Integrated Environmental Assessment and Management — Volume 00, Number 00—pp. 1–15
Received: 9 October 2023 | Revised: 1 February 2024 | Accepted: 7 March 2024

Health & Ecological Risk Assessment

Evaluation of the ecological risk of pesticide residues from the European LUCAS Soil monitoring 2018 survey

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Abstract

The 2018 LUCAS (Land Use and Coverage Area frame Survey) Soil Pesticides survey provides a European Union (EU)-scale assessment of 118 pesticide residues in more than 3473 soil sites. This study responds to the policy need to develop risk-based indicators for pesticides in the environment. Two mixture risk indicators are presented for soil based, respectively, on the lowest and the median of available No Observed Effect Concentration ($NOEC_{soil,min}$ and $NOEC_{soil,50}$) from publicly available toxicity datasets. Two further indicators were developed based on the corresponding equilibrium concentration in the aqueous phase and aquatic toxicity data, which are available as species sensitivity distributions. Pesticides were quantified in 74.5% of the sites. The mixture risk indicator based on the $NOEC_{soil,min}$ exceeds 1 in 14% of the sites and 0.1 in 23%. The insecticides imidacloprid and chlorpyrifos and the fungicide epoxiconazole are the largest contributors to the overall risk. At each site, one or a few substances drive mixture risk. Modes of actions most likely associated with mixture effects include modulation of acetylcholine metabolism (neonicotinoids and organophosphate substances) and sterol biosynthesis inhibition (triazole fungicides). Several pesticides driving the risk have been phased out since 2018. Following LUCAS surveys will determine the effectiveness of substance-specific risk management and the overall progress toward risk reduction targets established by EU and UN policies. Newly generated data and knowledge will stimulate needed future research on pesticides, soil health, and biodiversity protection. *Integr Environ Assess Manag* 2024;00:1–15. © 2024 The Authors. *Integrated Environmental Assessment and Management* published by Wiley Periodicals LLC on behalf of Society of Environmental Toxicology & Chemistry (SETAC).

KEYWORDS: Mixtures risk assessment; Pesticide monitoring; Pesticide residues; Risk indicator; Soil health

INTRODUCTION

Plant protection products (PPPs) represent an important input to farming systems. In the European Union (EU), approximately 450 active substances, most of synthetic chemical origin, are approved for use in PPPs (European Commission, 2023). Annual sales amount to 355 000 tons and have remained stable between 2011 and 2021 (Eurostat, 2023a). After application, most pesticides end up in the topsoil and their residual concentration in this matrix decreases over time according to their mobility and degradability rate. All pesticides used in agriculture in the EU have been assessed for their potential ecological risk before their market approval. A dual system is implemented, under which the European Food Safety Authority (EFSA) evaluates active substances and Member States (MS) evaluate and authorize commercial products at the national level. Such evaluations include environmental exposure assessments based on fate models to estimate concentrations of active substances and relevant metabolites in soil, groundwater, and surface water over time. The assessment framework

This article contains online-on Supporting Information.

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Published xxxx on wileyonlinelibrary.com/journal/ieam.

follows regulatory guidelines and tools designed for the prospective evaluation and approval procedures of single active substances (European Commission, 2002; European Food Safety Authority [EFSA], 2017b). The framework does not assess multiple exposures resulting from the application of many PPPs per season (Knillmann et al., 2021). After approval, no further monitoring is required to validate predicted environmental concentrations. Member States are responsible for routine monitoring of pesticide residues in food items placed on the market to verify compliance with legal limits (EFSA, 2022; European Parliament and Council of the European Union, 2005). Large-scale monitoring programs are also in place for pesticides in drinking water (Ministère des Solidarités et de la Santé, 2021), surface water, and groundwater for pesticides identified as priority pollutants under the Water Framework Directive (EEA, 2022). In contrast, soil monitoring data are relatively limited in scope (Froger et al., 2023; Pelosi et al., 2021; Silva et al., 2019; Vašíčková et al., 2019). All of these studies reported the occurrence of several compounds in one soil sample.

The EU Green Deal and its underpinning strategies—including the Chemicals Strategy for Sustainability (CSS; European Commission, 2020a), the Farm to Fork Strategy (European Commission, 2020b), the EU Soil Strategy for 2030 (European Commission, 2021a), and the EU biodiversity strategy for 2030 (European Commission, 2021b)—have set ambitious objectives to reduce pollution and associated adverse effects on ecosystems, biodiversity, and human health. The most prominent target is the proposed 50% reduction in the overall use and risk of chemical pesticides in the Farm to Fork Strategy. However urgent, the implementation of strategic targets in EU legislation was rejected by the European Parliament in 2023, partly due to perceived food security implications against the backdrop of geopolitical tensions affecting trade of staple food commodities (e.g., Ukrainian wheat). The same target of 50% pesticide risk reduction was recently established under the UN Convention on Biological Diversity (UN CBD, 2022).

These policy strategies corroborated the need for better indicators to assess progress toward the objectives. Under the CSS, the European Chemical Agency, the European Environment Agency, and the European Commission are developing a framework of indicators to monitor the drivers and impacts of chemical pollution and measure the effectiveness of chemicals legislation. This requires the establishment of indicators that quantify risk posed by chemicals in different environmental matrices. Key objectives of the CSS are to replace substances of concern as much as possible in the EU (e.g., carcinogens, endocrine disruptors, developmental neurotoxicants) with safe and sustainable alternatives to minimize all sources of human and environmental exposure with respect to ecosystems' carrying capacity of chemicals. It also highlights the need to address environmental mixtures of chemicals and their potential adverse ecological effects in regulatory risk assessments, and supports EU-wide environmental monitoring capacity (European Commission, 2020a).

The pesticide module of LUCAS Soil is the first continental-scale monitoring of pesticide residues in European agricultural soils (Orgiazzi et al., 2018, 2022). The module has become a component of LUCAS Soil, a pan-European scheme for assessing the characteristics of soils in relation to land cover and land use. LUCAS Soil Pesticides started as a pilot study with the 2015 LUCAS Soil survey (Silva et al., 2019) and was expanded in the 2018 survey (Orgiazzi et al., 2022). The 2018 survey also introduced a soil biodiversity assessment module.

In this study, we used monitoring data generated in the LUCAS 2018 survey and ecotoxicological data from curated public datasets to develop ecological risk indicators of pesticide residues in the soil. We discuss how such risk indicators can support EU policies with specific regard to (1) the use of indicators to monitor progress toward policy objectives and (2) the implications for the regulatory assessment of risks from combined exposure to several residues in the soil.

METHODS

LUCAS Pesticides monitoring data

Sampling design. In the LUCAS Soil 2018 survey, soil sampling was done in all EU MS (EU-27 + UK) using the same set of 25 947 locations sampled in 2015. The LUCAS sites selection methodology and field survey data are available at the EUROSTAT portal (Eurostat, 2023b). In a subset of the locations, the occurrence and concentration of 118 pesticide residues were measured. The selection of sampling locations was performed by applying a conditioned Latin hypercube sampling (CLHS) strategy (Minasny & McBratney, 2006) to LUCAS sites known to be cropland, where pesticide application is expected. The variables considered for selecting croplands of interest were (i) soil physical and chemical properties from LUCAS 2009 (Orgiazzi et al., 2018), (ii) topography (elevation, slope, and wetness index derived from Digital Elevation Model over Europe [EU-DEM]), and (iii) climate (1970–2000 average monthly precipitation and temperatures derived from WorldClim bioclimatic variables; Orgiazzi et al., 2022). Following the same strategy (i.e., CLHS), a selection of grassland sites and a more limited number of woodlands as controls were included. The 3473 monitoring locations included 2443 croplands, 993 grasslands, 35 forests, and two "other" soil sites. Target substances include in-use (approved) and legacy (not approved) active ingredients as well as some of their toxicologically relevant metabolites. The selection aimed at including the most relevant synthetic pesticides for use, toxicity, and policy priority, also considering results from the 2015 pilot study (Silva et al., 2019). Table S1 reports the full list with substance identifiers (CAS number and InChIKeys), category (e.g., herbicide, fungicide, insecticide), and EU regulatory status as of July 2023 (https://food.ec.europa.eu/plants/pesticides/eu-pesticides-database_en).

Samples were collected from a depth of 0–20 cm between 26 March and 2 December 2018, with 99% of the sites collected between 20 April and 20 October 2018. Samples were left to air dry before the bags were sealed. Details of sampling procedures are described in a technical report (Vieira et al., 2023). Sealed samples were sent to the EC Joint Research Centre and stored in the dark under ambient conditions. They were later sent to Wageningen Food Safety Research laboratories for analysis, which was performed between April 2021 and October 2022.

The 2018 LUCAS Pesticides survey is an extension of a smaller scale survey performed in 2015. In 2015, 76 substances were analyzed in samples from 300 sites across 10 EU MS covering six crop classes (Silva et al., 2019). All substances analyzed in 2015 were also analyzed in the 2018 survey. However, the overlap of sites sampled in 2015 and 2018 is limited to 73. The sampling design, including the sampling period, was equivalent in the two surveys, and the analysis was done by the same laboratory. Therefore, for this combination of substances and sites, it was possible to compare results (Vieira et al., 2023).

Analytical methods. A set of multiresidue methods based on gas chromatography-mass spectrometry (GC-MS/MS) and liquid chromatography–tandem mass spectrometry (LC-MS/MS) was developed and validated to analyze the presence and concentrations of the selected pesticide residues (Vieira et al., 2023). Method validation was performed in accordance with regulatory guidelines (European Commission, 2020c, 2021c). Following these procedures, the limit of quantification (LOQ) was defined and established as the lowest level for which it was demonstrated that trueness, precision, and identification criteria were still met. The LOQs were 0.001, 0.005, and 0.010 mg/kg (for 64, 38, and 15 pesticides, respectively), whereas for glyphosate it was 0.025 mg/kg. These LOQs are within the range of reported LOQs from the scientific literature, as reviewed by Sabzevari and Hofman (2022; Table S2), although some of the most recent studies reported lower values (e.g., Froger et al., 2023). Note that there is no harmonized procedure for determining the LOQ and that the approach used here is conservative. This partly explains the differences with other studies, for example, Froger et al. (2023), who used a signal-to-noise-based approach and reported lower LOQs. Analytical procedures are described in detail in the Supporting Information.

Compilation of physicochemical and phase partitioning data

A set of physicochemical and environmental partitioning data was collected as inputs to risk calculations and to aid the interpretation of results. The octanol–water partition coefficient (log$P$), dissociation constant (pK_a), and the soil–water partition coefficient normalized to organic carbon (K_{OC}) are compiled in Table S1. The substance properties collected for dichlorodiphenyldichloroethane (DDD), dichlorodiphenyldichloroethylene (DDE), dichlorodiphenyltrichloroethane (DDT), and endosulfan were attributed to the different isomeric forms analyzed. Experimental data were taken mostly from the EFSA OpenFoodTox database (EFSA, 2023) and the PPDB database (Lewis et al., 2016). Where experimental data were not available, gaps were filled using in silico models. Details of the experimental and estimated data are reported in Table S1.

Soil and aquatic toxicity data

Ecotoxicological data on pesticides can be found in various datasets curated by regulatory and scientific institutions. We identified four data sources based on their scientific and policy relevance: the EFSA OpenFoodTox database (EFSA, 2023), the USEPA ECOTOX database (USEPA, 2023), and the PPDB (Lewis et al., 2016). The Organisation for Economic Co-operation and Development (OECD) eChemPortal (OECD, 2023) was also searched but did not add any records (Vieira et al., 2023).

Toxicity data were extracted for the 118 target substances identified by CAS number (Vieira et al., 2023). Initially, the extraction targeted results from experimental chronic tests on in-soil organisms were expressed as No Observed Effect Concentrations (NOEC). A chronic NOEC is the highest concentration in a chronic dose–response study at which no significant effect is observed. Test organisms considered include taxonomic groups and species within the scope of EFSA guidelines on soil risk assessment (EFSA, 2017b; European Commission, 2002), corresponding to the OECD harmonized templates (OHT; https://www.oecd.org/ehs/templates/), "Toxicity to soil macro-organisms except arthropods" (OHT 50-1), and "Toxicity to soil arthropods" (OHT 50-2). As for physicochemical properties, toxicity data for different isomers were read-across, unless reported for specific isomeric variants.

Some filters and harmonization steps were performed in R to combine and process the datasets from different sources. The USEPA ECOTOX dataset included some acute NOEC records. These were excluded by applying a test duration cutoff value of 21 days. This cutoff would exclude chronic studies of predatory mites (*Hypoaspis aculeifer*; OECD 226), but no such record was present. A single NOEC, the lowest, was kept from studies of the same substance and species, thus excluding NOECs reported for different endpoints from the same study. Data fields needing harmonization included substance identifiers (CAS), species names, and units. Duplicate entries were identified based on identical CAS, endpoint name (NOEC), species, values, and unit and were removed from the dataset. For substances not covered in the dataset and with at least one detection in the monitoring survey, a targeted search of the scientific literature was performed. This led to the manual addition of prothioconazole-desthio and glyphosate's main metabolite aminomethylphosphonic acid (AMPA).

The resulting dataset of experimental records consisted of 190 rows and covered 81 of the 118 target substances (Table S2). For these 81 substances, chronic NOEC values were available for between 1 and 9 species (median case = 2). The list of taxonomic groups and species

 DOI: 10.1002/ieam.4917

represented in the dataset includes the following in the order of frequency: annelids (*Eisenia foetida*, *E. andrei*, *Aporrectodea caliginosa*, *A. longa*, *Perionyx excavatus*, *Allolobophora icterica*, *Lumbricus rubellus*, *L. terrestris*, *Enchytraeus albidus*), collembolans (*Folsomia candida*, *Heteromurus nitidus*, *F. fimetaria*), and mites (*Hypoaspis aculeifer*).

The experimental data gaps were filled with quantitative structure–activity relationship (QSAR) predictions using the earthworm NOEC model available in the VEGA QSAR platform (Benfenati et al., 2013). Estimates flagged as "low reliability" were excluded. For the remaining gaps, chronic NOECs were extrapolated from short-term (14 days) earthworm LC50 data retrieved from OpenFoodTox and PPDB. A factor of 10 (Frampton et al., 2006) was assumed for the extrapolation. At this point, chronic NOEC values were compiled for 96 substances, including 81 experimental data, three QSAR estimates, and 12 acute–chronic extrapolations (Table S1).

Aquatic toxicity data are more abundant than those for soil species because of more extensive regulatory testing requirements. In this case, we used data from Posthuma et al. (2019). In this study, species sensitivity distributions (SSDs) were derived by pooling chronic toxicity data from a variety of sources, including the USEPA ECOTOX, EFSA reports, and PPDB. This dataset contains 256 409 records, including 49 244 NOECs (SI 2 in Posthuma et al., 2019). This study provides SSD parameters derived using measured data or defined extrapolation schemes. Species sensitivity distribution parameters were retrieved for 111 substances. The number of taxa associated with each SSD ranged from 1 to 182, with an average of 23 (Posthuma et al., 2019).

Ecological risk to soil organisms

The minimum and median soil NOEC values ($NOEC_{soil,min}$ and $NOEC_{soil,50}$) were calculated from the compiled dataset (Table S1). For the derivation of $NOEC_{soil,min}$ "greater than" (>) values were excluded unless they were the only experimental record found for a given substance. They were included for the derivation of $NOEC_{soil,median}$, with the condition $NOEC_{soil,median} \geq NOEC_{soil,min}$. Risk quotients were calculated for each substance in each site by dividing the measured concentration by the $NOEC_{soil,min}$ and the $NOEC_{soil,50}$. Left-censored data ($C_{soil} < LOQ$) were set at 0. This risk quotient calculated on the $NOEC_{soil,min}$ was multiplied by an assessment factor of 5, in accordance with established guidance on soil risk assessment (European Commission, 2002). In EFSA regulatory assessments, the risk of a single substance is expressed as the ratio of toxicity (e.g., NOEC) and the predicted environmental concentration (PEC), known as toxicity–exposure ratios (TER). In-field soil PEC (mg/kg) are average concentrations calculated over depths ranging from 5 to 20 cm, depending on the type of crop, plowing practice, substance degradability, and exposure scenarios assessed. Two exposure scenarios are typically considered: a peak and a time-weighted average for various time windows (7–56 days) after the occurrence of the peak concentration, representing a short- and a long-term exposure scenario, respectively (EFSA, 2017a).

Equivalent risk to aquatic species

The risk indicator based on aquatic toxicity provides a complementary assessment based on a larger database (Posthuma et al., 2019), covering more substances (111 compared with 94), and a much greater taxonomic diversity.

Assuming partitioning equilibrium between the soil and a corresponding aqueous phase, the equivalent aqueous concentration, $C_{aq,eq}$, was calculated from the soil concentrations and the soil–water partition coefficient K_d. The K_d itself was estimated by the product of the fraction of organic carbon in the soil, *OC* (g/g), and the soil–water partition coefficient normalized to organic carbon (K_{OC}, L/kg; Equation 1):

$$C_{aq,eq} = \frac{C_{soil}}{K_d} = \frac{C_{soil}}{OC\ K_{OC}}. \quad (1)$$

The soil OC is available from the LUCAS survey. In sites where OC measurements were not available (1%), they were estimated (De Rosa et al., 2024). The measured or estimated K_{OC} values are reported in Table S1 with the sources and algorithms used for estimations.

The aquatic risk indicator is the ratio of the $C_{aq,eq}$ and the 5th and 50th percentiles of the SSD of chronic aquatic NOECs ($HC_{X,i}\,SSD_{aq[NOEC]}$). The 50th percentile is reported as SSD midpoint μ (Posthuma et al., 2019). The 5th percentile was derived as (Equation 2):

$$HC_5\,SSD_{aq\,[NOEC]} = SSD\ \mu + z_{0.05} \times SSD\ \sigma, \quad (2)$$

where $SSD\ \sigma$ is the standard deviation and $z_{0.05}$ is the inverse of the normal distribution calculated at the 5th percentile (−1.645). In addition, in this case, left-censored data were set at 0. The equivalent aqueous concentrations and corresponding risk can be interpreted as a proxy for potential aquatic exposures and risks. The derived aquatic risk indicators represent a conservative long-term exposure scenario for edge-of-field surface water. In reality, transport from topsoils to freshwater ecosystems, for example, via surface runoff or groundwater transport, is limited because substances undergo degradation and dilution.

Mixture risk assessment

The overall toxic pressure of pesticide residues is the result of the combined effect of all toxic components of a mixture. In ecotoxicology, the sum of risk quotients (RQs), also referred to as the sum of toxic units when based on the same endpoint, is a commonly used model for low-tier mixture risk assessment (EFSA, 2019a). This is easily calculated by adding the RQs of all substances. The two soil risk indicators were defined as follows, with the inclusion of the

assessment factor of 5 on the indicator based on the $NOEC_{min}$ (Equations 3 and 4):

$$RI_{soil,NOEC\_min} = \sum_{i=1}^{n} \frac{C_{soil,i}}{NOEC_{soil\_min,i}} \times 5, \quad (3)$$

$$RI_{soil,NOEC\_50} = \sum_{i=1}^{n} \frac{C_{soil,i}}{NOEC_{soil\_50,i}}, \quad (4)$$

where $C_{soil,i}$ is the concentration of substance i of the mixture. A $RI_{soil,NOEC\_min}$ value below 1 indicates no expected adverse effects on non-target soil organisms. A $RI_{soil,NOEC\_50}$ value below 1 calculated indicates no expected adverse effects on 50% of the species tested. By including all data records in our dataset, the risk indicator based on median NOEC ($RI_{soil,NOEC\_50}$) provides a mean of comparison supporting the interpretation of results.

For the two aquatic risk indicators, the sum of aquatic RQs defines the mixture risk (Equation 5).

$$RI_{aq,SSD\_HCx} = \sum_{i=1}^{n} \frac{C_{aq_{eq},i}}{HC_{X,i}SSD_{aq[NOEC]}}. \quad (5)$$

For all indicators, left-censored concentration data (<LOQ) were considered 0. Because the NOEC values correspond to different types of studies depending on the substance, the risk indicators cannot be linked to specific species or endpoints. However, for soil toxicity, most underlying data are for earthworms and many refer to reproductive effects (Vieira et al., 2023).

RESULTS

General statistics on the occurrence

At least one pesticide residue was detected above the LOQ in 74.5% of the 3473 sites. Most samples (57.1%) had at least two different pesticide residues, 29.8% had more than five, whereas 11.1% had more than 10 pesticide residues (Vieira et al., 2023).

Most substances (77%, 91 out of 118) were quantified in at least one site. The median detection frequency of substances found in at least one site was 1.3% (46 sites). Thirteen substances were detected in more than 10% of sites, the most frequently detected being AMPA (54%), epoxiconazole (27%), tebuconazole (26%), pendimethalin (25%), diflufenican (21%), boscalid (20%), imidacloprid (18%), and glyphosate (16%).

Soil risk indicators

Results for the risk indicator $RI_{soil,NOEC\_min}$ (Equation 3) are shown in Figure 1 for all sites (upper panel) and zoomed in for the 100 sites with the greatest risk (lower panel). This risk indicator exceeds 1 in 483 sites, representing 14% of all monitoring sites. The long tail of low-risk sites comprises 23% of them with $RI_{soil,NOEC\_min}$ ranging between 0.1 and 1

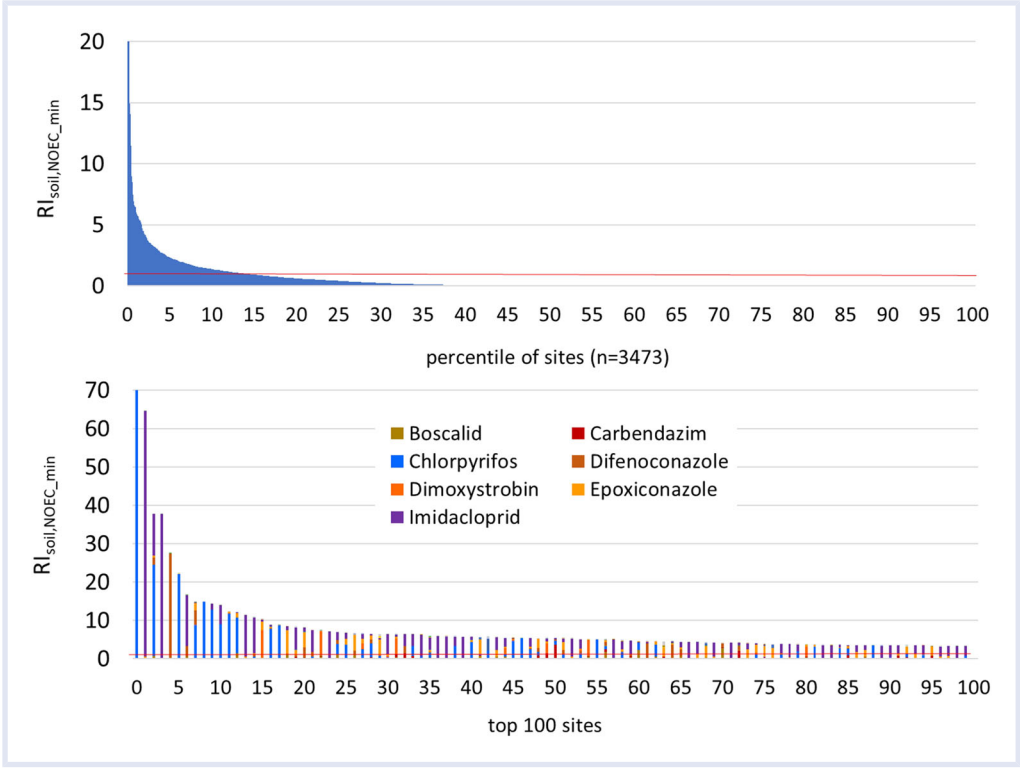

FIGURE 1 Results for the soil risk indicator $RI_{soil,NOEC\_min}$ for all sites (upper panel) and for the 100 sites at greatest risk (lower panel). The six highest values (upper panel) and the highest value (lower panel, $RI_{soil,NOEC\_min} = 271$) are cut at 20 and 70, respectively. Contributions from all substances are plotted. Data labels in the lower panel are shown only for substances with $RI_{soil,NOEC\_min} > 2.5$ at any site. The horizontal red line represents $RI_{soil,NOEC\_min} = 1$

 DOI: 10.1002/ieam.4917

and 63% with $RI_{soil,NOEC\_min} < 0.1$. In the top 100 sites with the greatest risk, the risk is, in most cases, driven by a single substance. Based on RQs for individual substances, 308 sites exceed the $RI_{soil,NOEC\_min}$ of 1. The insecticides imidacloprid and chlorpyrifos were the most recurrent toxicity drivers of the mixture, followed by the fungicides epoxiconazole, dimoxystrobin, and difenoconazole. One or few substances, up to three to five at most, drive the mixture risk at each site. Recurrent mixture combinations at risk-relevant levels include insecticides and fungicides (e.g., imidacloprid and epoxiconazole is a frequent combination), but also combinations of insecticides (e.g., chlorpyrifos and imidacloprid) and fungicides (Vieira et al., 2023).

The sum of RQs for the indicator based on the $NOEC_{median}$ ($RI_{soil,NOEC\_50}$) is obviously lower, with only five sites exceeding 1 (Figure 2). The substances contributing to this risk indicator are similar to those based on the $NOEC_{min}$, with imidacloprid and epoxiconazole being the most frequent toxicity drivers. A notable difference between these two scenarios is that the risk contribution of the insecticide chlorpyrifos is small based on $NOEC_{median}$. The difference between the $NOEC_{min}$ and the $NOEC_{50}$ for chlorpyrifos is particularly high (Table S1).

Overall, some insecticides and fungicides may pose a risk to soil organisms in a fraction of the surveyed sites. Herbicides, including glyphosate and its metabolite AMPA, contribute less to the risk, despite being detected most frequently and at the highest concentrations (Vieira et al., 2023).

Aquatic risk indicators

The sum of equivalent aquatic RQs is significantly higher than that for the soil risk indicators, especially when based on the 5th percentile of the SSDs (Figures S1 and S2). In this scenario, the risk indicator $RI_{aq,SSD\_HC5} = \Sigma(C_{aq}/SSD\ HC_5)$ exceeds 1 in 39% of all sites, with 43 sites (1.2%) exceeding 100. The main toxicity drivers are the insecticides imidacloprid and the herbicide metolachlor. Compared with the soil risk indicators, herbicides and particularly metolachlor contribute to ecotoxicological risk. Such a difference is explained mostly by the inclusion of primary producers, mainly algae, in the toxicity data underpinning the SSDs. The indicator based on median values of the aquatic SSD yields lower values (1.3% of sites with $RI_{aq,SSD\_HC50} > 1$) with more substances contributing to it (Figure S2). Herbicides, mainly metolachlor and prosulfocarb, contribute the most, followed by fungicides (e.g., difenoconazole, dimoxysrobin). As for the soil risk indicators, the mixture risk is determined by the contribution of one or a few substances.

Characterization of toxicity drivers

The four indicators provide complementary insights into pesticide risk distribution profiles in LUCAS soils. Their sensitivity is different when looking at different percentile levels and environmental media (terrestrial vs. aquatic). In contrast, the shape of the distribution, the limited number of toxicity drivers and, in several cases, the identity of the substances driving the risk are more consistent. Table 1 provides the list of the most prominent substances ranked by the sum across

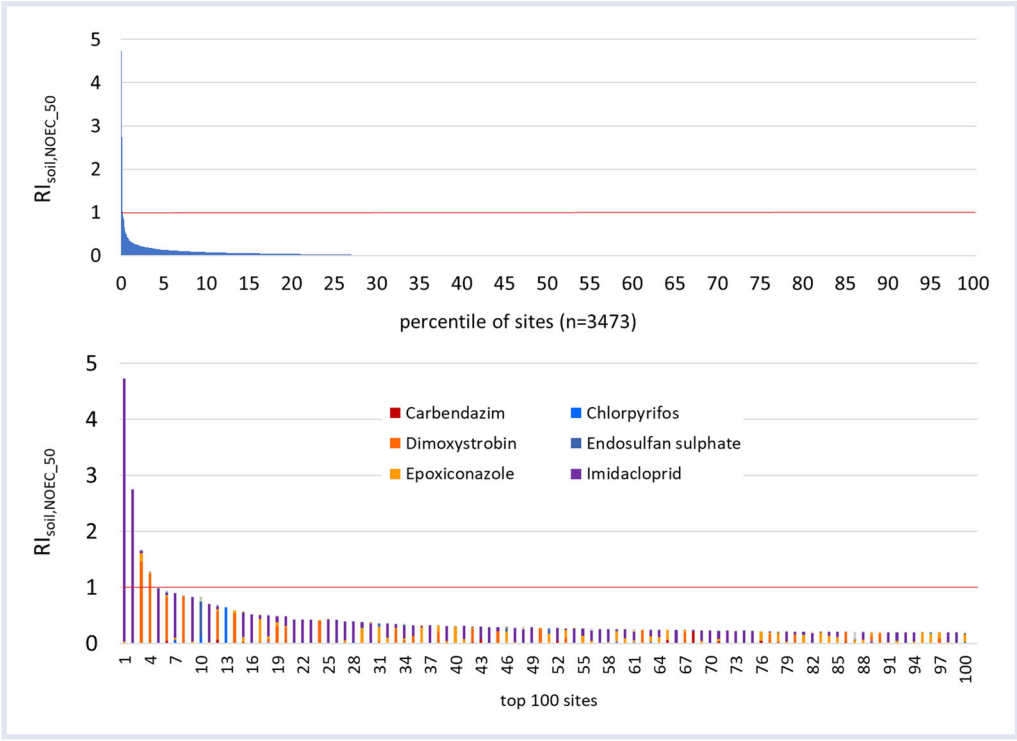

FIGURE 2 Results for the risk indicator $RI_{soil,NOEC\_50}$ for all sites (upper panel) and for the 100 sites at greatest risk (lower panel). Contributions from all substances are plotted. Data labels in the lower panel are shown for substances with $RI_{soil,NOEC\_50} > 0.5$ at any site. The horizontal red line represents $RI_{soil,NOEC\_50} = 1$

TABLE 1 Target toxicity profiling of the top 10 risk ranked substances according to any of the four risk indicators: (A) $RI_{soil,NOEC\_min}$, (B) $RI_{soil,NOEC\_50}$, (C) RI_{aq,SSD_HC5}, (D) RI_{aq,SSD_HC50}

			Risk ranking							
		Substance	A	B	C	D	$NOEC_{soil,min}$ endpoint	Substance group	Target mode of action	Reference
	Sterol biosynthesis inhibition	Epoxiconazole	3	2	4	4	Reproduction	Triazole	Demethylation inhibitor	FRAC
		Difenoconazole	4	51	7	3	Reproduction	Triazole	Demethylation inhibitor	FRAC
		Cyproconazole	9	31	24	14		Triazole	Demethylation inhibitor	FRAC
		Fluquinconazole	11	7	42	34		Triazole	Demethylation inhibitor	FRAC
Mitotoxic	Mitochondrial respiration inhibition	Dimoxystrobin	7	3	18	5		Oximino-acetamide	Quinone outside inhibitor	FRAC
		Azoxystrobin	14	18	11	9		Methoxy-acrylate	Quinone outside inhibitor	FRAC
		Boscalid	6	39	10	7		Pyridine-carboxamide	Succinate dehydrogenase inhibitor	FRAC
Cytotoxic	Cell division inhibition	Carbendazim	5	4	17	27	Growth reproduction mortality	Benzimidazole	Inhibition of ß-tubulin assembly in mitosis	FRAC
		Pendimethalin	19	12	8	23	Reproduction	Dinitroaniline	Inhibition of microtubule assembly	HRAC
Phototoxic	Photosynthesis inhibition	Terbuthylazine-desethyl			9	11		Triazine	D1 serine 264 binder	HRAC
		Atrazine-desethyl			48	8		Metabolite of atrazine (triazine)	D1 serine 264 binder	HRAC
		Linuron	24	17	14	10	Mortality	Urea	D1 serine 264 binder	HRAC
Cytotoxic	Cellular metabolism	Prosulfocarb	26	16	20	1		Thiocarbamate	Inhibition of very long-chain fatty acid synthesis	HRAC
		Metolachlor	17	10	2	2		Chloroacetamide	Inhibition of very long-chain fatty acid synthesis	HRAC
Neurotoxic	Modulation of acetylcholine metabolism	Imidacloprid	1	1	1	6	Reproduction	Neonicotinoid	Nicotinic acetylcholine receptor (nAChR) competitive modulator	IRAC
		Clothianidin	8	6	3	25	Growth	Neonicotinoid	Nicotinic acetylcholine receptor (nAChR) competitive modulator	IRAC
		Chlorpyrifos	2	5	5	12		Organophosphate	Acetylcholinesterase (AChE) inhibitor	IRAC
		Diazinon	67	64	6	33	Growth	Organophosphate	Acetylcholinesterase (AChE) inhibitor	IRAC
		AMPA	10	9	56	45		Metabolite of glyphosate	Not yet completely clarified	HRAC
		DDE p,p′-	15	8	12	15		Organochlorine	Not yet completely clarified	

Note: The ranking is based on the sum of the indicators for all sites. Substances are grouped according to the reported target mode of action and knowledge of the corresponding Adverse Outcome Pathway.

DOI: 10.1002/ieam.4917

all sites of each of the four indicators. Remarkably, seven out of the top 10 ranking substances according to the $RI_{soil, NOEC\_min}$ are currently (June 2023) not approved in the EU under Regulation (EC) No. 1107/2009. Six of these were approved in 2018. Only carbendazim was not approved at the time of sampling. Its approval status expired in 2014 and has not been renewed since then. This fungicide is also a degradation product of thiophanate-methyl, which was still approved in 2018 (discontinued by 2021). The top three high-risk substances, imidacloprid, chlorpyrifos, and epoxiconazole, were withdrawn from the list of approved active ingredients in 2020: imidacloprid due to identified risk to bees (European Commission, 2018), chlorpyrifos due to human health concerns on its genotoxic potential and developmental neurotoxicity (EFSA, 2019b), and epoxiconazole due to the applicant withdrawing its application. Several high-risk substances feature in the top 20 across all four indicators. This is the case of imidacloprid, chlorpyrifos, epoxiconazole, dimoxystrobin, DDE *p*,*p*′, metolachlor and azoxystrobin. The presence on this list of the DDT metabolite DDE, banned in the EU since 1986, confirms the long-term legacy of persistent organic pollutants (organochlorine compounds). In addition to the extreme case of DDE, with a half-life in the range of several years (Thomas et al., 2008), most of the high-risk substances also demonstrated high persistence in soil dissipation studies, that is, a half-life geometric mean of 174 days for imidacloprid, 362 days for epoxiconazole, and 256 days for boscalid (PPDB data; Lewis et al., 2016). Reported half-lives of chlorpyrifos are somewhat shorter but highly variable (ECHA, 2022).

A complementary risk ranking, based on the frequency of occurrence of each substance as contributing to more than 5% of the risk, is shown in Figures S3–S6. This alternative ranking gives relatively more weight to substances that contribute to the risk in many sites, including low-risk sites (epoxiconazole, AMPA), compared with substances that are detected less frequently but often contribute to relevant risk levels when present (e.g., chlorpyrifos, metolachlor).

The target mode of action (MoA) of the top risk-ranking substances is reported in Table 1. Mode of action classification was compiled using information from the international industry association CropLife's Resistance Action Committees for insecticides (IRAC), fungicides (FRAC), and herbicides (HRAC; CropLife, 2023) considering existing ontologies (https://aopwiki.org/) and previous MoA profiling of PPP ingredients (Carnesecchi, Toma, et al., 2020). A prominent MoA of the toxicity drivers in LUCAS soils is the inhibition of sterol biosynthesis by triazole fungicides (e.g., epoxiconazole, difenoconazole, cyproconazole, and fluquinconazole) targeting the cell membrane integrity by inhibiting C14 demethylation during sterol formation. Mitochondrial respiration inhibition by inhibiting quinone outsider or succinate dehydrogenase is another MoA of fungicides from different substance groups. The main MoAs of herbicides driving the risk in this study are photosynthesis inhibition with terbuthylazine-desethyl, atrazine, and linuron and the inhibition of very long-chain fatty acid synthesis by prosulfocarb and metolachlor. Finally, neurotoxicity via modulation of nicotinic acetylcholine receptor (nAChr) by the neonicotinoids (imidacloprid, clothianidin) and via acetylcholinesterase (AChE) inhibition by the organophosphate insecticides (chlorpyrifos, diazinon) is the major target MoA of the insecticides. Although the MoA of AMPA has not yet been fully clarified and deserves more research, it has been proposed that AMPA could interfere with chlorophyll synthesis by inhibiting glycine decarboxylase (Gomes et al., 2022).

Aggregation by country

Risk indicators calculated at each site can be aggregated by geographical region. Here, we present the soil risk indicator based on the $NOEC_{min}$ ($RI_{soil, NOEC\_min}$, Equation 3) for potential use in policy indicator frameworks. Figure 3 shows the distribution of the indicator at the EU and country levels. Aggregation by country reveals differences but no clear geographical patterns. The number of monitoring sites differs significantly across countries and, when too small ($n < 10$), the distribution is not reported in Figure 3. European Union and country-level aggregations are suitable for the analysis of temporal trends. The relatively small overlap of sampling sites monitored in 2015 and 2018 enables a first partial comparison (Figure 3, lower panel). Across the 73 sites monitored in both campaigns, the risk indicator increased slightly. Sampling design and analytical methods were consistent between the two campaigns, but environmental factors, pest pressure differences, or crop selection over the two years could have a notable effect on exposure and persistence. Despite the limited scope of the comparison, no progress is apparent between 2015 and 2018 toward meeting the proposed 50% reduction target in pesticide use and risk by 2030.

Aggregation by crop type

Aggregation by crop types (Figure 4) revealed that risk was greater in sugar crops, followed by vegetables and melons, root and tuber crops, and fruit and nuts. Cereals, the most abundant crop type, have the most sites with $RI_{soil,NOEC\_min}$ exceeding 1 in absolute terms, but the percentage of exceedances reflect a median scenario. Results for grassland, forest, and shrubland sites provide valuable information about the level of background contamination. As expected, occurrence and risks are lowest in grassland sites and in the few forests and scrubland sites surveyed (Vieira et al., 2023). However, the $RI_{soil,NOEC\_min}$ is greater than 0.01 in approximately 10% of grassland sites.

DISCUSSION

Filling the knowledge gap

LUCAS Soil Pesticides and the indicators presented address knowledge gaps in an area that has been neglected over the years. Major data gaps have been identified in the effort to characterize EU soil health, specifically regarding what soil pollution is concerned (European

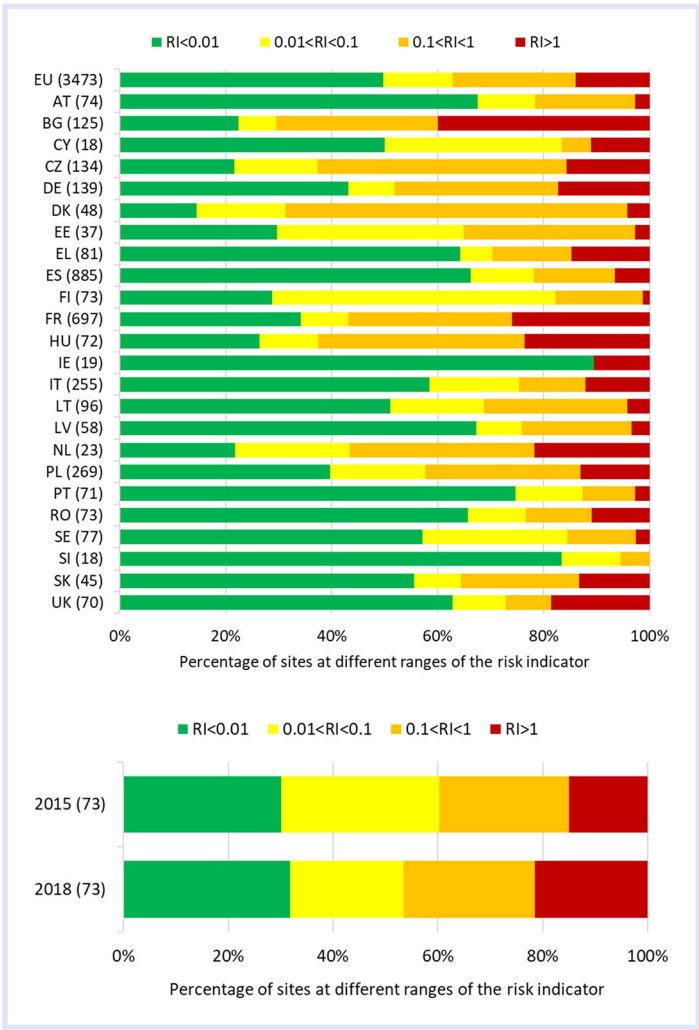

FIGURE 3 Upper panel: distribution of the soil risk indicator $RI_{soil,NOEC\_min}$ in European Union countries (UK included as of 2018). The number of sampling sites is given in parentheses. Countries with fewer than 10 sampled sites are not shown (Belgium, Croatia, Malta, and Luxembourg). Lower panel: comparison of the indicator for the same combination of substances and sites monitored in 2015 and 2018

Commission, 2022b). The indicators developed here are supported by the EU soil observatory (EUSO, https://esdac.jrc.ec.europa.eu/euso/euso-dashboard) and the Information platform on chemical monitoring data (IPCHEM, https://ipchem.jrc.ec.europa.eu/). These well-established platforms encourage data transparency and reusability while complying with the EU data protection policy. The 2015 and 2018 monitoring datasets are stored in IPCHEM (https://ipchem.jrc.ec.europa.eu/). Regarding ecotoxicity data, the derived compilation (Table S2 and derived reference values [Table S1]) integrates records from publicly available curated datasets. In this case, data retrieval and integration required some filtering and harmonization. The implementation of the CSS-One Substance One Assessment initiative, including the development of the EU Common Data Platform on Chemicals, will provide further opportunities to enhance data sharing and reuse.

A modeling study (Pistocchi et al., 2023) of similar scale and scope (148 active ingredients), but focused on surface water, estimated mixture risk exceedances at 4.3% of sites, based on the sum of RQs using the median of aquatic NOECs from the SSDs by Posthuma et al. (2019). This can be compared with the 1.3% exceedances derived from our corresponding indicator (RI_{aq,SSD_HC50}, Figure S2). The comparison of the most frequent risk contributors identified in the two studies demonstrates commonalities (e.g., chlorpyrifos, imidacloprid, chlorpyrifos, imidacloprid, dimoxystrobin, boscalid) but also notable differences (Figure 6 in Pistocchi et al., 2023). Synthetic pyrethroids (i.e., deltamethrin, cypermethrin) are prominent risk contributors in EU streams. Deltamethrin, the only pyrethroid monitored in LUCAS, is never found above the LOQ of 0.005 mg/kg. The opposite is observed for some azole fungicides (e.g., epoxiconazole). Differences can be explained by different emission and exposure profiles of substances between soil and streams (Vieira et al., 2023).

Notable monitoring studies of pesticide mixtures have been done in soils, especially in France. In a recent monitoring study of 111 substances in 47 sites (mostly arable land), 13% of the sites were considered at risk based on

 DOI: 10.1002/ieam.4917

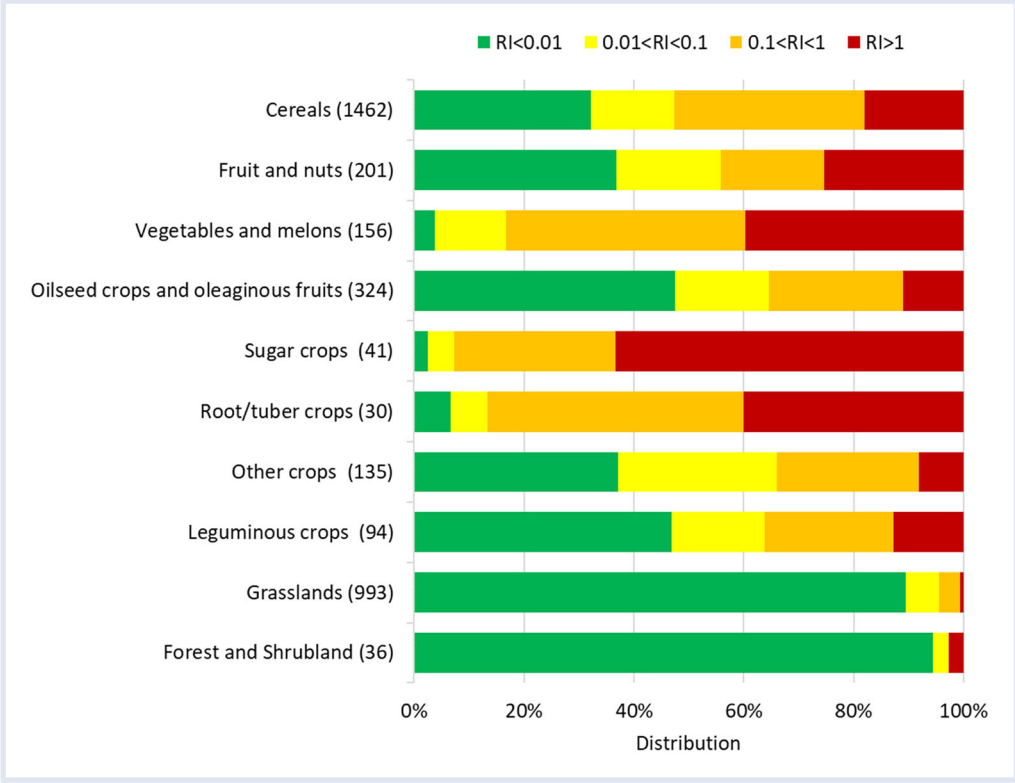

FIGURE 4 Distribution of the soil risk indicator $RI_{soil,NOEC\_min}$ by crop type in the European Union. The number of sites is given in parentheses. FAO (Food and Agriculture Organisation of the United Nations) crop type classification is used (category "Root/tuber crops with high starch or inulin content" is abbreviated as "root/tuber crops")

earthworm NOEC data (Froger et al., 2023). A similar study targeting 31 active ingredients in 180 French soil sites (Pelosi et al., 2021) concluded that 46% of the sites were at high risk, based on a similar risk assessment approach. Both studies corroborated major risk contributions from the fungicides dimoxystrobin, boscalid, cyproconazole, epoxiconazole, and from imidacloprid among the insecticides. In the Czech Republic, triazines and triazole fungicides, especially epoxiconazole, were found to be the main toxicity contributors among 68 substances monitored across 75 arable soil sites, with 35% of the sites found to be at risk (Vašíčková et al., 2019). Overall, these studies report higher levels of risk, but the findings are remarkably consistent with results from LUCAS regarding the identified drivers of toxicity. Differences in the reported risks can be explained by the use of higher assessment factors (e.g., 10 on chronic earthworm NOECs), to differences in the sampling design (LUCAS includes more grassland sites), and to some extent to the lower LOQs. Our choice of the assessment factor of 5, compared with 10 mostly used in the studies mentioned, considers the broader taxonomic coverage of our dataset and is consistent with current regulatory guidance.

Implications for the regulatory framework

The derived risk indicators provide retrospective evidence that complements the EU regulatory framework on pesticides. Differences in the scale and the scenario assessed do not allow a meaningful direct comparison of the results with regulatory assessments performed by registrants and EFSA. Nevertheless, the indicators presented provide an insight into the level of protection achieved in the field. Considering that regulatory assessments for soil are typically calibrated on the most sensitive species tested, the first indicator, based on the soil $NOEC_{min}$, is most closely related to protection objectives for non-target soil organisms. The extent of risk exceedances over the entire dataset, 14% of sites with $RI_{soil,NOEC\_min} > 1$, represents the actual level of protection achieved under the current risk management framework. The indicator also allows screening for many chemicals while highlighting primary risk drivers. By shedding light on substances of particular concern, the results can inform targeted policy interventions.

Combined (unintended) exposure to several pesticide residues remains a regulatory gap. EFSA published a harmonized guidance on risk assessment of combined exposure to multiple chemicals (EFSA, 2019a). However, in practice, detailed methods and models require further refinement for environmental scenarios. In this regard, a new scientific framework has been developed for bees for multiple chemicals as well as multiple stressors, in addition to in silico models providing means to fill data gaps and building on preliminary schemes for grouping substances for their potential combined effects on pollinators (Carnesecchi et al., 2019; Carnesecchi, Toropov et al., 2020; More et al., 2021).

The analysis performed here reflects a conservative mixture risk assessment. The simple sum of RQs is considered a

suitable low-tier approach to assessing the combined effects of environmental mixtures. As a pragmatic precautionary approach, EFSAand other agencies, including OECD and WHO, recommended this as the default model, as it "*can be readily applied by comparing exposure doses or concentrations with reference values derived from toxicity data (such as no effect or effect concentrations) often available in public databases*" (EFSA, 2019a).

Results from this study confirm earlier observations that the combined risk of environmental mixtures is driven by one or a few substances (EFSA, 2019a). Where several substances contribute to the risk at any given site, the combined risk is greater if toxic effects occur via a common MoA or by targeting common organs. Within the limited number of substances contributing to the risk, commonalities in MoA and target organs point to a greater likelihood of combined effects (i.e., through dose addition). Instead, for substance groups with dissimilar MoAs, response addition would be the most appropriate model to estimate the mixture risk (EFSA, 2019a). Mechanistic understanding of the effects of pesticides on non-target species supports the causal link between mixture exposure and associated adverse outcomes, such as decreased reproduction and mortality. In this context, the Adverse Outcome Pathway (AOP) framework can be useful in supporting the grouping of substances and in identifying shared key events (EFSA, 2021). An AOP describes the pathway from a molecular initiating event (MIE), that is, the interaction between the substance and the biological target through subsequent key events (KEs) at the molecular, cellular, tissue, and organ levels up to an adverse outcome in an individual organism or at the population level (Ankley et al., 2010). Several AOPs can form an AOP network by converging on the same adverse outcome or by sharing MIEs or other node KEs (Knapen et al., 2018). For example, AOPs based on a MIE of AChE inhibition leading to acute mortality were developed by computing information from numerous studies investigating diverse organophosphate and carbamate insecticides covering a wide range of species at different life stages (Russom et al., 2014).

For the two soil risk indicators, toxicity data refer to a few apical endpoints (reproduction impairment, growth, mortality) in most cases on the same taxonomic group (earthworms). In addition to information on the target MoA (Table 1), major gaps remain regarding potentially associated AOPs, including those triggered by non-target MoAs. Linking mechanistic information with commonly investigated endpoints remains a challenge. In practice, however, it has been reported that applying dose addition or response addition for ecotoxicity endpoints does not usually lead to substantial differences (EFSA, 2019a; Kortenkamp et al., 2009).

A baseline for monitoring progress toward policy objectives

Recent EU policy strategies on soil health, pollution, and chemicals have emphasized the importance of characterizing current baselines and defining risk thresholds for comparison with policy objectives. Improved pesticide risk indicators can be integrated into specific EU legislation (e.g., on the sustainable use of pesticides [European Commission, 2022a]) and into broader policy frameworks (e.g., CSS, Zero Pollution Monitoring, and Outlook). There is a gap between the state and trend of soil pollution in the EU and the most recent policy ambitions. Improving soil health, which includes soil pollution by definition, is an objective of the Soil Strategy (European Commission, 2021a), the zero pollution action plan (European Commission, 2021d) and, more recently, of the proposed directive on soil monitoring and resilience (European Commission, 2023).

Under the CSS, reducing the use of substances of concern by using safe and sustainable substitutes is part of a broader objective to restore health and environment to a sound state. The development of an indicator as a proxy for the ecological risk of pesticides tracks the extent of pesticide contamination above safe levels over time. Such an indicator can measure the impact of upstream actions such as regulatory bans, restrictions, promotion of the use of biopesticides, and other integrated pest management measures.

Considering that one or a few substances drive the total risk, targeted risk management of such substances can lead to substantial reductions in the risk indicators. Based on outcomes from EFSA assessments since 2018, several substances have been withdrawn from the EU list of approved active ingredients, including several substances topping the risk ranking (Table 1). Results from the next monitoring survey, covering the period 2022–2023, will track the effect of these interventions and of potential uses resulting from emergency authorizations. In fact, the evolution of exposure levels of no-longer approved substances, for example, imidacloprid and epoxiconazole, remains an issue, as exemplified by the case of the now discontinued emergency authorizations of imidacloprid for sugar beet cultivation (European Court of Justice, 2023).

For future assessments, potential bias must be avoided or minimized at every step, from the sampling design to the selection of substances and their toxicity data used in the calculations. A representative number of sampling sites per country, a transparent algorithm for selecting sampling locations, and a defined temporal scheme for sample collection are essential. The selection of substances must cover the most relevant pesticides used over time as well as their transformation products. Understanding how the sector responds to risk management interventions is important not only to contextualize results, but also to identify and add to the target list any relevant new or existing substances replacing those that have been phased out. These methodological aspects require some refinement and further standardization and automation.

Bridging soil ecotoxicology and biodiversity assessment

LUCAS Soil Pesticides adds to the large, interdisciplinary knowledge base of the LUCAS program. Pesticide exposure and risk information can be associated with data from other

 DOI: 10.1002/ieam.4917

LUCAS modules. For example, the database can be combined with LUCAS Soil Biodiversity, a DNA-based assessment of soil biodiversity (for both microorganisms and animals). As a first step, an evaluation of the pesticide residues, both as occurrence, and concentration, may facilitate the exploration of when and how an anthropic factor (i.e., pesticide application) may play a role in shaping soil microbial and animal communities at continental scale. Such an analysis may be applied not just at the taxonomical level (i.e., effects on species richness and diversity) but also at the functional level to determine if the presence and/or concentration of pesticide residues affects certain types of beneficial soil organisms, including, for example, symbiotic bacteria and fungi. Furthermore, the proposed risk indicators may be used to explore possible links with, once again, the diversity and functional metrics of soil-living communities. Chemical stressors may be combined with non-chemical stressors to investigate the contribution of several ecological stressors on biodiversity (van Gestel et al., 2021). The final aim would be to strengthen the evidence base at the interface between ecotoxicology and soil ecology, supporting policymakers in the implementation and monitoring of chemical, environmental, and agricultural policies.

Study limitations

Every target monitoring is limited in scope, and LUCAS Soil Pesticides 2018 is no exception. By taking advantage of the established LUCAS Soil monitoring program, the pesticide monitoring campaign covered an unprecedented spatial scale including all 27 EU Member States and the UK. Samples, however, were collected at a single point in time at each site. The time of sampling spanned more than eight months. Sampling started and ended at different times in each country, broadly following the plant growing season, considering practical and logistic constraints. Monitoring data come with neither information on the farming system (conventional, organic, integrated pest management) nor on the application of pesticides in the period before sampling. Pesticide applications follow seasonal patterns depending on crop type, climatic conditions, and agricultural practices. Residues in the soil are further influenced by weather conditions, especially rainfall events and temperature (Bento et al., 2016). Because such factors strongly influence residue levels at individual sites, results are most meaningful when averaged across the entire dataset.

Sampling depth influences results as well as storage and preservation. Air-dried, sealed samples were stored in the dark under ambient conditions for more than three years before analysis. As the targeted substances are not highly volatile, losses due to volatilization during air exposure were not expected. To the best of our knowledge and experience, in dried soil and protected from light, pesticides are stable (minimal/no microbial activity, no hydrolysis, no photolysis) even under ambient conditions. However, no storage stability assessment was performed between sampling and analysis; therefore, losses cannot be excluded.

From an analytical perspective, for all substances with a $NOEC_{min}$, the LOQ is always less than $NOEC_{min}$ (Table S1), indicating that the analytical precision is sufficient to detect $NOEC_{min}$ exceedances. With the single exception of abamectin, the LOQ is always at least a factor 10 lower than the $NOEC_{min}$, indicating that underestimation of soil risk indicators due to quantification limits is modest. This is not always the case for the aquatic risk indicators. The equivalent LOQ, expressed as the soil LOQ divided by the K_d, is not always sufficient to quantify risk-relevant concentrations, potentially underestimating the risk. For a typical soil with $OC = 2.5\%$, the equivalent aqueous LOQ is greater than the SSD HC_5 and the SSD HC_{50} for 13 and two substances, respectively.

The choice of using long-term (chronic) data for the derivation of risk indicators was driven by the exposure scenario and the need to cover most of the 118 monitored substances. However, taxonomic diversity is low for soil toxicity data. For instance, microorganisms such as fungi and bacteria are not represented. Toxicity tests on microorganisms target functional endpoints (mineralization, nitrification) and are generally not particularly sensitive to pesticide exposure. Soil NOEC data are, in many cases, available only for annelids (mostly earthworms) and, for a subset of substances, for collembola and mites.

At least one experimental NOEC was available for most of the substances. The three databases, from which we extracted the experimental data, implement different approaches of data quality control. For example, inclusion in OpenFoodTox implies that the study was evaluated for inclusion in EFSA opinions, whereas the study records in ECOTOX do not go through the same level of regulatory scrutiny. Some NOEC may be defined as the only or the highest concentration tested. These are reported as "greater than" (>) in OpenFoodTox and in PPDB, and were excluded from our compilation of $NOEC_{min}$ values, unless they were the only record available. Data gaps were partly filled with in silico estimates using the VEGA QSAR model or estimated from short-term toxicity tests. Substances contributing the most to the risk generally have a relatively large set of experimental data. For few substances, diverging data point to greater uncertainty in the risk estimates. For example, the two NOECs found for boscalid and difenoconazole, for reproductive effects on *F. candida* and *E. foetida*, vary by a factor of 1000 and 500, respectively (Table S2).

The dataset for aquatic species has a broader coverage of the substances and of the species tested. For both soil and aquatic risk indicators, data gaps are unlikely to influence the results. Of the 22 substances with no soil NOEC, nine were not detected. The remaining 13 are rarely detected above the LOQ, for example, chloridazon, making a modest contribution to the aquatic risk (Table S1).

Differences in exposure duration introduce additional uncertainty in the toxicity dataset. For soil NOECs, reported exposure durations range from 14 to 56 days. For aquatic data, exposure duration of chronic NOECs in the SSDs

depend on the species group. Standard exposure durations are 72–96 h for algae, 21 days for crustaceans, and 28 days for fish. However, NOECs from studies with shorter durations are also included. The lower limit ranges between one day (algae) and 4–7 days (fish; Posthuma et al., 2019).

The methodology underpinning the risk indicators inherits the limitations of the conventional regulatory risk assessment paradigm. In mixture risk calculations, calculating cumulative RQs using endpoints (e.g., $NOEC_{min}$) from different species reflects a worst-case scenario. Site-specific ecological conditions and protection goals are not considered. Further refinement of the indicator could consider the location of monitoring sites relative to specific habitats or Natura 2000 areas, where a higher level of protection is desired. Ongoing research, as proposed by the scoping study by Andres et al. (2022), is exploring how the risk could be modulated based on such spatial characteristics linking the risk to the desired protection level based, for example, on the sensitivity or biodiversity interest of habitats.

The future of LUCAS Soil Pesticides

The LUCAS program is essential for evaluating the impact of agriculture on the environment in Europe. The LUCAS Soil systematic campaigns achieve the long-term sustainability of data flows, which is critical for providing added value to policy. As part of the LUCAS Soil survey, LUCAS Soil Pesticides fills an important knowledge gap. The risk indicators presented support the implementation of EU as well as international policies establishing specific risk reduction targets, namely the regulation on the sustainable use of pesticides in the EU and the UN CBD. The latest LUCAS Soil survey was completed in 2023. Although laboratory analysis is currently ongoing, the final results are expected by 2024. Inputs from policy and scientific stakeholders are expected to update the scope of future surveys and possibly refine the methodology.

The proposed directive on soil monitoring and resilience (European Commission, 2023) sets a more stable legal mandate for the LUCAS Soil monitoring program. The proposal includes a soil pollution component, but monitoring is mandatory only for metals. Although pesticide residues are not mentioned explicitly, EU Member States can report such data if included in national monitoring programs. LUCAS Soil Pesticides could be included later in the directive.

AUTHOR CONTRIBUTION

Antonio Franco: Conceptualization; methodology; formal analysis; writing—original draft (lead); review and editing (equal). **Diana Vieira, Laure-Alix Clerbaux**: Formal analysis (supporting); original draft (supporting); review and editing (equal); **Edoardo Carnesecchi, Jean Lou Dorne**: Formal analysis (supporting); review and editing (equal); **Ruud van Dam, Vera Silva, Maeva Labouyrie, Julia Koeninger, Jeanne Vuaille, Joana Lobo Vicente, Alberto Orgiazzi**: Original draft (supporting); review and editing (equal). **Arwyn Jones**: Supervision; review and editing (equal).

ACKNOWLEDGMENT

The LUCAS Survey is coordinated by Unit E4 of the Statistical Office of the European Union (EUROSTAT). The LUCAS Soil sample collection is supported by the Directorate-General Environment (DG-ENV), Directorate-General Agriculture and Rural Development (DG-AGRI) and Directorate-General Climate Action (DG-CLIMA) of the European Commission. The authors acknowledge the assistance of Fabrizio Briganzoli (Unisystems) for the analysis of ecotoxicity data, and Alberto Pistocchi (JRC) and Hans Mol (WFSR) for reviewing the manuscript before submission to the journal. There are no funders to report.

CONFLICT OF INTEREST

The authors declare no conflicts of interest.

DATA AVAILABILITY STATEMENT

Pesticide residues monitoring data from the 2015 and 2018 surveys are stored in IPCHEM (https://ipchem.jrc.ec.europa.eu/#showmetadata/LUCASPESTICIDES2018) and also in the European Soil Data Centre (https://esdac.jrc.ec.europa.eu/). The dataset is subject to provisions of the EU General Data Protection Regulation (GDPR) and is therefore not public. Data aggregated by country can be provided upon request from jrc-ipchem-support@ec.europa.eu. Physicochemical, phase partitioning, and toxicity thresholds used in the calculations are reported in Table S1. Raw toxicity data records are retrieved from public repositories or estimated, as referenced in the article. The full compilation of raw data records used to derive toxicity thresholds is reported in Table S2. The R script used for the derivation of the toxicity thresholds is available at https://code.europa.eu/fraanto/jrc-chemical-risk-indicators.

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SUPPORTING INFORMATION

Figures S1 and S2 present results for the risk indicators developed for aquatic species based on the 5th and 50th percentile of hazard concentration of species sensitivity distributions (SSD HC_5 and HC_{50}) derived from aquatic No Observed Effect Concentrations (NOEC) data. Figures S3–S6 present risk rankings of target substances using different criteria based on soil (Figures S3 and S4) and aquatic toxicity (Figures S5 and S6). Table S1: List of substances monitored in the LUCAS Soil Pesticides 2018 survey with substance identifiers (CAS and InChiKey), limit of quantification (LOQ in mg/kg dw), information on the European Union regulatory status in July 2023 (with date of expiry of approval for the not-approved substances), and physicochemical and toxicity parameters used in the calculations. Further details and sources on the compilation of the datasets are described in the Supporting Information Word file. Table S2. Full collection of soil chronic toxicity records

 DOI: 10.1002/ieam.4917

used for the derivation of risk indicators, with database and reference as reported in the original database.

Analytical methods and details on substance data. Section 1 "Analytical methods" provides a more detailed description of the three analytical approaches used. Section 2 explains in more detail the sources and derivation of substance input data (physicochemical, soil–water partitioning, and toxicity parameters) reported in Table S1.

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