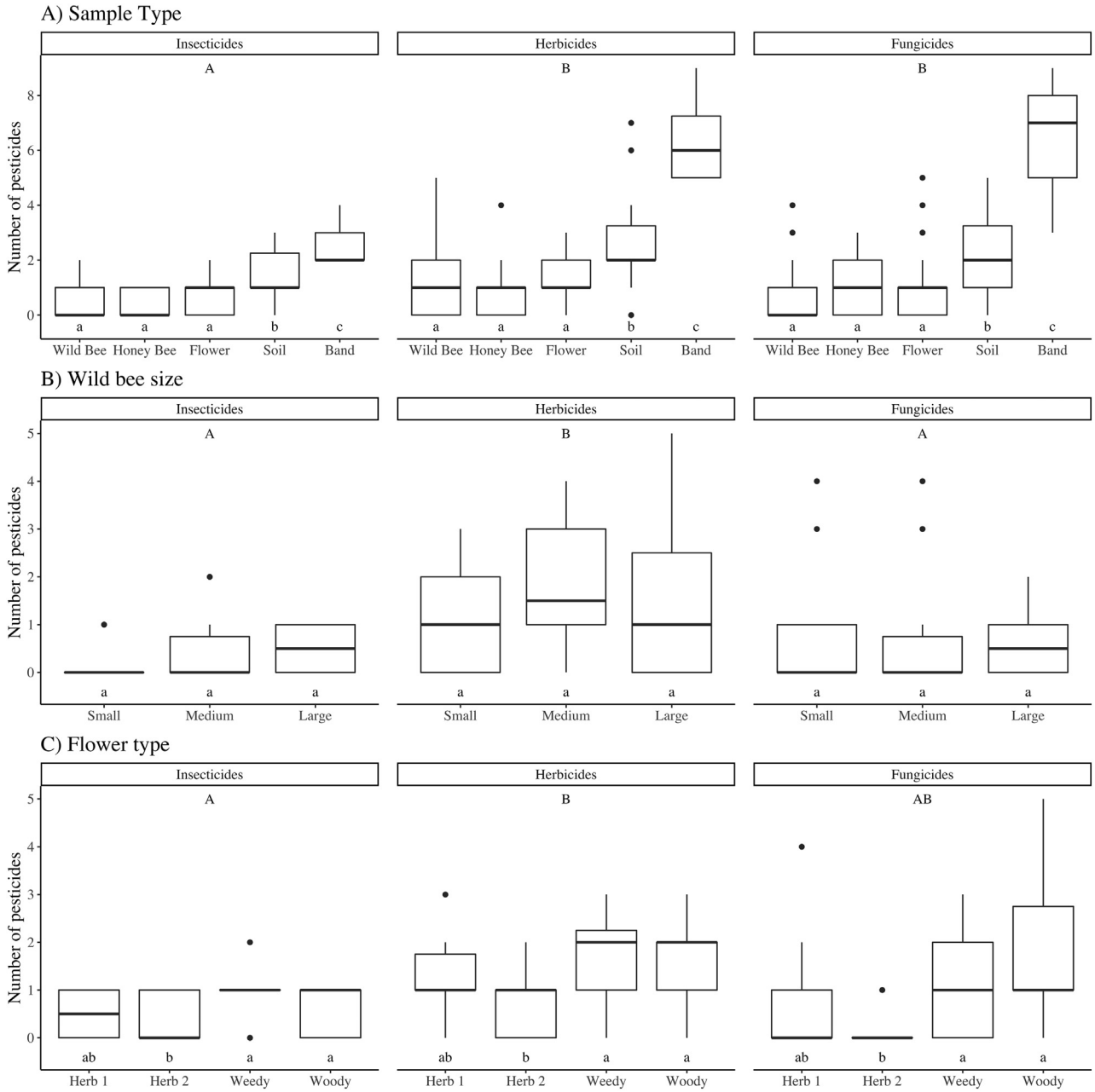


Fig. 2. Number of pesticides detected in relation to pesticide type for (A) sample type, (B) wild bee size, and (C) flower type. Bold lines depict medians, boxes depict 25th and 75th percentiles, whiskers depict 95th percentiles and dots depict outliers. Differences in mean number of pesticides between pesticide categories are indicated by capital letters ($P < 0.01$). Differences in mean number of pesticides within sample types, wild bee sizes, and flower types are indicated by lower case letters ($P < 0.01$). Note that the mean number of herbicides and fungicides, were also marginally different for herb 1 and herb 2 ($P = 0.07$).

flowers for all pesticide types (Fig. 3A); in contrast, insecticide and fungicide concentrations detected in honey bees were similar to those detected in flowers.

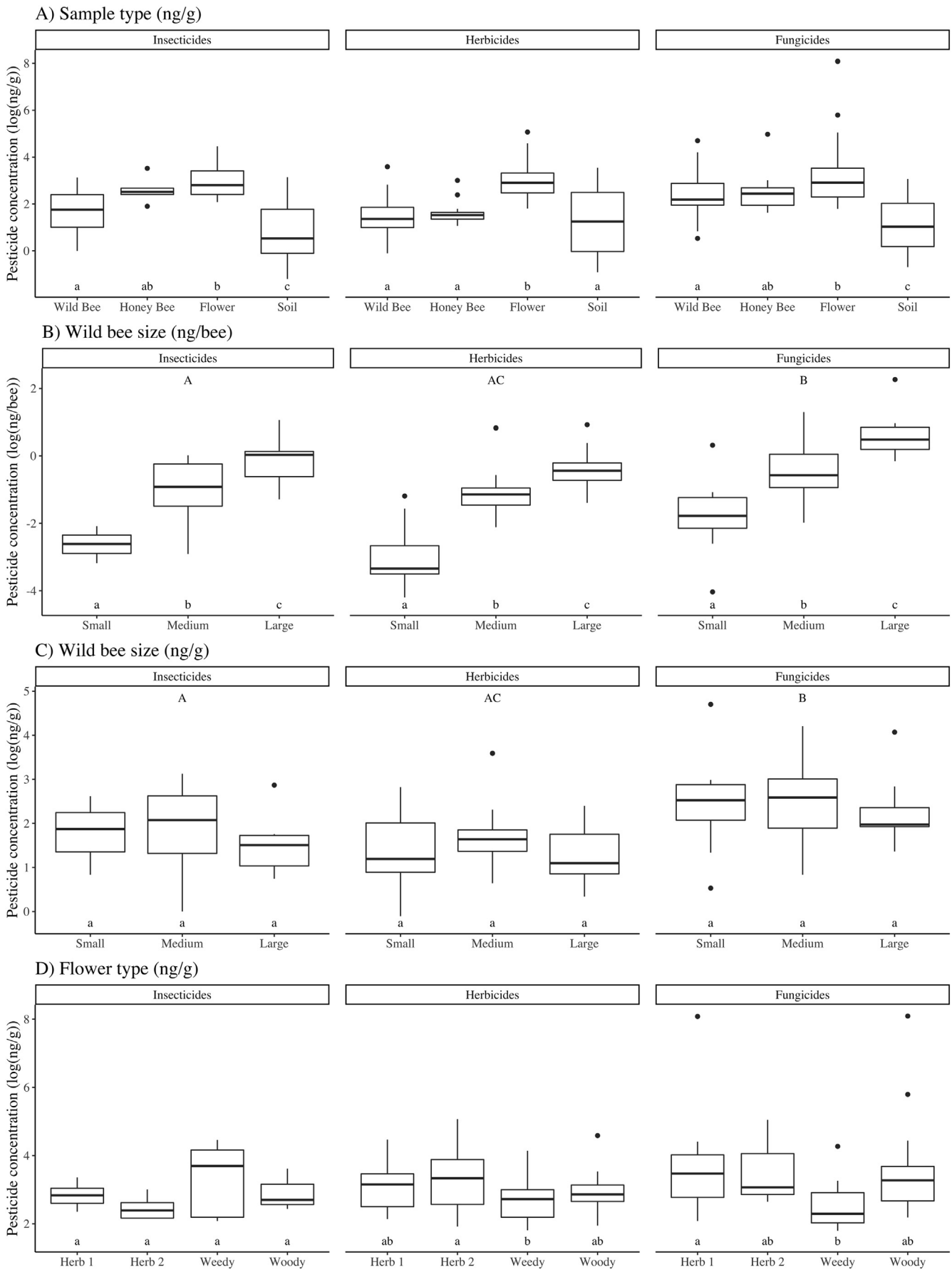
3.3.2. Wild bee size

For data from wild bees, mean pesticide residue per bee (ng/bee) increased with increasing bee size ($\chi^2_{\text{wild bee size}} = 180.35$, df = 2, $P < 0.001$; Fig. 3B) and differed by pesticide type ($\chi^2_{\text{pesticide type}} = 29.27$, df = 2, $P < 0.001$), with no significant interaction between these factors ($\chi^2_{\text{interaction}} = 1.84$, df = 4, $P = 0.765$). Although mean pesticide concentration (ng/g) did not differ with wild bee size ($\chi^2_{\text{wild bee}} = 1.33$, df = 2, $P = 0.514$; Fig. 3C), it did differ by pesticide type ($\chi^2_{\text{pesticide type}} = 21.60$, df = 2, $P < 0.001$); again, there was no significant interaction between these factors ($\chi^2_{\text{interaction}} = 1.15$, df = 4, $P = 0.887$).

3.3.3. Flower type

For data from flowers, there was a significant interaction between pesticide type and flower type ($\chi^2_{\text{interaction}} = 19.40$, df = 6, $P = 0.004$), and

Fig. 3. Pesticide concentrations detected in relation to pesticide type and (A) sample type, (B) wild bee size (ng/bee), (C) wild bee size (ng/g) and (D) flower type. Bold lines depict medians, boxes depict 25th and 75th percentiles, whiskers depict 95th percentiles and dots depict outliers. Differences in mean concentration between pesticide categories are indicated by capital letters, for cases when no interaction was present ($P < 0.05$). Differences in mean concentration within sample types, wild bee sizes, and flower types for each pesticide type are indicated by lower case letters ($P < 0.05$).


A) Sample type (ng/g)
Insecticides
Herbicides
Fungicides
Pesticide concentration (log(ng/g))
Wild Bee
Honey Bee
Flower
Soil
B) Wild bee size (ng/bee)
Pesticide concentration (log(ng/bee))
A
AC
B
Small
Medium
Large
C) Wild bee size (ng/g)
D) Flower type (ng/g)
Herb 1
Herb 2
Weedy
Woody

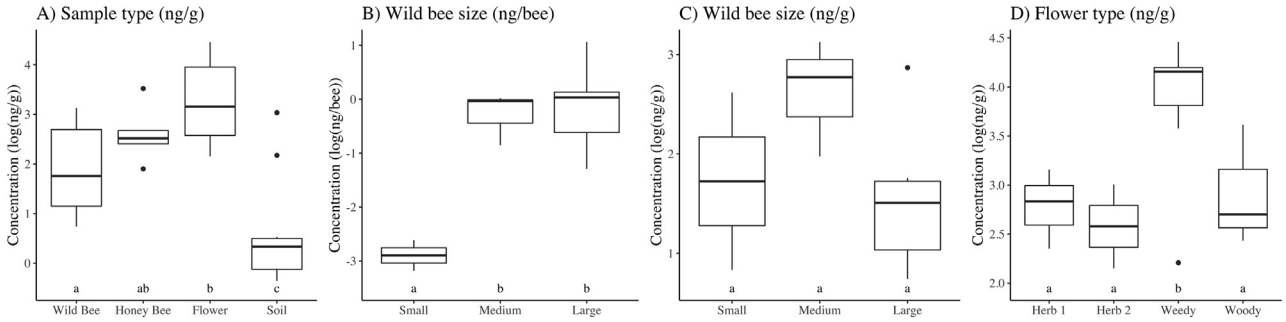


Fig. 4. Concentration of bifenthrin detected in relation to (A) sample type (ng/g), (B) wild bee size (ng/bee), (C) wild bee size (ng/g) and (D) flower type (ng/g). Bold lines depict medians, boxes depict 25th and 75th percentiles, whiskers depict 95th percentiles and dots depict outliers. Differences in mean concentration are indicated by lower case letters ($P < 0.05$).

thus we analyzed each pesticide type separately. The mean concentration of pesticides detected differed among flower types for all three pesticide types ($\chi^2_{insecticides} = 8.94$, $\chi^2_{herbicides} = 9.85$, $\chi^2_{fungicides} = 10.04$ with df = 3 and $P < 0.05$ for all; Fig. 3D). Insecticide concentrations varied the least across flower types, but the concentration of both herbicides and fungicides in weedy plants tended to be lower than in planted herbaceous plants (Fig. 3D).

3.3.4. Bifenthrin

In the post facto bifenthrin analysis, mean bifenthrin concentration differed significantly among sample types ($\chi^2 = 84.85$, df = 3, $P < 0.001$). Bifenthrin concentration generally increased from soil, to bees, to flowers (Fig. 4A). For wild bees with residues expressed as ng/bee, bifenthrin residues increased with wild bee size ($\chi^2 = 23.49$, df = 2, $P < 0.001$; Fig. 4B). In contrast, when analyzed for bifenthrin concentration expressed as ng/g, there was no difference between bee sizes ($\chi^2 = 3.64$, df = 2, $P = 0.162$; Fig. 4C). Bifenthrin concentration differed by flower type ($\chi^2 = 25.51$, df = 3, $P < 0.001$) and was higher in weedy plants than other flower types (Fig. 4D). Comparative analyses showed a significant association between bifenthrin concentrations in honey bees and those in both woody plants ($\chi^2 = 10.35$, df = 1, $P = 0.001$) and planted herb 1 ($\chi^2 = 4.87$, df = 1, $P = 0.027$), while bifenthrin concentrations in wild bees were significantly associated with those in planted herb 2 ($\chi^2 = 5.59$, df = 1, $P < 0.018$) and marginally associated with those in weedy plants ($\chi^2 = 3.82$, df = 1, $P < 0.051$).

4. Discussion

Strong evidence shows that field border plantings (e.g., hedgerows and forb strips) are effective techniques for restoring and maintaining pollinator communities in intensive agricultural regions (Albrecht et al., 2020; Cohen et al., 2021; Kremen et al., 2019; but see Nicholson et al., 2020). However, these plants may also harbor pesticides that are detrimental to bees (Botías et al., 2015; David et al., 2016). We found that the number of pesticides detected was greater in aerial and soil samples than in flowers or bees. Pesticide concentrations generally increased from soils to bees to flowers, but varied considerably among flower types, and pesticide residues were higher in larger wild bees than smaller wild bees when expressed as ng/bee. While many studies have focused on neonicotinoid insecticides alone, we detected many other pesticides in each component of our study system, including those that are known to be harmful to bees on their own and synergistically with other pesticides (Iverson et al., 2019). Given that studies across multiple years are more robust and our measurements were collected over only one year, this study can serve as a useful framework for future research. Collectively our findings suggest that (1) bee-attractive, flowering field border plants contain a number of pesticides that are applied to crops in the greater landscape, (2) different bee types (honey bee versus wild bee) and wild bee sizes exhibited different patterns of exposure, and (3) in addition to neonicotinoids, bees are exposed to a mixture of other pesticides under field conditions that can have negative sublethal effects on pollinators.

4.1. Pesticide exposure and field border plants

The flowers that we sampled from the field border of agricultural crops harbored several pesticides. This follows other studies on field border plants that have also found that these plants contain pesticides even though these products have not been applied directly to field border plants (Botías et al., 2015; David et al., 2016; Long and Krupke, 2016). Pesticides can be deposited on plants indirectly via aerial drift or dust (Krupke et al., 2012), and, in fact, passive aerial samplers had the highest number of pesticides observed in our study. Soils had the second highest number of pesticides; these can include persistent compounds that were applied in prior years (e.g., DDT). Water soluble pesticides such as neonicotinoids that have systemic properties can also be taken up by plants from the soil (Bonmatin et al., 2015). Thus, pesticides may be present on the surface of plant structures (as residue deposits) via aerial drift and/or within plant tissues and products (such as pollen, nectar and guttation fluid) through uptake from soils even in the absence of direct applications (Botías et al., 2016, 2015). Consequently, the pesticides detected in our flower samples likely came from various sources, including drift (aerial) and/or uptake (soil).

Some of the pesticides detected in our study were likely applied to crops grown at some distance from the field sites. For example, the herbicides clomazone, propanil, and thiobencarb were observed in our field sites but are applied exclusively to rice in California (California Department of Pesticide Regulation, 2018; Hladik et al., 2020). While 75% of our field sites were directly adjacent to rice fields (California Department of Pesticide Regulation, 2018), this finding suggests that the exposure of bees to pesticides needs to be considered at the landscape scale, not just the field scale.

Our study shows that woody and weedy plants have a greater number of pesticides than planted herbaceous plants. This finding may reflect the tendency for woody plants to be present in the landscape for longer than herbaceous plants, which may allow the former to accumulate more pesticides. It is unclear why weedy plants at our field sites had a greater number of pesticides than the intentionally planted herbaceous plants, but other studies have also found higher levels of pesticides in non-cultivated plants (Long and Krupke, 2016). Differences in pesticide concentration between flower types did not show a consistent pattern across pesticide types. Taken together, this difference suggests a multifaceted interplay of pesticide chemistry and plant anatomy and physiology and indicates that studies of acquisition of specific active ingredients across different plant types are needed to better understand flowers as a route of pesticide exposure for bees.

The frequency of detection and concentration of bifenthrin, specifically, was strikingly different among flower types. Bifenthrin was detected in all eight weedy plant samples (*Centaurea solstitialis*) and in seven of eight woody samples (*Heteromeles arbutifolia*), during sampling period two, yet in only one to two of the other flower samples. Bifenthrin concentrations

were also significantly higher in the flowers of weedy plants than all other plants, driven largely by its presence in yellow star-thistle (*Centaurea solstitialis*). Given that bifenthrin is a non-systemic pesticide, and is not translocated through plants, it is possible that the difference in the frequency of detection across flower types indicates that the residues from aerial drift are unevenly distributed or degraded, potentially due to differing plant heights/architecture and/or their location in the landscape. Additionally, the association of bifenthrin concentrations detected in honey bees and wild bees varied by flower type, with honey bee concentrations associated with nearly the opposite flower types as wild bee concentrations. These findings highlight the importance of considering plant-pollinator relationships in regard to pesticide exposure for different bee and plant species within a landscape.

4.2. Effects of bee biology and ecology

The factors contributing to pesticide exposure for bees in agricultural landscapes are complex. Both soil and aerial samples had a greater number of pesticide types than bees and flowers (Fig. 2A); bees had a similar number of pesticides as flowers (Fig. 2A), with concentration in flowers generally higher than in bees (Fig. 3A). These results suggest that bees may be exposed to more pesticides than can be detected in their bodies as residues (e.g., due to metabolism); however, even when pesticides are not detected in bee bodies, there may be effects from these pesticides (Ward, 2020). The difference in pesticide concentrations between bees, flowers, and soil (Fig. 3A) may suggest that adult bees are exposed to greater concentrations of pesticides while foraging than nesting. Further, the results are consistent with two non-exclusive explanations - either they are not exposed to the full pesticide load presented in flowers and/or detoxification in bees leads to lower concentrations than in flowers. In addition to being exposed to pesticides while foraging, bees can encounter pesticides during most activities that they engage in: during flight (via aerial sprays or the dust from planting of treated seeds), while walking or resting on the surface of plants and soils, during nesting (in soil, leaves, resins, wax and other materials), and while feeding on pollen that may contain pesticides while developing as larvae (Boyle and Sheppard, 2017; Gradish et al., 2019; Sgolastra et al., 2019). As our samples consisted of composites of bee species with various nesting strategies, future studies might consider comparing pesticide profiles in flowers and soil to that of cavity nesting versus soil nesting bee species to better understand the relationship between exposure via foraging versus nesting. Since we did not analyze the concentration of pesticides in the silicone bands per unit mass, we cannot directly compare concentrations in bees to those in air. However, we suspect that exposure from flying through dust or drift patches would be an infrequent occurrence. While our results suggest that flowers are an important source of pesticide exposure, our findings also underscore the difficulty of generalizing sources of exposure for bees as a whole.

We expected to detect a greater number of pesticides in honey bees compared to wild bees given that bee keepers transport honey bee hives over large distances from different landscapes and geographic regions to pollinate crops in California. Instead, we found no difference in the mean number of pesticides detected in wild versus honey bees and that the total number of pesticides detected exclusively in wild bees was greater than that for honey bees. While this outcome may reflect the greater number of individuals collected per sample for wild bees compared to honey bees, the same pattern was evident in samples for large-bodied wild bees, which had on average fewer individuals per composite sample than honey bees. It is likely that differences in life history traits between wild bees and honey bees affect routes of exposure and thus the number and composition of pesticides encountered by different bee species.

A number of traits are relevant to pesticide exposure for bees including: sociality, fecundity, nest type (e.g., substrate, materials and period), flower preferences, adult and larval food, body size, and other anatomical and physiological characteristics (e.g., how pollen is transported and metabolism) (Sgolastra et al., 2019). In this study, we capitalized on the fact that bee size is correlated with foraging distance to analyze pesticide exposure for bees with differing foraging ranges (Greenleaf et al., 2007). Despite being intensively managed, California has a greater diversity of crops compared to other regions like the Midwestern United States where similar studies have been conducted (e.g., Main et al., 2020). We expected that larger bees that forage over greater distances would be exposed to a greater number of pesticides. In our study, the number of pesticides detected in wild bees did not differ by bee size, which contrasts with results from other studies. For example, individual bees collected from non-crop patches in a region dominated by cotton revealed a general trend toward increased neonicotinoid detections in bee genera with greater average body mass (Longing et al., 2020). The effects of body size and foraging distance on pesticide exposure may be more pronounced in areas with reduced crop diversity, although we caution against drawing strong conclusions given that the number of bees in our composite samples was negatively correlated to bee size. We suggest that in areas of greater crop diversity, the number of pesticides detected may also increase with bee size if composite samples had been standardized by number rather than mass of bees.

Larger bees in our study did appear to experience greater exposure to pesticides than smaller bees. This trend was apparent when concentration was expressed as pesticide residue per bee (ng/bee), but not when it was expressed as pesticide concentration per unit body mass (ng/g). Theories on how body size may affect exposure levels are mixed. Since body size and surface area are inversely related, smaller bees may experience greater contact exposure per unit body mass than larger bees. Furthermore, the inverse relationship between body size and mass-specific metabolic rates (Heinrich, 1993) suggests that smaller bees may experience greater oral exposure because they ingest more nectar and/or pollen per unit body mass than large bees (Sgolastra et al., 2019). Yet, for example, Botías et al. (2017) found that bumble bees with lower mass had lower amounts of pesticide residue and suggested this may result from smaller bees consuming less than larger bees. The inverse relationship between body size and mass-specific metabolic rates may also mean that smaller bees detoxify pesticides more quickly, so while their exposure may be higher, it is possible that detection frequency and possibly sensitivity could be lower in smaller bees. Taken together bee biology and ecology likely lead to differences in pesticide exposure, detection ability, and pesticide sensitivity that are relevant not just to lethal effects, but also to sublethal effects. These differences highlight the drawback of categorizing bees as a single and uniform entity, best represented by honey bees as a model organism, when it comes to risk assessment from pesticide exposure.

4.3. Beyond neonicotinoids

Neonicotinoids were not frequently detected in our samples, though they are applied in the region (e.g., aerial spray on walnuts, seed treatments for sunflower: California Department of Pesticide Regulation, 2018; Long et al., 2019). This outcome may reflect a lower usage of this class of insecticides in our study region as compared to studies of bees in areas that are dominated by neonicotinoid-treated crops (e.g., oil-seed rape, corn, soybeans: David et al., 2016; Botías et al., 2015; Main et al., 2020). While neonicotinoid research has been highly beneficial in revealing the necessity of updating pesticide risk assessments, especially with regard to (1) sublethal effects and (2) consideration of a greater range of bee species (Sgolastra et al., 2020), more research is needed to understand bee exposure and sensitivity to a greater range of pesticides that are currently used in the field.

Bifenthrin was the most frequently detected insecticide in our samples and was detected alongside the fungicide, myclobutanil. Bifenthrin can be harmful to bees on its own (Sanchez-Bayo and Goka, 2014) and is known to react synergistically with the triazole SBI fungicides difenoconazole and myclobutanil, increasing its toxicity to bumble bees (Iverson et al., 2019). No bee samples collected had concentrations near the LD_{50} for bifenthrin, as would be expected because only live bees were collected; however, at least one wild bee composite had a TU = 0.20. While TU < 0.20 is unlikely to contribute to acute toxicity, it may contribute to chronic toxicity, sublethal effects or synergistic effects with other compounds that increase its toxicity (von der Ohe and de Zwart, 2013). Myclobutanil was

present in two samples that also contained bifenthrin, suggesting that synergistic effects may have occurred. In addition, difenoconazole was detected in silicone band samples, providing additional evidence that interactions among pesticides may have occurred at our field sites.

Fungicides on their own can also be directly harmful to bees. For example, myclobutanil, which was observed in our bees, has been found to inhibit detoxification pathways in honey bees (Mao et al., 2017). Chlorothalonil was detected in many sample types including wild bees but not honey bees. Chlorothalonil was not detected at a level of lethal concern in our samples (<90 ng/g in flowers and <17 ng/g in bees); however, in general chlorothalonil has been associated with bumble bee (*Bombus impatiens*) colonies that produce fewer workers and smaller queens (Bernauer et al., 2015) and found to be strongly associated with pathogen presence (*Nosema bombi*) in declining bumble bee species at field realistic levels (McArt et al., 2017). Further, insecticides that are not generally considered toxic to bees have been shown to increase in toxicity in the presence of fungicides. For example, we detected chlorantraniliprole and propiconazole together in one small wild bee sample and also in one medium wild bee sample. While the relatively new diamide insecticide chlorantraniliprole has low toxicity to bees, a recent study found increased honey bee larval mortality and adult toxicity when chlorantraniliprole exposure was combined with the fungicide propiconazole (Wade et al., 2019). Collectively, these findings underscore the importance of considering multiple pesticides and pesticide mixtures when assessing effects on wild bees. Several previous studies have reported a range of pesticides in bees or bee related samples, including several of the same insecticides, herbicides, and fungicides detected in our study (Botías et al., 2017; David et al., 2016; Hladik et al., 2016; Krupke et al., 2012; Main et al., 2020; Mullin et al., 2010). To date, however, pesticide risk assessments in the United States typically consider only one compound at a time, indicating that additional work is needed to assess potential interactions and synergistic effects at the field scale (Sgolastra et al., 2020).

5. Conclusion

Much remains to be learned about routes of pesticide exposure, frequency of exposure, and toxicity in relation to conservation of wild bees and honey bees foraging in agricultural landscapes. Our study demonstrates that bee-attractive field border plants in agricultural areas harbor many pesticides, despite the fact that pesticides are not applied directly to these plants. Additionally, as the number and concentration of pesticides differ among plant types, bee exposure through foraging must inevitably be related to plant-pollinator dynamics. The pesticides detected in floral, aerial, and soil samples suggest that bees are exposed to many more pesticides than those detected in their bodies. Furthermore, some pesticides (e.g., bifenthrin) were detected in bee bodies at potential levels of concern, particularly for sublethal effects, as well as in combination with other pesticides that can synergistically increase its toxicity. Thus, studies that analyze only a subset of pesticides (e.g., neonicotinoids alone) and a subset of bee species (emphasis on honey bees) may overlook evidence for larger ecological consequences. While this study generates important insights for pesticide exposure of bees in an intensively managed agricultural landscape, more research is needed to understand the sensitivity of a range of bee species from exposure to field realistic pesticide concentrations and pesticide mixtures at various scales.

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CRediT authorship contribution statement

Laura T. Ward: Conceptualization, Methodology, Software, Validation, Formal analysis, Investigation, Resources, Writing - original draft, Visualization, Supervision, Project administration, Funding acquisition. **Michelle L. Hladik:** Conceptualization, Investigation, Resources, Data curation, Writing - review & editing, Visualization, Supervision, Funding acquisition. **Aidee Guzman:** Methodology, Software, Formal analysis, Writing - review & editing. **Sara Winsemius:** Conceptualization, Investigation, Resources, Writing - review & editing, Project administration. **Ariana Bautista:** Investigation, Writing - review & editing. **Claire Kremen:** Conceptualization, Methodology, Validation, Writing - review & editing, Funding acquisition. **Nicholas J. Mills:** Conceptualization, Methodology, Validation, Formal analysis, Writing - review & editing, Supervision.

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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