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Temporal and spatial trends of imidacloprid-related hazards in France

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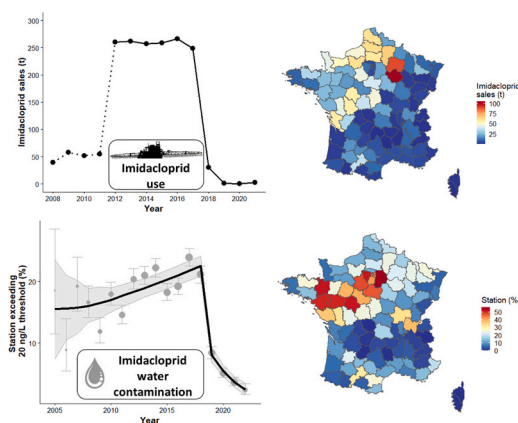
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HIGHLIGHTS

- Imidacloprid (neonicotinoid) is impactful to biodiversity but their temporal and spatial use remains little known.
- We characterise the temporal and spatial use of imidacloprid and its water contamination in France (ban since 2018).
- Imidacloprid use was higher use in northern and western France, relied on cereal and beet crops area.
- Water contamination indicated that imidacloprid has contaminated the environment and increased the risk to biodiversity.
- Results will help to identify priority areas for mitigation and restoration measures.

GRAPHICAL ABSTRACT



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ABSTRACT

Neonicotinoids are the top-selling insecticides worldwide. Because of their method of use, mainly to coat seeds, neonicotinoids have been found to widely contaminate the environment. Their high toxicity has been shown to be a major concern in terms of impact on biodiversity, and the use of these insecticides has been associated with population declines of species in different countries. Despite the widespread recognition of the risk of neonicotinoids to biodiversity, their temporal and spatial use remains poorly known in many countries. Yet this information is essential to address the potential impacts of these pesticides on biodiversity and to inform measures to establish protected areas or biodiversity restoration. The present study relied on a large publicly available dataset to characterise the temporal and spatial use in France of imidacloprid, the most widely used neonicotinoid worldwide, as well as analysed water contamination surveys between 2005 and 2022 to assess the contamination of the environment. The results show that imidacloprid was the main neonicotinoid used in France over the study period. This use was spatially structured, with higher use in northern and western France, particularly related to

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cereal and beet crops area. The water contamination survey indicated that imidacloprid has widely contaminated the environment and consequently increased the risk to biodiversity, especially in counties crossed by the Loire, Seine and Vilaine rivers. This risk increased between 2005 and 2018 due to the higher use of imidacloprid and decreased sharply after 2018 due to its ban, although it was reauthorized by derogation for sugar beet in 2021. This study is the first assessment of imidacloprid pressure on biodiversity in France and shows the spatial and temporal correlation between agricultural practices and the freshwater contamination level. These results will help to identify priority areas for mitigation and restoration measures.

1. Introduction

Since their introduction in the 1990s, neonicotinoids have become the world's best-selling insecticides (Sparks and Bryant, 2021). This success can be explained by their broad spectrum of activity (due to their action against the nicotinic acetylcholine receptors common to all insects), systemic plant protection, low cost, long-lasting effects and versatility of application (Elbert et al., 2008). Today, neonicotinoid-coated seeds and foliar applications are still among the most effective and costless options for protecting crops against plant pests and are therefore widely used as prophylactics (Schulz et al., 2021). Furthermore, neonicotinoids are effective at very low doses, with lethal doses for most target pests below 10 ng/insect (Dadé et al., 2020; Jiang et al., 2018).

While seed coating has been promoted as a low-risk practice for the environment, neonicotinoids have been shown to contaminate both terrestrial and aquatic environments, consequently affecting biodiversity (Douglas and Tooker, 2016; Hallmann et al., 2014; Li et al., 2020; Main et al., 2018; Morrissey et al., 2015; Pelosi et al., 2021; Pisa et al., 2017; Silva et al., 2019; Wagner, 2019). In fact, only 2 % to 20 % of the seed-coating neonicotinoids are actually taken up by crops, with the rest remaining in the soil or leaching onto surface water (Goulson, 2014; Sánchez-Bayo, 2014). Neonicotinoids persist in the soil (> 150-day half-life for the majority, Bonmatin et al., 2015) – they have been found in soil up to five years after being banned in several countries (Froger et al., 2023; Wintermantel et al., 2020). Furthermore, neonicotinoids are soluble in water, which facilitates their transport through and contamination of both aquatic and terrestrial ecosystems, even in remote areas where neonicotinoids are not in use (Pelosi et al., 2021; Van Dijk et al., 2013). Neonicotinoids have been found in environments where these molecules have not been applied, such as grasslands, hedgerows and organically farmed fields (Pelosi et al., 2021). Due to their widespread contamination of the environment and their efficiency in killing non-target insects at very low doses (Eisenback et al., 2010; Krupke and Long, 2015), neonicotinoids are currently identified as a major cause of insect and other invertebrate decline in various parts of the world, in both terrestrial and aquatic environments (Forister et al., 2016; Hallmann et al., 2014; Li et al., 2020; Liess et al., 2021; Pisa et al., 2017). This in turn makes them a significant factor contributing to the decline of insect-feeding species such as birds and fish (Hallmann et al., 2014; Yamamuro et al., 2019), in addition to their sub-lethal effects on many organisms (Eng et al., 2019; Main et al., 2018).

Despite the recognition of the risk of neonicotinoids to biodiversity, their temporal and spatial use remains poorly known in many countries. Among the few existent studies, Douglas and Tooker (2015) and Li et al. (2020) have described such patterns in the USA. Such information is essential to assess the potential impacts of these pesticides on biodiversity and to establish protected areas or biodiversity restoration measures. To begin to fill this gap in France, the present study used publicly available data to characterise the temporal and spatial use of neonicotinoids in the country. We focused on imidacloprid, the first neonicotinoid authorised on the market and, to date, the most widely used in different regions of the world (Simon-Delso et al., 2015). Imidacloprid residues are the most common neonicotinoid residues found in arable soils in France (Froger et al., 2023; Pelosi et al., 2021) and in Europe (Silva et al., 2019). It is also the main insecticide found in

earthworms (Pelosi et al., 2021) and is frequently detected in rodents (Fritsch et al., 2022) as well as in honeybees (Daniele et al., 2018). Imidacloprid has been marketed in France since 1991; it was since then regulatorily banned in the EU for flowering crops in 2013, and for all crops in 2018 (https://eur-lex.europa.eu/eli/reg_impl/2018/783/oj). However, annual exemptions were granted for sugar beet in 2021 and 2022. Eventually, all neonicotinoids have been completely banned for all crops in European Union in 2023, but this decision is still challenged by several agricultural unions.

The first step of the present study was to characterise the spatial and temporal trends of imidacloprid use in France. To this end, we used pesticide sales information, and to identify the spatial pattern we also investigated the relationship between crop distribution and imidacloprid use, as the latter is highly dependent on crop type (Douglas and Tooker, 2015). In a second step, we investigated the relationship between imidacloprid use and environmental contamination by this molecule using pesticide analysis of water samples from French rivers. Water contamination by pesticides has been shown to reflect pesticide use in surrounding agricultural landscapes (Gauroy and Carlier, 2011; Hunt et al., 2006) and has been linked to biodiversity decline in several countries (Hallmann et al., 2014; Thunissen et al., 2022).

2. Materials and methods

2.1. Pesticide sales/purchases

To assess the temporal and spatial use of imidacloprid in France, we used the 'French national database of sales of plant protection products by authorised distributors' (NDS-D, <https://ventes-produits-phytopharmaceutiques.eaufrance.fr/>). This national database collected annual sales, per French county, declared by distributors from 2008 to 2021 (last year of data availability) for all types of pesticides (insecticides, herbicides, etc.) and the application methods (seed coating or spraying). The counties represent 96 administrative divisions in continental France and overseas (see Fig. 1 for their location). The reporting of sales of seed treatment products has only been required by French law since 2012. Consequently, data on pesticide sales, including sales of imidacloprid before 2012, is incomplete. Sales of pesticides were reported as quantities of active ingredient or of commercial formulations. As bill of sale and actual use might occur in different locations, we used a second database that reported the postal code of the purchaser. This data was available from 2013 to 2021 (<https://ventes-produits-phytopharmaceutiques.eaufrance.fr/>). We extracted sales for all neonicotinoids (NNI).

2.2. Authorisation for use and type of land use

To identify the crop types for which imidacloprid was authorised between 2008 and 2022 as well as the method of application and the date of withdrawal from the market, we used the 'Ephy database' (<http://ephy.anses.fr/>) provided by the French National Food Safety Agency (ANSES). Table S1 shows the commercial pesticide products containing imidacloprid authorised in France with their registration and withdrawal dates, as well as the crops targeted and the method of application.

A second database was extracted from 'Agreste' (<https://agreste.agriculture.gouv.fr/>), the French agricultural statistics database. This

database provides agricultural land use for crops or crop types belonging to different agricultural sectors: arable crops, vegetables, arboriculture and viticulture, as well as their yields at county level from 2000 to 2020. In this analysis, we did not consider imidacloprid used in forestry because of its reduced usage in this sector (see Table S1 for products authorised in forestry, and Fig. S3 for a description of their use over time).

2.3. Surface water contamination

Data on surface water contamination by imidacloprid was extracted from the French national database 'Naiade' (<https://naiades.eaufrance.fr/bienvenue-naiades>). Imidacloprid measurements were performed at 8001 stations from 2005 to 2022 (with an average of 2363 stations per year; min–max = 607–3511). Sampling stations were mainly located on watercourses (96.7 %, rivers, streams, ...) and water bodies (3.3 %, reservoirs, lakes, ponds, etc.). On average, sampling was repeated for a period of 5.3 years per station (min–max = 1–18 years). A total of 300,081 water analyses were performed, with an average of 7.1 samples per year and station (min–max = 1–79). Imidacloprid concentrations reported in water samples were below the limit of detection (LOD) in 1.2 % of samples, between the LOD and the limit of quantification (LOQ) in 90.3 % of samples, and above the LOQ in 8.5 % of samples. However, LOD and LOQ limits were not homogeneous between stations both because water samples were analysed by different laboratories and because analytical procedures have improved since 2005. The LOD varied between 0.3 and 100 ng/L and the LOQ varied between 1 and 250 ng/L. The number of stations sampled per year is shown in Fig. S1.A and their spatial location in Fig. S1.B, as well as the number of counties with the presence of water sampling according to year in Fig. S1.C, and the number of years of sampling per county in Fig. S1.D. We also extracted water sampling data for other neonicotinoids to ensure that imidacloprid was the main NNI contaminating surface water in France.

2.4. Evaluation of threat to biodiversity

We then used this water contamination data to assess the threat to biodiversity. As most of the measurements were below the LOD or LOQ (91.6 % of the dataset), we could not correctly estimate the imidacloprid concentration in surface water (Tekindal et al., 2017). To circumvent this problem, we drew on previous studies conducted under natural conditions that identified a threshold imidacloprid concentration in water above which imidacloprid had adverse effects on biodiversity. We chose a threshold of 20 ng/L, as this concentration in surface water has

been found to be negatively associated with bird abundance (>20 ng/L; Hallmann et al., 2014), with aquatic taxa abundance and richness (> 17 ng/L; Schmidt et al., 2022) and with macroinvertebrate abundance (>13 ng/L; Van Dijk et al., 2013). Some analyses reported imidacloprid concentration with a LOD or LOQ > 20 ng/L, which made it impossible to determine if imidacloprid concentration was superior or inferior to 20 ng/L. For example, if the LOQ in the analysis was 30 ng/L, we could not determine if the sample was between 20 and 30 ng/L or lower than 20 ng/L. For this reason, we removed all the water samples for which the analytical procedure granted a LOD/LOQ >20 ng/L. Eventually, 269,106 water samples were retained for further analysis (32,949 were removed, i.e. 11 % of the original dataset).

2.5. Stream map, water flow, precipitation and soil type

Precipitation and stream width are known to modify pesticide concentration in surface waters (Halbach et al., 2021; Vormeier et al., 2023). To account for this effect, precipitation datasets were retrieved from the European Climate Assessment & Dataset (<https://www.ecad.eu/>), which provides daily precipitation and temperature records with a resolution of 25 × 25 km at the European scale. We approximated the width of streams by the flow records, using the HydroRIVERS data-stream (Lehner and Grill, 2013), which provides a global stream map as well as an estimate of their stream flow. Water diffusion in the environment also depends on the soil type, the characteristics of which were extracted from GIS Sol (<https://www.gissol.fr/>), which provides soil type maps at the national scale with a resolution of 2500 × 2500 m across France. Soils were classified into six types: alluvial, loamy, rocky, calcareous, sandstone and sandy (Baize and Girard, 2009).

2.6. Analysis

2.6.1. Characterisation of the temporal and spatial use of imidacloprid in France

We first characterised the temporal and spatial use of imidacloprid. For the temporal pattern, sales of imidacloprid per year were calculated and compared to other NNI sales, indicating that imidacloprid was the main NNI used in France. Then a graph was created to compare NNI sales for different years. For the spatial pattern, imidacloprid sales between 2008 and 2021 were calculated per county and a density map was created showing the location of imidacloprid sales.

2.6.2. Relationship between imidacloprid sales and land use

We identified the spatial use of imidacloprid sales by linking land use

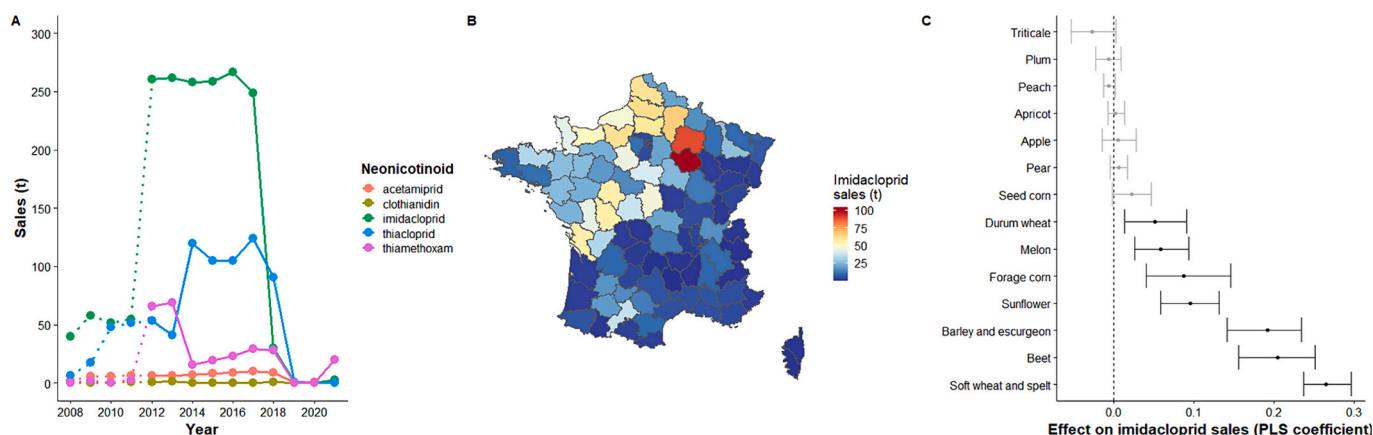


Fig. 1. (A) Temporal variation of neonicotinoid sales in France between 2008 and 2021. Green curve indicates imidacloprid sales, orange = acetamiprid, yellow = clothianidin, blue = thiacloprid, pink = thiamethoxam. Incomplete data on seed-coating sales before 2012 is represented by dotted lines. (B) Spatial variation of imidacloprid sales in France. Imidacloprid sales between 2008 and 2022 were calculated for each county. Gradient from blue to red indicates imidacloprid sales from lowest to highest quantity. (C) Relationship between land use and imidacloprid sales between 2008 and 2017. Relative effects of main crop type with imidacloprid authorisation on imidacloprid sales were obtained by partial least squares regression (PLS). Bias-corrected and accelerated confidence intervals are shown.

and imidacloprid sales using partial least squares (PLS) analyses. PLS is a multivariate analysis that can handle multiple explanatory variables and collinearity (Bertrand et al., 2016; Carrascal et al., 2009). The relevant scale was considered to be the county. We analysed imidacloprid sales between 2008 and 2017 per county and per year and used these values (imidacloprid sales per county) as a dependent variable in the PLS. We restricted this analysis to data between 2008 and 2017, as imidacloprid was banned for most crops in 2018 (see Table S1). The areas of crops with imidacloprid-use authorisation gathered from the Ephy database (see Table S1) were used as explanatory variables. To avoid over-parameterisation, we selected the ten largest crops in terms of surface area for each main agricultural sector (arable crops, vegetable crops, tree crops), retaining 14 crops with imidacloprid authorisation: common wheat and spelt, barley, durum wheat, triticale, sunflower, beet, seed corn, forage corn, melon, pear, apple, apricot, peach and plum. These 14 crops accounted for 77.5 % of the area under cultivation in France between 2008 and 2017. As sales data of imidacloprid before 2012 was incomplete, a robustness analysis was performed by repeating the same analysis using only data from 2012 to 2017.

2.6.3. Characterisation of the temporal and spatial threat of imidacloprid for biodiversity in France

In a second step, we characterised the temporal and spatial variation of imidacloprid risk to biodiversity using water contamination sampling results. We first described the evolution of imidacloprid risk to biodiversity between 2005 and 2022 using a binomial regression with multiple change points analysis (Lindeløv, 2020). This method was selected because of possible break points in the temporal sequence of water contamination. First, the highest imidacloprid concentration values at each station per year were extracted, to see if any of the monitored rivers were exposed to levels potentially harmful to biodiversity. Then, we reported the number of stations per county with an imidacloprid level above the threshold of 20 ng/L as the number of stations per county, i.e. the percentage of stations per county exceeding the 20 ng/L threshold. In this analysis, the percentage of stations per county exceeding the 20 ng/L threshold was used as the dependent variable. Year was used as an explanatory variable.

Then we characterised the spatial variation of imidacloprid risk by estimating the proportion of stations exceeding the 20 ng/L imidacloprid threshold for each county between 2005 and 2022. An additional analysis was performed to identify the rivers with the highest neonicotinoid contamination levels by using a generalised linear mixed model (GLMM). Rivers were defined as all major streams and their influents flowing into oceans and seas. The water samples came from 666 different rivers. We focused on the ten largest rivers in France (representing 77.4 % of the total length of the national watercourse); other rivers were grouped in a category named 'other rivers'. The probability of imidacloprid levels exceeding the 20 ng/L threshold was used as the dependent variable. The river was used as a categorical variable and the year as a random variable to account for the measurement (repetitions in time). A quasi-binomial distribution was used in this model due to the overdispersion of the percentage data.

2.6.4. Relationship between biodiversity risk and imidacloprid use

Finally, we linked the risk to biodiversity of the use of imidacloprid using generalised linear models (GLMs). The percentage of stations per county exceeding the threshold was estimated each year at county scale and used as a dependent variable. Imidacloprid sales per county corresponding to the year of water sampling was used as an explanatory variable. In this model, soil type, annual mean precipitation, and mean river flow per county and their interaction with imidacloprid sales were also added. For soil type, only the dominant soil type within a county was considered. A dominant soil type represents on average 36.4 % of county area (min–max = 14.4 %–91.4 %). A sensitivity analysis was performed using only counties with a dominant soil >25 % of the county area. This did not modify the results (see Fig. S8).

As the cumulative use of imidacloprid over the years can lead to contamination (Wintermantel et al., 2020), we also investigated the effect of cumulative sales over the two, three, four and five previous years on the probability of the station exceeding the 20 ng/L imidacloprid threshold in four other GLMs. These models were compared on the basis of R-squared (R^2). The Akaike information criterion (AIC) was not used for this comparison because it is not available for the 'quasi'-family. The models were compared using the same dataset between 2013 and 2021 to have the same number of observations per model. Soil type, annual mean precipitation, and mean river flow per county and their interaction with imidacloprid sales were also added in these models as explanatory variables.

All analyses were performed using R.4.3.0. The 'plsRglm' package was used for PLS analysis, the package 'stats' for GLMs, the package 'glmmPQL' for GLMMs and the package 'mcp' for multiple change points regression analysis (Lindeløv, 2020). Climatic variables were extracted with the 'climateExtract' package. The counties Paris, Seine-Saint-Denis, Haut-De-Seine and Val-de-Marne were grouped together for all analyses (sales, water sampling) because of their small surface area. For GLMs and GLMMs, the distribution of model residuals was visually checked for normality and homoscedasticity. Imidacloprid sales data was square-root transformed to reduce the skewness of the distribution and improve the normality and homoscedasticity of the model residuals. Collinearity between explanatory variables was checked using the variance inflation factor (VIF) and none was found ($VIF < 1.7$).

3. Results

3.1. Temporal and spatial use of imidacloprid in France

Imidacloprid was indeed the neonicotinoid with the largest sales in France until 2018, when it was banned (Fig. 1.A). Three distinct phases of sales were observed: between 2008 and 2011, with a mean level of active ingredients of 51.0 t per year and per county (+/-SD = 8 t), followed by an increase in sales between 2012 and 2017 (mean sales = 258.9 t per year, SD = 5.9 t), and then a fall in sales after 2017 (mean = 8.6 t, SD = 14.4 t, Fig. 1.A).

Sales were unevenly distributed across France, with higher amounts in the north and northwestern part of the country (Fig. 1.B). This was mainly explained by areas cultivated for cereal crops, mainly soft wheat and barley, followed by beetroot, and to a lesser extent by corn, sunflower and melon, but not tree crops (apple, peach: Fig. 1.C; see also Fig. S2 for the location of beet, soft wheat and barley crops in France). Imidacloprid sales were mainly represented by the commercial products Gaucho 350 and Gaucho DUO Fs, which were used on cereal crops, and to a lesser extent Imprimo, which was mainly used on sugar beet (Fig. S3, Table S1). When the analysis was restricted to sales between 2012 and 2017, we found the same results (see Fig. S4). Sales per county during the 2008–2011 period were strongly correlated to the sales per county between 2012 and 2017 (Pearson correlation test, $\rho = 0.64$, $df = 91$, $p\text{-val} < 0.001$). Imidacloprid sales were strongly correlated to the purchaser location (Pearson correlation test, $\rho = 0.91$, $df = 835$, $p\text{-value} < 0.001$), suggesting that most of the products sold were applied close to the purchase location.

3.2. Temporal and spatial contamination of surface water

Imidacloprid was the main NNI found in surface water in France (Fig. S5). On average, 16.2 % of stations showed an exceedance of the 20 ng/L threshold for imidacloprid at least once during the year in water samples between 2005 and 2021. Contamination of surface water by imidacloprid was stable between 2005 and 2011 and increased from 15 % in 2012 to 22 % in 2018, and then dropped after 2018 to reach 2.4 % in 2022 (Fig. 1.A).

Contamination of surface water also significantly varied in space (Fig. 2.B). Higher water contamination was found in counties through

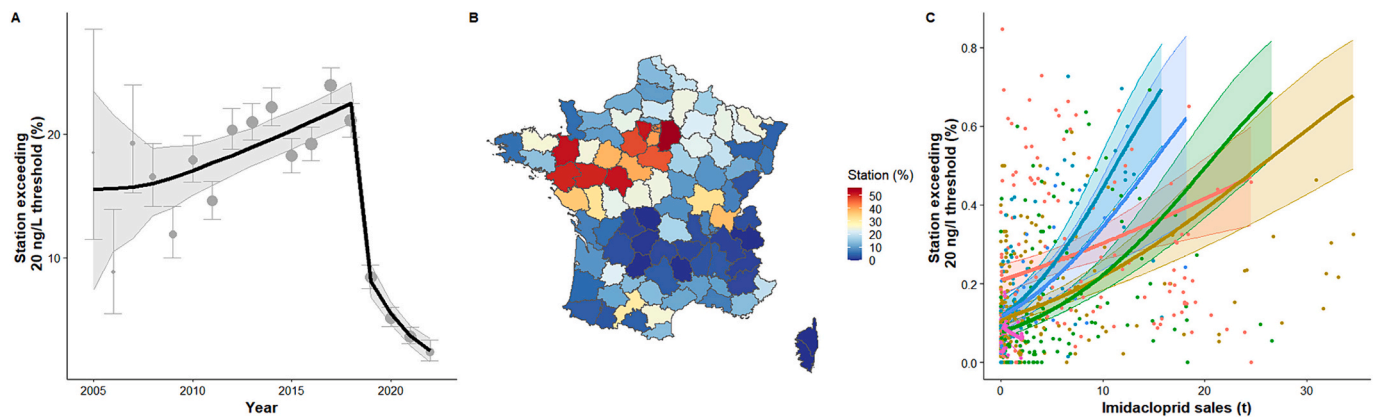


Fig. 2. (A) Yearly % of stations exceeding the 20 ng/L imidacloprid threshold. Black line represents relationship obtained by multiple change points binomial regression analysis. Grey area represents interval of prediction. (B) Map of % of average stations exceeding 20 ng/L threshold of imidacloprid during the year between 2005 and 2018. Colour gradient from blue to red indicates % stations exceeding 20 ng/L threshold from lowest % to highest %. (C) Relationship between % stations exceeding 20 ng/L threshold of imidacloprid and imidacloprid sales per county according to dominant soil type of county. Imidacloprid sales are cumulative sales over two years as this better explains % stations exceeding 20 ng/L threshold (see ‘Methods’ and ‘Results’). Coloured lines represent relationship predicted by generalised linear models between imidacloprid sales and % stations exceeding 20 ng/L threshold: red = loam soil type; green-blue = alluvial; blue = sandstone; green = rock; yellow = calcareous, pink = sand. Shaded areas represent 95 % confidence intervals of prediction.

which the Vilaine, Loire and Seine rivers flow (Fig. 2.A, see Fig. S6.A for location of main rivers in France). Additional analysis confirmed that the rivers Vilaine, Loire and Seine and their tributaries had higher water contamination levels than other rivers (Fig. S6.B). The level of water contamination was significantly and positively correlated with imidacloprid sales per county, and especially with the cumulative sales of imidacloprid over the previous two years (GLMM, $\chi^2 = 176.6$, $df = 1$, $p < 0.001$). The model with two years of cumulative imidacloprid sales and the model with three years of cumulative sales had similar results for the proportion of stations exceeding 20 ng/L ($R^2 = 30.7\%$ and 31.2% respectively for two and three years of cumulative sales), explaining this better than the models with one year ($R^2 = 26.1\%$), four years ($R^2 = 29.4\%$) or five years ($R^2 = 26.4\%$) of cumulative sales. Interestingly, the relationship between two years of imidacloprid sales and the proportion of stations exceeding 20 ng/L depended on soil type (interaction between two years of imidacloprid sales and soil type, $\chi^2 = 39.1$, $df = 5$, $p < 0.001$), precipitation ($\chi^2 = 4.0$, $df = 1$, $p = 0.046$) and average river flow rate ($\chi^2 = 9.4$, $df = 1$, $p = 0.002$). Higher positive correlation was found between imidacloprid sales and proportion of stations contaminated in counties with alluvial, sandstone and rock soil-type dominance than in counties with other soil types (Fig. 2.C). Cumulative precipitation also increased the percentage of contaminated stations per county (Fig. S7.A). Counties with high-flow rivers showed greater water contamination (Fig. S7-B).

4. Discussion

Our analysis showed that the use of imidacloprid was spatially structured, with the highest use in northern and western France, where it was mainly used on cereal and sugar beet crops. The water pollution analysis showed that imidacloprid use resulted in wide environmental contamination, increasing the risk to biodiversity, especially in the counties crossed by the Loire, Seine and Vilaine rivers. This risk increased between 2005 and 2018 due to increased use of imidacloprid and decreased after 2018 when the compound was banned.

During the study period, imidacloprid was the most widely used neonicotinoid in France and the most commonly found in surface water, as has been observed in other regions of the world (Simon-Delso et al., 2015). Imidacloprid sales in France were comparable to those reported in the Netherlands and USA relatively to agricultural area: around 250 t of imidacloprid sold in 2014 in France for 29 million ha of agricultural area (i.e. 8.3 t/millions ha), around 3400 t of neonicotinoids sold in

2014 in the USA for 408 million ha of agricultural area (i.e. 8.6 t/millions ha, Li et al., 2020), and around 6.3 t of imidacloprid sold in 2006 in the Netherlands for 1.8 million ha of agricultural area (i.e. 3.5 t/millions ha) (Hallmann et al., 2014).

In France, the use of imidacloprid had a strong spatial pattern linked to the spatial distribution of soft wheat, barley and beet crops. Neonicotinoids in wheat and beet were mainly used to control aphids and associated virus diseases such as barley yellow dwarf in cereals (McKirdy, 1996). These results differ from the only previous study that has investigated imidacloprid use per crop type, which showed a stronger link to maize and soybean crop areas in the USA (Douglas and Tooker, 2015). This difference can be explained by the different crop dominance in the two countries (FAOSTAT, 2014). The relationship between the quantity of imidacloprid sold per hectare and the area of crops associated with its use indicated an applied quantity of 28.7 g per ha (min–max = 27.2–29.7 g/ha) between 2012 and 2017. This ratio is lower than the maximum dosage recommended by the French Agency for Food, Environmental and Occupational Health and Safety (<https://ephy.anses.fr/>). For example, for Gaucho 350, the dose recommended by hectare is between 84 and 140 g/ha for a sown density of 120–200 kg of wheat per ha. This suggests that imidacloprid was applied only on a limited percentage of crop fields and may vary between crop types. For example, in the USA, Douglas and Tooker (2015) estimated that 87 % to 100 % of maize fields and 34 % to 44 % of soybean fields used neonicotinoid seed coating.

The increase in the use of imidacloprid between 2008 and 2018 in France coincides with an increase in its use in other countries, such as the USA, the UK and Sweden (Goulson, 2013; Li et al., 2020; Simon-Delso et al., 2015). Our analysis found that this increase resulted in higher water contamination, particularly in the Loire, Seine and Vilaine rivers, and highly depended on imidacloprid sales in the counties crossed by these rivers. However, this relationship was modulated by the type of soil in the county as well as the annual precipitation and river flow. Water pollution by pesticides is known to depend on soil type, river flow and precipitation (Gramlich et al., 2018). Imidacloprid occurrence and concentration in surface waters has been found to increase in wet weather (Batikian et al., 2019) due to its high water solubility (Bonmatin et al., 2015). Soil with a high percentage of sand such as alluvial or sandstone soils are known to increase the mobility of neonicotinoids in the environment (Bonmatin et al., 2015), which was confirmed by our results, which showed higher contamination in counties with alluvial and sandstone soil. We found that imidacloprid water contamination

also increased in large streams. Previous studies have shown opposite results, with higher pesticide concentration in small streams (Halbach et al., 2021). It is possible that the indicator used to estimate the width of the river in our study, i.e. its flow rate, reflected the speed of diffusion of the water rather than its quantity, which could explain this difference.

It should be noted that although imidacloprid contamination of water decreased after its ban in 2018, its presence was still detectable in rivers after four years (on average, 4.9 % of samples exceeded 20 ng/L between 2019 and 2022), confirming its long persistence in the environment (Froger et al., 2023; Wintermantel et al., 2020). The re-authorization by derogation of imidacloprid for sugar beet does not appear to have led to an increase in water contamination (3.0 % in 2021–2022 during derogation period in sugar beet and 6.8 % in average between 2019 and 2020). This can be explained by the limited spatial distribution of sugar beet in France (see Fig. S2.A for spatial distribution of sugar beet in France) and by lower use compared to the period with authorization (258 t in average between 2012 and 2017 versus 2.5 t in 2021).

The large body of existing literature on the negative impact of imidacloprid – as with other neonicotinoids – leaves little doubt on the potential relationship between its use and biodiversity decline (Mamy et al., 2023; Pisa et al., 2017). This study is the first in France to provide a temporal and spatial description of imidacloprid use, allowing the identification of the potential risk to terrestrial and aquatic biodiversity across the country. However, our study only takes imidacloprid into account, and therefore only estimates a minimal risk to biodiversity, as the toxicity of imidacloprid is added to or synergized with that of other pesticides (Zhang, 2022). Over the study period, the use of imidacloprid in France was comparable in terms of quantity to that applied in countries with similar agricultural practices, and where imidacloprid has been shown to be linked to the decline of several taxa (Hallmann et al., 2014; Li et al., 2020). Consequently, the results of this study allow the first assessment and mapping of the threat of imidacloprid to biodiversity in France, showing the spatial correlation between freshwater contamination level and agricultural practices. Considering the transformative changes to intensive agriculture necessary to cope with ongoing biodiversity loss (Pe'er et al., 2022; Rigal et al., 2023), risk maps could be a useful tool to identify priority areas for mitigation and restoration measures. They can provide results-based evidence of the often overlooked environmental costs of pesticide use.

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CRediT authorship contribution statement

Thomas Perrot: Writing – original draft, Methodology, Formal analysis, Conceptualization. **Jean-Marc Bonmatin:** Writing – original draft, Validation, Methodology, Conceptualization. **Hervé Jactel:** Writing – original draft, Methodology, Conceptualization. **Christophe Leboulanger:** Writing – original draft, Methodology, Conceptualization. **Robin Goffaux:** Writing – original draft, Supervision, Project administration, Funding acquisition. **Sabrina Gaba:** Writing – original draft, Methodology, Conceptualization.

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.

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.173950>.

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