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Pesticides in France: ten years of combined exposure to active substances in land, air and surface water

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Spatial and temporal fine-scale data of exposure to pesticide in the environment are of great need to environmental and health research. The omnipresence of pesticides in the environment is attested to by numerous analyses, but the availability of data remains inadequate, preventing temporal analyses on large spatial scales such as the national level, where agricultural policies are implemented or adopted. We have compiled data on the purchase of more than a hundred active substances with measurements of pollution of these substances in the air and surface water to propose a map of exposure to the most dangerous active substances between 2013 and 2022 in metropolitan France. We provide a technical validation of the exposure index using a dataset constructed from infield surveys. The combined exposure index is designed to be updated annually, and we anticipate that this dataset will provide first-rate information for conservation and health research.

Background & Summary

Industrial agriculture is one of the main drivers of biodiversity decline[1] and has significant impacts on human health[2,3]. Industrial agriculture is defined by increased use of synthetic inputs (pesticides and fertilisers), extensive mechanisation, fewer farmers[4], and landscape simplification[5]. Some of these characteristics can be tracked directly using agricultural statistics (e.g. size and number of farms, productivity per worker) or spatial data analysis (e.g. landscape simplification, disappearance of hedgerows). Conversely, data on the use of pesticides and the environmental pollution they cause remains scarce[6], in particular due to statistical secret and private data protection[7]. Yet, pesticides are designed to reduce certain species or groups of species, and the link between some classes of substances and the decline of certain species, even those not targeted initially by these products, as well as their consequences on human health, is increasingly well documented[3,8,9]. The availability of interpreted data from open databases and the mapping in time and space of exposure to these substances is therefore critical for conservation and health research.

The term “pesticide” actually refers to commercial formulations containing active substances (AS) used to control organisms considered as pest. AS correspond to the compounds whose toxicity is directly assessed. The production of reliable data on pesticide exposure is therefore complexified by the existence of two possible proxies: the quantity of the commercial product and the quantity of active substance actually applied. This distinction is all the more important as recent active substances are effective at lower quantities, which may lead to a reduction in the quantities used but not in the pollution of the environment and hazards to health. As a result, research has proposed a number of indices for comparing pesticide use between countries or over time, without identifying an optimal indicator[10]. Using the quantities of commercial products, the treatment frequency index (TFI)[11] provides an initial estimate of pesticide pressure on the environment by standardising products by the registered dose per hectare, using data from farmers’ declarations[12]. It is therefore an index that provides direct information on agricultural practices, but it does not explicitly account for variations in toxicity among active substances. The pesticide load index, which combines negative impacts on human health and ecosystems, and

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accumulation in the environment[13], and the total applied toxicity index, which focuses on toxicity to different taxa[14], have been proposed to overcome these limitations[10].

However, commercial product formulations are often not accessible and what ends up being measured in the environment are AS. This is why other indices have been proposed based directly on AS, in particular the quantity of AS (which is a limited index due to the different efficacy of each substance), the number of dose units (NODU) which standardises the quantity of each AS by a dose unit reflecting the different efficacy of each AS, and finally the treatment intensity index (TII)[15] which standardises the NODU per hectare and is one of the most widely used indicator based on AS. The advantage of working on AS is that it is possible to combine declared use or purchase data with the pollution background measured in other compartments such as the atmosphere and surface water, which also affects ecosystems beyond the area of pesticide application[16]. It also makes it possible to account for the levels of toxicity of each AS that differ by order of magnitudes.

In this paper, we propose a standardised dataset of the exposure to the most environmentally harmful AS (i.e. toxic or carcinogenic, mutagenic, reprotoxic) in metropolitan France in local use, air and surface water to provide a proxy of the global pesticide exposure in ecosystem over 10 years (2013–2022). To the best of our knowledge, this is the first dataset available in Europe on a national scale with a spatial accuracy of below 10 km and temporal coverage, offering a significant advance in the dynamics of pesticide exposure assessment[17].

Methods

Pesticide use. We use sold pesticide per postal code retrieve from the national bank of plant protection products[18] which is a robust proxy of pesticide use[19] in particular in France[7]. The amount of active substance (AS, in kg) is provided for each postal code between 2013 and 2022. We focus on the 183 active substances mandatory to report from 2013 to 2022 among the 335 AS reported. The obligation to report an AS are retrieved from the ministerial decrees available at https://www.legifrance.gouv.fr/.

Several spatialisation methods have been proposed for these data[7,20] to account in particular the fact that farm siege can be in a different postal code than the treated fields. However, only the simple method attributing pesticide quantities to the postal code where it is reported allows to propose an open dataset. Moreover, comparison between models and field use have shown the reliability of this simple model[7]. A way to improve the accuracy of this spatialisation is to disaggregate the pesticide quantity sold at a postal code pc to the municipalities using the share of the total cultivated area (TCA) that the municipality represent in the postal code.

We use the TCA by municipality obtained from the national agricultural census via the national database on territories[21] to calculate the sold quantity (SQ) of each AS at the municipality level. We then account for the difference in AS effectiveness to standardise the SQ of each AS by using the standard dose SD. To obtain the SD of each AS, we use the registered dose by AS provided for each commercial product and each usage[22] and we select the registered dose of the AS for the commercial product with only this substance as SD for this substance. In our case, we cannot distinguish by usage as the information on substance apply to crops is not available, we therefore use the minimal possible standard dose for each substance[15]. To provide a comparable unit of pesticide use across municipalities, we finally compute an extension of the treatment intensity index ($eTII$) which account for the different effectiveness of AS and the area treated[15]. In the treatment intensity index, the SD of each AS is used to standardised the quantity of the substance, the sum of this ratio for the N active substances being divided by TCA, originally at the national level. With $eTII$, we calculate the quantity of AS sold, standardised by their effectiveness and the area a of the municipality m, per year y between 2013 and 2022 for the 183 active substances as follows (Eq. 1):

$$eTII_{m,y} = \frac{TCA_m}{TCA_{pc}} \frac{\sum_{n=1}^{N} \frac{SQ_{n,y}}{SD_n}}{a_m} \tag{1}$$

with TCA the total cultivated area of municipality m and postal code pc, SQ_n the sold quantity of AS n, SD_n the standard dose of AS n and a_m the area of municipality m.

$eTII$ also corresponds to the original TII at the postal code level, weighted by the share of agricultural land in the municipality.

Pesticide air pollution. As a proxy of pesticide air pollution, we use the Phytatmo database which compile the measurements of 321 gaseous and particular active substances in the air in 205 stations between 2002 and 2022[23]. The concentration of each substance in each site is provided in ng.m^{-3}. In the Phytatmo database, the concentration is set to zero if the measurement is below the detection threshold, and half of the quantification threshold if the measurement is between the detection and the quantification threshold. Stations are heterogeneously distributed over metropolitan France (Fig. S1). We therefore use a kriging interpolation to cover the metropolitan territory with a external drift that relates the presence of AS in the air with the local use of AS. To do so, for each year, among the 341 monitored AS, we focus on the 101 AS measured in at least 10 stations and also available in the 183 AS of the sold pesticide database. We produce an exponential variogram model to implement the regression kriging with the quantity of the AS sold at the municipality level as an explanatory variable to optimise the kriging weights. The predictions are provided in ng.m^{-3} on a 1 × 1 km grid of metropolitan France between 2013 and 2022.

Pesticide water pollution. As a proxy of pesticide water pollution, we use the NAIADES database of surface water quality[24] between 2013 and 2022. Geographical data on the 35,803 measurement stations in metropolitan France are provided by the SANDRE[25]. Among the 355 monitored AS, we focused on the 156 AS also present in 183 AS from the sold pesticide databases. Conversely to the air measurements, an interpreted concentration is not

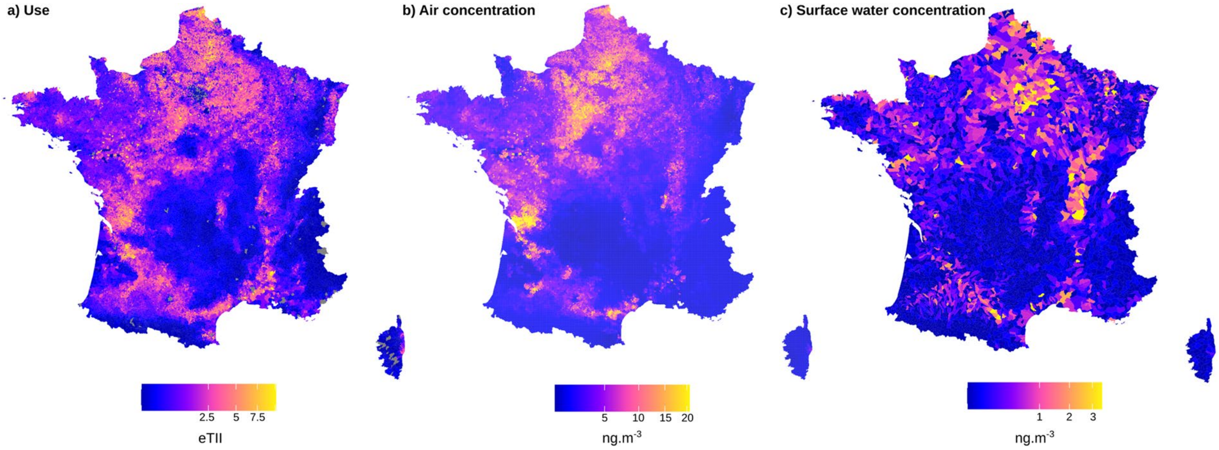

Fig. 1 Average maps for pesticide use with toxic or carcinogenic, mutagenic, reprotoxic effects in terms of active substance use (extended Treatment Intensity Index from 175 AS), active substance concentration in the air (in ng.m^{-3} from 99 AS) and in water (in ng.m^{-3} from 145 AS).

initially available and we therefore construct a conservative concentration estimate. To do so, we select only measurements classified as correct by the provider and made in the water fraction. We set the concentration to zero when the measurement is below the detection threshold. For each AS, we retrieve the lowest detection threshold across all stations and we remove the trace measurements (i.e. between detection and quantification thresholds) for which the detection threshold is higher than the lowest. This prevents from biases due to change in detectability across years or stations (as the variability in data producers is high). The concentration for the remaining trace measurements is set to the detection threshold (i.e. the minimal possible concentration). Measurements for which the concentration is higher than the quantification threshold and smaller than the saturation threshold are also kept. This results in a measurement dataset of 9,374 station (Fig. S1). We then average the concentration to each AS by catchment area[26] and each year.

The geographical datasets produced for each AS and each year in use, air and water correspond to the first product of the database presented in this paper.

Pesticide exposure. In order to provide an assessment to pesticide exposure for ecosystems and populations, we specifically focus on the AS know to be the most harmful to biodiversity and health, and therefore classified as toxic under the diffuse pollution charge. This refers to AS classified between 2008 and 2018 as toxic, highly toxic, carcinogenic, mutagenic, reprotoxic and since 2019 as carcinogenic, mutagenic, reprotoxic, with acute toxicity to health and target organs, with acute or chronic toxicity for aquatic environment[18]. This represents 175 AS for pesticide use, 99 for air pollution and 145 for water pollution (Table S1). We produce a map and a geographical dataset of use by aggregating the eTII as well as a map and a geographical dataset of air pollution and water pollution by aggregating the concentration of the CMR AS for each year between 2013 and 2022. These 30 (3 types × 10 years) aggregated maps and datasets correspond to the second product of the database presented in this paper.

We then produce three average maps (with their geographical datasets) for use, air pollution and water pollution. We use them to provide a combined exposure map to CMR AS. To do so, we first standardise the values of each average maps between 0 and 1 using the double of the maximal observed value. This way, if new maps show values higher than the current maximum in the future, the same scale can still be produce and comparison with current map will still be relevant. We then compiled those three standardised average maps to produce the combined exposure maps (following the method of the naturalness index, another environmental combined index[27]). These three average maps (Fig. 1) and the combined map (Fig. 2) correspond to the third and final product of the database presented in this paper.

Data Records

The dataset is available on Dryad[28]. It is divided in three sub dataset: (1) "Active substances in use, quantity, air and water", (2) "Yearly exposure to active substance in use, air and water", (3) "Combined exposure to active substance in use, air and water".

The "Active substances in use, quantity, air and water" is composed of four gpkg files built on the same structure (Table 1) and the "Active substance to report" csv file providing the Chemical Abstracts Service (CAS) number of each active substance mandatory to report between 2013 and 2022. The "Yearly exposure to active substance in use, air and water" and "Combined exposure to active substance in use, air and water" dataset are composed of one gpkg file each (Table 2).

Technical Validation

We validate the exposure map with an independent dataset providing the treatment frequency index (TFI) by municipality produced by Solagro[29,30]. This dataset is constructed from in-field surveys on agricultural practices and provide the average TFI with and without biocontrol for the 34,817 metropolitan municipalities in 2020 and 2021 (Fig. S2). As the exposure map is build from CMR AS, we use TFI without biocontrol. Spearman

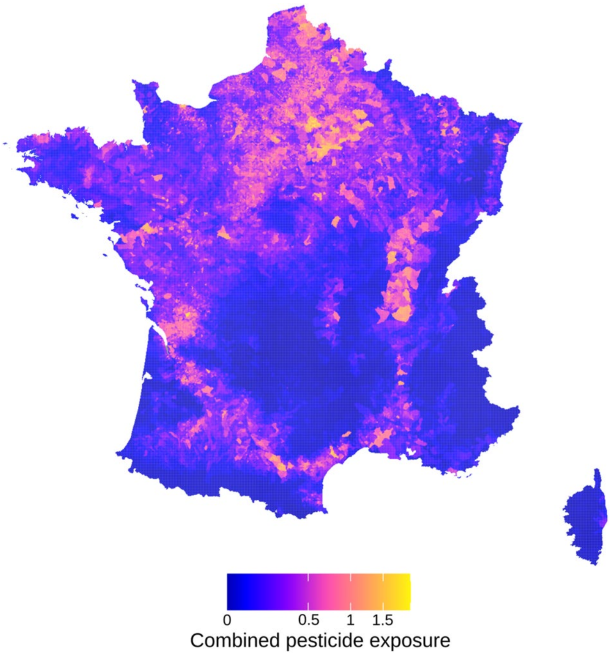

Fig. 2 Average level of combined pesticide exposure through active substance use, and concentration in air and water between 2013 and 2022. Standardised scale between 0 and 1.5 on historical data.

Field	Definition
geo_id	Polygon ID: Municipality code for use and raw quantity data, 1x1km polygon ID for air data and catchment area for water data
active substance name	Quantity of each active substance mandatory to report in the pesticide sale: Treatment Intensity Index for use data (183 different active substances), kg for raw quantity data (183 different active substances), concentration in ng.m^{-3} for air (101 different active substances) and water data (156 different active substances)
year	Year of the monitoring (2013 to 2022)
geom	Polygon geographic information (coordinate reference system RGF93/Lambert-93)

Table 1. Data record contained in the "Active substances in use, quantity, air and water" datasets.

Field	Definition
id	Polygon ID
year	Year of the monitoring (2013 to 2022)
area	Polygon size in m^2
mean_concentration_air	Summed concentration of the 99 toxic, carcinogenic, mutagenic, reprotoxic active substances in the air (ng.m^{-3})
mean_concentration_scale_air	Summed concentration of the 99 toxic, carcinogenic, mutagenic, reprotoxic active substances in the air (ng.m^{-3}), scale to the maximum observed value
mean_tii	Summed treatment intensity index of of the 175 toxic, carcinogenic, mutagenic, reprotoxic active substances used
mean_tii_scale	Summed treatment intensity index of of the 175 toxic, carcinogenic, mutagenic, reprotoxic active substances used, scale to the maximum observed value
mean_concentration_water	Summed concentration of the 145 toxic, carcinogenic, mutagenic, reprotoxic active substances in the surface water (ng.m^{-3})
mean_concentration_scale_water	Summed concentration of the 145 toxic, carcinogenic, mutagenic, reprotoxic active substances in the surface water (ng.m^{-3}), scale to the maximum observed value
all_pesticide_exposure	Combined exposure in toxic, carcinogenic, mutagenic, reprotoxic active substances from the scaled concentrations in air and water and treatment intensity index, designed to vary between 0 and 3 (historical data between 0 and 1.5)
geom	Polygon geographic information (coordinate reference system RGF93/Lambert-93)

Table 2. Data record contained in the "Yearly exposure to active substance in use, air and water" dataset. Data record in the "Combined exposure to active substance in use, air and water" are similar exepted from the year column, and summed values are replaced by average values.

correlation between combined exposure and TFI values at the 1×1 km grid is 0.78. We also perform a comparison between the combined exposure and TFI map at a the scale of small agricultural regions[31] (dividing France in homogenous agricultural areas of 770 km^2 on average) to limit the bias due to the difference in initial accuracy (Fig. 3). We find a spearman correlation of 0.83 between combined exposure and TFI at this scale. We

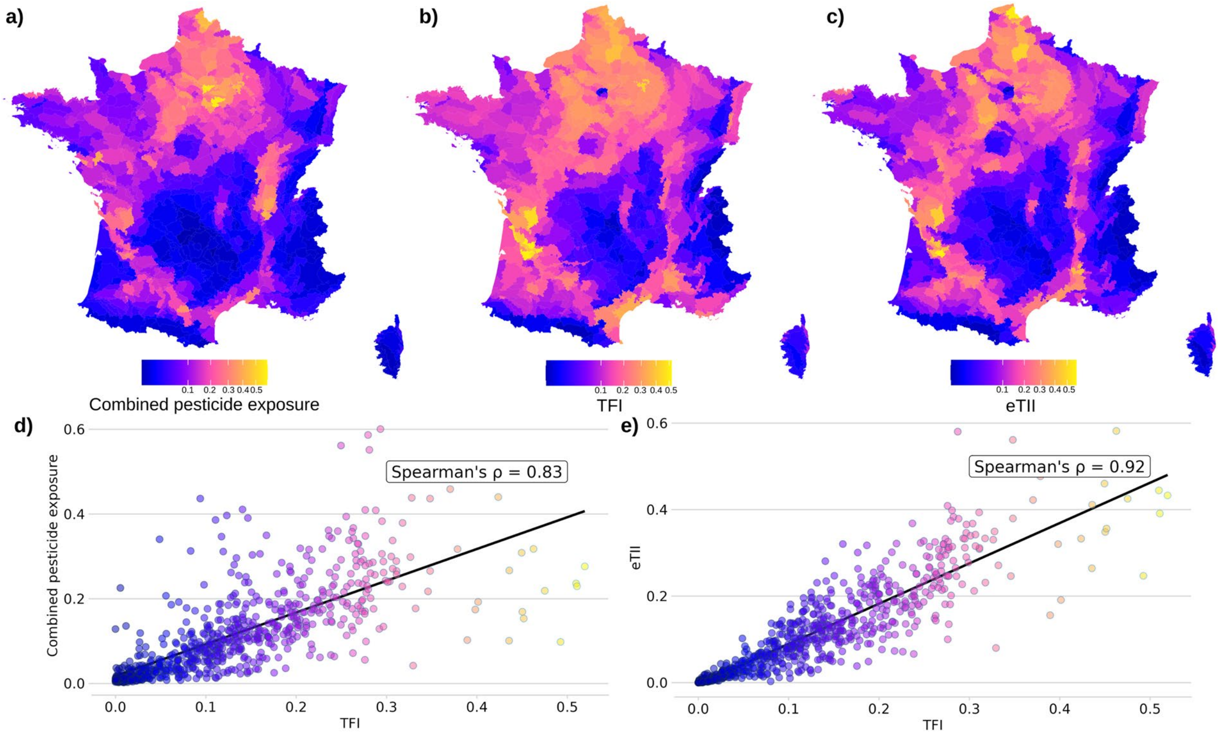

Fig. 3 Comparison between maps at the scale of the small agricultural regions between (**a**) the average combined pesticide exposure, (**b**) the treatment frequency index (TFI) produced from in-field survey and (**c**) the extended treatment intensity index (eTII). Spearman's correlations are provided between (**d**) combined pesticide exposure and TFI and (**e**) eTII and TFI.

finally perform a comparison at scale of the small agricultural regions between TFI and eTII that should be even stronger as eTII is directly related to AS use. We find a spearman correlation of 0.92 between eTII and TFI at this scale. These results confirm the adequacy between the combined exposure map, its component directly related to AS use (eTII) and TFI which is constructed from field surveys (Fig. 3).

Usage Note

The datasets produced to estimate the use and concentration of pesticide active substances (AS) in the air and water can be used at three levels:

- annual data per active substance are available for the analysis of a particular pollutant by combining data from the three environments or by carrying out a separate analysis per environment. Information on the quantity of AS in kg is also available, and if necessary this value can be used to construct other elaborate indices such as the total applied toxicity[14].
- annual data on average exposure in each environment to the AS most harmful to health and the environment (e.g. toxic, carcinogenic, mutagenic or reprotoxic) are expected to be very useful for environmental or health studies requiring an estimate of pollution by synthetic pesticides in a particular environment.
- combined average annual exposure data for environmental or health studies requiring a general estimate of pollution by pesticides harmful to health and the environment. As an example, in France, 27.7% of areas benefiting from special protection (Natura 2000[32]) have a combined exposure to pesticide that is higher than the national average (Fig. 4a) and 50.5% of the population (in 2022[33]) lived in places where the combined pesticide exposure was higher than the national average (Fig. 4b).

Inter-annual comparisons can be made (Fig. 5) while acknowledging that these data allow changes to be monitored for the most toxic active substances with a mandatory declaration of quantities sold between 2013 and 2021. It may no longer be compulsory to declare the sale of certain active substances present in one year in subsequent years because the sale of the substance is no longer authorised (9% of active substances subject to mandatory declaration in 2013 were no longer be so declared in 2021). Conversely, the sale of certain AS in recent years was not mandatory before these substances came onto the market (14% of AS subject to mandatory declaration in 2021 were not so in 2013). A variable proportion of AS is therefore not taken into account each year in the datasets presented. The inter-annual variation in the quantities of AS tracked in the datasets presented here thus reflects only part of the variation in the total quantity of AS in the environment. In addition, measuring pesticide exposure using AS alone remains conservative because total exposure is accentuated by coadjuvants which improve the effectiveness of AS and do not take into account their potential interaction (i.e. cocktail effect).

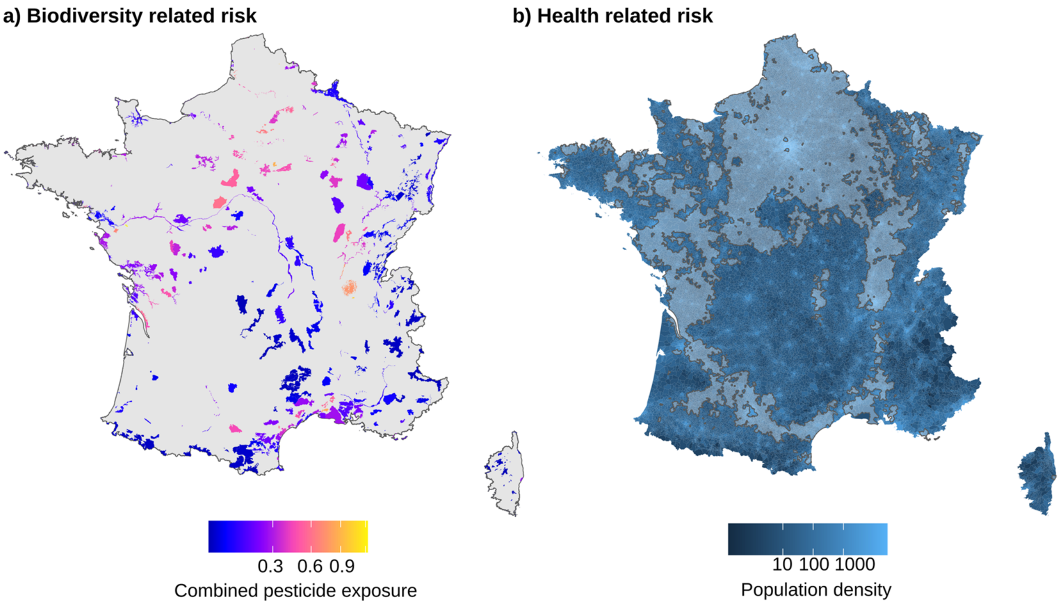

Fig. 4 Combined pesticide exposure in (**a**) special protection zones (Natura 2000 protected areas) and (**b**) in relation with population density, the whited area corresponds to municipalities whose combined exposure to pesticides is higher than the national average.

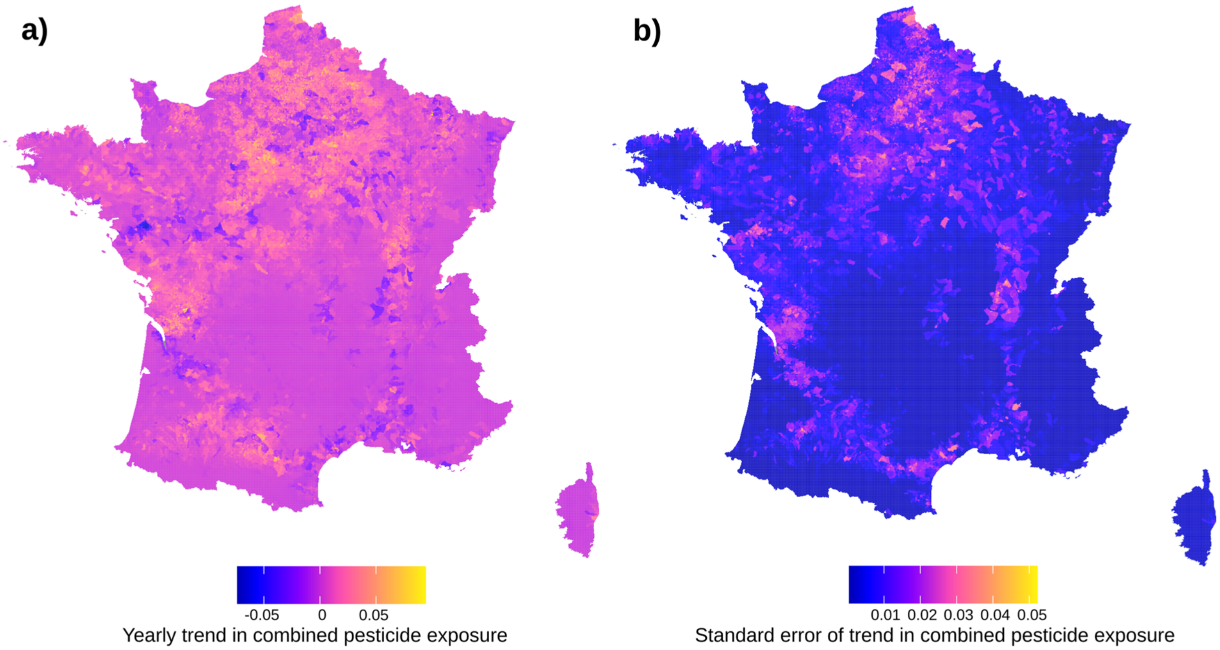

Fig. 5 (**a**) Yearly linear trend in combine pesticide exposure between 2013 and 2022 and (**b**) associated standard error.

Regarding the use of data in space, we recommend using the data at a scale no finer than that of municipalities (16 km^2 on average) or catchment areas (89 km^2 on average), i.e. a spatial resolution of 4 to 9 km.

Code availability

The code for production the active substance datasets was processed using the R software[34] version 4.3.1 and is available on Zenodo (https://doi.org/10.5281/zenodo.14198724).

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Author contributions

S.R. developed the conceptualisation and methodology, curated the data and wrote the original draft, T.P. curated the data and made a substantial contribution to the writing.

Competing interests

The authors declare no competing interests.

Additional information

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