Environmental Toxicology and Chemistry—Volume 43, Number 8—pp. 1836–1843, 2024
Received: 21 November 2023 | Revised: 9 January 2024 | Accepted: 24 April 2024

Environmental Toxicology

First Detection of Thiamethoxam in a Free-Ranging Insectivorous Bird After its Agricultural Use Ban in Spain

Marina Klaas-Fábregas,[a,1] Pilar Gómez-Ramírez,[a,*,1] Lola Fernández-Gómez,[b] Jesus Alfonso i Prieto,[c] Isabel Garrido,[d] Juana Cava,[d] Carmen M. Martínez-Escudero,[d] Jose Fenoll,[d] and Juan M. Pérez-García[b]

[a]Area of Toxicology, Department of Social Health Sciences, University of Murcia, Murcia, Spain
[b]Department of Applied Biology, Centro de Investigación e Innovación Alimentaria (CIAGRO-UMH), Universidad Miguel Hernández de Elche, Elche, Spain
[c]Societat Ilicitana d'Ornitología, Elche, Spain
[d]Instituto Murciano de Investigación y Desarrollo Agrario y Medioambiental (IMIDA), Murcia, Spain

Abstract: Neonicotinoids are insecticides used worldwide in phytosanitary and biocidal products and veterinary pharmaceuticals. Recently, some restrictions and bans have been imposed due to their adverse effects on nontarget invertebrates, including pollinators. Although they may have direct and indirect effects on wild vertebrates, few studies have assessed exposure to these compounds in wild birds, so our knowledge remains limited. In the present pilot study we have assessed the prevalence of seven neonicotinoid insecticides and some of their metabolites in whole blood samples from 19 European roller (*Coracias garrulus*) nestlings and five adult common kestrels (*Falco tinnunculus*) in an area treated with neonicotinoids to control the palm weevil (*Rynchophorus ferrugineus*) in southeastern Spain. One European roller nestling born in a palm tree was positive for thiamethoxam, with a concentration of 2.26 ng mL^{-1}, but no residues of neonicotinoids or their metabolites were found in adult common kestrels. Future studies are needed to elucidate potential exposure to neonicotinoids at different times of the year. To our knowledge, this is the first report of the presence of thiamethoxam residues in whole blood of a wild bird species after its ban in Spain. *Environ Toxicol Chem* 2024;43:1836–1843. © 2024 The Authors. *Environmental Toxicology and Chemistry* published by Wiley Periodicals LLC on behalf of SETAC.

Keywords: Biomonitoring; Common kestrel; European roller; Insecticide; Wildlife toxicology

INTRODUCTION

Neonicotinoids are systemic insecticides used in many pest control products and applied by a variety of methods (Jeschke et al., 2011), which has led to a widespread use worldwide (Simon-Delso et al., 2015). Due to adverse effects on pollinators (Goulson et al., 2018), in 2013 the European Union restricted the use of imidacloprid, thiamethoxam, and clothianidin for plant protection products (European Commission, 2013), and currently their use is only allowed in permanent greenhouses (European Commission, 2018b, 2018c, 2018d). However, these compounds are still authorized for professional use as biocides, for example, in Spain (Spanish Minister of the President, 2002), as is dinotefuran (European Commission, 2015). Thiacloprid authorization expired in 2020, and it has not been renewed (European Commission, 2020). Finally, nitenpyram has never been approved in the European Union (European Commission, 2022). Currently, acetamiprid is the only neonicotinoid authorized for outdoor phytosanitary use in the European Union (European Commission, 2018a, 2018e). Although these regulations are likely to decrease environmental exposure and, therefore facilitate the recovery of affected species, the effects of such bans still need to be evaluated using an integrative approach. Such an evaluation is needed at the present time, due to the divided opinions on this issue. It should be noted that bans and restrictions of pesticides often open a new exposure scenario, whereby approximately 30% of the products are sold illegally (African, Caribbean, and Pacific Group of States–European Union Joint Parliamentary Assembly [ACP–EU JPA], 2018). This illegal trade is global, moving compounds to many countries, but mainly to developing ones. In fact, in 2018, the European Union published a resolution on the impact of the illegal trade in phytosanitary products, seeds, and other agricultural inputs on the economies of the ACP countries (ACP–EU

This article includes online-only Supporting Information.

[1]These authors contributed equally to this work.
* Address correspondence to pilargomez@um.es
Published online 21 May 2024 in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.5899

JPA, 2018), in an attempt to take action to stop it. In the case of neonicotinoids, the permission for indoor use can be relevant, because the products remain available at market, leaving an open door for emergency authorizations. In fact, in 2020 and 2021, 11 European countries (including Spain) were granted emergency use authorizations for neonicotinoids on sugar beet (European Food Safety Authority, 2021). In addition, several neonicotinoids are still authorized and marketed for veterinary and biosanitary use, which means they are a source of exposure to wildlife.

An increasing number of studies have proved the toxicity of neonicotinoid insecticides to birds, causing immunotoxicity, endocrine disruption, and neurotoxicity in game farm (Gul et al., 2019; Siddiqui et al., 2007) and wild birds (Lopez-Antia et al., 2013, 2015; Pandey & Mohanty, 2015). Such behavioral impairments have adverse effects on body weight and migration due to prolonged stopovers (Eng et al., 2019). Indirect effects are related to a reduction in prey richness and abundance, leading to food deprivation in insectivorous species (Hallmann et al., 2014; Kuechle et al., 2022). Indeed, recent studies have associated the decline of insectivorous birds with high concentrations of neonicotinoids in the environment (Hallmann et al., 2014; Li et al., 2020).

Exposure of wild birds to neonicotinoid insecticides has been commonly described in liver of granivorous birds feeding on coated seeds (MacDonald et al., 2018; Millot et al., 2017). However, insectivorous birds are also susceptible to neonicotinoid contamination when feeding on insects that have been exposed to these compounds (Byholm et al., 2018; Humann-Guilleminot et al., 2021). In predatory birds, the compounds found are the more persistent ones, such as thiacloprid or imidacloprid (Humann-Guilleminot et al., 2021; Taliansky-Chamudis et al., 2017). More recently, residues have also been detected in seabird feathers (Distefano et al., 2022).

In the southeast of Spain lies one of the most extensive palm grove areas in Europe, declared a UNESCO World Heritage Site because of its historical interest. However, since 2005, an outbreak of the palm weevil (*Rynchophorus ferrugineus*) introduced by imported palm trees poses a significant threat necessitating several measures, including the preventive use of neonicotinoids such as imidacloprid, thiamethoxam (most recommended to control this insect), and acetamiprid (Dembilio & Jaques, 2012; Estevez et al., 2011; Ferry & Gómez, 2007; José Javier Sigüenza Murcia, Technical Department Baobab Viveros S.L., personal communication, May 30, 2022). Nevertheless, the risk of neonicotinoids to wildlife inhabiting this area, particularly breeding birds, has never been assessed. To study wild bird exposure to imidacloprid, acetamiprid, thiamethoxam, and other neonicotinoids, we conducted a pilot study in 2021 and 2022 using two species, the European roller (*Coracias garrulus*) and the common kestrel (*Falco tinnunculus*). Some of the neonicotinoid metabolic transformation products were included to facilitate their detection. These species were selected as candidate sentinels due to their high insect consumption during the breeding season and their occupation of palm trees from different habitats, which may result in varying exposure levels to neonicotinoids. The common kestrel inhabits urban areas, and the European roller is found in agricultural areas.

MATERIALS AND METHODS

Study species

The European roller is a medium-sized migratory bird that breeds in the Iberian Peninsula, mainly from May to July. It primarily feeds on insects and occasionally preys on small rodents (Avilés & Parejo, 2002; Catry et al., 2019). Apart from insects, nestlings also consume other organisms such as spiders and gastropods (Avilés & Parejo, 1997). The common kestrel is a small falcon, resident in the Iberian Peninsula. It is a generalist predator that can feed on a wide variety of prey such as insects, micromammals, reptiles, and small birds, among others (Martínez-Padilla, 2016; Orihuela-Torres et al., 2017). Both bird species are considered endangered in Spain according to the National Red Book (Spanish Ornithological Society, 2021).

Study area

Our study area encompassed the city of Elche (Alicante Province, Spain) and surrounding areas, which include El Hondo Natural Park and Santa Pola salterns Natural Park, two Special Protection Areas for Birds in southeastern Spain (38°11′N 0°41′W; Figure 1). Apart from the palm groves in the urban area, the landscape is composed mainly of irrigated crops and palm and pomegranate trees, alternating with natural patches of Mediterranean thermophilus scrubland, and buildings. No greenhouses are found within a 10-km radius of the study area.

Sample collection

During the study period, a total of 19 European roller nestlings (2021, $n=7$; 2022, $n=12$), approximately 20 days old, were hand captured from seven natural nests located in the agricultural areas south of the city of Elche in the surroundings of the two Natural Parks. Adult kestrels (2021, $n=3$; 2022, $n=2$) were captured in the field, in the palm groves of the urban area of Elche, using Bal-Chatri traps baited with mouse (Autonomic Government permits for capture N/Ref.: PM/10.589/vj). Because this was a pilot study, the number of samples was limited, to avoid unnecessary disturbance to the two endangered species, and to obtain a first approximation of exposure to the pesticides.

In both species, blood samples (<500 μL for kestrels and 300 μL for rollers) were collected from the brachial vein, using a syringe and needle, and placed in an Eppendorf tube with heparin as the anticoagulant, following the method described by Espín et al. (2021) and the guidelines of the European Union Directive (European Commission, 2010) for animal experimentation (approved by the Ethical Committee for Animal Experimentation at the University of Murcia; code 657/2020). All samples were immediately transported on ice to the laboratory

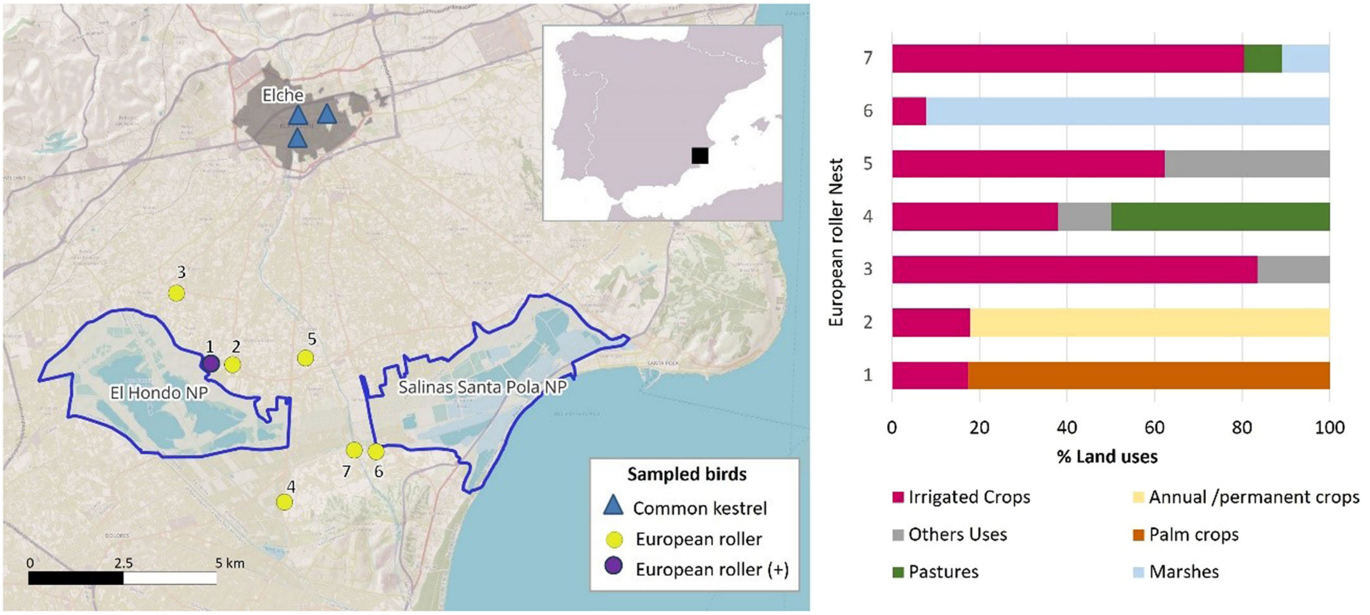

FIGURE 1: Location of the study area in southeast Spain. Circles and triangles indicate the sampling sites for European rollers and common kestrels, respectively. Percentages of land uses corresponding to each roller nest are shown beside the map.

within hours, where they were stored and preserved at −20 °C until analysis.

Sample preparation

All blood samples were thawed immediately before processing and homogenized by vortexing. The extraction technique was performed according to the methods described by Taliansky-Chamudis et al. (2017) and Rial-Berriel et al. (2020), with slight modifications. Briefly, 500 µL of 1% formic acid (Probus®), in acetonitrile (Lab-Scan®) was added to each sample (76–150 µL of whole blood) in an Eppendorf tube and vortexed for 30 s, at 35 hertz (VX-200; LabNet). The samples were then homogenized in an ultrasonic bath (Labsonic®) during 20 min at room temperature. After this step, a combination of salts consisting of 150 mg magnesium sulfate and 37.5 mg sodium acetate (SupelCo; Sigma-Aldrich) was added to each sample, which was then vigorously shaken for 20 s by vortex, then by hand for 1 min, and finally centrifuged at 1538 g for 5 min. After centrifugation, the supernatant was collected, filtered using a syringe with a nylon filter (pore size 0.22 µm and diameter of 13 mm), and then placed in a 2-mL chromatographic vial for chromatographic analysis.

Neonicotinoid analysis

The vials containing the filtered supernatant were sent to the laboratory of the Instituto Murciano de Investigación y Desarrollo Agrario y Medioambiental, Murcia, Spain, under cold conditions for processing using the technique described by Martínez et al. (2022). Seven neonicotinoids (imidacloprid, thiamethoxam, thiacloprid, acetamiprid, clothianidin, dinotefuran, and nitenpyram) and some of their transformation products (6-chloronicotinic acid, hydroxy-imidacloprid, imidacloprid-urea, imidacloprid-olefin, thiamethoxam-urea, thiacloprid-amide, acetamiprid-acetate, and acetamiprid-desmethyl) were analyzed using an high-perfomance liquid chromatography system (consisting of a vacuum degasser, autosampler, and binary pump; Agilent Series 1200, Agilent Technologies) equipped with a reversed-phase C8 analytical column of 150 × 4.6 mm and 5-µm particle size (Zorbax Eclipse XDB-C8) and an Agilent G6410A triple quadrupole mass spectrometer equipped with an electrospray ionization (ESI) interface operating in positive ion mode. The mobile phases A and B were acetonitrile and 0.1% formic acid in water, respectively. The gradient program started with 10% A, constant for 5 min, followed by a linear gradient to 100% A after 35 min. After this 35-min run time, 10 min of post run time followed using an initial 10% of A. The flow rate was held constant (0.7 mL min^{-1}) during the entire process, and 10 µL of sample was injected in every case. Fragmentor voltages and collision energies applied to the compounds, along with their retention times and their optimized selected reaction monitoring transitions, are described in the Supporting Information, Table S1. Calibration curves were analyzed by spiking the pesticides (imidacloprid, thiamethoxam, thiacloprid, acetamiprid, clothianidin, dinotefuran, and nitenpyram) at 0.5 to 20 ng mL^{-1} levels into blank blood samples obtained from healthy hens (*Gallus gallus domesticus*) from the Laboratory Animals Section of the University of Murcia, with authorization code CEEA 177/2015, in three replicates. Linearity was >0.9, limits of quantification were 0.5 ng mL^{-1} for imidacloprid, thiamethoxam, thiacloprid, acetamiprid and clothianidin, 5 ng mL^{-1} for dinotefuran, and 2 ng mL^{-1} for nitenpyram; limits of detection were 0.16 ng mL^{-1} for imidacloprid, thiamethoxam, thiacloprid, acetamiprid, and clothianidin, 1.67 ng mL^{-1} for dinotefuran, and 0.67 ng mL^{-1} for nitenpyram; recoveries ranged from 89% to 172%, except for nitenpyram, which was approximately 30% at all the spike concentrations.

Study of land use composition

To determine the relation of land use composition in the area of the sampled birds to the detection frequency and blood concentrations of neonicotinoids, we studied the composition around nests. This analysis was carried out only for European rollers, because all the kestrels sampled were breeding adults captured within the city of Elche, and their hunting territories are within the urban area, (no difference in land composition among kestrels). We established a radius of 150 m around each roller nest as a surrogate of their home range (Avilés & Costillo, 1998), and land use composition was determined using CORINE Land Cover (European Environment Agency, 2018) in QGIS 3.4 (QGIS Development Team, 2022).

RESULTS AND DISCUSSION

Neonicotinoid residues in European roller and common kestrel

Among all the compounds analyzed, only thiamethoxam was found in one European roller nestling, at a concentration of 2.26 ng mL^{-1} and a prevalence of 5% ($n = 19$). No neonicotinoid residues or their metabolites were found in common kestrel samples.

Despite this low prevalence, the mere fact of the detection is remarkable, given the restricted use in agricultural areas. Therefore, this finding may indicate the presence of other neonicotinoids in the environment, specifically thiamethoxam, 3 years after its restriction by the European Union to the in permanent greenhouses (European Commission, 2018b, 2018c, 2018d).

Although the route of exposure remains unknown, the most likely scenario for the positive chick is the ingestion of prey contaminated with thiamethoxam. These insects, provided by the parents, would have been exposed via biocidal products, illegal practices for phytosanitary uses, or environmental residues of thiamethoxam. As mentioned previously in the *Introduction*, this compound was considered very effective against the palm weevil. Although the half-life of the molecule in the environment varies depending on the type of application, thiamethoxam is not expected to accumulate in the soil, but to degrade in approximately 1 year (Hilton et al., 2016). Ingestion of treated seeds is unlikely, not only because of the regulatory restrictions, but also because of the insectivorous diet of the European roller. However, the diet of nestlings may be complemented with amphibians (Meschini et al., 2009) or reptiles (Pérez-García et al., 2022), and indeed, these preys have been found to be exposed to neonicotinoids in the field (Keller, 2021).

The absence of thiamethoxam in the two siblings of the positive nestling can be explained by the rapid metabolism of this compound (4 h half-life), as observed in toxicokinetic studies in Japanese quail (*Coturnix japonica*; Pan et al., 2022b). Although thiamethoxam is quickly metabolized into clothianidin (in less than 10 h), none of the samples contained residues of this molecule, probably due to the similar rapid elimination of the metabolite (Pan et al., 2022b). In addition, the rare occurrence of positive animals may indicate that their exposure to thiamethoxam is sporadic rather than continuous.

This is one of the few reports of thiamethoxam detection in whole blood samples from a free-ranging bird, as shown in Table 1, a summary of research on thiamethoxam exposure in wild birds. The concentration of thiamethoxam in our study is similar to that found in cirl buntings (*Emberiza cirlus*; Fuentes et al., 2023), and higher than in other wild bird species (Table 1).

Although it was not possible to perform a statistical analysis due to the low prevalence, it is noteworthy that the European roller chick with thiamethoxam hatched in an area with a predominance of palm tree plantations (82.5% in the 150-m buffer used as the home range proxy; Supporting Information, Table S2). This is in contrast to the other nests, which were surrounded by palm trees interspersed with other crops such as olives, pomegranates, and/or abandoned crops and saline vegetation fields (Supporting Information, Table S2). Therefore, the predominance of palm trees in the habitat of the home range used by the parents of the positive nestling could increase the risk of exposure to the neonicotinoids used against the palm weevil.

The absence of imidacloprid and acetamiprid in kestrels is particularly noteworthy. These substances were used in the city of Elche, mainly against palm weevil, until 2018 (imidacloprid and thiamethoxam) and in 2018 to 2022 (acetamiprid; José Javier Sigüenza Murcia, Technical Department Baobab Viveros S.L., personal communication, May 30, 2022). This could be due to the time that had elapsed between treatments and sample collection. In the year of the sampling, acetamiprid was applied in February through tree injection and spraying in the urban area of Elche (José Javier Sigüenza Murcia, Technical Department Baobab Viveros S.L., personal communication, May 30, 2022), and kestrels were trapped in June. To our knowledge, the toxicokinetics of acetamiprid in birds are unknown. However, acetamiprid is expected to dissipate quickly in the environment, with a dissipation half-life in water of 4 to 7 days, and a half-life in soil of 10 to 23 days, depending on concentration and moisture (Pitam et al., 2013). For these reasons, if sampling had taken place shortly after neonicotinoid applications in the study area, there would have been some residues in kestrels blood. On the other hand, negative results do not imply an absence of exposure to neonicotinoids. As just mentioned, the persistence of neonicotinoids in the environment varies among molecules and types of environments (Bonmatin et al., 2015), and the current toxicokinetic literature suggests a rapid metabolism in birds (Bean et al., 2019; Pan et al., 2022b). Consequently, the exposure to neonicotinoids at other times during their lifetime cannot be excluded. In this sense, feathers may be useful because they can reflect the exposure during the whole growth period, in contrast to plasma or blood concentrations (Dauwe et al., 2005; Espín et al., 2016; Humann-Guilleminot et al., 2019).

TABLE 1: Overview of studies that investigated thiamethoxam exposure in wild birds

Species	Thiamethoxam residues	Matrix	Reference
Rufous hummingbird (*Selasphorus rufus*) Anna's hummingbird (*Calypte anna*)	1.47 ng mL^{-1}	Cloacal fluid	Bishop et al. (2018)
Rufous hummingbird Anna's hummingbird Calliope hummingbird (*Selasphorus calliope*) Black-chinned hummingbird (*Archilocus alexandri*)	<LOD (0.063 ng mL^{-1})	Cloacal fluid	Bishop et al. (2020)
Sandwich tern (*Thalasseus sandvicensis*) Mediterranean gull (*Ichthyaetus melanocephalus*)	0.36 ± 0.03 ng g^{-1} 0.16 ± 0.02 ng g^{-1}	Feathers (mean ± SD)	Distefano et al. (2022)
Northern bobwhite (*Colinus virginianus*)	<LOQ (0.50 ng g^{-1})	Liver	Ertl et al. (2018)
Blackbird (*Turdus merula*) Cirl bunting (*Emberiza cirlus*) Common nightingale (*Luscinia megarhynchos*) Gray partridge (*Perdix perdix*) Montagu's harrier (*Circus pygargus*)	<LOD (0.012 pg µL^{-1}) 2.59 ± 1.37 ng mL^{-1} 0.06 ng mL^{-1} 23.73 ng mL^{-1} 1.64 ± 0.46 ng mL^{-1} <LOD (0.012 ng mL^{-1})	Blood	Fuentes et al. (2023)
Anna's hummingbird Black-chinned hummingbird	<LOD (0.74 ng g^{-1}) <LOQ (2.2 ng g^{-1}) <LOD (0.74 ng g^{-1})	Feather rinsate Carcass Feather rinsate Carcass	Graves et al. (2019)
Tricolored blackbird (*Agelaius tricolor*)	<LOD (0.74 ng g^{-1})	Whole liver (carcass)	Graves et al. (2023)
House sparrow (*Passer domesticus*)	Organic farm: 0.10 ± 0.56 ng g^{-1} Integrated production: 0.87 ± 2.77 ng g^{-1} Conventional: 3.48 ± 9.16 ng g^{-1}	Feathers (mean ± SD)	Humann-Guilleminot et al. (2019)
Alpine swift (*Tachymarptis melba*) Barn owl (*Tyto alba*)	<LOQ (0.03 ng mL^{-1})	Feathers Plasma Food boluses Feathers	Humann-Guilleminot et al. (2021)
Wild turkey (*Meleagris gallopavo silvestris*)	110–160 ng mL^{-1}	Liver (range)	MacDonald et al. (2018)
Common kestrel (*Falco tinnunculus*) Barn owl	<LOD (0.80 ng mL^{-1})	Blood	Rial-Berriel et al. (2020)
Eurasian eagle owl (*Bubo bubo*)	<LOQ (10 ng mL^{-1})	Blood	Taliansky-Chamudis et al. (2017)
Northern bobwhite Scaled quail (*Callipepla squamata*)	<LOQ (3.42 ng g^{-1})	Blood	Turaga et al. (2016)

<LOD = below the limit of detection; <LOQ = below the limit of quantification.

Risk assessment

The interpretation of concentrations of contaminants found in wildlife is usually difficult. First, the toxicity of neonicotinoids to the species under study remains unknown. In addition, the 50% lethal dose varies among bird species, including for thiamethoxam (Addy-Orduna et al., 2019; Mineau & Palmer, 2013). However, controlled exposure studies in Japanese quail found that thiamethoxam can induce hepatotoxicity and oxidative DNA damage at plasma concentrations of approximately 2 to 20 ng mL^{-1} (Pan et al., 2022a). Therefore, further field studies should include biomarkers of effects to improve risk assessment for these species.

Populations of both the European roller and the common kestrel are declining in Spain (Spanish Ornithological Society, 2021). The main threats to the European roller are the conversion and intensification of agricultural practices, habitat degradation, the disappearance of nesting sites, and the use of pesticides (Kovacs et al., 2008). For the common kestrel, the current major threats have been the exposure to pesticides, shooting, electrocution, collision with wind turbines, and traffic accidents (Martínez-Padilla, 2016). Neonicotinoid exposure may be contributing to this decline, with an impact on their fitness, reproduction, and survival (Eng et al., 2019).

CONCLUSIONS

This is the first study on neonicotinoid insecticides in the European roller and the common kestrel in southeastern Spain after their ban, to provide information on their prevalence in wild vertebrate species. Thiamethoxam residues in whole blood of a free-ranging bird are seldom reported, so our study contributes to the limited research available. Further research is required to elucidate the risk of exposure of wild birds to neonicotinoids in the southeast of Spain, and might include feathers, plasma, and faeces as matrices, monitoring different age stages, including other insectivorous species, analysis of biomarkers, and attempting to sample shortly after neonicotinoid application in the palm groves.

Supporting Information—The Supporting Information is available on the Wiley Online Library at https://doi.org/10.1002/etc.5899.

Acknowledgments—This article is the outcome of a collaboration between the Area of Toxicology of the University of Murcia, the Ecology Area of the University Miguel Hernández, the Societat Ilicitana d'Ornitologia, the Instituto Murciano de Investigación y Desarrollo Agrario y Medioambiental, José Javier Sigüenza Murcia, Technical Department Baobab Viveros S.L., and the City Hall of Elche. We thank all the investigators and volunteers who helped in the capture and sampling, and F. Sola for laboratory assistance. Our research was partially funded by Ayuntamiento de Elche under an agreement with the University Miguel Hernández of Elche entitled "Estudios para el conocimiento de la Biodiversidad del Municipio de Elche."

Conflict of Interest—The authors declare no conflicts of interest.

Disclaimer—The funders had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, or in the decision to publish the results.

Author Contribution Statement—**Marina Klaas-Fábregas**: Investigation; Visualization; Writing—original draft. **Pilar Gómez-Ramírez**: Conceptualization; Methodology; Validation; Formal analysis; Data curation; Investigation; Supervision; Resources; Visualization; Writing—review & editing. **Lola Fernández-Gómez**: Investigation; Resources; Writing—review & editing. **Jesus Alfonso i Prieto**: Resources; Methodology; Investigation. **Isabel Garrido**: Methodology; Data curation; Investigation; Validation; Writing—review & editing. **Juana Cava**: Formal analysis; Data curation; Validation; Writing—review & editing; Investigation. **Carmen M. Martínez-Escudero**: Methodolog; Data curation; Validation; Writing—review & editing; Investigation. **Jose Fenoll**: Methodology; Data curation; Validation; Writing—review & editing; Resources; Investigation. **Juan M. Pérez-García**: Conceptualization; Methodology; Formal analysis; Writing—review & editing; Data curation; Investigation; Project administration; Resources; Visualization; Funding acquisition; Supervision. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement—Data are available on request to the corresponding author (pilargomez@um.es).

REFERENCES

African, Caribbean, and Pacific Group of States–European Union Joint Parliamentary Assembly. (2018). Resolution on the impact of the illegal trade in phytosanitary products, seeds and other agricultural inputs on ACP countries' economies. *Official Journal of the European Union, C415*, 6–10.

Addy-Orduna, L. M., Brodeur, J. C., & Mateo, R. (2019). Oral acute toxicity of imidacloprid, thiamethoxam and clothianidin in eared doves: A contribution for the risk assessment of neonicotinoids in birds. *Science of the Total Environment*, *650*, 1216–1223. https://doi.org/10.1016/j.scitotenv.2018.09.112

Avilés, J. M., & Costillo, E. (1998). Selection of breeding habitats by the roller (*Coracias garrulus*) in farming areas of the southwestern Iberian Peninsula. *Die Vogelwarte*, *39*(4), 242–247.

Avilés, J. M., & Parejo, D. (1997). Dieta de los pollos de carraca (*Coracias garrulus*) en una zona mediterránea (Extremadura, Suroeste de España). *Ardeola*, *44*(2), 235–237.

Avilés, J. M., & Parejo, D. (2002). Diet and prey type selection by Rollers *Coracias garrulus* during the breeding season in southwest of the Iberian Peninsula. *Alauda*, *70*(1), 227–230.

Bean, T. G., Gross, M. S., Karouna-Renier, N. K., Henry, P. F. P., Schultz, S. L., Hladik, M. L., Kuivila, K. M., & Rattner, B. A. (2019). Toxicokinetics of imidacloprid-coated wheat seeds in Japanese quail (*Coturnix japonica*) and an evaluation of hazard. *Environmental Science & Technology*, *53*(7), 3888–3897. https://doi.org/10.1021/ACS.EST.8B07062

Bishop, C. A., Moran, A. J., Toshack, M. C., Elle, E., Maisonneuve, F., & Elliott, J. E. (2018). Hummingbirds and bumble bees exposed to neonicotinoid and organophosphate insecticides in the Fraser Valley, British Columbia, Canada. *Environmental Toxicology and Chemistry*, *37*(8), 2143–2152. https://doi.org/10.1002/etc.4174

Bishop, C. A., Woundneh, M. B., Maisonneuve, F., Common, J., Elliott, J. E., & Moran, A. J. (2020). Determination of neonicotinoids and butenolide residues in avian and insect pollinators and their ambient environment in western Canada (2017, 2018). *Science of the Total Environment*, *737*, 139386. https://doi.org/10.1016/j.scitotenv.2020.139386

Bonmatin, J. M., Giorio, C., Girolami, V., Goulson, D., Kreutzweiser, D. P., Krupke, C., Liess, M., Long, E., Marzaro, M., Mitchell, E. A., Noome, D. A., Simon-Delso, N., & Tapparo, A. (2015). Environmental fate and exposure; neonicotinoids and fipronil. *Environmental Science and Pollution Research*, *22*(1), 35–67. https://doi.org/10.1007/s11356-014-3332-7

Byholm, P., Mäkeläinen, S., Santangeli, A., & Goulson, D. (2018). First evidence of neonicotinoid residues in a long-distance migratory raptor, the European honey buzzard (*Pernis aviporus*). *Science of the Total Environment*, *639*, 929–933. https://doi.org/10.1016/j.scitotenv.2018.05.185

Catry, I., Sampaio, A., Silva, M. C., Moreira, F., Franco, A. M., & Catry, T. (2019). Combining stable isotope analysis and conventional techniques to improve knowledge of the diet of the European roller *Coracias garrulous*. *Ibis*, *161*(2), 272–285. https://doi.org/10.1111/ibi.12625

Dauwe, T., Jaspers, V., Covaci, A., Schepens, P., & Eens, M. (2005). Feathers as a nondestructive biomonitor for persistent organic pollutants. *Environmental Toxicology and Chemistry*, *24*(2), 442–449. https://doi.org/10.1897/03-596.1

Dembilio, Ó., & Jaques, J. A. (2012). Bio-ecology and integrated management of the red palm weevil, *Rhynchoporus ferrugineus* (Coleoptera: Curculionidae), in the region of Valencia (Spain). *Hellenic Plant Protection Journal*, *5*, 1–12.

Distefano, G. G., Zangrando, R., Basso, M., Panzarin, L., Gambaro, A., Volpi Ghirardini, A., & Picone, M. (2022). The ubiquity of neonicotinoid contamination: Residues in seabirds with different trophic habits. *Environmental Research*, *206*, 112637. https://doi.org/10.1016/j.envres.2021.112637

Eng, M. L., Stutchbury, B. J. M., & Morrissey, C. A. (2019). A neonicotinoid insecticide reduces fueling and delays migration in songbirds. *Science*, *365*, 1177–1180. https://doi.org/10.1126/science.aaw9419

Ertl, H. M., Mora, M. A., Boellstorff, D. E., Brightsmith, D., & Carson, K. (2018). Potential effects of neonicotinoid insecticides on northern bobwhites. *Wildlife Society Bulletin*, *42*(4), 649–655. https://doi.org/10.1002/wsb.921

Espín, S., Andevski, J., Duke, G., Eulaers, I., Gómez-Ramírez, P., Hallgrimsson, G. T., Helander, B., Herzke, D., Jaspers, V. L. B., Krone, O., Lourenço, R., María-Mojica, P., Martínez-López, E., Mateo, R., Movalli, P., Sánchez-Virosta, P., Shore, R. F., Sonne, C., van den Brink, N. W., … García-Fernández, A. J. (2021). A schematic sampling protocol for contaminant monitoring in raptors. *Ambio*, *50*, 95–100. https://doi.org/10.1007/s13280-020-01341-9

Espín, S., García-Fernández, A. J., Herzke, D., Shore, R. F., van Hattum, B., Martínez-López, E., Coeurdassier, M., Eulaers, I., Fritsch, C., Gómez-Ramírez, P., Jaspers, V. L. B., Krone, O., Duke, G., Helander, B., Mateo, R., Movalli, P., Sonne, C., & van den Brink, N. W. (2016). Tracking pancontinental trends in environmental contamination using sentinel raptors—What types of samples should we use? *Ecotoxicology*, *25*, 777–801. https://doi.org/10.1007/s10646-016-1636-8

Estévez, A., Ferry, M., & Gómez, S. (2011). Endoterapia en palmeras. Estudio de la eficacia y persistencia de tiametoxam en tratamientos preventivos contra el picudo rojo. *Phytoma*, *226*, 42–49.

European Commision. (2010). Directive 2010/63/EU of the European Parliament and of the Council of 22 September 2010 on the protection of animals used for scientific purposes Text with EEA relevance. *Official Journal of the European Union, L276*, 33–79.

European Commission. (2013). No 485/2013 of 24 May 2013 amending Implementing Regulation (EU) No 540/2011, as regards the conditions of approval of the active substances clothianidin, thiamethoxam and imidacloprid, and prohibiting the use and sale of seeds treated with plant protection products containing those active substances. *Official Journal of the European Union, L139*, 12–26.

European Commission. (2015). 2015/416 of 12 March 2015 approving dinotefuran as an active substance for use in biocidal products for product-type 18. *Official Journal of the European Union, L68*, 30–32.

European Commission. (2018a). 2018/113 of 24 January 2018 renewing the approval of the active substance acetamiprid in accordance with Regulation (EC) No 1107/2009 of the European Parliament and of the Council concerning the placing of plant protection products on the market, and amending the Annex to Commission Implementing Regulation (EU) No 540/2011. *Official Journal of the European Union, L20*, 7–10.

European Commission. (2018b). 2018/783 of 29 May 2018 amending Implementing Regulation (EU) No 540/2011 as regards the conditions of approval of the active substance imidacloprid. *Official Journal of the European Union, L132*, 31–34.

European Commission. (2018c). 2018/784 of 29 May 2018 amending Implementing Regulation (EU) No 540/2011 as regards the conditions of approval of the active substance clothianidin. *Official Journal of the European Union, L132*, 35–39.

European Commission. (2018d). 2018/785 of 29 May 2018 amending Implementing Regulation (EU) No 540/2011 as regards the conditions of approval of the active substance thiamethoxam. *Official Journal of the European Union, L132*, 40–44.

European Commission. (2018e). 2018/1129 of 13 August 2018 approving acetamiprid as an existing active substance for use in biocidal products of product-type 18. *Official Journal of the European Union, L205*, 4–7.

European Commission. (2020). 2020/23 of 13 January 2020 concerning the non-renewal of the approval of the active substance thiacloprid, in accordance with Regulation (EC) No 1107/2009 of the European Parliament and of the Council concerning the placing of plant protection products on the market, and amending the Annex to Commission Implementing Regulation (EU) No 540/2011. *Official Journal of the European Union*, *L8*, 8–11.

European Commission. (2022). Active substance nitenpyram. EU Pesticides database. https://ec.europa.eu/food/plant/pesticides/eu-pesticides-database/active-substances/?event=as.details&as_id=1386

European Environment Agency. (2018). Copernicus land service—CORINE land cover. European Environment Agency. https://land.copernicus.eu/pan-european/corine-land-cover/clc2018

European Food Safety Authority (EFSA). (2021). Evaluation of the emergency authorisations granted by Member State Finland for plant protection products containing clothianidin, imidacloprid or thiamethoxam. *European Food Safety Authority*, *18*(11), e6959. https://doi.org/10.2903/sp.efsa.2021.EN-6959

Ferry, M., & Gómez, S. (2007). El picudo rojo de la palmera datilera: Gravedad de la plaga en España y necesidad de un cambio radical y urgente de estrategia en la lucha. *Phytoma*, *184*, 1–4.

Fuentes, E., Gaffard, A., Rodrigues, A., Millet, M., Bretagnolle, V., Moreau, J., & Monceau, K. (2023). Neonicotinoids: Still present in farmland birds despite their ban. *Chemosphere*, *321*, 321. https://doi.org/10.1016/j.chemosphere.2023.138091

Goulson, D., 232 signatories. (2018). Call to restrict neonicotinoids. *Science*, *360*(6392), 973. https://doi.org/10.1126/science.aau0432

Graves, E. E., Jelks, K. A., Foley, J. E., Filigenzi, M. S., Poppenga, R. H., Ernest, H. B., Melnicoe, R., & Tell, L. A. (2019). Analysis of insecticide exposure in California hummingbirds using liquid chromatography-mass spectrometry. *Environmental Science and Pollution Research*, *26*, 15458–15466. https://doi.org/10.1007/s11356-019-04903-x

Graves, E. E., Meese, R. J., & Holyoak, M. (2023). Neonicotinoid exposure in tricolored blackbirds (*Agelaius tricolor*). *Environmental Science and Pollution Research*, *30*, 15392–15399. https://doi.org/10.1007/s11356-022-23290-4

Gul, S. T., Khan, A., Ahmad, M., Ahmad, H., Saleemi, M. K., Naseem, M. N., & Bilal, M. (2019). Immuno-toxicological effects of different sub-lethal doses of thiamethoxam (TMX) in broiler birds. *Toxin Reviews*, *38*(3), 200–205. https://doi.org/10.1080/15569543.2018.1435554

Hallmann, C. A., Foppen, R. P. B., van Turnhout, C. A. M., de Kroon, H., & Jongejans, E. (2014). Declines in insectivorous birds are associated with high neonicotinoid concentrations. *Nature*, *511*, 341–343. https://doi.org/10.1038/nature13531

Hilton, M. J., Jarvis, T. D., & Ricketts, D. C. (2016). The degradation rate of thiamethoxam in European field studies. *Pest Management Science*, *72*(2), 388–397. https://doi.org/10.1002/ps.4024

Humann-Guilleminot, S., Clément, S., Desprat, J., Binkowski, Ł. J., Glauser, G., & Helfenstein, F. (2019). A large-scale survey of house sparrows feathers reveals ubiquitous presence of neonicotinoids in farmlands. *Science of the Total Environment*, *660*, 1091–1097. https://doi.org/10.1016/j.scitotenv.2019.01.068

Humann-Guilleminot, S., Laurent, S., Bize, P., Roulin, A., Glauser, G., & Helfenstein, F. (2021). Contamination by neonicotinoid insecticides in barn owls (*Tyto alba*) and alpine swifts (*Tachymarptis melba*). *Science of the Total Environment*, *785*, 147403. https://doi.org/10.1016/j.scitotenv.2021.147403

Jeschke, P., Nauen, R., Schindler, M., & Elbert, A. (2011). Overview of the status and global strategy for neonicotinoids. *Journal of Agricultural and Food Chemistry*, *59*(7), 2897–2908. https://doi.org/10.1021/jf101303g

Keller, P. (2021). Detection of neonicotinoids in northern leopard frog (*Rana pipiens*) brains. *Honors Thesis*, University of South Dakota. 132. https://doi.org/10.1016/j.scitotenv.2021.152424

Kovacs, A., Barov, B., Orhun, C., & Gallo-Orsi, U. (2008). International species action plan for the European roller *Coracias garrulus garrulus*. Birdlife International for the European Commission.

Kuechle, K. J., Webb, E. B., Mengel, D., & Main, A. R. (2022). Seed treatments containing neonicotinoids and fungicides reduce aquatic insect richness and abundance in midwestern USA–managed floodplain wetlands. *Environmental Science and Pollution Research*, *29*, 45261–45275. https://doi.org/10.1007/s11356-022-18991-9

Li, Y., Miao, R., & Khanna, M. (2020). Neonicotinoids and decline in bird biodiversity in the United States. *Nature Sustainability*, *3*, 1027–1035. https://doi.org/10.1038/s41893-020-0582-x

Lopez-Antia, A., Ortiz-Santaliestra, M. E., Mougeot, F., & Mateo, R. (2013). Experimental exposure of red-legged partridges (*Alectoris rufa*) to seeds coated with imidacloprid, thiram and difenoconazole. *Ecotoxicology*, *22*, 125–138. https://doi.org/10.1007/s10646-012-1009-x

Lopez-Antia, A., Ortiz-Santaliestra, M. E., Mougeot, F., & Mateo, R. (2015). Imidacloprid-treated seed ingestion has lethal effect on adult partridges and reduces both breeding investment and offspring immunity. *Environmental Research*, *136*, 97–107. https://doi.org/10.1016/j.envres.2014.10.023

MacDonald, A. M., Jardine, C. M., Thomas, P. J., & Nemeth, N. M. (2018). Neonicotinoid detection in wild turkeys (*Meleagris gallopavo silvestris*) in Ontario, Canada. *Environmental Science and Pollution Research*, *25*, 16254–16260. https://doi.org/10.1007/s11356-018-2093-0

Martínez, C. M., Garrido, I., Flores, P., Hellín, P., Contreras, F., & Fenoll, J. (2022). Ozonation for remediation of pesticide-contaminated soils at field scale. *Chemical Engineering Journal*, *446*, 137182.

Martínez-Padilla, J. (2016). Cernícalo vulgar—*Falco tinnunculus*. A. Salvador & M. B. Morales (Eds.), *Enciclopedia Virtual de los Vertebrados Españoles*.Museo Nacional de Ciencias Naturales.

Meschini, A., Massa, B., & Bruno, M. (2009). Dieta, ritmi di foraggiamento ed importanza degli anfibi durante l' allevamento dei pulli di Ghiandaia marina *Coracias garrulus* nella Maremma laziale. *Alula XVI*, *1–2*, 249–251.

Millot, F., Decors, A., Mastain, O., Quintaine, T., Berny, P., Vey, D., Lasseur, R., & Bro, E. (2017). Field evidence of bird poisonings by imidacloprid-treated seeds: A review of incidents reported by the French SAGIR network from 1995 to 2014. *Environmental Science and Pollution Research*, *24*, 5469–5485. https://doi.org/10.1007/s11356-016-8272-y

Mineau, P., & Palmer, C. (2013). *The impact of the nation's most widely used insecticides on birds*. American Bird Conservancy.

Orihuela-Torres, A., Rosado, D., Perales, P., & Pérez-García, J. M. (2017). Feeding ecology of the common kestrel *Falco tinnunculus* in the South of Alicante (SE Spain). *Revista Catalana d'Ornitologia*, *33*, 10–16.

Pan, Y., Chang, J., Wan, B., Liu, Z., Yang, L., Xie, Y., & Xu, P. (2022a). Integrative analysis of transcriptomics and metabolomics reveals the hepatotoxic mechanism of thiamethoxam on male *Coturnix japonica*. *Environmental Pollution*, *293*, 118460.

Pan, Y., Chang, J., Xu, P., Xie, Y., Yang, L., Hao, W., Li, J., & Wan, B. (2022b). Twenty-four hours of thiamethoxam: In vivo and molecular dynamics simulation study on the toxicokinetic and underlying mechanisms in quails (*Coturnix japonica*). *Journal of Hazardous Materials*, *427*, 128159. https://doi.org/10.1016/j.jhazmat.2021.128159

Pandey, S. P., & Mohanty, B. (2015). The neonicotinoid pesticide imidacloprid and the dithiocarbamate fungicide mancozeb disrupt the pituitary-thyroid axis of a wildlife bird. *Chemosphere*, *122*, 227–234. https://doi.org/10.1016/j.chemosphere.2014.11.061

Pérez-García, J. M., Ferrández, M., Botella, F., Gracià, E., & Orihuela-Torres, A. (2022). Range extension of the ocellated skink, *Chalcides ocellatus* (Forsskål, 1775), in southern Spain provides a warning on its invasion potential. *BioInvasions Record*, *11*(2), 560–570. https://doi.org/10.3391/bir.2022.11.2.28

Pitam, S., Mukherjee, I., & Kumar, A. (2013). Evaluation of environmental fate of acetamiprid in the laboratory. *Environmental Monitoring and Assessment*, *185*, 2807–2816. https://doi.org/10.1007/s10661-012-2750-6

QGIS Development Team. (2022). QGIS Geographic Information System. Open-Source Geospatial Foundation Project. http://qgis.org

Rial-Berriel, C., Acosta-Dacal, A., Zumbado, M., & Luzardo, O. P. (2020). Micro QuEChERS-based method for the simultaneous biomonitoring in whole blood of 360 toxicologically relevant pollutants for wildlife. *Science of the Total Environment*, *736*, 139444. https://doi.org/10.1016/j.scitotenv.2020.139444

Siddiqui, A., Choudhary, M., Goriya, H. V., Bhavsar, S. K., & Thaker, A. M. (2007). Evaluation of immunotoxic effect of short-term administration of quinalphos and imidacloprid in white leghorn cockerels. *Toxicology International*, *14*(1), 15–19.

Simon-Delso, N., Amaral-Rogers, V., Belzunces, L. P., Bonmatin, J. M., Chagnon, M., Downs, C., Furlan, L., Gibbons, D. W., Giorio, C., Girolami, V., Goulson, D., Kreutzweiser, D. P., Krupke, C. H., Liess, M., Long, E., Mcfield, M., Mineau, P., Mitchell, E. A., Morrissey, C. A., … Wiemers, M. (2015). Systemic insecticides (neonicotinoids and fipronil): Trends, uses, mode of action and metabolites. *Environmental Science and Pollution Research*, *22*, 5–34. https://doi.org/10.1007/s11356-014-3470-y

Spanish Ministry of the President. (2002). Real Decreto 1054/2002, de 11 de octubre, por el que se regula el proceso de evaluación para el registro, autorización y comercialización de biocidas. Boletín Oficial del Estado, 247,

Spanish Ornithological Society. (2021). Libro Rojo de las aves de España. N. López-Jiménez (Ed). SEO/BirdLife.

Taliansky-Chamudis, A., Gómez-Ramírez, P., León-Ortega, M., & García-Fernández, A. J. (2017). Validation of a QuECheRS method for analysis of neonicotinoids in small volumes of blood and assessment of exposure in Eurasian eagle owl (*Bubo bubo*) nestlings. *Science of the Total Environment*, *595*, 93–100. https://doi.org/10.1016/j.scitotenv.2017.03.246

Turaga, U., Peper, S. T., Dunham, N. R., Kumar, N., Kistler, W., Almas, S., Presley, S. M., & Kendall, R. J. (2016). A survey of neonicotinoid use and potential exposure to northern bobwhite (*Colinus virginianus*) and scaled quail (*Callipepla squamata*) in the rolling plains of Texas and Oklahoma. *Environmental Toxicology and Chemistry*, *35*(6), 1511–1515. https://doi.org/10.1002/etc.330

