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Pesticide residues with hazard classifications relevant to non-target species including humans are omnipresent in the environment and farmer residences

Vera Silva [a,1], Lingtong Gai [a,1,*], Paula Harkes [a], Gaowei Tan [a], Coen J. Ritsema [a], Francisco Alcon [b], Josefa Contreras [b], Nelson Abrantes [c], Isabel Campos [c], Isabelle Baldi [d], Mathilde Bureau [d], Florian Christ [e], Daniele Mandrioli [f], Daria Sgargi [f], Igor Pasković [g], Marija Polić Pasković [g], Matjaž Glavan [h], Jakub Hofman [i], Esperanza Huerta Lwanga [a], Trine Norgaard [j], Zuzana Bílková [i], Rima Osman [a], Chrow Khurshid [a], Irene Navarro [k], Adrián de la Torre [k], Paloma Sanz [k], María Ángeles Martínez [k], Jonatan Dias [l], Hans Mol [l], Gerrit Gort [m], Daniel Martins Figueiredo [n], Paul T.J. Scheepers [o], Vivi Schlünssen [p], Anne Vested [p], Abdallah Alaoui [e], Violette Geissen [a]

[a] Soil Physics and Land Management Group, Wageningen University & Research, Netherlands
[b] Agricultural Engineering School, Universidad Politécnica de Cartagena, Spain
[c] CESAM and Department of Biology, University of Aveiro, Portugal
[d] Univ. Bordeaux, INSERM, BPH, U1219, F-33000 Bordeaux, France
[e] Institute of Geography, University of Bern, Hallerstrasse 12, 3012 Bern, Switzerland
[f] Cesare Maltoni Cancer Research Center, Ramazzini Institute, Italy
[g] Department of Agriculture and Nutrition, Institute of Agriculture and Tourism, K. Huguesa 8, 52440 Poreč, Croatia
[h] Agronomy Department, Biotechnical Faculty, University of Ljubljana, Jamnikarjeva 101, 1000 Ljubljana, Slovenia
[i] RECETOX, Faculty of Science, Masaryk University, Brno, The Czech Republic
[j] Department of Agroecology, Aarhus University, Blichers Allé 20, 8830 Tjele, Denmark
[k] Unit of POPs and Emerging Pollutants in Environment, Department of Environment, CIEMAT, Madrid, Spain
[l] Wageningen Food Safety Research (WFSR), part of Wageningen University & Research, Wageningen, The Netherlands
[m] Biometris, Wageningen University, The Netherlands
[n] Institute for Risk Assessment Sciences, Utrecht University, Utrecht, The Netherlands
[o] Radboud Institute for Biological and Environmental Sciences, Radboud University, Nijmegen, the Netherlands
[p] Department of Public Health, Research Unit for Environment, Occupation and Health, Danish Ramazzini Centre, Aarhus University, Aarhus, Denmark

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ABSTRACT

Intensive and widespread use of pesticides raises serious environmental and human health concerns. The presence and levels of 209 pesticide residues (active substances and transformation products) in 625 environmental samples (201 soil, 193 crop, 20 outdoor air, 115 indoor dust, 58 surface water, and 38 sediment samples) have been studied. The samples were collected during the 2021 growing season, across 10 study sites, covering the main European crops, and conventional and organic farming systems. We profiled the pesticide residues found in the different matrices using existing hazard classifications towards non-target organisms and humans. Combining monitoring data and hazard information, we developed an indicator for the prioritization of pesticides, which can support policy decisions and sustainable pesticide use transitions. Eighty-six percent of the samples had at least one residue above the respective limit of detection. One hundred residues were found in soil, 112 in water, 99 in sediments, 78 in crops, 76 in outdoor air, and 197 in indoor dust. The number, levels, and profile of residues varied between farming systems. Our results show that non-approved compounds still represent a significant part of environmental cocktails and should be accounted for in monitoring programs and risk

* Corresponding author at: Soil Physics and Land Management Group, Wageningen University & Research, Droevendaalsesteeg 4, 6708 PB Wageningen, The Netherlands.
E-mail addresses: lingtong.gai@wur.nl, sprint@wur.nl (L. Gai).
[1] Share the first authorship on this work.

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assessments. The hazard profiles analysis confirms the dominance of compounds of low-moderate hazard and underscores the high hazard of some approved compounds and recurring "no data available" situations. Overall, our results support the idea that risk should be assessed in a mixture context, taking environmentally relevant mixtures into consideration. We have uncovered uncertainties and data gaps that should be addressed, as well as the policy implications at the EU approval status level. Our newly introduced indicator can help identify research priority areas, and act as a reference for targeted scenarios set forth in the Farm to Fork pesticide reduction goals.

1. Introduction

Synthetic pesticides, introduced into the agricultural sector in the 1940 s to decrease yield losses caused by pests and diseases, have become a foundation for modern agriculture (Pimentel, 1996). Since then, thousands of active substances have been approved in the European Union (EU) for pest control. Over time, many of these were banned due to knowledge gained *a posteriori* on their high persistence and/or toxicity to non-target species (EASAC, 2023a; OJL, 2023). At the beginning of 2021, the year when the sampling campaign related to this study took place, there were 489 active substances approved for pesticide use in Europe, of which over 340 were synthetic (EU, 2021). Pesticide use in Europe has been stable over the last decade, with almost 346,000 tonnes of pesticides sold in the 27 member states of the EU in 2020 (EUROSTAT, 2023).

Although there is a rigorous pesticide pre-market entry risk assessment procedure in Europe, European-wide post-market monitoring programs, and pesticide-related quality benchmarks, are only defined for drinking water and food (Carrasco Cabrera et al., 2023; EC, 1998; EC, 2000; EC, 2005; EFSA, 2013; EU, 2008). Monitoring data for other environmental matrices are rather scattered (Kruse-Plaß et al., 2021; M. Figueiredo et al., 2022; Peris et al., 2022; Zaller et al., 2022). However, there is substantial progress being made concerning soils: after Silva et al. (2019) investigated the contamination of European agricultural soils by pesticide residues using 317 samples from the 2015 LUCAS survey (Land Use and Coverage Area frame Survey), a pesticide module was included in the 2018 LUCAS program. This module is expected to be expanded in the 2022 LUCAS program (Orgiazzi et al., 2022; Vieira et al., 2023). Moreover, on July 5th 2023, the EU proposed a new Soil Monitoring Law to protect and restore soils (EC, 2023c). Although limited, environmental monitoring data indicate that mixtures of pesticide residues are the rule rather than the exception. Harmonized and large-scale monitoring data on (mixtures of) pesticide residues is urgently needed to support the assessment of the risk posed by pesticides to ecosystems health. Such a point is highlighted in the protection goals approach suggested by EFSA (2016). Ultimately, this kind of data is also relevant for human health assessments. Although diet is known to be the major exposure route to consumers, environmental exposure may add to that. For farm workers and residents close to agricultural fields, environmental exposure may add more significantly to the overall pesticide exposure (HBM4EU, 2022). In fact, the International Labour Organization of the United Nations has now included pesticides in the top ten chemical exposures of concern for workers (ILO, 2021).

In 2020, the European Commission announced two pesticide reduction targets as part of its Farm to Fork strategy: a reduction of 50 % in the use of and risk from chemical pesticides by 2030, and a reduction of 50 % in the use of the more hazardous pesticides by 2030 (EC, 2020). More hazardous pesticides refer to candidates for substitution (CfS) - i.e., active substances to be substituted or removed when a viable pest control alternative is available (EC, 2023b). The first reduction target is monitored using the Harmonised Risk Indicator 1, HRI1 (EU, 2009), which accounts for pesticide sales and pesticide hazard properties. The second target is monitored via sales of CfS (EC, 2017). Despite concerns about the suitability of these indicators, such as the fact that actual pesticide use is not accounted for, or the increasing situation of pesticides used in derogation (PAN, 2021), these are, at least for now, the official tools to assess progress. Recent data from the European Commission (EC, 2023a) show a decreasing trend in the use of and risk from chemical pesticides (14 %), and in the use of CfS (26 %). However, concrete action plans to guarantee that the Farm to Fork pesticide targets are reached are missing. These may include generic recommendations per crop system, or more profound changes such as conversion to organic farming, or severe restrictions on the pesticides available on the market (Gauthier, 2020; Silva et al., 2022). Monitoring and risk data could support policymakers in such decisions (Chow et al., 2020; United Nations Environment Programme, 2022; Vijver et al., 2017). In this paper, we propose a method for prioritization of pesticides by integrating occurrence and distribution data with hazard classifications (Chow et al., 2020; Huber et al., 2022; Silva et al., 2019). Some pesticide prioritization indicators have been developed over the last few years (Tsaboula et al., 2016; Tsaboula et al., 2019a; Tsaboula et al., 2019b; Carazo-Rojas et al., 2018 and Vryzas et al., 2020). The prioritization strategies and the indicator applications are highly variable among studies, see Vryzas et al., (2020) for an overview. The approach and goal of the EC-HRI1 indicator is the closest to ours. The HRI1 indicator, applied to evaluate progress toward the Farm to Fork 50 % use and risk reduction goal, divides pesticides into a few hazard/approval status-related groups, and accounts for sales of active substances. The monitoring and hazard data considered in our indicator was generated and compiled under the H2020 SPRINT project umbrella (https://sprint-h2020.eu/). The specific objectives of this study were to:

a) assess the occurrence and levels of pesticide residues in soil, crop, water, sediment, indoor dust, and outdoor air. Samples originated from 10 European countries, covering main European crop types, and conventional and organic farming systems. Previous works have shown that samples from organic farms are not necessarily pesticide-free, often presenting a mix of persistent and currently use pesticides (from historical pesticide use and off-site contamination, respectively);
b) characterize hazards for non-target organisms including humans, based on intrinsic hazard properties of the pesticide residues detected in the different matrices; and
c) introduce a prioritization indicator for pesticides based on occurrence and existing hazard classifications. Higher scored compounds are considered to be of higher interest for pesticide use and risk reduction actions.

2. Methods

2.1. Field campaign

This study describes the results from 625 environmental samples collected during the 2021 growing season, across 10 Case Study Sites (CSS; Fig. 1). These CSS covered the main crop types in Europe (viticulture, horticulture, root crops, fruits trees, olives, cereals, and oil plants) and the three EU-pesticide regulatory zones (North, Central and South) (Silva et al., 2021). The size of the sampling region varied among CSS, due to CSS characteristics and the feasibility of meeting the SPRINT monitoring plan requirements (SPRINT, 2021). We sampled one crop class per CSS, the one with the highest abundance or relevance for the study design. In each CSS, we sampled a similar number of organically and conventionally managed fields – hereinafter referred to as organic fields (OF) and conventional fields (CF). The OF were managed as such

for at least 5 years before our sampling campaign. The CF follow Integrated Pest Management principles [IPM, compulsory in the EU since 2014 (EC, 2009)].

We collected soil, crop, and outdoor air samples from CF and OF, indoor dust samples from the farmers households, and water and sediment samples from waterbodies connected to or close to the sampled fields. The number of samples collected per matrix and CSS is given in Table 1. A brief overview of the sampling methods is presented in *Supplementary Material*. For outdoor air, passive sampling was employed, using a sampler developed by TIEM technic, described in Kruse-Plaß et al. (2021). This device contained two sampling units: i) a polyurethane foam (PUF) disk enclosed in a sampler dome, open to air but protected from sun, rain, and particulate matter, that captures (semi) volatile pesticides from the gas phase, and ii) four polyester filters (PEF), positioned underneath the dome and openly exposed to air, that captures particulate matter for analysis of glyphosate and AMPA.

Soil, water, sediment, indoor dust, and outdoor air samples were collected in the middle of the growing season, crops were collected at harvest time. The middle of the growing season corresponded to the moment when most of the pesticide applications of the year were already carried out. The middle of the growing season varied across CSS, based on specific crop-region timelines. For most CSS this corresponded to the end of May-beginning of June. For CSS2 and CSS5 with a later growing season, this corresponded to September-October.

2.2. The list of analytes

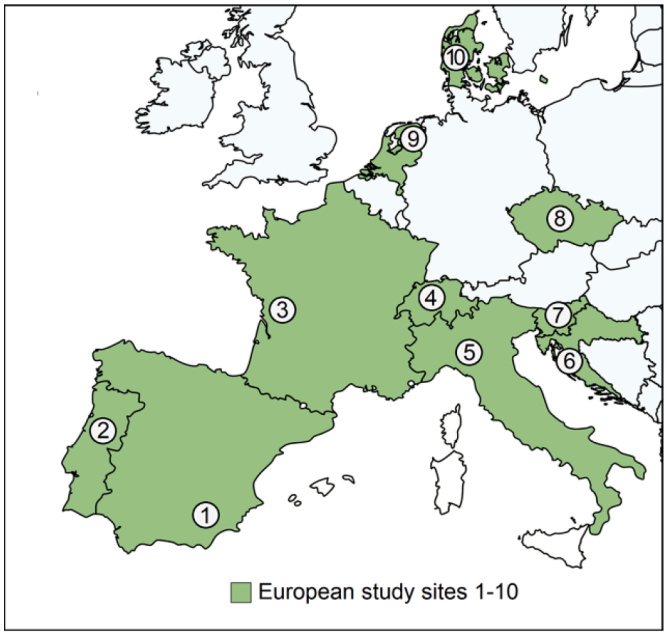
During the initiation phase of the field campaign, a target list of 209 pesticide residues to be measured in all samples was established (*Table SM1)*. The rationale behind the 209 analytes list was presented in Silva et al. (2021). Briefly, this considered information on (possible) used pesticides for plant protection in each CSS situation, known occurrences in the environment and food, and results from a full scan screening of soil, sediment and dust collected from the CSS regions prior to the sampling campaign. For practical reasons, analytes not amenable to multi-residue methods had to be excluded, except for glyphosate and AMPA. The list included 164 active substances of plant protection products, 44 transformation products, and 1 synergist (piperonyl butoxide). Of the 164 active substances tested, 118 were approved for plant protection use in Europe (29 of which CfS), and 46 were non-approved. Approval status refers to European status (retrieved from the EU pesticide database) as of 01/01/2021. Transformation products were linked to parent compound approval status (30 approved compounds – 5 of which are CfS, and 14 non-approved). This list covers mostly synthetic organic pesticides but also natural substances: azadirachtin, pyrethrins I/II, spinetoram, spinosad (spinosyn A&D).

The analytes in the various matrices were measured by quantitative multi-methods based on LC-MS/MS and GC–MS/MS. For glyphosate/AMPA, separate dedicated methods were used. All methods were validated before sample analysis (EURL, 2021). A brief overview of the analytical methods is presented in the *Supplementary Material*. A detailed description will be provided in separate, matrix-specific papers (soil, sediment and indoor dust papers are currently in the peer-review process). However, it was not always possible to measure the 209 analytes in all matrices/samples. Some compounds were excluded because analytical reference standards were not available or because of low analytical performance (e.g., recoveries < 70 %). We ended up analysing 198 pesticide residues in indoor dust, 196 in sediment, 193 in water, 192 in soil and plants, and 161 in outdoor air. In total, 142 residues were analysed in all six matrices.

The limit of detection (LOD), i.e., the lowest level at which the pesticide residues can be detected and identified, was used as the analytical and data analyses reporting limit. The LOD values varied per compound and matrix (see *Table SM1*). In indoor dust samples, LODs varied from 0.012 to 38 μg kg^{-1} (median: 0.38), in water from 0.001 to 14.6 ng/L (median: 0.96), in sediment from 0.010 to 4.44 μg kg^{-1} (median: 0.28), and in soil and crops from 0.33 to 16.67 μg kg^{-1} (median soil: 1.33, median crop: 0.67). TIEM PUF disks and PEF filters were analysed by an external lab that provided only limits of quantification (LOQ). These ranged from 0.008 to 0.100 μg $sample^{-1}$ (median: 0.01). Air concentrations (in ng/m^{-3}(−|-)) could not be derived by calculation due to lack of information on the diffusion behavior of the target pesticides and transformation products.

2.3. Further characterization of the analytes

Basic properties of the 209 analytes were compiled (*Table SM2*) to support compound-specific evaluations. Such data was extracted from



Case study site	Region	Crop
1. Spain (SP)	Cartagena	Vegetables - Broccoli
2. Portugal (PT)	Bairrada	Vine - Grapes
3. France (FR)	Bordeaux	Vine - Grapes
4. Switzerland (CH)	Canton Bern	Orchards - Apple, plum, strawberry, pears, cherries
5. Italy (IT)	Po region	Vegetables - black cabbage, peppers, radicchio, green cabbage, salad, broccoli
6. Croatia (HR)	Istria	Olive trees
7. Slovenia (SL)	Central zone	Maize
8. Czech Republic (CZ)	Central zone	Oilseed rape, sunflower, poppy, mustard
9. The Netherlands (NL)	Groningen and Friesland	Potatoes
10. Denmark (DK)	Central Zone	Cereals spring barley, winter wheat, winter rye, oats

Fig. 1. Case Study Site (CSS) locations and main characteristics. Green represents sampled countries, the white numbered circles represent the CSS sampling regions. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Table 1
Number of samples collected across the Case Study Sites per farming system. CF = conventional fields, OF = organic fields, n = number of samples, SP – Spain; PT – Portugal; FR – France; CH – Switzerland; IT – Italy; HR – Croatia; SL – Slovenia; CZ – Czech Republic; NL – the Netherlands; DK – Denmark.

	SP		PT		FR		CH		IT		HR		SL		CZ		NL		DK	
	CF	OF	CF	OF	C F	OF	CF	OF	CF	OF	CF	OF	CF	OF	CF	OF	CF	OF	CF	OF
Soil (n = 201)	10	10	12	8	7	10	10	10	10	10	10	10	10	10	11	13	10	10	10	10
Crop (n = 193)	10	10	12	8	5	8	10	10	10	10	10	10	10	10	11	10	10	10	10	9
Air outdoor (n = 20)	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Indoor dust (n = 115)	5	4	4	1	6	7	5	7	2	5	9	7	6	6	7	7	9	7	5	6
Water (n = 58)	7		8		6		5		6		3		6		8		6		3	
Sediment (n = 38)	0		8		5		5		0		3		6		3		5		3	

the Pesticide Properties DataBase [PPDB, see (Lewis et al., 2016) for details on the database] and cover: i) solubility in water at 20 °C (mg L^{-1}); ii) octanol–water partition coefficient at pH 7, 20 °C, logP; iii) soil degradation (DT_{50} typical, days); iv) water–sediment degradation (DT_{50}, days); v) soil adsorption coefficient (kd and Koc), vi) vapour pressure at 20 °C (mPa), and vii) Henry's Law Constant at 25 °C (Pa m^3 mol^{-1}).

The hazard of tested residues to non-target organisms was also retrieved from the PPDB, in April 2023. All terrestrial and aquatic ecotoxicology entries available in the PPDB, in terms of organisms, exposure routes, and duration of exposure, were considered. We compiled the qualitative information available in the database high, moderate, low hazard. Such classes are defined via comparison of standard toxicological dose descriptors (EC_{50}/LC_{50}/NOEC values) with endpoint-specific thresholds. The toxicological descriptors, and the endpoints covered in the database, are in line with those required by EFSA. The data is mostly all verified data used for regulatory purposes, and the thresholds considered are consistant with EU regulatory thresholds (PPDB, 2023). In cases of 'no data available' in PPDB, we looked for the missing information on individual residues in the EFSA conclusion reports (accessible via), and applied PPDB thresholds. This is mostly because recent EFSA data may not been integrated into PPDB at the time we retrieved the hazard information from the database. We also considered the possibility of some PPDB entries not being updated correctly towards EFSA reports, but when we compared PPBD and EFSA data for mammals, these were a rather good match (despite the fact that PPDB has additional data sources). The data originating from the EFSA reports (7 % of the cells in *Table* SM3) is presented in yellow highlighted cells. Our final hazard dataset still includes some gaps as we could not find EFSA reports for long-banned compounds.

We also compiled qualitative data for eleven specific human health issues: carcinogenic, genotoxic, endocrine disruptor, reproduction/development effects, acetyl cholinesterase inhibitor, neurotoxicant, respiratory tract irritant, skin irritant, skins sensitizer, eye irritant, and phototoxicant. For each one of these issues PPDB provides one of the following classifications: 'yes, known to cause a problem', 'possible, status not identified', or 'no, known to not cause a problem'. 'No data' available situations may happen here as well. Since literature is not universally uniform in the way these health issues are addressed, PPDB uses a 'weight-of-the-evidence' approach erring on the side of caution (PPDB, 2023). Other classifications systems, and even other databases, may be considered more adequate for human-specific assessments, but not explored here as this was not the primary focus of the current study.

2.4. Data analysis

2.4.1. Numbers, type, and levels of pesticide residues

This overview focuses on multi-analyte and multi-matrix analyses. Analysis over analytes was performed with boxplots, on the total number of residues and total/cumulative pesticide concentration in individual samples. The number of pesticides gives an indication of the complexity of the mixtures, the total pesticide concentration of overall levels of contamination. This last parameter, although rather simplistic and possibly overly aggregated, is being increasingly used in sample and site characterizations (Bhandari et al., 2020; Tang and Maggi, 2021; Vieira et al., 2023; Froger et al., 2023), being potentially relevant for the development and interpretation of chemical/health status thresholds.

Analysis over matrices was performed by multi-matrix distribution heatmaps, and prioritization indicator curves. Both approaches rely on the frequency of detection and the median concentrations of individual analytes across the matrices. The frequency of detection of a pesticide residue *x* was calculated as the total number of entries where it was ≥ LOD divided by the number of samples where tested. Total pesticide concentrations, and the median concentrations, as well as the 5th and 95th percentile concentrations of individual compounds (introduced in 2.4.2 section), were calculated accounting only ≥ LOD entries.

The type of residues found in the samples was evaluated based on their approval status in Europe and the hazard profile. The total number and median concentration of approved, CfS and not approved residues per matrix is presented in simple stacked column charts. For the case of simplicity, metabolites/transformation products were attributed to the same classes as parent compounds. The hazard analyses (presented in 100 % stacked column charts) involved a simple correspondence between the matrices and the (eco)toxicological data compiled from the PPDB (Fig. 2). We identified the organisms expected to be directly or indirectly affected by the presence of contaminants in a certain matrix, and explored the hazard profile of the pesticide residues detected in that matrix to those organisms.

Statistical analyses were performed for the numbers and total pesticide concentrations, between different farming systems within soil, crop, outdoor air, and indoor dust datasets. The different number of analytes and sampling schemes across matrices hamper further statistical analyses. The CF and OF statistical comparisons were made using Linear Mixed Models (LMM) and Generalized Linear Mixed Models (GLMM). For the total pesticide concentration, a normal distribution was tested, after log-transformation and addition of 0.1 (Sokal & Rohlf, 1995). This value was added to consider all samples in the analyses, including those where no analytes were found ≥ LOD (i.e.,'pesticide free' samples). Since the crop dataset contained many 'pesticide-free' samples, a GLMM with the Tweedie distribution and log link was employed to analyse the total pesticide concentration. The number of pesticides was analysed using GLMM's assuming a Poisson distribution. For the comparison of farming systems, the GLMM included Conventional/Organic classification as fixed effect, and country and country by farming system interaction as random effects. An additional random effect was introduced to handle observations from the same field. This was done to avoid that a higher pesticide use, or even misuse of pesticides, in some fields becoming a confounding factor in such overarching assessment. Differences between farm types were tested, either with approximate F-tests using the Kenward-Roger method (in case of LMM's) or Wald tests (in case of GLMM's). The statistical models were

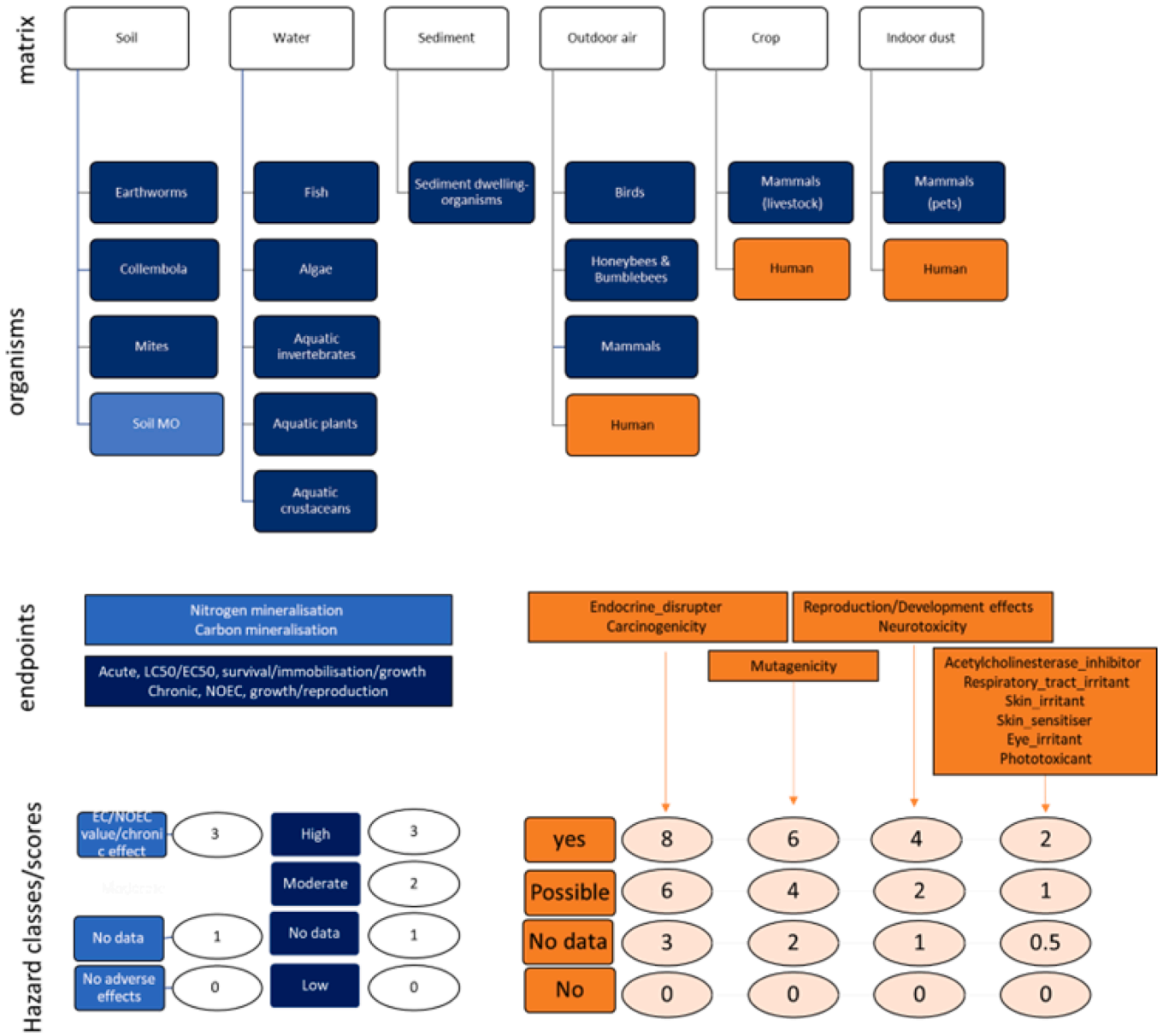


Fig. 2. Correspondence between the sampled matrices, non-target organisms, endpoints, and hazard classes and hazard scores (inside the oval shapes). EC_{50} = Effect concentration, 50 % (the concentration of a chemical that can be expected to cause a defined nonlethal effect in 50 % of the tested population); LC_{50} = Lethal Concentration, 50 % (concentration required to kill half the tested population). NOEC = No Observed Effect Concentration (greatest level or concentration of a substance, found by observation or experiment, which causes no detectable effect). AChE = Acetylcholinesterase. Shapes colored in blue refer to ecosystem assessment, in orange to human assessment. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

run in R v4.2.1 (R Core Team, 2021), using packages lme4 (Bates et al., 2015), glmmTMB (Brooks et al., 2017) and emmeans (Lenth, 2022). Model assumptions were checked using diagnostic residual plots obtained with package DHARMa (Hartig, 2022).

2.4.2. Pesticide prioritization indicator (PPI)

A new pesticide prioritization indicator was developed based on occurrence data and pesticide hazard profiles. The indicator was calculated as follows:

$$\text{PPI}_X \text{ for ecosystem} = \sum_{i=1}^{6} (FDi^{*}Ci^{*}HHSi) \quad (1)$$

$$\text{PPI}_X \text{for human} = \sum_{i=1}^{3} (FDi^{*}Ci)^{*}HHSHIi \quad (2)$$

$$\text{PPI}_X \text{for matrix} = FD^{*}C^{*}HHS \quad (3)$$

$$\text{Cumulative PPI for matrix} = \sum_{i=analyte\ 1}^{analyte\ 209} PPIi \quad (4)$$

where x = pesticide (residue) being considered; FD = frequency of detection of pesticide x in the matrix being considered; C = median concentration of pesticide x in the matrix being considered; HHS = highest hazard score of the residue x among organisms related to the matrix being considered; 1 = crop, 2 = outdoor air, 3 = indoor dust, 4 = water, 5 = sediment, 6 = soil; and HHSHI = highest hazard score of the residue x among the eleven specific human health issues considered in the study. For matrix-specific assessments (Eq. (3), Figures SM4-13), 5th and 95th percentile concentrations were also accounted for to display uncertainty.

Due to the highly variable nature and severity of hazards/effects covered by ecosystem and human domains, ecosystem and human prioritization assessments are presented as two stand-alone systems. For the ecosystem assessment, we attributed a 0 to 3 score to the PPDB hazard classes (low, no data, moderate and high, respectively). This scoring system, introduced by Silva et al. (2022), makes no differentiation among organisms or endpoints. By considering the highest hazard score for the pesticide residues among the organisms of each matrix, our prioritization indicator value reflected a worst-case scenario. Accounting for the hazard scores for all standard organisms (versus the most sensitive one per matrix) could lead to a biased value, as organisms' representation is highly variable among matrices (see number of dark blue boxes in Fig. 2). For the human assessment, we used a scoring system adapted from Dabrowski et al. (2014) and Valcke et al. (2005). This system ranges from 0 to 4, 0 to 6 or 0 to 8 depending on the health issue considered. However, it covers only highly severe effects (i.e., carcinogenic, endocrine disrupter, mutagen, reproductive/development, neurotoxicant). We added a 0 to 2 scale for the lower severity effects such as skin irritant or skin sensitizer (see Fig. 2 for further details and hazard scores attribution per health issue).

PPI values were plotted in two ways. The first, truer to the indicator name, involved sorting residues based on increasing PPI values. This allows us not only to rank but also to define and numerically compare the relevance of different pesticide residues for ecosystem and human health assessments (Eqs. (1) and (2)). This is expected to be particularly relevant for pesticide reduction measures. The second, more applied manner, involved plotting the PPI values in matrix cumulative curves (Eqs. (3) and (4)). This allows us to quantify the current pesticide pressure on ecosystems and humans, quantify contribution of individual residues towards the bulk situation, and define benchmark values for each matrix.

2.4.3. Sensitivity analyses

Uncertainty was addressed in the different analyses covered in this study. First, due to a considerable number of < LOD entries across the analytical datasets we performed imputation of such values as sensitivity analysis. Imputation was performed at matrix level, for compounds detected in at least 40 % of the samples. This threshold was chosen based on previous studies that have imputed pesticide concentrations across different matrices (Fuhrimann et al., 2022; Oerlemans et al., 2021). We imputed left-censored values based on truncated distribution. Imputation was done using maximum-likelihood (Lubin et al., 2004) based on log10 transformed pesticide values and country as a predictor. This estimation accounts for both correlation and distribution of all pesticide data applying the R package "survival". Medians of truncated distributions (i.e., non-imputed data, accounting ≥ LOD values only) were compared to the medians of full distributions (non-imputed + imputed data) using the Mann-Whitney *U* test. Further details on the imputation exercise and the imputation results themselves are presented in Supplementary Material.

Then, considering the substantial amount of "no data available" entries in the hazard dataset, we prepared additional ecosystem- and human-PPI curves where we attributed a hazard score of 0 ('no adverse effect') or of 2–8 ('high' or 'yes', depending on the endpoint considered, see Fig. 2) to this hazard class. Such curves, presented in Supplementary Material, aim to represent the best- and worst-case scenarios (i.e. current "no data available" entries turn out to be low or high hazard, respectively). The shape of these curves and list of residues (presented in the Supplementary Material) with the highest PPI values (assumed to be of higher concern) corroborate those in the main manuscript.

Finally, in the matrix-specific curves (Figures also in *Supplementary material*), PPI values derived from situations where 'no hazard data available' and low persistence compounds were highlighted graphically. This because 'no hazard data available' residues can be ranked differently if the hazard data becomes available, and residues of low persistence are considered to be potentially less urgent since frequency and levels are expected to be markedly reduced after the sampling time/growing season. In these curves, the PPI values are presented with horizontal grey bars. The bar limits correspond to PPI scores associated with the 5th and 95th percentile concentration of the pesticide residue under consideration.

3. Results

3.1. Number, type and levels of pesticide residues found

Analytical results revealed the co-occurrence of pesticide residues in most of the samples, with the number and total pesticide concentration varying across matrices and farming systems. In soil, the primary sink of pesticides in agricultural fields, we detected 100 different pesticide residues, 96 in soils CF (51 approved for pesticide use on the European market, 23 CfS, and 22 not approved), 49 residues in soils from OF (23 approved, 11 CfS, 15 not approved; Figs. 3 and 4). Forty-five residues were found in both CF and OF soils. The numbers of residues in a sample varied between 0 and 21 in CF soils (median = 8), and 0–12 in OF soils (median = 3; Fig. 3). 99 % of the CF soils contained pesticide residues; 96 % contained mixtures of 2 or more residues. The OF soils presented slightly lower figures: 95 % of OF soils had at least 1 pesticide residue, 79 % had mixtures of residues. p,p'-DDE was the only compound detected in more than 50 % of samples, in both CF and OF soils (*Figure SM1-3*). Total pesticide concentrations reached values as high as 28,678 μg kg^{-1} in CF soils, and 5,458 μg kg^{-1} in OF soils. CF soils presented a median value of total pesticide concentration 8 times higher than OF soils (253 vs 31 μg kg^{-1}). AMPA and dieldrin were the compounds with the highest median concentration in CF and OF soils, respectively (*Figure SM1-3*).

In crops, we detected 78 pesticide residues; 76 in CF samples (43 approved, 16 CfS, 17 not approved), 25 in OF samples (9 approved, 7 CfS, 9 not approved). Twenty-three residues were common to both farming systems. In samples from CF, we found a maximum of 18 residues per sample (median = 3) and in samples from OF, we found a maximum of 5 residues per sample (median = 0). Over 63 % of tested crop samples contained at least 1 pesticide residue, 43 % had mixtures of residues. The percentage of samples containing pesticide residues was higher for CF than for OF (86 vs 40 %). Metalaxyl-M and dieldrin were the most common compounds in CF and OF samples, respectively (*Figure SM1-3*). In samples from CF, total pesticide concentration reached 951 μg kg^{-1} (median 23.7 μg kg^{-1}), and in samples from OF, maximum total pesticide concentration was 228 μg kg^{-1} (median 0.0 μg kg^{-1}). Pyrimethanil and cypermethrin had the highest maximum concentrations in both CF and OF samples, respectively.

In the outdoor air, we found a total of 76 residues. All samples presented pesticide residues (minimum 3, maximum 26). In CF samples, a total of 65 residues were found (40 approved, 18 CfS, 7 not approved; minimum–maximum 7–26 residues/sample, median 16 residues/sample). In OF samples, the number was slightly lower with a total of 53 residues (35 approved, 8 CfS, and 10 not approved; 3–26 residues $sample^{-1}$, median 10 residues $sample^{-1}$). Remarkably, 42 residues were common to CF and OF samples. Glyphosate was found in all PEF filters, and AMPA in 75 % of the samples. Pendimethalin was the most common compound in the PUF disks. Pirimicarb and metalaxyl-M were the compounds with the highest median concentration in the PUF-CF and PUF-OF samples, respectively (pirimicarb: 5.0 μg $sample^{-1}$, frequency 10 %; metalaxyl-M: 2.0 μg $sample^{-1}$, frequency: 20 %). The median and maximum total pesticide concentration in CF samplers was 2 to 3-fold higher than in OF samplers (median: 2.6 vs 0.8 μg $sample^{-1}$, maximum: 26 vs 14 μg $sample^{-1}$).

In indoor dust samples, 197 residues were detected, out of the 198 that were tested in this matrix (exception: chlorpyrifos-methyl-desmethyl). 195 residues were found in dust from conventional farmer farmhouses (106 approved, 34 CfS, 54 not approved, 1 synergist), 190 in organic farmer farmhouses (103 approved, 32 CfS, 54 not approved, 1

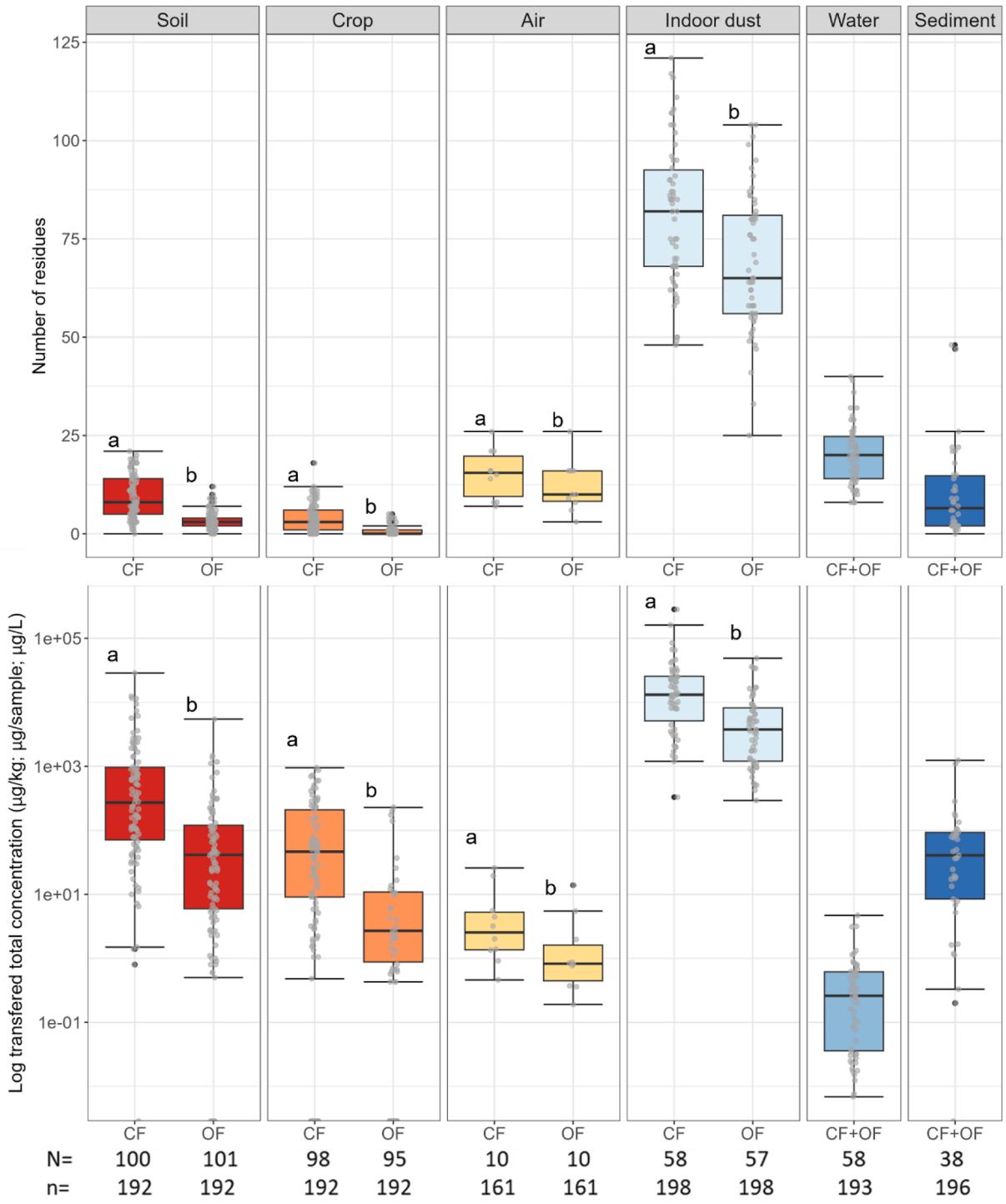


Fig. 3. Number of pesticide residues detected (top), and total pesticide concentration measured (bottom) in the different matrices, across all Case Study Sites. Detection refers to number of residues ≥ limit of detection, which vary among residues and matrices (see Table SM2). Concentration units vary among matrices: µg kg^{-1} for soil, plant, indoor dust and sediments; µg $sample^{-1}$ for air; and µg/L for water. The boxplots represent core data (25 % percentile, median, 75 % percentile; whiskers = 1.5 times the interquartile range, or minimum/maximum; black dots = outliers), the grey dots represent individual samples. N = number of tested samples, n = number of tested pesticide residues. Different letters (a and b) indicate significant differences between farming systems.

synergist). All indoor dust samples presented mixtures of pesticide residues. In conventional farmhouses, we found a minimum of 48 residues, a maximum of 121, and a median of 82. In organic farmhouses, we found a minimum of 25 residues, a maximum of 104, and a median of 65. Fludioxonil, hexachlorobenzene, imidacloprid, and piperonyl butoxide were found in all the 115 indoor dust samples. Twenty other compounds were found in more than 90 % of the samples (*Figure SM1-3*). In this matrix, we found total pesticide concentrations as high as 283, 000 µg kg^{-1}. Glyphosate and 2,4-D had the highest median concentrations in conventional and organic farmhouses, respectively.

In water samples, a total of 112 residues were detected (57 approved, 19 CfS, 35 not approved, 1 synergist). All water samples presented pesticide residues (minimum 8, maximum 40, median 20). Hexachlorobenzene and lindane were found in all water samples, and dieldrin, glyphosate and p.p'-DDE in almost all samples (note the different LODs among pesticide residues and matrices). Total pesticide

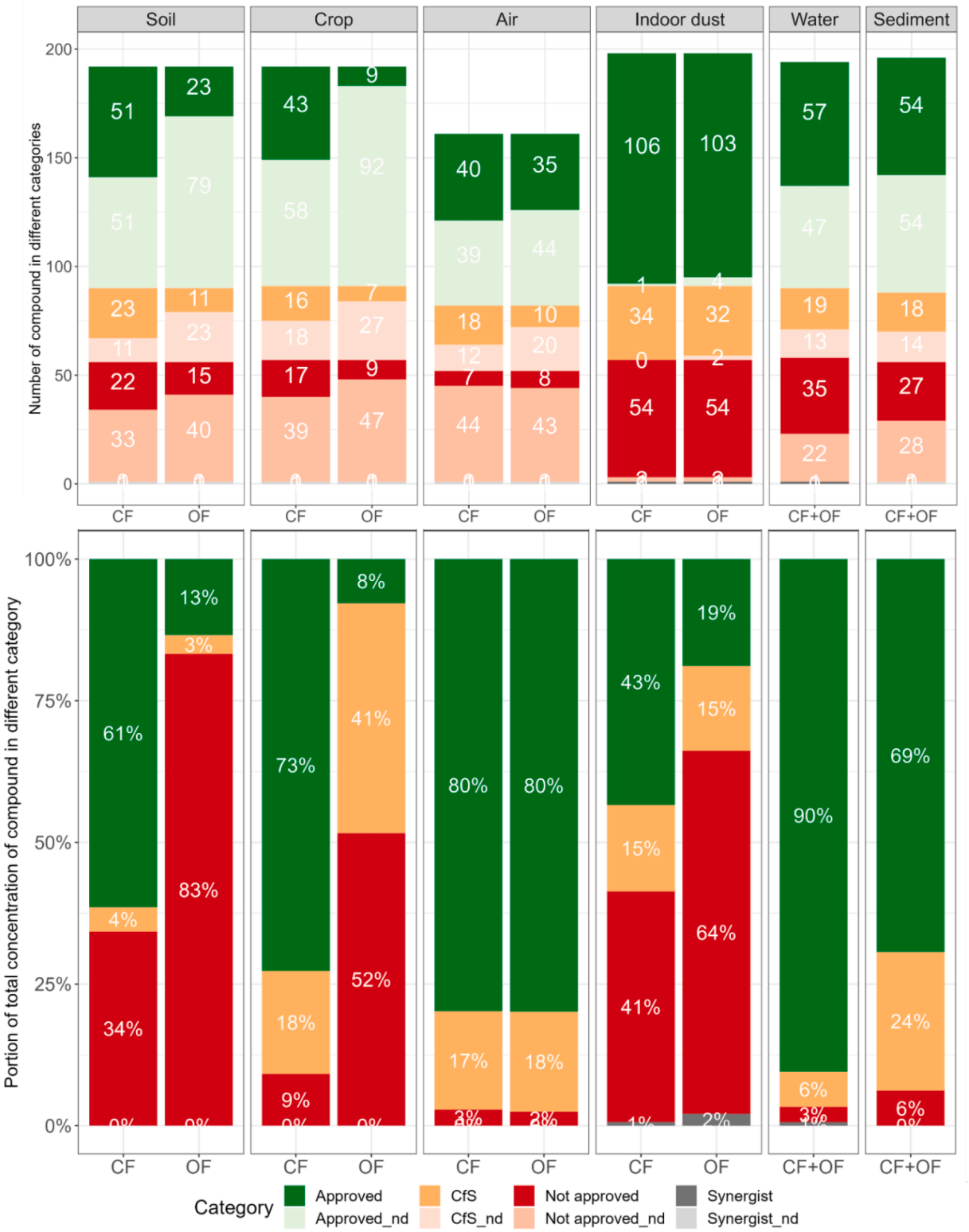


Fig. 4. Type of compounds found per matrix (top), and respective contribution to total pesticide concentration (bottom). The upper stacked columns cover the total number of residues tested and detected according to their approval status in the EU market: approved (A), candidates for substitution (CfS) and not approved (NA). The bottom columns show the median concentration of A, CfS, NA classes (here only detected residues are accounted).

concentration ranged from < 1 to 4.7 μg/L. Glyphosate was found in the highest frequency and concentration in this matrix. Azoxystrobin-O-demethyl, spinetoram, dimethenamid-P, fenhexamid, haloxyfop-P and fluroxypyr had slightly higher levels than glyphosate but were found in only a few samples.

In the sediment of surface water bodies, 99 pesticide residues were detected (54 approved, 18 CfS, 27 not approved). One fifth of the residues found in sediments were 'sediment-specific' (the remaining residues had also been found in waters and/or soils – the biggest sources of residues to sediments; *Figure SM1*). One sample was 'pesticide-free'; 3

samples had a single residue, and the remaining 34 samples had mixtures of residues. Glyphosate and its main metabolite AMPA were the only compounds present in more than 50 % of the tested sediments. The number of residues in a sample ranged from 0 to 48, with a median of 6. The total pesticide concentration reached a maximum of 1,241 μg kg^{-1}. The median total pesticide concentration (across samples and water bodies) was 37 μg kg^{-1}. Cyprodinil and dicamba were the compounds with the highest levels, followed by spinosyn A, AMPA, and difenoconazole.

When individual matrices data were aggregated, the extent of pesticide contamination was even more obvious (*Figures SM1-3*). Aggregation revealed that altogether, 86 % of the 625 tested samples presented pesticide residues, 76 % had mixtures. For samples collected during the middle of the growing season, only 7 were 'pesticide-free' (one soil sample from CF, five soil samples from OF and one sediment sample). Crops, collected at harvest time had a high frequency of 'pesticide–free' samples (71 out of 193 samples). The aggregation showed that 31 pesticide residues were found in all six matrices, 26 residues in 5 matrices, 23 residues in 4 matrices, 41 residues in 3 matrices, 44 residues in 2 matrices, and 43 residues in a single matrix (*Figure SM1-3*). There were 38 residues found only in indoor dust, 4 only in water, and 1 only in air samples. Finally, cross-matrix analyses, such as that depicted in Figures SM1-3, provide some insights into preferential accumulation matrices for different pesticide residues (for instance, soil for DDTs and dieldrin), and compound accumulation factors among matrices [highly variable per compound, with, for instance, water:sediment median concentration ratios varying from < 0.01 (DDTs, Hexachlorobenzene) to > 320 (Metobromuron)].

3.2. Hazard profile of the residues found

3.2.1. Hazard to terrestrial and aquatic species

Most pesticide residues found in the soil samples present low to moderate hazard to the terrestrial species used in standardized ecotoxicological assays. Nevertheless, despite their small proportion, there are compounds in soil (and on the European market) that may harm terrestrial organisms and, consequently, terrestrial ecosystem services. See for instance that 67 % of the approved residues found in CF soils, and 52 % of CfS residues, are highly hazardous to mites (Fig. 5). Some residues are highly hazardous to multiple soil organisms (acetamiprid, chlorpyrifos and chlorpyrifos-methyl; *Table SM3*). The hazard profiles vary only slightly between CF and OF samples. Although most of the pesticides that we tested were synthetic, and despite the higher proportion of not approved residues found in OF than in CF soils, approved and CfS compounds were detected in OF soils.

Eleven out of the 76 pesticide residues found in outdoor air present high hazards to birds and/or honeybees (acute). Information for honeybees (chronic) and bumblebees is surprisingly limited, at least for these 76 residues (Fig. 5). The duration of exposure to the pesticide residues seems to be a crucial aspect for birds, with most residues being classified as low hazard when exposure is acute, and high or moderate hazard if exposure is chronic. Chlorpyrifos, acetamiprid and cyproconazole are hazardous to birds in both acute and chronic contexts. Most of the residues detected in outdoor air are of moderate or unknown hazard to bees. Chlorpyrifos, chlorpyrifos-methyl, deltamethrin, cypermethrin, indoxacarb, fenvalerate, lambda cyhalothrin and phosmet are highly hazardous to bees. The proportion of hazardous compounds is much lower in approved and CfS panels than in the not approved panel, highlighting the legislative efforts made over the last years to protect pollinators.

Most of the hazard entries retrieved for the residues found in water indicate a moderate hazard to aquatic organisms. There are, however, recurrent 'no data available' situations, especially for not approved compounds, aquatic crustaceans, and chronic endpoints. Nearly 35 % of the residues found (39 out of 112) are highly hazardous to at least one of the 8 aquatic endpoints considered (Fig. 5). Over 19 % of the residues (21 out of 112) are of high hazard to more than one of the 5 aquatic organisms considered.

The hazard of the compounds found in sediments to sediment-dwelling organisms is largely unknown (see percentage of 'no data available' ranging from 22 to 74 %; Fig. 5). Lack of hazard data occurs mostly for not approved compounds and transformation products but extends to approved and high frequency/level compounds (like cyprodinil, dicamba, or glyphosate). The available data show a rather large proportion of high hazard compounds in this matrix (across endpoints and approval status). Bifenthrin, chlorantraniliprole, chlorpyrifos, epoxiconazole, imidacloprid and thiacloprid presents high hazard to all three sediment endpoints, and five other residues present high hazard to two endpoints (deltamethrin, flupyradifurone, indoxacarb, and lambda-cyhalothrin; *Table SM3*).

3.2.2. Hazard to animals (surrogate to livestock and pets)

Harm to other animals was explored since crops containing pesticide residues can be used as feed, poor air quality may affect livestock health, and the presence of pesticide residues in indoor dust may affect indoor pets' health. Hazard information for mammals is relatively complete compared to other non-target organisms (Figs. 5–7). Mammal hazard data indicates a high vulnerability of these organisms to pesticides, especially if exposed to CfS compounds, via diet, and long duration exposures (Fig. 6*)*.

Fifty-four out of the 78 pesticide residues found in crops have high hazard for mammals. Bifenthrin and chlorpyrifos, among the top 20 frequency compounds in crop samples, are classified as highly hazardous for all three mammalian endpoints considered in the PPDB/EFSA assessments. Eight other residues were marked as 'high hazard' for two endpoints, and low, moderate, or no data in the other (this last case refers to lambda-cyhalothrin and emamectin; *Table SM3*). Like in soil, CF and OF crop hazard profiles were similar. This is because the lists of residues found in CF crops and in OF crops (that overlap only partially) cover all hazard classes, in a surprisingly similar balanced way.

Over 65 % of the residues found in indoor dust (129 out of 197) are highly hazardous to mammals; 106 of these are known to be of high hazard to one endpoint, 18 to 2 endpoints, and 5 to all 3 endpoints (bifenthrin, chlorpyrifos, esfenvalerate, fipronil and methiocarb). These numbers could increase once more ecotoxicological hazard data becomes available. Note that 2 compounds presented one 'high' and two 'no data' entries (dieldrin and propoxur). Three compounds presented two 'high' and one 'no' data entry (emamectin, methiocarb sulfoxide and lambda-cyhalothrin), and 14 residues had no mammal hazard information. CF and OF-indoor dust hazard profiles were also almost the same because of the identical list of residues found in CF and OF dust samples.

Over 75 % of the pesticide residues found in outdoor air (57 out of 76 tested) are highly hazardous to mammals. 47 pesticide residues are highly hazardous for 1 endpoint, 9 compounds for 2 endpoints, and chlorpyrifos for all 3 endpoints. We could not find any mammal hazard information of phosmet oxon which was detected in air. OF profiles presented in general slightly lower percentages of high hazard class than CF profiles (exception: CfS panel, endpoint: chronic exposure).

3.2.3. Hazard to humans

Over 64 % of the pesticide residues found in crops (50 out of 78) have been linked to at least one out of the 11 adverse human health effects covered in the PPDB, 50 % are linked to multiple adverse effects (*Table SM3 – the compiled hazard dataset)*. Individual residues are linked to a maximum of five effects (thiacloprid, p,p'-DDT, p,p'-DDE, pendimethalin, Thiophanate-methyl and pirimicarb). Dieldrin, the most common residue inOF crop samples, is linked to 4 endpoints, all with high severity: carcinogenicity, endocrine disruptions, reproductive/development effects, and neurotoxicity. Metalaxyl-M, the most common in CF crop samples, is only known to be a skin and eye irritant. The most common effects linked to the pesticide residues found in the crop

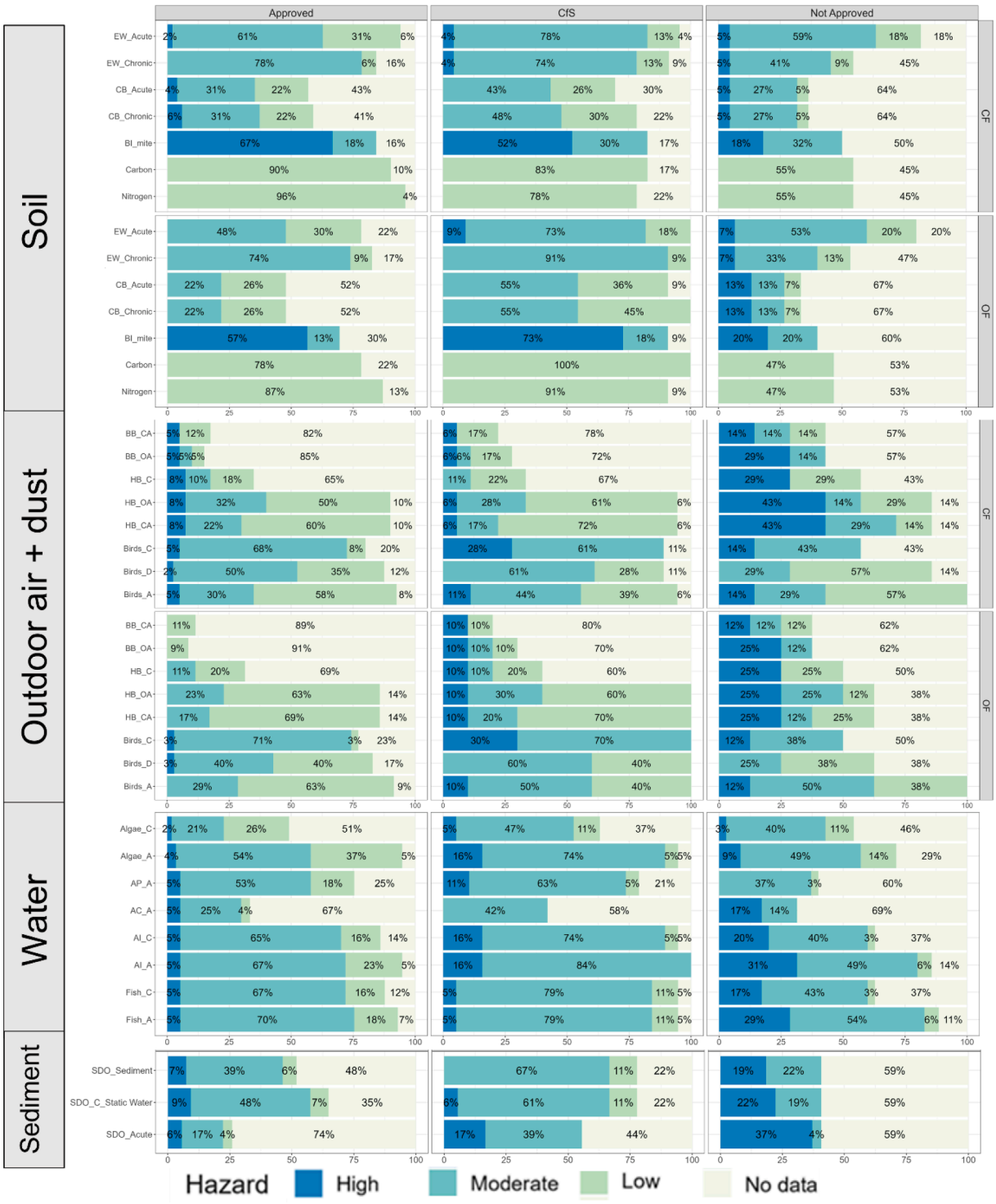


Fig. 5. Hazard profile of the pesticide residues detected in soils, outdoor air, water and sediments for terrestrial and aquatic organisms. For soil and air we have two hazard profiles, one related to the pesticide residues found in the samples from conventional fields (CF), the other related to the pesticide residues found in the samples from organic fields (OF). CfS = Candidates for Substitution; EW = Earthworms, CB = Collembola, BI = Beneficial Insects - predatory mite; Carbon = soil micro-organisms related to carbon mineralisation; Nitrogen = soil micro-organisms related to nitrogen mineralisation. Birds_A = Birds - Acute; Birds_D = Birds - Short term dietary; Birds_C = Birds - Chronic; HB_CA = Honeybees Contact acute; HB_OA = Honeybees Oral acute; HB_C = Honeybees Chronic; BB_OA = Bumblebees Oral acute; BB_CA = Bumblebees Contact acute; Fish_A = Fish - Acute; Fish_C = Fish - Chronic; AI_A = Aquatic invertebrates - Acute; AI_C = Aquatic invertebrates - Chronic; AC_A = Aquatic crustaceans - Acute; AP_A = Aquatic plants - Acute; Algae_A = Algae - Acute; Algae_C = Algae - Chronic. SDO_Acute = Sediment dwelling organisms - Acute; SDO_C_Static Water = Sediment dwelling organisms - Chronic static, water.

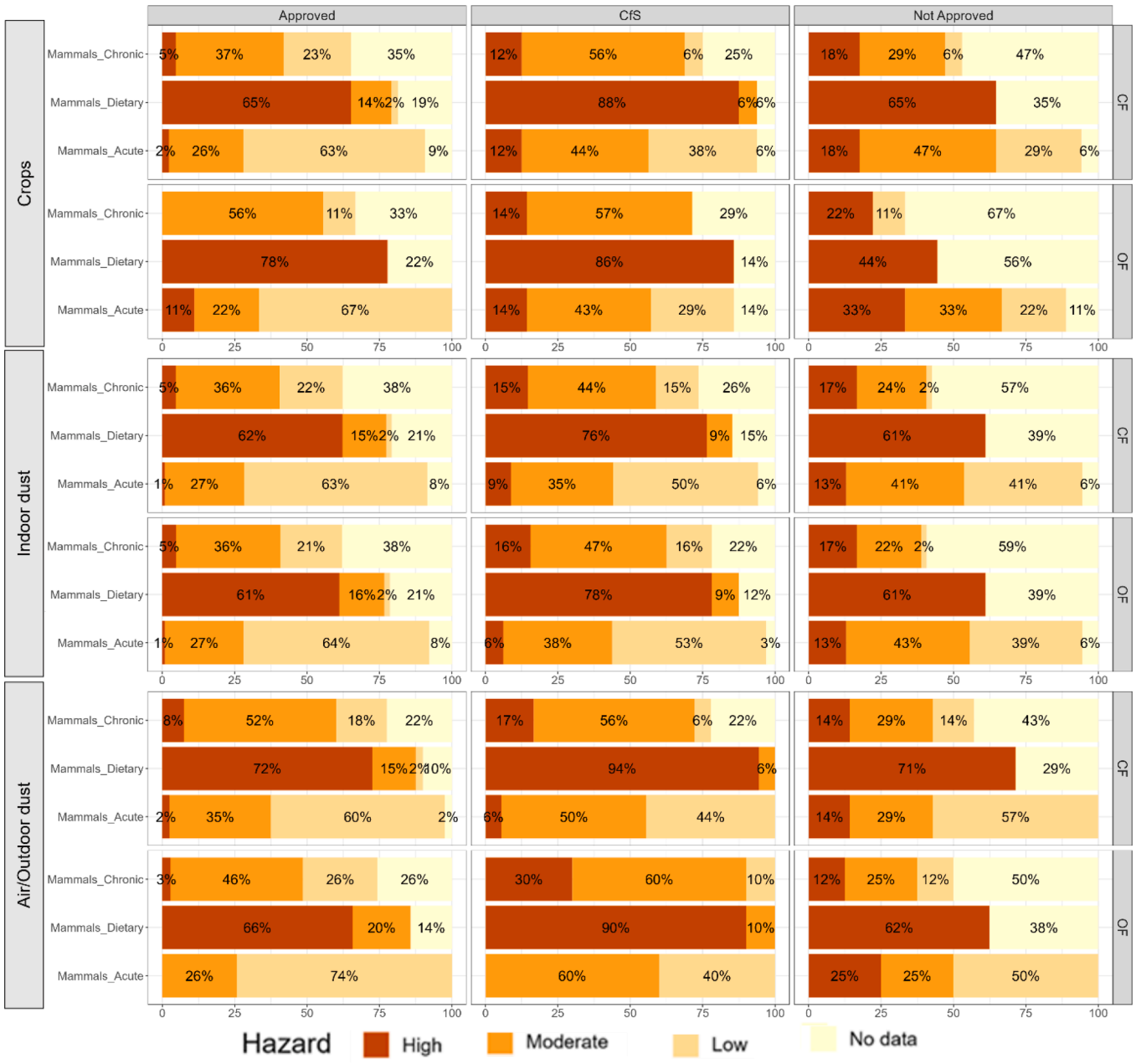


Fig. 6. Hazard profile of the pesticide residues detected in crops, indoor dust and outdoor air for mammals, a surrogate for livestock and pets. CfS = Candidates for Substitution. Note we have two hazard profiles per matrix, one related to the pesticide residues found in the samples from conventional fields (CF), the other related to the pesticide residues found in the samples from organic fields (OF).

samples are eye and skin irritations, as well as reproductive/developmental effects (Fig. 7). However, we observed "yes" entries for 10 out of the 11 human endpoints considered, the exceptions being phototoxicant. Although pesticide-effect associations are seen across farming systems and approval status, the proportion of compounds with highly severe effects has been drastically reduced due to regulations (see e.g., 59–78 % of not approved compounds vs 0–11 % of approved and CfS compounds linked to neurotoxicity). At the same time, it is important to stress the high proportion of approved and CfS compounds marked with 'possibly' (especially at carcinogenic and reproductive/development effects endpoints), and 'no data available' entries (Fig. 7).

Nearly 66 % of the residues found in indoor dust (131 out of 197) are linked to adverse human health effects (*Table SM3*), 43 % of the residues (84 out of 197) are linked to high severity effects. A maximum of 7 effects are associated with a single residue (chlorothalonil). 2,4-D, the most common residue in OF indoor dust samples, is linked with 5 endpoints (endocrine disrupter, reproductive/development effects, neurotoxicity, respiratory tract irritant, and eye irritant). Glyphosate, the most common residue in CF indoor dust samples, is only known to be an eye irritant, according to PPDB/EFSA. It is also a possible carcinogenic, endocrine disruptor and may cause reproduction/development effects. As mentioned above, under the indoor dust-mammals section, the very high overlap between the list of pesticide residues found in CF and in OF indoor dust samples, results in almost perfectly matched CF and OF human hazard profiles.

Over 76 % of the residues found in the air samples (58 out of 76 tested) have been linked to human health effects, over 49 % (37 out of 76) of the residues have been linked to highly severe effects (*Table SM3*). Like for indoor dust, one single residue is linked to a maximum of 7 effects (chlorothalonil). Pendimethalin, the most common residue in air

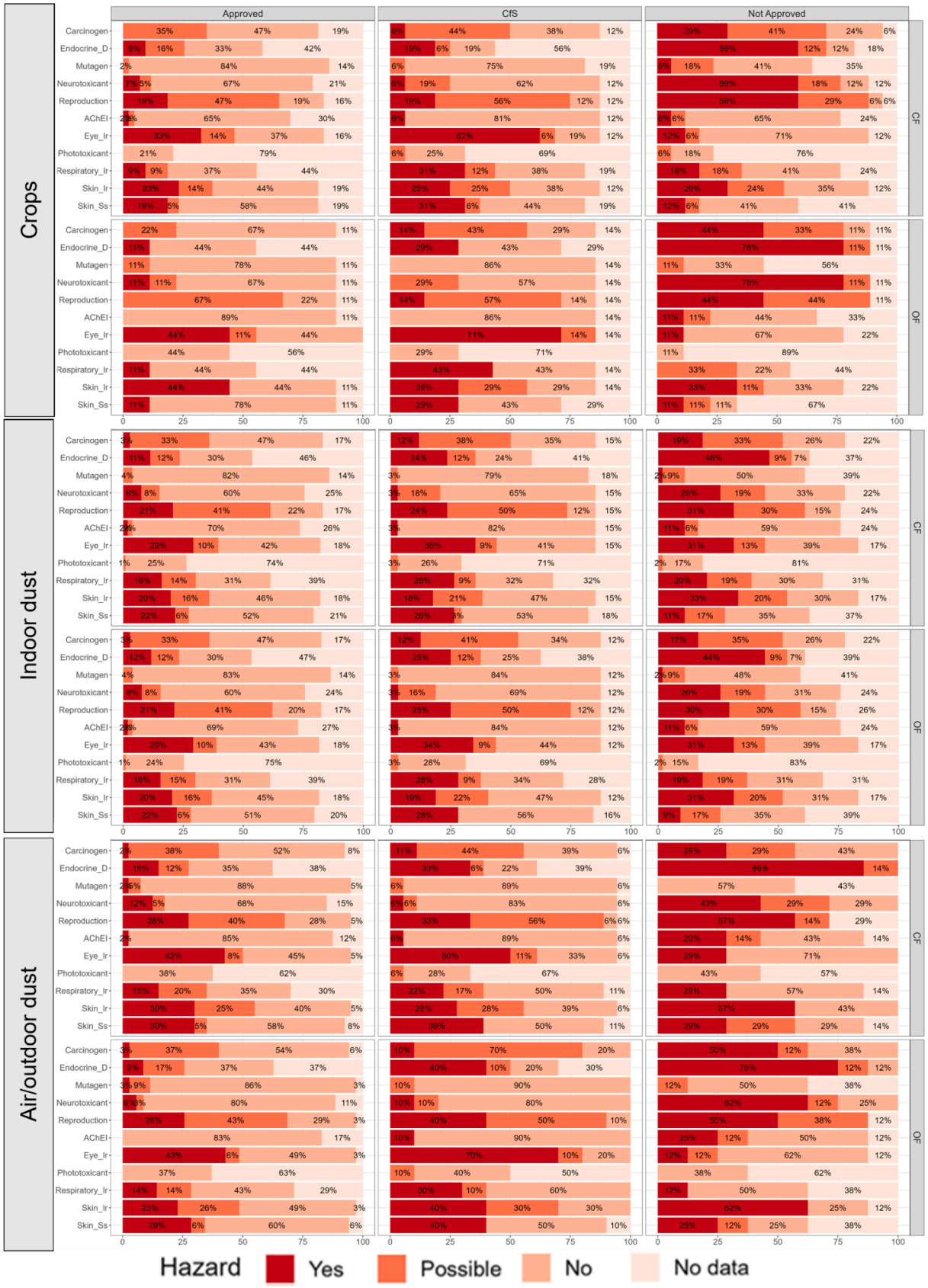


Fig. 7. Hazard profile of the substances found in crops, outdoor air and indoor dust samples for human. Abbreviations. CfS = Candidates for substitution. Endocrine_D = Endocrine Disruptor; AChEi = Acetyl Cholinesterase Inhibitor; Ir = Irritant; Ss = Sensitiser.

samples, is marked "yes, known to cause a problem" in 5 endpoints (reproductive/development effects, respiratory tract irritant, skin irritant, eye irritant, and skin sensitizer), and "possible" for 3 others (carcinogenic, endocrine disrupter and phototoxic). Reproductive/developmental effects and endocrine disrupter have the highest expression of "yes" indications among the highly severe effects. Reproductive/developmental effects and carcinogenicity have the highest expression of "possible" indications, being 38 % and 46 % of the detected residues respectively. CF and OF profiles are once again rather similar (Fig. 7).

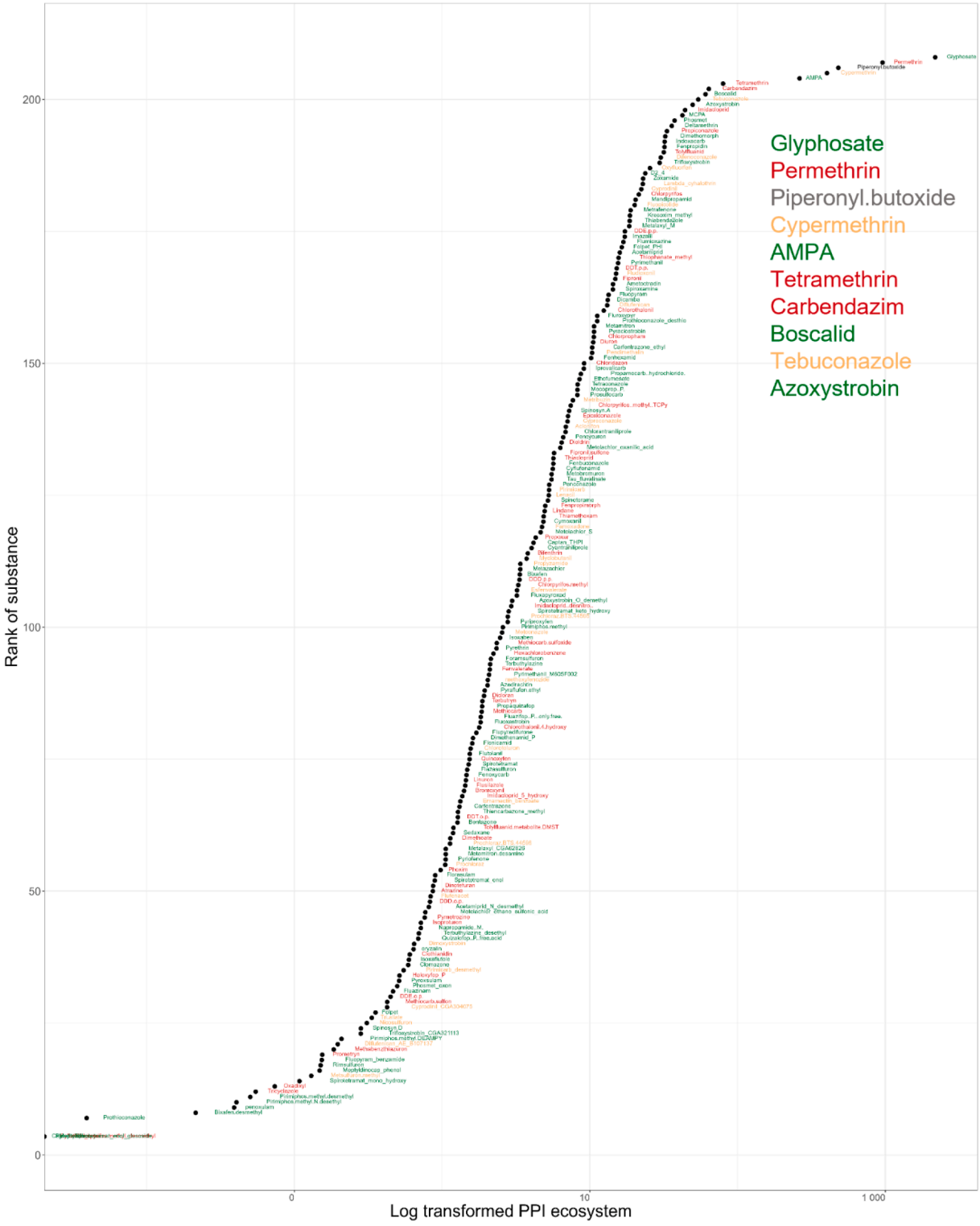


Fig. 8. Pesticide Prioritization Indicator (PPI) for ecosystems. Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

3.3. Pesticide prioritization indicator (PPI)

Monitoring and hazard data were combined to develop a prioritization indicator of pesticides for ecosystem and human health assessments (Figs. 8 and 9, respectively). 202 out of the 209 pesticide residues covered in this study pose some hazard to ecosystems and/or human health. The exceptions are captan, chlorpyrifos-desethyl, chorpyrifos-methyl-desmethyl, cyfluthrin, meptyldinocarp, pyrethin II and spirotetramat-enol-glucoside. Chlorpyrifos-desethyl was not possible to be measured on any of the matrices covered in this study. Glyphosate had the highest PPI values, for both ecosystem and human assessments, a consequence of its very high frequencies and levels across matrices.

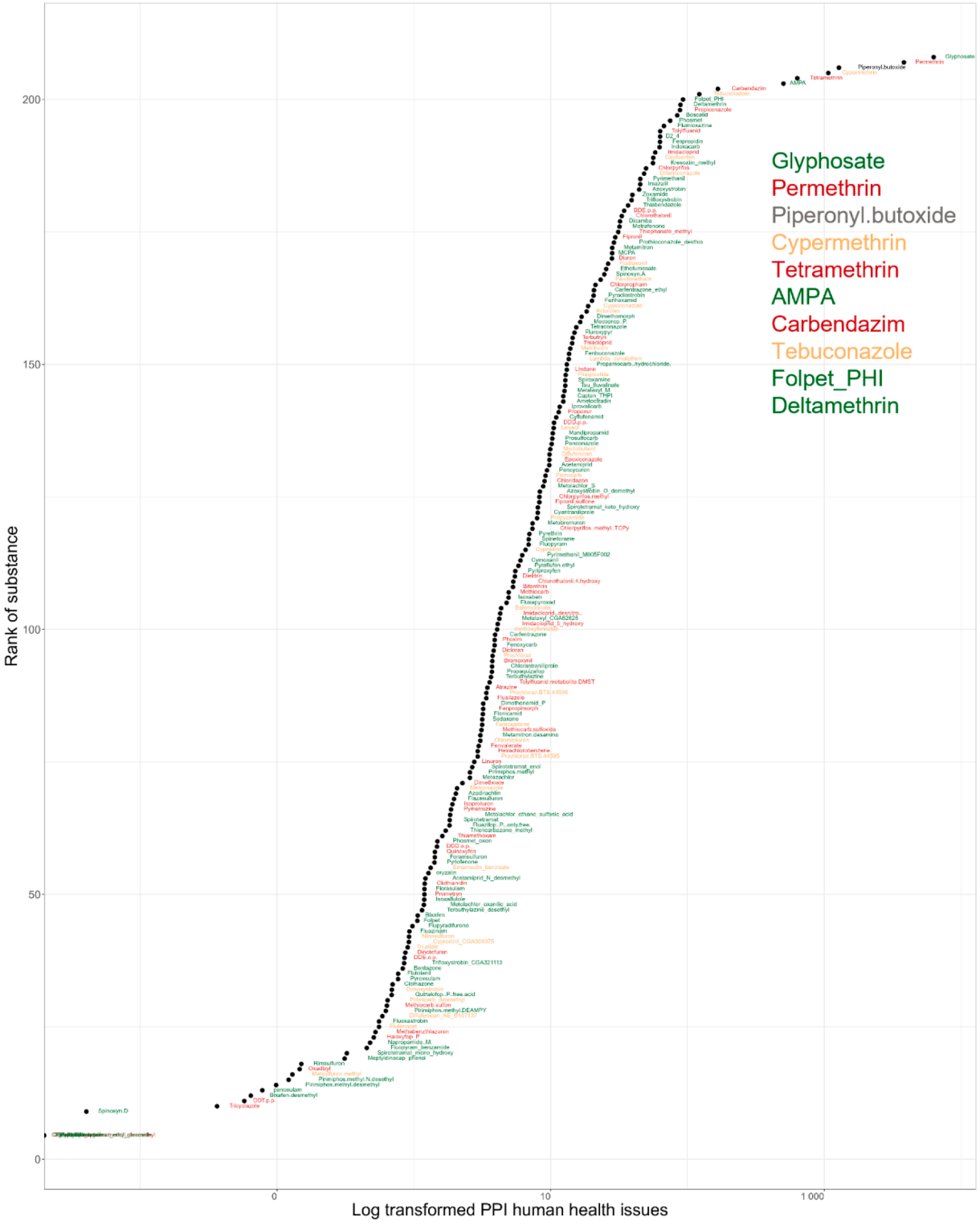


Fig. 9. Pesticide Prioritization Indicator (PPI) for humans. Residues are sorted from lowest to highest PPI values. The name of pesticide residues in red indicates not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The list on the top right corner corresponds to the 10 compounds with the highest PPI values. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The not approved compound permethrin had the second highest PPI values, due to its highly hazardous character for multiple non-target organisms, and links to high severity human endpoints. The lowest PPI values were observed on transformation products (bixafen desmethyl, pirimiphos-methyl-N-desethyl, pirimiphos-methyl-desmethyl, spirotetramat-mono-hydroxy, meptyldinocap phenol) and not approved substances (tricyclazole, DDT and Oxadixyl) which had low frequencies/levels and highly incomplete hazard profiles. Ecosystem and human curves show a mix of not approved, CfS, and approved substances. The sum of ranking positions in both curves shows 24 residues of top concern (sum of rank positions < 50): glyphosate, permethrin, pirimicarb, cymoxanil, folpet, cypermethrin, pendimethalin, metalaxyl-M, AMPA, phthalimide, piperonyl butoxide, kresoxim-methyl, tetramethrin, phosmet, prothioconazole desthio, fludioxonil, azoxystrobin, tebuconazole, trifloxystrobin, penconazole, metolachlor (S), pirimicarb desmethyl, zoxamide, and chlorothalonil). The analysis of matrix

Fig. 10. Cumulative Pesticide Prioritization Indicator (CPPI) for each matrix. The red dots indicate not approved compounds, orange denotes candidates for substitution and green indicates approved compounds. The names of the 10 compounds with the highest PPI values per panel are marked at top left or bottom right corners. For further details on the curves, including uncertainty analyses, see Figures SM4-SM13. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

specific curves (Fig. 10) shows a higher number of top concern substances, with 46 pesticide residues listed across the panels (only the names of the compounds with the 10 highest PPI values were presented in the figures). Glyphosate is present in 9 panels out of 10; metalaxyl M, Folpet_PHI, cypermethrin, pirimicarb, AMPA, and trifloxystrobin in 4; and boscalid, carbendazim, tebuconazole, pendimethalin, dimethomorph, kresoxim methyl, Folpet and cymoxanil in 3. Eleven residues are present in 2 panels, the remaining 21 residues in one. Most of the compounds with the highest PPI values are approved residues (min 3 – max 9/panels). In indoor dust, there are however more not approved residues. The shape of the ten curves is rather similar, and all of them contain a mix of approved, CfS and not approved substances. To highlight the much higher CPPI value of indoor dust than of other matrices (remember the higher number of compounds and higher pesticide concentrations in indoor dust samples). The CPPI values of indoor dust were highly affected by glyphosate - its PPI represents 41 % of CPPI for humans, 51 % for CPPI for mammals. In water, glyphosate also had a > 40 % contribution to CPPI. Other curves present a more balanced contribution of residues, with the compound with highest PPI contributing between 15 and 28 % to the CPPI value.

4. Discussion

4.1. Occurrence of pesticide residues

This paper explores pesticide occurrence across 6 matrices, in 10 European countries, covering both conventional and organic production. The number of residues, total pesticide concentration, and mixture composition were highly dependent on the matrix and farming system. Although we accounted for the impact of CSS and crops in our statistical analyses, analyses concerning CSS and crop differences were considered to be more appropriate for matrix-specific publications (e.g., Navarro et al., 2023).

The number of pesticide residues, and total pesticide concentrations were significantly higher in soils from CF than OF. OF soils contained mostly persistent, long-banned pesticides. CF soils contained also recently banned and currently approved pesticides. Such trends have already been observed by Geissen et al. (2021) and Riedo et al., (2021). The presence of banned compounds in soils, and connected matrices, alert for long-lasting impacts of pesticides, which should be accounted for in risk assessment (as baseline contamination), and field management. Re-evaluation of pesticides persistence is also advised. Direct comparisons between pesticide figures among studies are limited due to the different crops covered, different sampling times, and analytical details. Transition/conversion to organic farming at the landscape level is expected to minimize off-site contamination of OF and OF products, and lead to an overall higher sustainability of organic practices (Smith et al., 2020). Hopefully, the increasing evidence and information concerning soil contamination by pesticide residues, and of associated risks and impacts (Beaumelle et al., 2023; Klátyik et al., 2023), will lead to the development of soil quality benchmarks. Despite the current research and regulatory focus on soils (EC, 2021; EC, 2023c; Vieira et al., 2023), we are still limited to only a few, member state specific, and mostly POPs-related, benchmark values (Carlon, 2007). This hampers interpretation of monitoring data and facilitates worsening soil quality status.

Over 63 % of crop samples contained pesticide residues (82 % of CF samples, 37 % of OF samples). EFSA's latest report (Carrasco Cabrera et al., 2023), with a much larger sample size, crop and country coverage than the present study, found smaller percentages of samples containing pesticide residues (44.3 % overall, 17.2 % in organic). This same EFSA report indicates a MRL exceedance of 3.9 %, a value possibly exacerbated because of the sampling strategy employed, and the inclusion of products from outside the EU. Our crop results, presented on dry weight basis, cannot be directly compared with EU-MRLs, based on the product 'as is'. Literature data on the typical moisture content of each of the crop/product tested could be used in the conversion but subsequent inferences on food safety would be linked to high uncertainties. To point out that the establishment of MRLs is done at an individual pesticide level, not yet taking into account that one may be exposed to multiple pesticide residues at the same time (EC, 2005). Setting MRLs for total pesticide concentrations [like in the drinking water regulation (EU, 2020)], and considering adapting MRL defining metrics to address the possibility of additive/synergistic effects among pesticide residues, could lead to higher consumer safety (Carvalho, 2017; Li and Fantke, 2023).

Almost all our water samples contained mixtures of pesticide residues. The water samples originated from surface water bodies; most are expected to be just used in the irrigation of agricultural fields, but some are possibly used recreationally, or as drinking water supply. The EU 2008/105 EC regulation, on environmental quality standards (EQS) for surface waters (EU, 2008), covers a few pesticides. Our results did not exceed such EQS values. The drinking water directive (EU, 2020) sets a limit of 0.1 μg/L for individual pesticides/metabolites, and of 0.5 μg/L for total pesticide concentrations. The former was occasionally exceeded (<1% individual compound entries), the latter was often exceeded (38 % of the samples). We sampled small water bodies in regions with high intensity agricultural activity. Percentages and levels of pesticides are usually lower in larger rivers and lakes due to longer distances to agricultural fields, and dilution from non-agricultural waters (Halbach et al., 2021; Mohaupt et al., 2020). It is important to stress that i) the number and type of compounds in the mixtures is not accounted for in existing water EQS (SCHEER, 2023), and that ii) the EU has no sediment EQS, although these are known to be a sink and source of contaminants (Chiaia-Hernández et al., 2022). The EC requests Member States to establish EQS for sediment and/or biota at national levels (EU, 2008), but centralization of such values is necessary for large-scale assessments. Several studies had already pointed out the problematic of transport of pesticides with runoff, in dissolved and particulate phase, exploring, for instance, the impact of land management on transported values, and concentration-discharge dynamics (Commelin et al., 2022; Lefrancq et al., 2017; Vormeier et al., 2023). Creation of buffer areas around water bodies, and more effective soil erosion control could reduce the impact of agriculture on water bodies, improving their chemical, and likely also their ecological status (Carter, 2000).

Despite the low number of outdoor air samples in this study, our results align with the findings of the European studies looking at pesticides in outdoor air (Degrendele et al., 2016; Estellano et al., 2015; Kreuger J and Lindström, 2019; Kruse-Plaß et al., 2021; Lu et al., 2000). The findings from Kruse-PlaB et al. (2021) and Kreuger and Lindström (2019) alert us to the possibility of medium-range transport of pesticide residues, which are often considered negligible (Figueiredo et al., 2022). Since our TIEM devices were installed at the edge of the agricultural fields, and the surrounding areas were not mapped, and wind direction and speed not explored, inferences on transport distance would be too speculative. It should still be pointed out that there are no pesticide benchmarks for this matrix, and that a higher implementation of precision application techniques and protective measures (like avoiding pesticide applications in windy conditions) could lead to an improvement in air quality.

Finally, of all matrices tested, indoor dust contained the highest number of pesticide residues and highest pesticide concentrations. The higher figures in this matrix may be partially related to the fact that our analytes list included 32 dual use residues (plant protection product and biocide). Previous studies have already reported high diversity, frequency and concentration of pesticides in indoor dust. These being positively associated with the proximity of agricultural areas, pesticide use indoors, and the preparation of pesticide tank mixes in homes (Degrendele et al., 2022; Kuiper et al., 2022; Mu et al., 2022; M. Figueiredo et al., 2022). This last point, together with the fact that agricultural workers can bring pesticides indoors via shoes, clothing, skin, or hair may explain the higher concentrations found in the CF households.

The magnitude of the findings in this matrix is of potential concern for three reasons. First, farm workers may not be aware of the importance of taking protective measures before entering homes (namely changing clothes and taking a shower). Second, our understanding of pesticide degradation in an indoor environment, especially in shaded areas, is very limited. Third, the real contribution of inhalation and dust ingestion routes are not properly explored, nor are the risks of such highly complex mixtures to animal and human health.

4.2. Hazardous characteristics of the pesticide residues

The hazard dataset compiled and explored in section 3.3 highlights i) the presence of a significant number of hazardous compounds in European soils, water bodies, air, and farmers' households; and ii) 'no data available' situations are rather common, especially for non-approved compounds, which are a major contributor to the cocktails found across matrices; and iii) an increase in lower hazard compounds and a reduction of higher hazard compounds on the market, as a result of multiple regulatory efforts, and regular re-evaluation of the approval status of pesticides (EASAC, 2023b).

Our hazard dataset is based on verified data used for regulatory purposes. Europe, with the strictest pesticide regulatory system in the world, relies on EFSA pesticide risk assessment reports to decide on the active substances allowed on the EU market. The EFSA reports cover the impacts of single active substances on humans and other non-target species. The impacts are inferred from standard ecotoxicological tests and modelling exercises, using a few standard organisms, endpoints and conditions (Ockleford et al., 2017), in a much-simplified version of post-approval, 'real life' exposure scenarios (Walder et al., 2022; Beaumelle et al. ,2023; Holmes et al., 2018). More et al., (2019), PARC (Partnership for the Assessment of Risks from Chemicals, and the EuroMix project are making substantial progress towards more comprehensive assessments. This includes the development of new test procedures for multi-chemical exposure by grouping chemicals into common assessment groups and exploring interactions among these groups. New testing guidelines should reflect real-life field conditions by accounting for background contamination of persistent pesticide residues and dynamics associated with pulse-based but longer-term exposures.

The impacts of the environmental mixtures found in this study on terrestrial and aquatic ecosystems are currently unknown. These impacts are also difficult to predict based on the methodological approaches currently available for risk assessment, and monitoring and hazard/risk data limitations, even for individual pesticide residues. Evaluations based on straight forward comparisons between our measured concentrations and regulatory, NOEC values could be misleading, due to temporal variations in pesticide content and limited NOEC values. AMPA for example (the compound with the highest median concentration in CF soils) present much lower levels in the European soils than the NOEC value for earthworms presented in the glyphosate conclusion report. Such report includes a single NOEC value for AMPA-earthworms, from an assessment with *Eisenia fetida.* Native earthworm species such as *Aporrectodea caliginosa* and *Lumbricus terrestris* are however more sensitive to pesticides than the standard species (Pelosi et al., 2013). Furthermore, more comprehensive assessments, like our CPPI exercise, may lead to different outcomes. Difenoconazole for example, with medium–low concentrations in soil, exceed ocasionally its *E. fetida* NOEC value (EFSA, 2011) However, due to its also medium–low frequencies in soil, and low toxicity to other soil organisms, such compound seems to be of lower concern than AMPA (Figure SM4).

The uncertainty surrounding the effects of the detected mixtures also applies to humans. Although we know that humans are exposed to pesticides in multiple ways, and comprehensive exposure predictions are done for pre-market risk evaluations (Dabrowski et al., 2014), only uptake by food is monitored. And even so, food safety, monitored via pesticide-specific MRL and acceptable daily intake values (EC, 2005), is likely underestimating real risks to consumers by not accounting for mixtures. All routes of exposure should be accounted for in assessments, and the findings translated into regulatory limits and action plans to reduce risks to farm workers, residents living in agricultural areas, and consumers. Finally, on top of the above-mentioned concerns and recommendations concerning the representativity of the risk assessments, the implementation of the precautionary principle in their evaluation should be addressed. This principle dictates that in the case of uncertainty or suggestion of harm, measures to protect ecosystems and human health should be adopted (EC, 2009). The high number of reports of adverse effects from approved substances (de Montaigu and Goulson, 2021; Kaila et al., 2021; Syromyatnikov et al., 2020; van Bruggen et al., 2021) and the increase in derogations over the last years (PAN, 2023), suggest a dangerously high flexibility surrounding the implementation of the precautionary principle which should be explored and rectified.

4.3. Meeting the Farm to Fork pesticide reduction goals

In this paper, we present a first attempt for a pesticide prioritization indicator (PPI) based on a comprehensive assessment of reallife occurrence combined with data from existing hazard classifications. It was developed using occurrence data from the SPRINT 2021 field campaign, qualitative hazard information from PPDB, ecosystem hazard scores from Silva et al. (2022), and human hazard scores based on Dabrowski et al. (2014) and Valcke et al. (2005). The indicator was developed for re-evaluation/regulatory contexts, but it is expected to be also useful in the development of pesticide use scenarios, and on design of (eco)toxicological tests. The PPI values and related curves can easily be updated to include new monitoring and hazard data and can be adapted to more complex inferences. These may include different weighting factors for different exposure routes, or for quantity and quality of data available/considered, or even inclusion of other human-hazard classes more suitable for human-specific assessments (Burtscher-Schaden et al., 2022; Damalas and Eleftherohorinos, 2011; RIVM, 2019; Salomon et al., 2012).

Our PPI indicator follows a rationale similar to the EC pesticide indicators but has the advantage of accounting for individual pesticide characteristics and figures. The EC HRI1divides pesticides into four hazard/approval status-related groups, each one with a different Weighting Factor, WF: "low-risk active substances" - WF1, "substitution candidates" - WF16, "not approved active substances" - WF64, or "approved active substances that do not fall into any other group" - WF8. Our hazard dataset, and hazard profile analyses, show that hazardous characteristics vary strongly among pesticides of the same group, especially WF8, corroborating some existing concerns that using a single group WF can lead to misleading assessments (Burtscher-Schaden, 2022). HRI1, which accounts for sales of active substances, does not provide information about real use of pesticides, and is not sensitive to the fact that lower risk, lowerpersistence substances often have higher application rates. Our indicator addresses these challenges by accounting for frequency of detection and levels of different pesticide residues in different matrices. Our monitoring approach adds valuable information to pesticide marketing data, since it is also applicable to pesticide transformation products, which our data, and other previous studies (Chow et al., 2020; Huber et al., 2022; Silva et al., 2019), show to be a substantial part of pesticide cocktails in environmental and biological matrices.

Comprehensive datasets and analyses like the ones generated in this study are expected to trigger more discussions surrounding pesticide use and risks, a higher engagement of actors in food production, and developing concrete action plans to meet the Farm to Fork reduction targets. However, investing in signalling transitions and systemic changes may be more important and effective than focusing on an exact target number. In our opinion, the use of the most hazardous pesticides, and of several approved substances with high PPI values (Figs. 7–10), should be reduced beyond the 50 % target. We also suggest re-evaluating

the derogations process, and accounting for existing background contamination data before extending use authorization of substances.

4.4. Strengths and limitations

The current study is unique in its comprehensive approach. It covers dominant crops, in conventional and organic farming systems in a variety of climatic zones across Europe. The methods applied for sample collection, storage and shipment, were harmonized and described in standard operating procedures. The overall study protocol was published before the start of the field work (Silva et al., 2021). The study is also unique in its attempt to analyse a large number of pesticide residues in a broad variety of matrices according to standardized methods for treatment and analysis, by experienced and reference laboratories.

On the other hand, its large-scale and multi-matrix aspect led to some limitations. First, there are some uncertainties on the occurrence results associated to the fact that we have a single sampling campaign. Frequency of detection of pesticide residues, individual and total pesticide concentration, and mixture compositions are likely to change during the year, and between years (Figueiredo et al., 2021; Martínez-Megías et al., 2023). Our analytical data provides a snapshot of the contamination in CSS regions in the middle of the growing season. This sampling strategy may not reflect all early applications and does not cover late applications. Our findings, as with all occurrence findings, are dependent on the interval between pesticide application and sampling, on persistence of the pesticide residues considered, and of analytical power. The establishment of regular and comprehensive monitoring programs for pesticide residues can help understand temporal and spatial variability, and patterns. Such data would allow legislators, policy makers, and water-agri-nature managers to access and monitor EU soil, water, air quality, the efficacy of pesticide use restriction measures, and support decisions on land use and agricultural practices (Bach et al., 2020; Hvězdová et al., 2018).

Second, due to the short time frame to carry out a complex sampling campaign (see Silva et al., 2021 for details on other covered matrices), while accounting for the coronavirus pandemic safety guidelines at the sampling time, we focused on sample and data collection in the selected fields. Mapping all agricultural areas in each region, producing the same or different crops to those sampled, and collecting information on their management, including on pesticide use, could have provided valuable additional information for the interpretation of results. Modelling could be used too, to explore for instance connectivity of sampled fields and water bodies, and the factors affecting pesticide distribution. SPRINT is applying such approaches in some CSS to better explain the monitoring results.

Third, although rather comprehensive, we had a targeted list of analytes. Having analysed more residues (although much more costly, and time consuming) could have resulted in an even higher number of detections and higher total pesticide concentrations. These parameters are also strongly affected by decisions around the reporting limits. Establishing a unique reporting limit, applicable to all analytes and matrices, would allow direct comparisons among results (especially in terms of frequency of detection). We chose however to provide the most comprehensive monitoring assessment possible, providing low level exposure data for future risk assessments. It is also important to note that some simplifications were made surrounding the characterization of analytes. Due to the large spatial coverage and number of residues assessed in the study, we considered European approval status of residues (a more accurate analysis of not approved figures could be possible if derogations, emitted at Member States level, were explored), and analytes were treated as independent, and application decontextualized residues (parent-metabolite associations, and relationships between application records collected from selected fields and analytical findings are being explored in matrix specific papers).

Finally, in the hazard profiles section, we linked the sampled matrices to standard matrix related organisms [the same strategy is applied in tier 1 of EFSA risk assessments; (EFSA, 2013)]. We did so to provide the full hazard profile of the substances found, though recognizing that some routes of exposure may be more likely or relevant for the organisms. Birds and bees, for instance, are more likely to be exposed to pesticides via feed than air. More tailored eco-toxicological and modelling studies, like the ones now being conducted in SPRINT, can clarify the contribution of individual routes to total pesticide effect. Simplifications were also done in the human analysis. The existence of alternative, more complex hazard score systems have been mentioned before. The RIVM approach (RIVM, 2019) is particularly interesting, accounting for the disability weights introduced by Salomon et al. (2012). Moreover, crops, outdoor air, and indoor dust were equally weighted in the human assessments while these matrices likely result in different internal exposures (HBM4EU, 2022). This was done to avoid biased interpretations, as there are still some uncertainties about exposure contributions for certain residues, and on mixtures impacts. At the same time, we recognize some limitations from using only our data in the assessment. SPRINT crop residues data were used while much more comprehensive dietary intake data is available at EFSA, number of outdoor air samples was very limited, and since indoor dust was only collected from farmers' homes, it may be less applicable for the general population. Integration of different monitoring data and the establishment of even more comprehensive monitoring programs can lead to more accurate assessments.

5. Conclusions

In the present study, we showed that mixtures of pesticide residues are omnipresent in European agricultural environments, and in farmers' households. The chemical dataset, unique in terms of spatial, matrix, and compound coverage, highlights the need for post-approval monitoring programs, and of the definition of benchmarks or quality values for mixtures (across matrices, and ideally farming system-specific). Most of the pesticide residues detected in soil, water, sediment, crops, outdoor air, and indoor dust samples are hazardous for non-target organisms including humans, yet little is known about health risks posed by environmentally relevant mixtures. Pre-market entry risk assessment strategies should be re-evaluated to ensure that they consider all relevant exposure pathways and background contamination, making adequate allowance for uncertainties (in line with the precautionary principle). Pesticide use and risk reduction strategies should take the current situation and pesticides distribution data and hazard as baseline. Together, including the insights from pesticide prioritization curves, these could facilitate: i) better decision-making concerning pesticides approved in the European market, 2) the development of tailored transition plans, and 3) to the establishment of higher levels of protection for humans and the environment.

CRediT authorship contribution statement

Vera Silva: Conceptualization, Data curation, Formal analysis, Investigation, Writing - original draft, Writing - review & editing. **Lingtong Gai:** Conceptualization, Data curation, Formal analysis, Visualization, Software, Writing - original draft, Writing - review & editing. **Paula Harkes:** Conceptualization, Visualization, Investigation. **Gaowei Tan:** Data curation, Writing – review & editing. **Coen J. Ritsema:** Conceptualization, Funding acquisition, Writing – review & editing. **Francisco Alcon:** Investigation, Writing - review & editing. **Josefa Contreras:** Investigation, Writing - review & editing. **Nelson Abrantes:** Investigation, Writing - review & editing. **Isabel Campos:** Investigation, Writing - review & editing. **Isabelle Baldi:** Investigation, Writing - review & editing. **Mathilde Bureau:** Investigation, Writing - review & editing. **Florian Christ:** Investigation, Writing - review & editing. **Daniele Mandrioli:** Investigation, Writing - review & editing. **Daria Sgargi:** Investigation, Writing - review & editing. **Igor Pasković:** Investigation, Writing - review & editing. **Marija Polić Pasković:**

Investigation, Writing - review & editing. **Matjaž Glavan:** Investigation, Writing - review & editing. **Jakub Hofman:** Investigation, Writing - review & editing. **Esperanza Huerta Lwanga:** Investigation, Writing - review & editing. **Trine Norgaard:** Investigation, Writing - review & editing. **Zuzana Bílková:** Investigation, Writing - review & editing. **Rima Osman:** Investigation, Writing - review & editing. **Chrow Khurshid:** Investigation, Writing - review & editing. **Irene Navarro:** Investigation, Writing - review & editing. **Adrián de la Torre:** Investigation, Writing - review & editing. **Paloma Sanz:** Investigation, Writing - review & editing. **María Ángeles Martínez:** Investigation, Writing - review & editing. **Jonatan Dias:** Investigation, Writing - review & editing. **Hans Mol:** Investigation, Writing - review & editing. **Gerrit Gort:** Formal analysis, Writing - review & editing. **Daniel M. Figueiredo:** Formal analysis, Writing - review & editing. **Paul T.J. Scheepers:** Writing - review & editing. **Vivi Schlünssen:** Project administration, Writing - review & editing. **Anne Vested:** Project administration, Writing - review & editing. **Abdallah Alaoui:** Project administration, Writing - review & editing. **Violette Geissen:** Conceptualization, Funding acquisition, Writing – original draft, Writing – review & editing.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

The following are the Supplementary data to this article:

- Word file: Description of sampling methods; description of pesticide analytical methods; results of sensitivity analysis by left-censored data imputation and sensitivity analysis for best/worst case in terms of 'no data' entries; figures SM1-3 heatmap of approved, CfS and not approved compounds; figures SM4-13 matrix specific CPPI curves.
- Excel file 1: Limits of detection of different pesticide residues across matrices.
- Excel file 2: Basic characteristics of pesticide residues analysed.
- Excel file 3: Hazard dataset.

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