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# Neonicotinoids: Still present in farmland birds despite their ban

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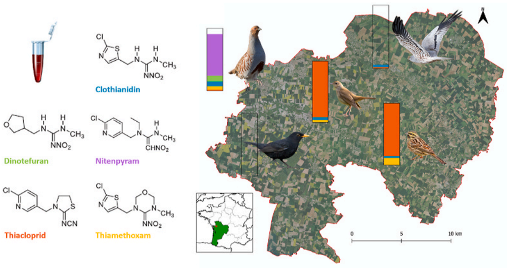
## HIGHLIGHTS

- Neonicotinoids (neonics) quantification in blood of wild birds is scarce.
- Neonics are found in passerine birds, grey partridges and Montagu's harriers' blood.
- Clothianidin, thiacloprid and thiamethoxam, banned in France since 2018, are found.
- Dinotefuran and nitenpyram, used in veterinary care, are found in grey partridges.
- Wild fauna exposure questions the persistence of neonics in the environment.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Neonicotinoids (neonics) are the most widely used insecticides worldwide and are considered to be of low risk to non-target organisms such as vertebrates. Further, they are reported to be rapidly excreted and metabolized, reducing their potential toxicity. Nevertheless, growing evidence of adverse effects of neonics on farmland bird species raise questions about the purported harmless nature of these pesticides. We attempted to search for pesticide residues in species of different trophic levels and at different life stages, by using multiple bird monitoring programs on a Long-Term Socio-Ecological Research (LTSER) platform. Three passerine birds—the blackbird (*Turdus merula*), cirl bunting (*Emberiza cirlus*), and common nightingale (*Luscinia megarhynchos*)—that feed on seeds and invertebrates were monitored during their reproductive period, and the grey partridge (*Perdix perdix*) that feeds on seeds was monitored during its wintering period. We also monitored chicks of an apex predator—the Montagu's harrier (*Circus pygargus*)—that preys mostly upon common voles but also upon insects. We found that the birds' blood samples showed presence of residues of five neonics: three banned since 2018 in France—clothianidin, thiacloprid, and thiamethoxam—and two—dinotefuran and nitenpyram—used for veterinary purposes only. While none of these neonics was detected in blackbirds, all were present in grey partridges. Clothianidin was detected in all species, except blackbirds. Concentrations of the three banned neonics were similar or higher than concentrations found in birds monitored elsewhere before the ban. These findings raise questions about the persistence of neonics within the environment and the mode of exposure to wild fauna. Future investigations on the sublethal effects of these neonics on life-history traits of these farmland birds may

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help in providing a better understanding of the effects of exposure of bird populations to these insecticides, and also to the consequent effect on human health.

## 1. Introduction

During the last century, the need for feeding the growing human population worldwide has led to an intensification of agricultural practices, including an extensive use of pesticides. Despite their use for millennia, several pieces of evidence implicate pesticides in the global decline in biodiversity (Wood and Goulson, 2017; Stanton et al., 2018; Moreau et al., 2022a). Moreover, other studies identify them as the cause of certain diseases in humans (Köhler and Triebskorn, 2013). As all living organisms in a given area share the same environment, protecting biodiversity by reducing pesticides inputs means also reducing humans' exposure to pesticides, and this is necessary to ensure our own health and safety (*One Health* concept). One way is thus to capitalize on wildlife monitoring schemes to better understand the risk of pesticide exposure for humans (Moreau et al., 2022a). In that respect, wild bird species are valuable candidates as they are well-monitored worldwide, being involved in long-term banding programs over decades or more, which have highlighted a global declining trend in several taxa and especially in farmland birds, e.g., in France, Sweden, United Kingdom, US, Europe, and North America (Wretenberg et al., 2006; Comolet-Tirman et al., 2015; Stanton et al., 2018; Rosenberg et al., 2019; Li et al., 2020; Burns et al., 2021; DEFRA, 2021). Pesticide use has been often identified as a major component responsible for this decline (Campbell et al., 1997; Geiger et al., 2010; Mineau and Whiteside, 2013; Chiron et al., 2014; Tassin de Montaigu and Goulson, 2020). Recent studies, for instance, showed a negative relationship between the abundance of northern bobwhites (*Colinus virginianus*) and their exposure to neonicotinoid insecticides (neonics) from 1978 to 2012 (Ertl et al., 2018). Similarly, a wide-panel dataset regarding neonics use and birds' population trends revealed a significant negative impact of this family of pesticides on avian biodiversity from 2003 to 2010 in the Netherlands (Hallmann et al., 2014) and from 2008 to 2014 in USA (Li et al., 2020).

Neonics are insecticides developed in the 1970s, with the first patent dating back to 1977 for nithiazine (rapidly abandoned due to its poor stability), followed by patents for imidacloprid and thiacloprid in 1985, nitenpyram in 1988, acetamiprid and clothianidin in 1989, thiamethoxam in 1992, and dinotefuran in 1994 (Tomizawa and Casida, 2005). Neonics target the neural function and are competitive agonists of nicotinic acetylcholine receptors (nAChR), which increase specificity to insects and not vertebrates (Tomizawa and Casida, 2003). In fact, in contrast to other insecticides such as carbamates and organophosphorous that also target the neural function by inhibiting the acetylcholinesterase (AChE) enzyme—an ubiquitous enzyme in the animal kingdom (review in Grue et al., 1997; Story and Cox, 2001; Walker, 2003; Mitra et al., 2011)—neonics were supposed to have minimal effects and a low toxicological impact on vertebrates and consequently on birds, due to the lower number of nAChRs they have and the lower sensitivity of their nAChRs compared to those of insects (Tomizawa and Casida, 2003, 2005; Ihara et al., 2017; Casida, 2018). Moreover, they were claimed to be rapidly metabolized and excreted, in a few hours (Bishop et al., 2018, 2020; Casida, 2018; Bean et al., 2019; English et al., 2021; Pan et al., 2022), but some reports question their fate along the trophic chain as they have been found in insectivorous birds, granivorous birds, piscivorous birds, and birds of prey (see references in the supplementary materials Table S1). Under experimental conditions, they seem to accumulate in the liver (Lopez-Antia et al., 2015a) and to be detectable in different organs and tissues, although they (at least for imidacloprid and thiamethoxam) seem to be rapidly cleared from birds' organism (Bean et al., 2019; Pan et al., 2022). Several studies showed various effects of neonics exposure on birds at different physiological levels (review in Gibbons et al., 2015 and Moreau et al., 2022a). Although neonics do not act directly on AChE, they may induce neuronal degeneration, which affects AChE activity (Abu Zeid et al., 2019; Rawi et al., 2019), altering more complex functions such as learning and migration behaviour (Eng et al., 2017) but also having sublethal effects on important functions of the organism such as the haematocrit, antioxidant defences, immunity, or fecundity (Lopez-Antia et al., 2013, 2015a; 2015b; Tokumoto et al., 2013; Hoshi et al., 2014; Mohanty et al., 2017; Humann-Guilleminot et al., 2019; Lv et al., 2020).

Since the 1990s, the use of neonics has become widespread, making them the most widely used class of insecticide worldwide, mostly for coating seeds, despite being shown to impact non-target species, including humans (Tomizawa and Casida, 2005; Casida and Durkin, 2013; Gibbons et al., 2015; Simon-Delso et al., 2015; Henry et al., 2015; Wood and Goulson, 2017; Casida, 2018; Thompson et al., 2020; Zhang et al., 2022). In Europe, despite an EU moratorium in 2014, neonics were still detectable in bee-attractive crop nectar until 2018 at least (Wintermantel et al., 2020). Indeed, their degradation in soil (measured by $DT_{50}$ which is the Detection Time 50% representing the time to detect a 50% decrease in pesticide concentration) can take quite a long time, up to more than 6900 days (i.e., 19 years for clothianidin; see Table 2 in Thompson et al., 2020). Therefore, banning harmful neonics does not necessarily eradicate the problem of exposure. In EU, dinotefuran and nitenpyram have never been considered for use in phytopharmaceutical products (PPP; July 2022, EU Pesticides database: https://food.ec.europa.eu/plants/pesticides_en) but are commonly used in veterinary medicine. In France, neonics are banned for outdoor use and in PPP since September 2018 (Décret n° 2018–675, July 2018), except for emergency authorized use of thiamethoxam and imidacloprid on sugar beet crops in production areas (EFSA, 2021). Nonetheless, the use of neonics is still allowed in veterinary medicine.

In the present study, we use different bird monitoring programs on a long-term socio-ecological research (LTSER) platform and a multi-residue analysis (Rodrigues et al., 2023) to evaluate the presence of neonics in an intensive farmland area where acetamiprid, clothianidin, imidacloprid, thiacloprid, and thiamethoxam were banned for 3 years for agricultural use. Nitenpyram and dinotefuran were also included in the screening to control for potential exposure of wild fauna to veterinary products. As the method was not specifically developed for neonics detection, acetamiprid and imidacloprid, were not detectable among the other 104 pesticide compounds. We selected five different species for their different ecology: (i) three passerine birds during their reproductive period: the blackbird (*Turdus merula*), cirl bunting (*Emberiza cirlus*), and common nightingale (*Luscinia megarhynchos*) that feed on seeds and invertebrates; (ii) the grey partridge (*Perdix perdix*) caught during its wintering period when it feeds only seeds; and (iii) an apex predator species, namely, the Montagu's harrier (*Circus pygargus*) that preys mostly upon common voles but also upon orthopterans. For this fifth species, we focused on chicks that are fed by their parents during the rearing period. Beyond the interest of their contrasted ecologies, farmland bird species including buntings, blackbirds or raptors have been shown to be exposed to neonics, however, a limited number were subject of measures in blood samples (Lennon et al., 2020a). Additionally, grey partridge is recognised to be a focal species for pesticide risk assessment (Millot et al., 2017; Bonneris et al., 2019). Here, we aimed at monitoring potential exposure of multiple bird species in an area where there is presumably no use of neonics for agricultural purposes for 3 years and where nitenpyram used in veterinary medicine has never been assessed to our knowledge. We thus collected blood samples from all individuals to determine the presence and measure the level of exposure to neonics. We selected blood (whole blood, i.e., red blood cells and plasma) for the analyses in order to focus on the effects of short-term exposure only

(Espín et al., 2016), so that potential exposure of the migratory species (i.e., common nightingales, Montagu's harriers, and blackbirds) at wintering areas can be neglected.

## 2. Materials and methods

### 2.1. Study area

The study site is located in southwestern France (46°11′N, 0°28′W, Fig. 1), in the Long-Term Socio-Ecological Research Zone Atelier Plaine & Val de Sèvres (LTSER ZAPVS), a 450 km$^2$ area where the soil occupancy and the agricultural practices are monitored each year since 1994 (Bretagnolle et al., 2018). In this intensive farming area, winter cereal crops accounted for ~41% (wheat: 33.8% and corn: 9.6%) of the area under cultivation; in addition, there were sunflower (10.4%), oilseed rape (8.3%), pea (2%), and meadows (13.5%) (average coverage between 2009 and 2016, Bretagnolle et al., 2018). In this area, organic farming (no pesticide use) is carried out in 18% of the agricultural area. Detailed data on pesticide applications was not available, however, until 2018 imidacloprid was used on cereal crops in the study area, thiacloprid, thiamethoxam and clothianidin for their part were mainly used in maize, oilseed rape and cereal crops. Different monitoring studies performed in this area showed the transfer of neonics in several compartments. For instance, imidacloprid was found in soils, oilseed rape nectar, earthworms, and small mammals in this area (Henry et al., 2015; Wintermantel et al., 2020; Pelosi et al., 2021; Fritsch et al., 2022). Thiacloprid was detected in soils, earthworms and small mammals' hair while thiamethoxam was detected in nectar and soils, and clothianidin in nectar and small mammals' hair (Wintermantel et al., 2020; Pelosi et al., 2021; Fritsch et al., 2022). Acetamiprid has been detected in small mammals' hair (Fritsch et al., 2022) despite its use is mainly for market gardening such as tomatoes, squash, and melon cultures which are not present in a substantial surface of the study area. All five neonics are banned for agricultural use in France since 2018, except imidacloprid and thiamethoxam for emergency authorizations on sugar beet crops. Still, sugar beet crops are only present in this area for their first year (seed production), and the use of neonics is banned for this purpose. Therefore, at the moment when birds were caught (2020–2022, see section 2.2.1), no neonics were being used for agricultural purposes. However, imidacloprid, dinotefuran, and nitenpyram may be in domestic use as veterinary treatment for domestic animals (cats, dogs, ferrets, and bunnies). There is no known screening of nitenpyram in the study area and dinotefuran has only been investigated in one previous study but was not detected in small mammals' hair sampled (Fritsch et al., 2022).

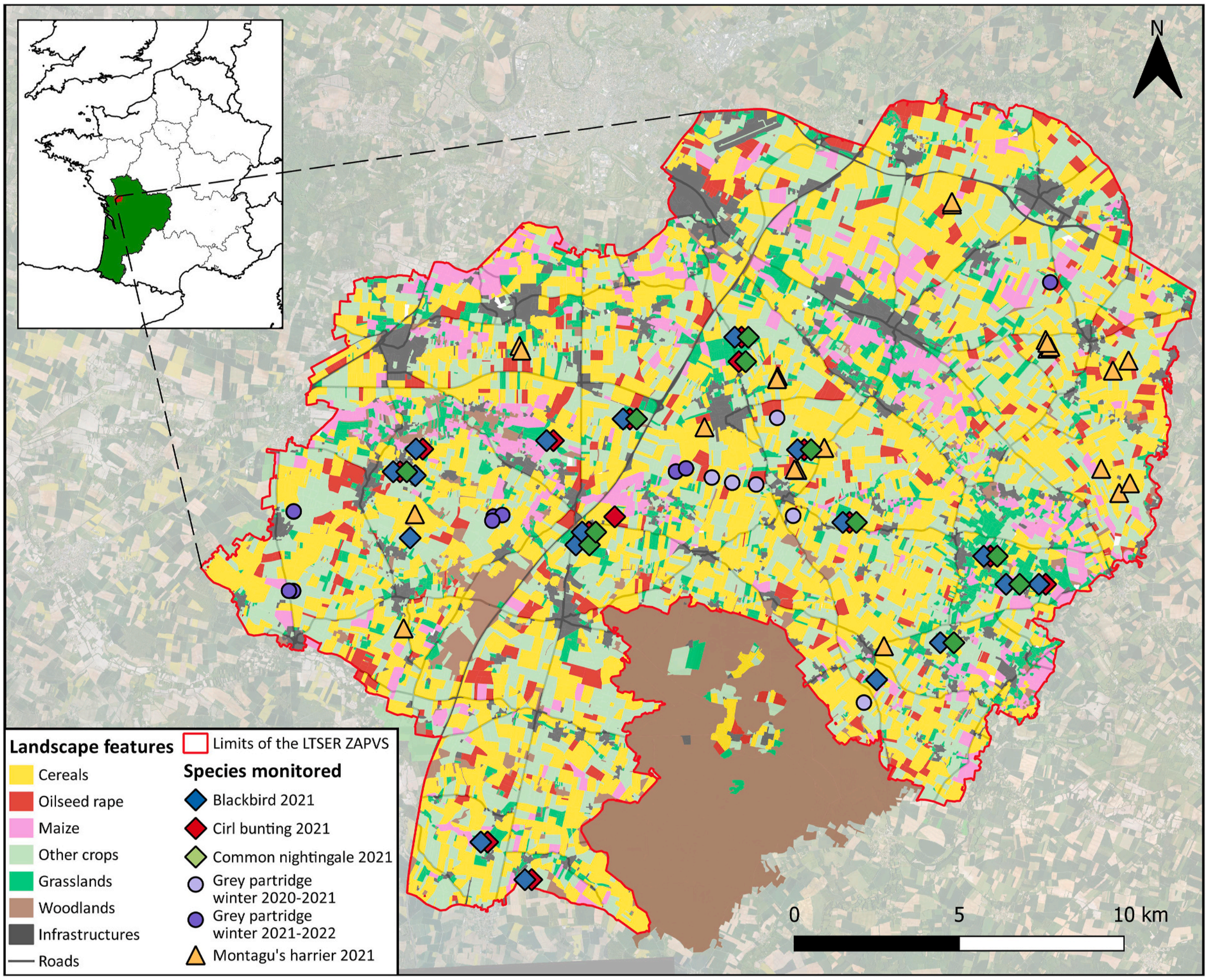

**Fig. 1.** Spatial distribution of birds' catching sites across the Long-Term Socio-Ecological Research Zone Atelier Plaine & Val de Sèvres (LTSER ZAPVS). Landscape features provided correspond to data available for 2021 from our GIS database. Infrastructures correspond to buildings, sport fields, cemeteries, locks, and bridges present in the study area. The five bird species monitored in the study area are blackbird (*Turdus merula*, $N_{2021} = 64$), cirl bunting (*Emberiza cirlus*, $N_{2021} = 31$), common nightingale (*Luscinia megarhynchos*, $N_{2021} = 34$), grey partridge (*Perdix perdix*, $N_{2020\text{-}2021} = 23$ and $N_{2021\text{-}2022} = 31$), and chicks of Montagu's harrier (*Circus pygargus*, $N_{2021} = 55$). N corresponds to the number of adults sampled, except in the case of Montagu's harrier where N corresponds to the number of chicks sampled.

### 2.2. Model species and blood collection

#### 2.2.1. Model species

*2.2.1.1. Adult passerine birds.* The passerine species were sampled during the reproductive period from mid-April to end of June 2021. Birds were caught using net-trapping, following the same methodology as Moreau et al. (2022b). Among the 17 species trapped in this area, we selected three, namely, cirl buntings (N = 31), blackbirds (N = 64), and common nightingales (N = 34) because these species were among the biggest of those captured, which allowed us to draw enough blood for analysis without risking any individual's health. They were also the most caught in the study area allowing us to make a large spatial screening.

*2.2.1.2. Adult grey partridges.* Grey partridges were caught during two consecutive years from November 2020 to February 2021 (winter 2020–2021) and from December 2021 to March 2022 (winter 2021–2022) in the study area. These individuals, even when caught in the wild, are gamebirds and have been probably raised in captivity before being released for hunting purposes; however, knowing the proportion of captive-born vs. wild-born partridges is almost impossible as banding before release is not mandatory. In winter, grey partridges are mostly herbivorous and granivorous, living in large coveys in winter crops and sleeping in ploughed fields at night. Thermal binoculars were used to spot them at nightfall and birds were then captured in the dark by dazzling them and using a landing net (i.e., a method inspired by Eurasian Woodcock catching technique; Williams, 2015). Blood samples were then collected, and birds were immediately released. A total of 23 and 31 partridges were caught during the 2020–2021 and 2021–2022 winters, respectively.

*2.2.1.3. Montagu's harrier chicks.* The Montagu's harrier has been monitored in the ZAPVS since 1994 (Bretagnolle et al., 2018). In this intensive agricultural area, they nest mainly on the ground of cereal crops and lay up to eight eggs (Arroyo et al., 1998; Millon et al., 2008). The incubation period lasts 29 days, and the rearing period is between 30 and 35 days (Arroyo et al., 2007). The mean productivity in this site is 2.05 fledglings per breeding attempt (Arroyo et al., 2004), this success depending mainly on the availability of its main prey, the common vole (*Microtus arvalis*) although in case of poor vole availability, harriers may also feed on orthopterans (Salamolard et al., 2000; Butet and Leroux, 2001). Blood samples of chicks that were 26 ± 2 days old were collected from June 2021 to early August 2021. Fifty-five chicks from 22 nests were sampled.

#### 2.2.2. Blood sampling procedure

For all species, blood samples were collected in 2021, and in 2020 and 2022 for grey partridges, on wild individuals included in different monitoring programs (see details in section 2.2.1). For all of them, blood sampling was conducted by puncturing the brachial vein using a sterile needle and using heparinized capillaries to collect 50 μL of blood. Blood samples were placed in Eppendorf tubes and kept refrigerated (0–5 °C), before being returned to the laboratory where they were stored at −20 °C for further analyses.

### 2.3. Neonics analysis

Neonic extractions were conducted following the method reported by Rodrigues et al. (2023; see also Table S2 in supplementary materials for a description of the neonics). Briefly, blood samples were defrosted and weighed, and a mixture of 2 mL of dichloromethane and ethyl acetate (1:1) was added to each sample, followed by homogenisation by using a vortex for 1 min. Extracts were then sonicated for 10 min. This sonication step was repeated three successive times. After each sonication step, a centrifugation step of 5 min was performed, and supernatants were collected, pooled, and then gently evaporated under a fume hood until a final volume of 500 μL. The extract was collected and stored at −20 °C until the analyses to determine pesticide levels were performed by liquid chromatography coupled to tandem mass spectrometry (LC/MSMS) using multiple reactions monitoring (MRM) for quantification.

LC/MSMS analyses were conducted with a Thermo Scientific TSQ Quantum Access Triple Quadrupole Mass Spectrometer operating in heated positive electrospray ionization mode (HESI+) coupled with a Thermo Accela 1250 pump and a Thermo Combi Pal autosampler. Analyses were performed on a Nucleodur $C_{18}$ Pyramid column (150 mm × 3 mm, 3 μm). Samples were analysed in the gradient mode using a mobile phase composed of water and acetonitrile with both containing 0.1% formic acid.

The multiresidue analysis comprised detection and quantification of 104 pesticide molecules, including five neonics—clothianidin, dinotefuran, nitenpyram, thiacloprid, and thiamethoxam—performed using the MRM detection mode. The source was operated in the positive ionization mode with a spray voltage of 4500 V and the same spray and capillary temperatures of 300 °C each. Nitrogen was used as the sheath and auxiliary gas (20 and 10 arbitrary units), while argon was used as the collision gas (1.5 arbitrary units). Two precursor product ion transitions for each analyte and internal standards were used for quantification. The transitions selected for MSMS analysis and retention times are shown in supplementary materials (Table S3). Data were acquired and processed using Excalibur software.

The limits of detection (LOD) and quantification (LOQ) represent three and ten times the ratio of the average noise height on either side of a known amount of a compound's peak to the peak height, respectively. The objective was to determine the minimum peak heights that can be used to distinguish a compound's peak from the noise on either side of the peak. The LOD and LOQ determined for each sample type are presented in Table S3. LODs varied from 0.001 to 0.012 pg $\mu L^{-1}$ and LOQs from 0.005 to 0.041 pg $\mu L^{-1}$.

## 3. Results

No neonics were detected in blackbirds. For the other species, all 5 neonics searched, namely, clothianidin, dinotefuran, nitenpyram, thiacloprid, and thiamethoxam were detected. A summary of the concentrations and occurrence of these neonics in our sampled bird species is provided in Table 1, and detailed neonicotinoid distribution among each individual is provided in Fig. 2. Clothianidin was found in all species (except blackbirds), with average concentrations ranging from 0.05 pg $\mu L^{-1}$ in common nightingales to 951.60 pg $\mu L^{-1}$ in Montagu's harriers. The number of individuals that exhibited concentrations higher than the LOQ ranged from ~5% among Montagu's harriers up to ~26% among grey partridges sampled during the 2021–2022 winter. No clothianidin was detected in grey partridges sampled in the 2020–2021 winter. Thiacloprid was detected in all cirl buntings and common nightingales but always at concentrations below the LOQ. It was detected in one Montagu's harrier chick (89.58 pg $\mu L^{-1}$) and one grey partridge (0.07 pg $\mu L^{-1}$) in the 2020–2021 winter. Thiamethoxam was detected in both passerine species and in grey partridges from both winters, with concentrations ranging from 0.06 pg $\mu L^{-1}$ in a common nightingale to 23.73 pg $\mu L^{-1}$ in a grey partridge from the 2020–2021 winter. The number of individuals that exhibited concentrations higher than the LOD ranged from ~4% among common nightingales up to 12% among cirl buntings. Dinotefuran and nitenpyram were only detected in grey partridges: in the first winter (2020–2021), ~13% and ~87% of the individuals exhibited dinotefuran and nitenpyram concentrations higher than the LOD, with the average concentrations being 6.20 and 23.10 pg $\mu L^{-1}$, respectively. In the second winter (2021–2022), dinotefuran and nitenpyram were detected with concentrations higher than the LOD in ~32 and ~94% of the individuals, respectively, with the

**Table 1**
Summary of the mean concentrations [c] with standard deviations (SD) and range in pg μL$^{-1}$ with their sample size (n) obtained by LC-MS/MS above the limits of detection (LOD) for each species. The sample size for each species is also provided (N). The percentage of samples above the LOD (n/N) is given in brackets (rounded to the nearest percent). Blackbirds are not referenced as no neonic was detected in the sampled individuals.

| | | Clothianidin | Dinotefuran | Nitenpyram | Thiacloprid | Thiamethoxam |
|---|---|---|---|---|---|---|
| **Cirl buntings (N = 34)** | | | | | | |
| | n | 5 (*15%*) | 0 | 0 | 34 (*100%*) | 4 (*12%*) |
| | [c] ± SD | 2.28 ± 1.55 | – | – | LOD < [c] < LOQ | 2.59 ± 1.37 |
| | range | 1.04–4.87 | – | – | – | 1.61–4.56 |
| **Common nightingales (N = 25)** | | | | | | |
| | n | 2 (*8%*) | 0 | 0 | 25 (*100%*) | 1 (*4%*) |
| | [c] ± SD | 0.05 ± 0.06 | – | – | LOD < [c] < LOQ | 0.06 |
| | range | 0.009–0.093 | – | – | – | – |
| **Grey partridges (N = 54)** | | | | | | |
| *Winter 2020/2021 (N = 23)* | n | 0 | 3 (*13%*) | 20 (*87%*) | 1 (*4%*) | 1 (*4%*) |
| | [c] ± SD | – | 6.20 ± 3.62 | 23.10 ± 11.33 | 0.07 | 23.73 |
| | range | – | 2.14–9.10 | 1.24–41.03 | – | – |
| *Winter 2021/2022 (N = 31)* | n | 8 (*26%*) | 10 (*32%*) | 29 (*94%*) | 0 | 3 (*10%*) |
| | [c] ± SD | 5.26 ± 4.10 | 7.85 ± 4.07 | 18.29 ± 10.84 | – | 1.64 ± 0.46 |
| | range | 1.92–14.26 | 3.32–16.61 | 1.78–43.53 | – | 1.23–2.14 |
| **Montagu's harriers (N = 55)** | | | | | | |
| | n | 3 (*5%*) | 0 | 0 | 1 (*2%*) | 0 |
| | [c] ± SD | 951.60 ± 1299.21 | – | – | 89.58 | – |
| | range | 194.48–2451.78 | – | – | – | – |

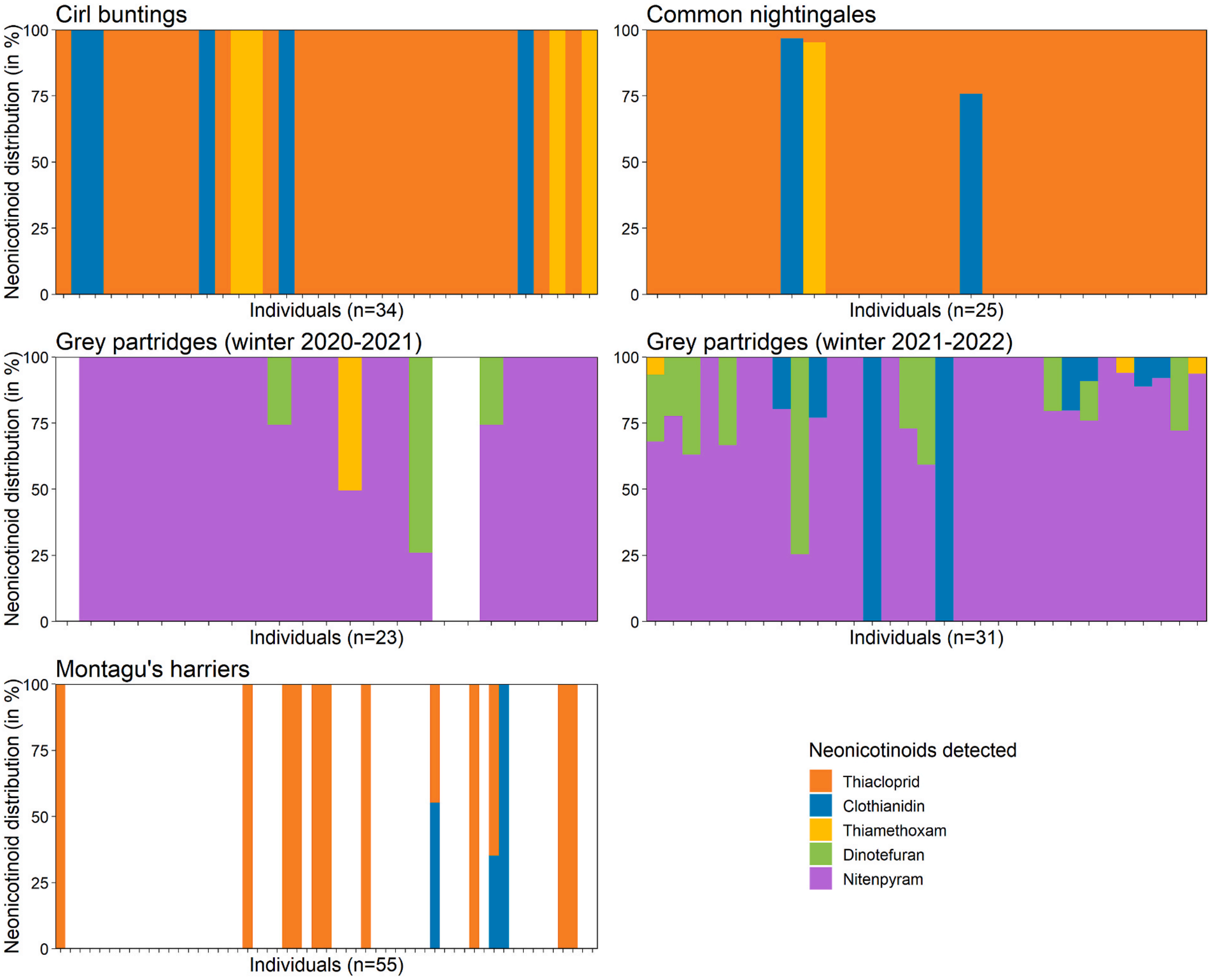

**Fig. 2.** Wild farmland birds' exposure to 5 neonicotinoids. Distribution of the different neonicotinoid molecules for each individual of each species are represented as one stacked bar. Distribution in percent was obtained from raw concentrations of each molecule in birds blood sampled.

average concentrations being 7.85 and 18.29 pg $\mu L^{-1}$, respectively.

## 4. Discussion

In the last few decades, neonics have attracted considerable interest (Klingelhöfer et al., 2022), especially because of the rising concern about their effects on non-target species such as honeybees (Wood and Goulson, 2017) and humans (Cimino et al., 2017; Han et al., 2018). Because of their chemical properties such as their half-life in the soil ($DT_{50}$), solvability, and leaching potential, and because of their extensive use for agricultural purposes, the assessment of their presence in farmlands is imperative (Thompson et al., 2020). Considering that no neonics were used on plants since 2018 in France and that the analytical method used here provided good sensitivity results (Rodrigues et al., 2023), we expected to find no or very low concentrations in bird blood samples as this matrix reflects short-term exposure (Espín et al., 2016). However, in the present study, we not only detected five out of seven neonics, three of which have been banned since 2018 in France (clothianidin, thiacloprid, and thiamethoxam) and two others (dinotefuran and nitenpyram) that are supposedly being used only for treating domestic pets (Table S2), but in some cases, at rather high concentrations. The remaining two neonics (imidacloprid and acetamiprid) are not discussed here as these compounds were not detectable with the multiresidue method used in this study. However, knowing the historical background of the study area and results from studies in the same area before neonics ban (see section 2.1), if titration had been possible, we would have expected to find large amounts of imidacloprid in birds' blood and probably no or very low concentrations of acetamiprid.

### 4.1. Monitoring neonics in wild birds

Neonics were increasingly studied during the last decade and several recent studies have focused on quantifying the level of exposure in wild birds. However, these studies differ from our work in several aspects. First, in these studies, several biological matrices—blood, carcasses, eggs, faecal pellets and cloacal fluids, feathers, liver, and/or gizzard—were considered (see Table S1 for detailed references). Correlations between neonics quantifications from different matrices can be found but are not systematic (reviewed in Espín et al., 2016). Indeed, pesticides are distributed in biological tissues with different temporal patterns. For instance, pesticide molecules are integrated into feathers during the moulting period (Humann-Guilleminot et al., 2022). The distribution of such neonic molecules in internal tissues such as the liver also depends on the chemical nature of the molecules and can be degraded if samples are taken on carcasses (Espín et al., 2016, and references therein). Then, a time lag might be observed between the exposure and the detection depending on the matrices used (Lennon et al., 2020b). Therefore, the results obtained from different matrices cannot be directly compared.

Our results can thus be reasonably compared to those of five previous studies that tested for neonics in whole blood (or plasma) samples from wild birds (Taliansky-Chamudis et al., 2017; Byholm et al., 2018; Hao et al., 2018; Lennon et al., 2020b; and Humann-Guilleminot et al., 2021; Table S4). For clothianidin, our concentration range was similar to those found by Lennon et al. (2020b), although the maximum value was lower: they found concentrations ranging from 0.5 (dunnocks) to 69, 300 pg $\mu L^{-1}$ (yellowhammers), whereas in our study, the values were 0.009 (common nightingale) to 2451.78 pg $\mu L^{-1}$ (Montagu's harrier chick). The maximum clothianidin concentration found by Humann-Guilleminot et al. (2021) was 0.34 pg $\mu L^{-1}$ in alpine swifts, which is similar to the lower values in the concentration range we obtained. Thiacloprid concentrations in all cirl buntings and common nightingales were below the LOQ, but concentrations in the grey partridges and the Montagu's harrier chicks were far higher than those found in honey buzzards by Byholm et al. (2018) and in white-crowned sparrows by Hao et al. (2018). They ranged from 0.012 to 0.031 and 0.0025 to 0.0031 pg $\mu L^{-1}$ for honey buzzards and white-crowned sparrows, respectively, and from 0.073 to 89.58 pg $\mu L^{-1}$ in our study (Table S4). This result suggests that the grey partridges and the Montagu's harrier chicks were exposed to high, or recent quantities of thiacloprid (see section 4.2 for further details). For thiamethoxam, the concentrations we found were above those obtained by Hao et al. (2018): they ranged from 0.06 (common nightingales) to 23.73 (grey partridges) pg $\mu L^{-1}$, whereas Hao et al. reported 0.0051 to 0.0337 pg $\mu L^{-1}$. Taliansky-Chamudis et al. (2017) detected imidacloprid in one Eurasian eagle owl (*Bubo bubo*) chick among the 30 sampled, but did not detect any other neonics, so comparisons with our results are not possible. The difference in the results despite all studies using blood samples may be because some of the studies considered whole blood samples (Taliansky-Chamudis et al., 2017; Byholm et al., 2018; our study), while others used only plasma (Hao et al., 2018; Lennon et al., 2020b; Humann-Guilleminot et al., 2021), inducing differences in the detectability of some molecules whose levels vary according to their water/lipid solubility, and affinity to different proteins (Rodrigues et al., 2023; Zhang et al., 2023). Moreover, all these studies fundamentally differ in their sensitivity as they rely on their own developed chemical analysis methods [although they are all derived from the same method, i.e., Quick, Easy, Cheap, Effective, Rugged, and Safe (QuEChERS); Anastassiades et al., 2003]. If we consider clothianidin for instance, LOD and LOQ in our study (0.01 and 0.04 pg $\mu L^{-1}$, respectively; Table S3) were 5 (LOQ) and 15 (LOD) times lower, respectively, than those (0.15 and 0.21 pg $\mu L^{-1}$, respectively) of Lennon et al. (2020b), while LOQ was similar to that (0.05 pg $\mu L^{-1}$) reported by Humann-Guilleminot et al. (2021). This may induce a difference in the number of neonic positive samples between studies, hence differences in average concentrations as well. For instance, applying the LOD of clothianidin from the study of Lennon et al. (2020b) would have led to non-detection of positive samples in common nightingales while we found 8% of individuals with concentrations above our LOD. It is not therefore easy to compare the results provided by the authors, especially when not all statistical values, such as the percentage of neonic detection among individuals or the mean concentration with its standard deviation, were obtained on the same basis (Table S4). As the use of the same methodology in all studies does not seem to be feasible, researchers should systematically report these values to allow direct comparisons.

### 4.2. Exposure of wild fauna to neonics

Previous studies reported the limitations of feather and internal tissue samples for determining timing of exposure to contaminants, so blood is considered most suitable for determining recent exposure (Espín et al., 2016; Lennon et al., 2020b). Neonics are supposedly "rapidly" excreted (in hummingbirds, English et al., 2021) and cleared from blood (24 h in quails), according to data regarding imidacloprid (Bean et al., 2019) and thiamethoxam (Pan et al., 2022); however, extrapolation to the behaviour of other neonics may not be reliable. Based on blood samples used for this study we cannot estimate long-term exposure, therefore, possible exposure of the migratory species at their wintering areas may be disregarded. However, thiamethoxam/clothianidin were found in grey partridges' eggs in another study (Bro et al., 2016), so in the case of the Montagu's harrier chicks, we cannot exclude the possibility of maternal transfer of neonics, i.e., from the mother to the eggs. In other words, all tested positive individuals in the present study were probably recently exposed to neonics. This is important because the birds were sampled three and four years after EU banned neonics for PPP, and no derogation is known in our study area conversely to other regions in France. Nonetheless, we found three out of the five PPP neonics in our samples, with concentrations similar to those in birds sampled elsewhere before the ban (Byholm et al., 2018). Taken together, our results strongly suggest that clothianidin, thiacloprid, and thiamethoxam are still present in farmlands and raise questions about the mode of exposure to birds. Thiamethoxam is metabolized into clothianidin in animals, plants, and soil (Nauen et al., 2003, review in Simon-Delso et al., 2015; Pan et al., 2022) potentially explaining why it is more commonly present than thiamethoxam. These neonics' $DT_{50}$ ranged from few days for thiacloprid up to several years for clothianidin (Table S2) in the soil, meaning that they can still be incorporated into the diet of several detritivores such as earthworms (Pelosi et al., 2022). If so, at least some blackbirds, which are omnivorous and feed on different invertebrate species during the breeding season, including earthworms which are known to bioaccumulate pesticides (Pelosi et al., 2022, 2021), should have tested positive, but were not. This surprising result could have been first seen as a technical failure; however, other pesticide molecules than neonics were detected in blackbirds' blood, so that an analysis problem can be discarded. The absence of neonics in blackbirds' blood might be the consequence of several processes. One of them could be a higher detoxification capacity of blackbirds, mediated by their ability at monopolizing high amounts of carotenoids, which are antioxidants, involved in sexual selection and known for their role in reducing oxidative stress (Møller et al., 2000, see Moreau et al., 2022a). Further analyses would be needed to explore this explanation. Granivorous birds, including grey partridges (as they were caught during the winter), cirl buntings, and common nightingales, which are omnivorous (mostly insectivorous during reproduction), tested positive. This suggests that the mode of exposure may include contaminated seeds from previous treated crops (Wintermantel et al., 2018) or organisms feeding on contaminated seeds. The higher concentrations found in Montagu's harriers, which are apex predators (mostly preying upon common voles and orthopterans), might be a result of biomagnification, i.e., the accumulation of toxic neonics along the trophic chain (Badry et al., 2020). In fact, clothianidin and thiacloprid, two compounds quantified in Montagu's harriers' blood, were found in hairs of small mammals from the area, reinforcing that view (Fritsch et al., 2022). Further investigations would be needed, and one way to do so would be using chicks' food pellets. These differences among species may also be attributable to the choices of individuals regarding their habitat and feeding resources for avoiding contaminated sources, as has been highlighted previously (McKay et al., 1999; Ruuskanen et al., 2020; Addy-Orduna et al., 2022). Another explanation relies on the detoxification processes in wild birds that are still poorly investigated and deserved further investigations (Moreau et al., 2022a). Indeed, some individuals could be more efficient than others at protecting themselves against toxic substances (Arnold et al., 2015).

Another important issue observed in our results is the high prevalence of dinotefuran and nitenpyram in grey partridges, although wildlife fauna is not supposed to be exposed to these as they are used only for pets and not farm animals, and consequently not for outdoor use in Europe, including France (Table S2). Even if grey partridges caught in this study area may originate mainly from captive breeding stocks (released for hunting purpose), the six registered veterinary medicines containing dinotefuran and nitempyram neonics that are authorized for use in France are not intended for treating any bird species or farm animal (http://www.ircp.anmv.anses.fr/index.aspx). Imidacloprid is included in 36 different speciality medications, making this neonic potentially more common even if not detectable here (see, for example, Perkins et al., 2021). Neonics are either used for topical applications on the skin (dinotefuran) or *per os* (nitenpyram) to treat flea infestations in cats and dogs typically; further, nitenpyram is expected to be eliminated within 48 h (Jeschke and Nauen, 2005; Rust, 2017). In Europe, veterinary regulatory processes are governed by the European Medicines Agency, which states that products for non-food animals are not supposed to be of major environmental concerns since these animals are treated individually with low concentrations of active neonics (CVMP/VICH, 2000). To our knowledge, these two neonics, investigated in few studies (Table S1), have only been detected in one sample of hummingbirds' feather rinsate (i.e., not within the organism; Graves et al., 2019), suggesting contact and not ingestion. In the present study, the substantial concentrations of dinotefuran and nitenpyram found in grey partridges but not in any other species may indicate possible exposure during their stay in farms before release. This implies either an illegal use of these substances in farms, as they are not included in veterinary medicines for farm animals, or an unintentional contact of partridges through multiple potential pathways. One of these could be direct contact and/or ingestion of farmer's dogs or cats' urine, or indirect through contaminated drinking water. However, neonics being presumably rapidly excreted and blood reflecting short-term exposure, individuals sampled here, if contaminated in farms before release, should not test positive. Thus, we cannot exclude in our case that partridges once in the wild could have directly ingested contaminated water with cat and/or dog urine and/or after pet baths (Teerlink et al., 2017; Diepens et al., 2023). Besides, partridges that commonly use field margins contrary to the other species, where pets walk and urinate, may have ingested these neonics while preening as the external surface of feathers are often contaminated and might even accumulate compounds (Pacyna-Kuchta, 2023). Indeed, there is growing evidence of veterinary products' transfer to the environment from dogs' hair and urine, and of secondary transfer to wildlife through nesting material or contaminated water (Diepens et al., 2023). Even though we cannot ascertain by which route the partridges were exposed to these neonics, our results indicate in line with Diepens et al. (2023), the urgent need for monitoring all pesticide or medicine compounds, irrespective of their intended use. Although treating domestic animals against ectoparasites might be of sanitary importance for public health (human and animal), these treatments may have the same adverse effects on wildlife as PPP (reviewed in Moreau et al., 2022a).

It should be noted that the interpretations given to the results provided in the present study are only assumptions and that the origin of birds' contamination remain unknown. Ongoing studies on neonics levels in soils and invertebrates of the study area since 2018 should help to provide further clarifications on birds' contamination pathways. Moreover, the method of multiresidue analysis used here could be improved to allow the detection of all neonics, including imidacloprid and acetamiprid, especially as imidacloprid was extensively applied in the crops of the study area and is still highly used in veterinary medicine.

### 4.3. Conclusions

Our results highlight several problems with the use of a class of chemicals that are among the most used worldwide. First, banning neonics for outdoor use does not prevent the exposure of wildlife fauna to them, at least a few years after ban. Although illegal use cannot be disregarded, it cannot explain on its own their ubiquity in wild birds from a wide ecological range, caught at the scale of the study site. This is an important element to consider in countries where neonics are still massively applied, considering their impact on both animals and humans (reviewed in Moreau et al., 2022a). Second, the detection of neonics used for domestic animals in wildlife fauna raises questions regarding the manner in which risk assessment for such applications is performed (Perkins et al., 2021; Diepens et al., 2023). For instance, to our knowledge, few studies have investigated the effects of dinotefuran and nitenpyram on wildlife fauna (Wang et al., 2018). Although clothianidin, dinotefuran, nitenpyram, and thiamethoxam are considered to be of low toxicity to birds, as indicated by their acute oral $LD_{50}$ (Table S2), thiacloprid, which was also found to be quite ubiquitous, is highly toxic to birds (Table S2). However, $LD_{50}$ is indicative of acute lethal toxicity under laboratory conditions for model species and is not necessarily an appropriate estimate of sublethal effects, as shown in the numerous studies that investigated the adverse effects of neonics on birds (Moreau et al., 2022a). Considering that very low residual levels of pesticides may have considerable sublethal effects on birds' reproduction (see Moreau et al., 2021, for example), the consequences of these exposures on bird populations as well as on human health (*One Health* concept) should be carefully considered.

## Ethical statement

This study was conducted conforming the French guidelines for the ethical use of animals in research (APAFIS#18557-2019010822312199v2, APAFIS#9465e201703101551625). We are grateful to the Nouvelle-Aquitaine Regional Agency of the Environment, Development and Housing for the official authorizations for the capture of passerines birds (DREAL/2019D/2323). Because Montagu's harrier is a protected raptor species, its handling was allowed and licensed by the Centre de Recherches sur la Biologie des Populations d'Oiseaux – Museum National d'Histoire Naturelle (license #1308 for Montagu's harriers).

## Credit author statement

EF, AG, AR, MM, VB, JM and KM conceived the ideas and designed the methodology; AG, JM and KM collected the data; EF, AR, MM and KM analysed the data; EF, AG, VB, JM and KM led the writing of the paper. All the authors contributed critically to the drafts and gave final approval for publication.

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## Declaration of competing interest

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

## Data availability

Data will be made available on request.

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## Appendix A. Supplementary data

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# Update

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Corrigendum

# Corrigendum to “Neonicotinoids: Still present in farmland birds despite their ban, Chemosphere, **321**, April 2023, 138091”

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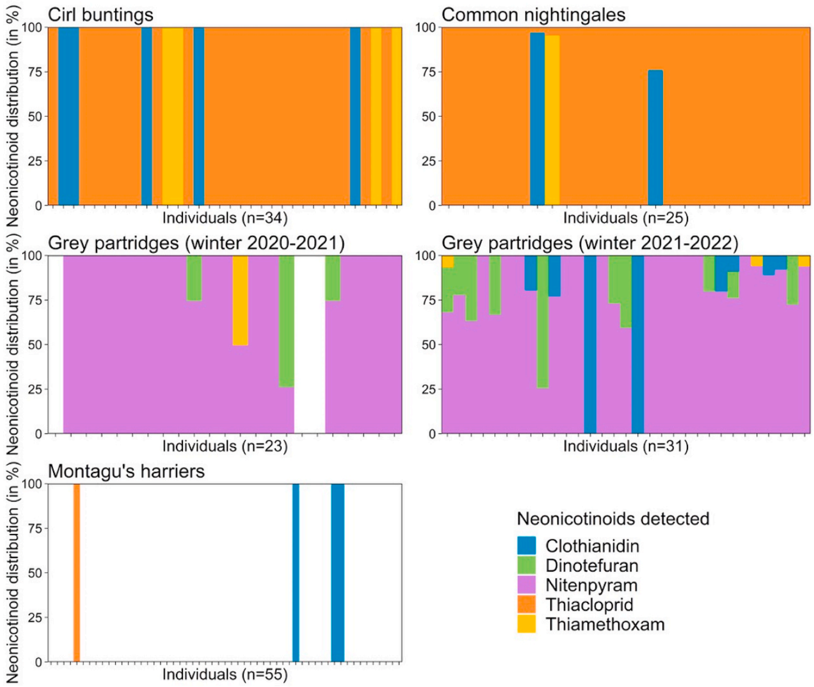
The authors regret a mistake on the **Figure 2** concerning Montagu’s harrier plot. Data used for plotting Montagu’s harrier contamination did not correspond to the data used for summary statistics of the results, due to a confusion in the data column used in R software. Consequently, the number of individuals with thiacloprid contamination appeared to be 12 in the plot while there is actually a single individual with a determined concentration of thiacloprid (given in **Table 1**).

Here is the correct **Figure 2**.

The authors would like to apologize for any inconvenience caused.



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