Pesticide residues in European sediments: A significant concern for the aquatic systems?

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ABSTRACT

The presence of pesticide residues in waterbed sediments poses a significant concern for aquatic ecosystems' health. This study examined pesticide contamination in sediments of 38 water bodies, embedded in agricultural-dominated regions, across eight European countries. Three indicators were targeted: occurrence, type, and concentrations of multiple pesticide residues in sediments. 196 pesticide residues (including degradation products) were tested in the sediment samples. The analytical results showed that only one sample was 'pesticide-free', three samples contained a single pesticide residue, and the remaining 34 samples contained mixtures of residues. Overall, 99 different residues were found in the sediments, with a maximum of 48 in a single sample. Twenty-seven out of the 99 detected residues were not approved for agricultural use at the time of sampling. The numbers of detected residues and pesticide levels varied among countries. AMPA, glyphosate and DDTs were the most common residues in sediment samples with frequencies of 76, 61, and 52%, respectively. The sediments from the Czech Republic had the highest pesticide concentrations, with total pesticide concentrations ranging between 600 and 1200 $\mu g\ kg^{-1}$. The lowest total pesticide concentrations were found in Slovenia, Switzerland, Croatia, and Denmark, ranging between 80 and 120 $\mu g\ kg^{-1}$. Sediments presented a mix of non-persistent and persistent compounds. Twelve of the detected pesticides are very persistent/stable in sediments, raising concerns about the long-term impacts of pesticides. Our study on the distribution of pesticide residues in European sediments provides valuable insights into the extent of pesticide contamination and possible risks of pesticides to water bodies' health. It also underlines the need for monitoring, research, and policy efforts to mitigate the impacts of pesticides, and to evaluate potential risks of re-use of dredged sediments.

1. Introduction

Pesticides play a vital role in modern agriculture by safeguarding crops from pests. The use of pesticides is linked to higher crop yields which helps meet the growing global demand for food (Foong et al., 2020; Cooper and Dobson, 2007a). By reducing crop damage, and

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avoiding the need for manual weed control, pesticides save farmers significant production costs and labour efforts (McErlich and Boydston, 2013). While these products offer several benefits (Cooper and Dobson, 2007b), their use may also pose serious risks to environmental and human health. Pesticide residues contaminate soil, water bodies, air, and affect non-target organisms such as beneficial insects, birds, and aquatic life (Brühl et al., 2021). Exposure to high levels or prolonged contact with certain pesticide residues may also lead to acute or chronic health issues for farm workers, and consumers (Wojciechowska et al., 2016).

When pesticides are applied in agricultural fields, they undergo several processes that can facilitate their transport to and accumulation in areas other than the treated fields. Nearby water bodies are particularly vulnerable to pesticide contamination (Campo et al., 2013; Kuster et al., 2008). Pesticide residues can be transported off-field through different pathways during or after application, especially via aerial dispersion, surface runoff, and leaching (Mishra et al., 2023; Loos et al., 2009). Some pesticide residues can volatilize into the air and be carried by wind currents, and in wind-eroded particles. Monitoring data in the air is rather limited but available data have corroborated the aerial transport of DEET, transfluthrin, piperonyl butoxide, and of other contaminants namely PCBs (Zaller et al., 2022). A recent study in Germany has reported that glyphosate, chlorothalonil, metolachlor, pendimethalin, and terbuthylazine were rather common in air (Kruse-Plaß et al., 2021). Recent research indicates that there has been an underestimation of the atmospheric transport and persistence of pesticides, emphasizing the necessity for enhancing their risk assessment (Mayer et al., 2024). Rainfall or irrigation can cause pesticide residues to be transported with the runoff water over the soil surface into rivers, lakes, and ponds. This is known to happen with e.g. atrazine, acetochlor, and metolachlor (Commelin et al., 2022; Antić et al., 2015). Pesticide residues can also infiltrate into the soil and reach groundwater sources. Atrazine and its metabolites, and bentazone are amongst the most problematic ones (EEA, 2024). Over time, groundwater sources may discharge into surface water bodies and contribute to sediment contamination (Calderon et al., 2016; Gimsing et al., 2019). It is important to note that the movement of pesticide residues from fields to water bodies varies depending on factors like soil type, landscape characteristics, pesticide properties, and management practices (Fairbairn et al., 2015).

Waterbed sediments are known sinks of contaminants, informing about the contamination from the watershed (Mohanavelu et al., 2022). Since waterbeds are an integral part of the aquatic ecosystem, pollution of waterbeds may have detrimental effects on the functions and health of the ecosystem (Marval et al., 2020). Some pesticide residues dissolve in water which allows them to remain mobile and potentially move to further locations with the water currents. Other pesticide residues have a higher affinity for organic matter and adsorb onto suspended particles, which eventually settle down and accumulate in the waterbed (Schipper et al., 2008; Burton, 2013). The persistence and impacts of pesticide residues in sediments depend strongly on their chemical properties. Some pesticide residues may degrade or break down relatively quickly while others retain their molecular integrity (and hence their physical, chemical, and functional characteristics) for longer, leading to long-term contamination/pollution. As mentioned above, pesticide residues, regardless of their persistence, can have undesired effects on non-target organisms such as fish, amphibians, and invertebrates (Sabra and Mehana, 2015; Calvo-Agudo et al., 2020; Gibbons et al., 2015). This can disrupt ecological balance and impair ecosystem services. A short overview of recent European studies addressing the contamination of sediments by pesticide residues is presented in *table SM1*. See also, for example, Oubiña et al., 2006, Carazo-Rojas et al., 2018, Liu et al., 2022, San Juan et al., 2023 and Damiani et al., 2023 non-European works and data.

The SPRINT project (on Sustainable Plant Protection Transition, https://sprint-h2020.eu/), funded under the EC program H2020, focuses on the impacts of pesticide residues on the ecosystem, crop, livestock, and human health, and on the transition towards more sustainable pesticide use. A key project activity related to the collection of environmental and biological samples is a European-wide survey for further analyses of pesticide residues and other health indicators (See Silva et al. (2021) for the study protocol). The primary aim of the current study is to assess the presence, type, and levels of pesticide residues in European waterbeds. The survey covers different regions, with different crops and pesticide regimes. The analytical results will enhance our understanding of pesticides' fate, persistence and potential consequences to the ecosystems. Ultimately, this knowledge may support decision-making, support sustainable land management practices, and contribute to the protection of aquatic ecosystem health.

2. Material and methods

2.1. Coverage of the assessment

As mentioned above, this study is linked to a comprehensive, holistic sampling campaign conducted in Europe, under the SPRINT project umbrella. This campaign, designed to assess the distribution and impacts of pesticides on environmental, plant, animal and human health, covered ten case study sites, all in different European countries. This was to cover the main crop types in Europe, and a diversity of agricultural landscapes and practices. The campaign occurred in 2021, being the exact timing of the samples' collection case study-specific; sampling was aligned to the middle of the growing season for each crop) when we expect the highest diversity and (bio)monitoring levels of pesticides. All samples but crop samples were collected at this time. Crop samples were collected at harvest time. The middle of the growing season corresponded to the end of May-begin June in almost all case studies. In the Spanish and Italian case studies (no sediments originating from these – see below) it corresponded to end September-begin October (Silva et al., 2021).

The sediment samples were collected from small reservoirs, lakes, ponds, rivers, or irrigation channels connected or in the vicinity of the sampled agricultural fields. The SPRINT sampling design aimed at the collection of three to eight sediment samples per case study, one per water body (Silva et al., 2021). Due to logistical difficulties, this was not always feasible (e.g. limited amount of water bodies in the study area, water bodies were too deep, etc). In the end, a total of 38 sediment samples were collected, originating from eight European countries. Details on the sediment sample locations are provided in Table 1. Details on the water bodies sampled in SPRINT are already presented in Navarro et al. (2024).

2.2. Sample collection and treatment

Each of the 38 samples analyzed for pesticide residues was a composite sample. Three sub-samples were collected at equidistant points along a transect representative of the water body. The sub-samples were collected with core samplers, from the top layer at a depth of 0–10 cm. Equal quantities of each sub-sample were placed into a bucket to create the composite sample per water body. The (composite) sediment samples were then split into 2 aliquots: one for the physico-chemical characterization, and the other for the determination of pesticide residues.

2.3. Sediment physico-chemical characterization

Sediment physico-chemical characterization was performed by each of the case study teams using harmonized methods (see supplementary materials for full details). In brief, pH measurements were performed following the protocol introduced by Van Reeuwijk (Van Reeuwijk, 2002), organic matter content was determined via the loss-on-ignition method at 550 °C (Jensen et al., 2018), and particle size distribution was measured via the hydrometer method (in HR and SL samples), laser diffraction (in NL, FR, DK, CH, CZ samples; Kristensen and Andersen,

Table 1

General information on the case studies where the sediment samples were taken.

Country	Country code	Covered region	Crop	Number of samples
Portugal	PT	Central zone	Vineyards	8
France	FR	Bordeaux	Vineyards	5
Switzerland	SH	Canton of Bern	Apple orchards	5
Croatia	HR	Istria	Olives	3
Slovenia	SL	Central zone	Maize	6
Czech Republic	CZ	All country	Oil plants	3
The Netherlands	NL	Groningen	Potatoes	5
Denmark	DK	North and Central	Spring barley and Winter wheat	3

1987; Goossens, 2019), or using a Coulter counter for the fine fraction (PT samples; Poppe et al., 2003). The basic characteristics of the 38 sediment samples are presented in *Table SM2*.

2.4. Determination of pesticide residues

Within the SPRINT project, a target list of 209 analytes was established (Silva et al., 2021). This list was targeted in all environmental samples. For sediments, the analytical methods were validated and considered fit for purpose for 196 compounds. This sub-list covers 164 active substances, 44 degradation products, and one synergist (piperonyl butoxide; *Table SM3*). 118 out of the 164 active substances were approved for use in plant protection product in the EU at the time of sampling, the remaining 46 were not approved (approval status refers to January 01, 2021, the beginning of the year of the sampling campaign). 172 out of the 196 compounds were analyzed by liquid chromatography with tandem mass spectrometry-based methods (LC-MS/MS), and the remaining compounds by gas chromatography-tandem mass spectrometry (GC-MS/MS; see Supplementary Material for further analytical details). The QuEChERS (quick, easy, cheap, effective, rugged, and safe) approach was adopted for the multi-residues methods LC-MS/MS and GC-MS/MS (Brondi et al., 2011; Köck-Schulmeyer et al., 2013; Zomer and Mol, 2015). The liquid chromatography-tandem mass spectrometry instrument used was a SCIEX Triple Quad™ 6500+ LC-MS/MS System. The gas chromatography-tandem mass spectrometry instrument was a 7010B MS coupled to a 7890B GC from Agilent Technologies (Mol et al., 2016). Details on the chemicals and reagents involved in the analyses are provided in the -supplemental materials.

The quality control of the analyses was performed following the SANTE/11813/2017 guidance document on analytical quality control and method validation procedures for pesticide analysis in food and feed (Working document on the summing up of LOQs in case of complex residue definitions, 2015; EC, 2015). The method was assessed for its LOD/LOQ, linearity, recovery, precision, and matrix effects. Multi-pesticide calibration standards were prepared for each analytical method based on a solution that combined the reference standards of all compounds included in the analysis.

2.5. Data analysis

Basic descriptives were performed at sample and matrix level. The sample-level analyses cover the number of residues per sample, pesticide levels per sample, and pesticide profile per sample. The matrix-level analysis included the percentage of pesticide-free and contaminated samples - i.e. with pesticide residues $\geq$ LOD (limit of detection), mixtures similarities evaluation, and a correlation matrix between overall pesticide findings and sediment basic characteristics. The LOD of the substances was used as the reporting limit (harmonized approach among SPRINT results, see e.g., Silva et al. (2023)). The LOD is the level at which signal-to-noise for both quantifier and qualifier ions are at least 3. 138 of the tested residues had a LOD between 0.01 and 0.5 µg kg^{-1}; 32 had a LOD between 0.50 and 1.0 µg kg^{-1}, and 24 had a LOD between 1.0 and 4.4 µg kg^{-1} (see SM excel Table 1 for compound-specific information). Pesticide approval status was retrieved from the EU pesticide database, and pesticide half-life time in sediment and persistence class thresholds, were retrieved from PPDB - Pesticide Properties DataBase (table SM3).

3. Results

3.1. Number of pesticide residues in sediment samples, and mixtures composition

The number and complexity of the mixtures in the sediment samples varied among countries (Fig. 1). The highest number of compounds was found in the sediment samples from the Czech Republic, this was for the maximum number of compounds (N: 48 in sample 4, S04) and the median/average number of compounds (median: 47, average: 39). The lowest number of compounds was found in Slovenia (min: 0 max: 6, average and median: 2; Fig. 1). Sample 15, from Slovenia, was the only 'pesticide-free' sample. Samples 16 and 19, also from Slovenia, and sample 23 from Switzerland contained a single pesticide residue (S16: Imidacloprid; S19: Prosulfocarb and S23: Metolachlor_S). The remaining 34 samples contained mixtures of residues. 13 samples contained 2–5 residues, 7 samples contained 6–10 residues, 7 samples contained 11–20 residues, 5 samples contained 21–30 residues, and the remaining 2 samples, from the Czech Republic, contained 47 and 48 residues. Czech Republic has the most complex mixtures (in terms of the number of residues, and type of compounds), while Croatia and Slovenia present rather simple mixtures (Figs. 1 and 2, and SM1).

Thirty-one different mixtures were detected among the sediment samples. A two-compound glyphosate-AMPA mixture was found in 4 samples, the remaining mixtures were unique (in their full composition, *Figure SM1*). There were however common combinations among mixtures, i.e. "mixture root, MR". Fourteen "2-compound-MR" were detected in ten or more samples (Glyphosate + AMPA root appears in 23 samples – 19 in combination with other compounds, AMPA + DDE p,p' root in 17 samples, etc; Fig. 3). Eight "3-compound-MR" were detected in ten or more samples (Glyphosate + AMPA + DDEpp root in 14 samples; glyphosate + DDDpp + DDEpp in 13 samples, etc; Fig. 3). Glyphosate and/or DDT residues (i.e. glyphosate/AMPA and/or DDDpp/DDEpp) were present in all of the most common MR.

3.2. Type of pesticide residues identified in the sediments

A total of 99 different pesticide residues were found above the respective LOD (*Table SM3*). 72 of the 99 were either approved active substances or degradation products of approved substances; remaining 27 were (related to) not approved substances. Glyphosate (approved herbicide) and its main metabolite AMPA were the two most frequently detected compounds. Glyphosate was present in 23 samples, originating from 7 countries (all countries except Croatia). AMPA was detected in 29 samples out of the same 7 countries. DDE p,p' (metabolite of the long-banned DDT) was the third most common compound, detected in 19 samples from 7 countries (in this case only not detected in Slovenian samples). These top 3 substances have very different profiles. According to PPDB, glyphosate and AMPA have high solubility in water and low bioaccumulation potential (octanol/water partition coefficient, Kow<
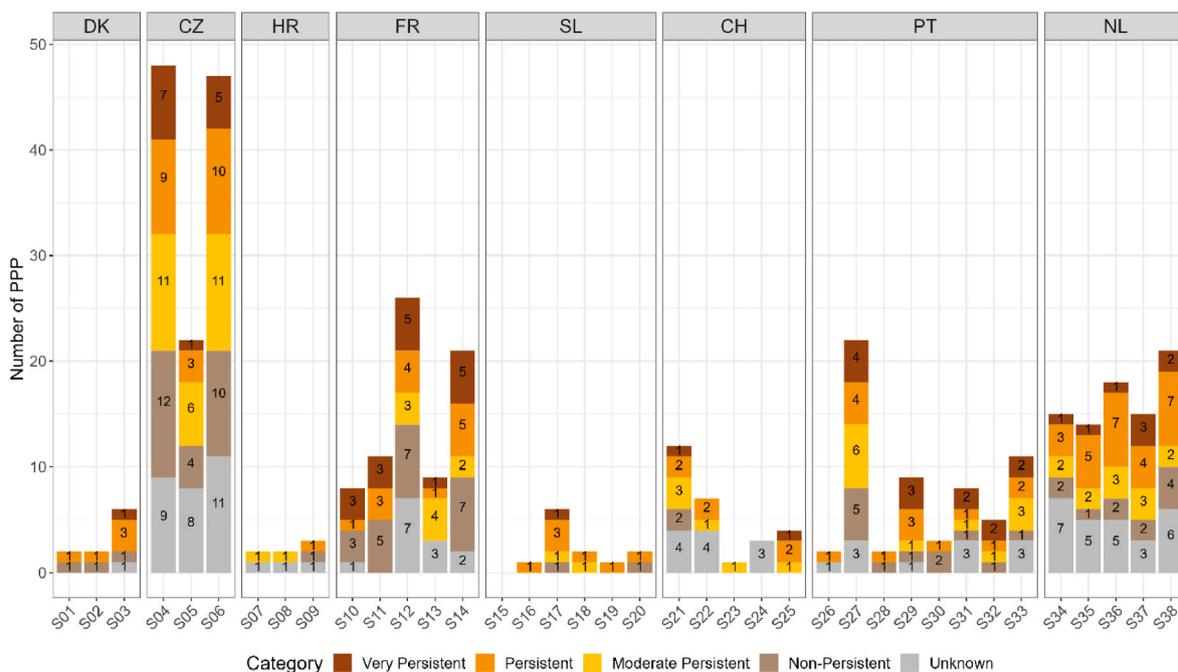
Fig. 1. Number of compounds detected in sediment samples across European Countries. Different colors represent different persistency classes in sediment, in line with PPDB classification (very persistent: $DT_{50\ WaterSediment} > 365$ days; persistent: $100\ d < DT_{50\ WS} \leq 365\ d$; moderate persistent: $30\ d < DT_{50\ WS} \leq 100\ d$; non-persistent: $DT_{50\ WS} \leq 30\ d$; unknown: $DT_{50\ WS}$ not known). DK: Denmark; CZ: Czech Republic; HR: Croatia; FR: France; SL: Slovenia; CH: Switzerland; PT: Portugal; NL: Netherlands.
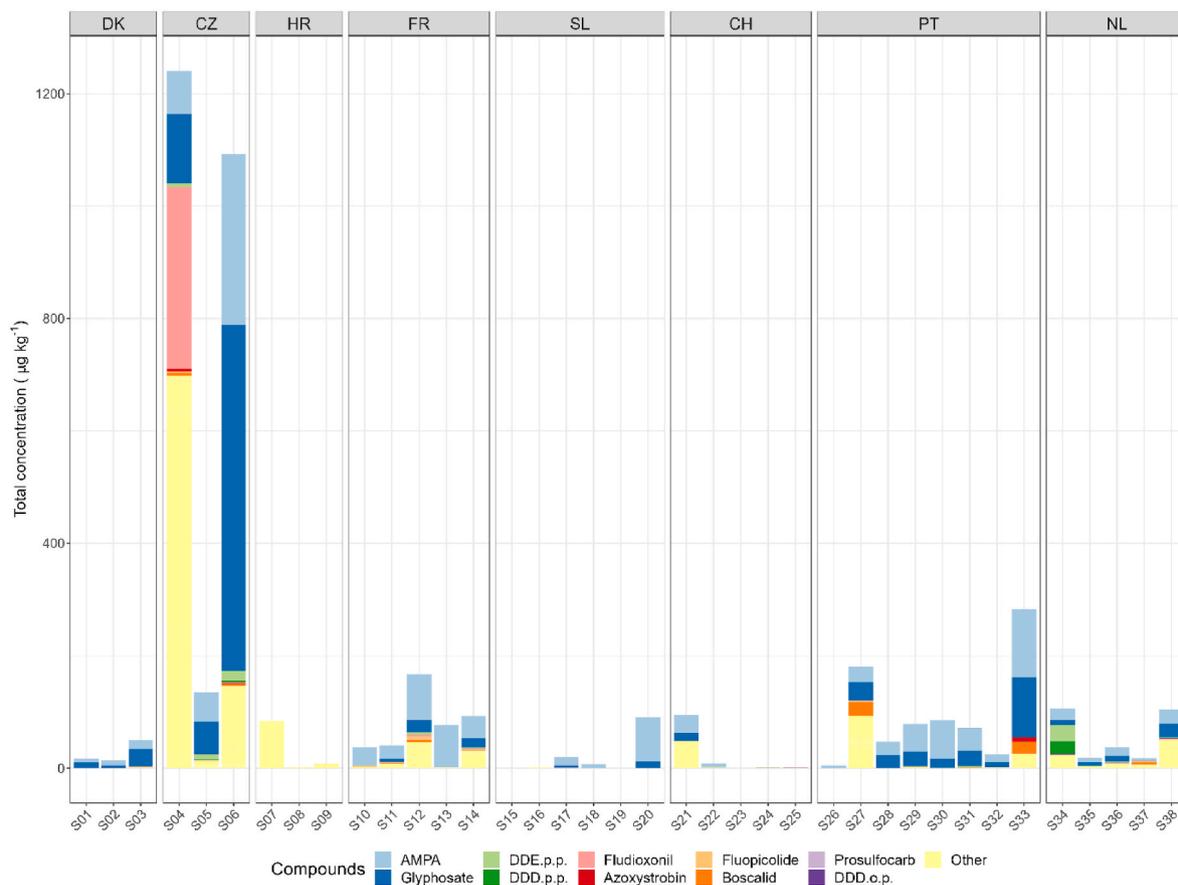

Fig. 2. Pesticide concentrations in the sediment samples ($\mu g\ kg^{-1}$). The 10 pesticides with highest concentrations are specified in the graph. The category 'others' reflects the sum of pesticide concentrations not included in the top 10. DK: Denmark; CZ: Czech Republic; HR: Croatia; FR: France; SL: Slovenia; CH: Switzerland; PT: Portugal; NL: Netherlands.
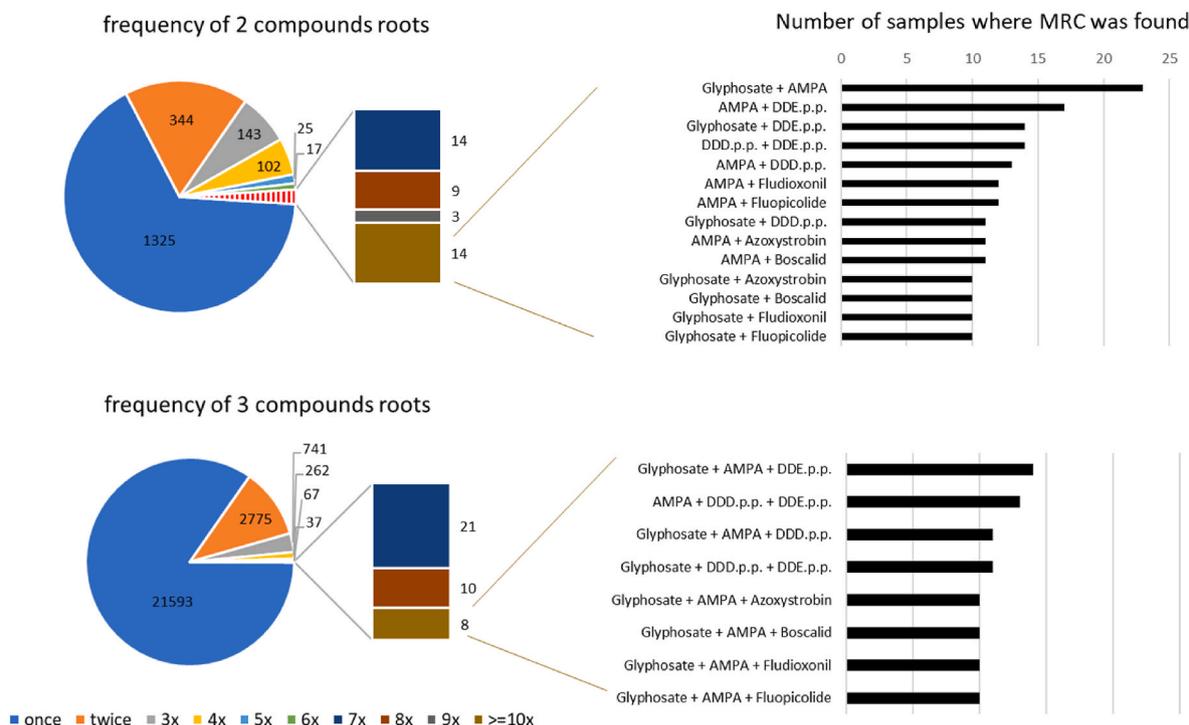
frequency of 2 compounds roots

Number of samples where MRC was found

frequency of 3 compounds roots

■ once ■ twice ■ 3x ■ 4x ■ 5x ■ 6x ■ 7x ■ 8x ■ 9x ■ >=10x

Fig. 3. Overview of similarities among pesticide mixtures. The pie charts on the left indicate how often a 2 or 3 compound combination was found among all 2 or 3 compounds possible combinations (from the pool of 196 tested compounds). The bar graphs on the right highlight the most common 2-/3-Mixture Root Compounds (MRC).

2.7), DDE has low solubility in water and a higher tendency to accumulate in sediments (Kow>3.0). Glyphosate is expected to undergo fast degradation in sediments, AMPA slow degradation and DDE is a rather stable compound, with very slow degradation. The remaining compounds had been detected mostly occasionally; 59 residues had frequencies of detection below 10%, 31 residues had frequencies of detection between 10 and 25%, and the remaining 6 residues had frequencies of detection between 25 and 50% (Fig. 2). See also table SM2 for interpretation on compound detections based on application information collected in the case studies.

The correlation matrix in Table 2 provides insights into the strength and direction of the associations between overall pesticide findings (number and total levels) and sediment characteristics. Despite the different origins of the samples, different numbers of samples per case study site/country, and other inherent variability factors linked to the samples, a strong and positive association exists between pesticide load and organic matter content. And, as somehow expected, samples with more residues present higher total pesticide contents.

3.3. Pesticide concentrations in the sediment samples

Cyprodinil and dicamba were the compounds with the highest (median) concentrations, of 116 and 83 μg kg^{-1}, respectively (Fig. 2). Spinosyn A, AMPA, difenoconazole, glyphosate, tebuconazole,

penconazole, and deltamethrin, represent the group with the second-highest levels, with median concentrations ranging from 10 to 32 μg kg^{-1}. The remaining 90 compounds had median concentrations below 10 μg kg^{-1}. The sediments from the Czech Republic presented also the highest total pesticide concentrations, ranging from 600 to 1200 μg kg^{-1}. This was followed by Portuguese, French, and Dutch samples, with total pesticide concentrations ranging between 200 and 800 μg kg^{-1}. The lowest total pesticide concentrations were found in Slovenia, Switzerland, Croatia and Denmark, ranging between 80 and 120 μg kg^{-1} (Fig. 2).

4. Discussion

In this study, we examined pesticide contamination in waterbed sediments at the European scale. Our findings corroborate off-site contamination by pesticides and that sediments can serve as sinks for pesticide residues. All sediment samples but one contained pesticide residues, in a mix of approved and non-approved substances, or metabolites. The pesticide-free sample was observed in Slovenia, in a region of maize production. This specific sample originates from the Slovenian water body with the greatest distance to agricultural fields (Navarro et al., 2024). Having information on soil types and connectivity/presence of discontinuity areas could also add to the discussion on Slovenian results. Other factors like land use, or the proportion

Table 2
Correlation matrix between variables under investigation. OM: organic matter.

	OM	Sand	Clay	Silt	Total_concentration	No_pesticide
pH	−0.13	0.32	−0.19	−0.27	0.00	−0.08
OM		−0.16	−0.19	0.24	0.63***	0.65***
Sand			−0.34*	−0.95***	0.14	0.03
Clay				0.03	−0.05	−0.17
Silt					−0.13	0.02
Total_concentration						0.87***

***: $p < 0.001$; *: $p < 0.05$; no * sign: not significant ($p \geq 0.05$).

of conventional and organic farms in the surrounding area/watershed do not seem to explain it as the water sample from this water body has tested positive for some of the analytes (Navarro et al., 2024). At the same time, it is important to stress that, although rather comprehensive, we had a targeted list of analytes. So it is possible that our pesticide-free sample, like some/all the other samples in the study, may present other not analyzed pesticides.

Glyphosate and AMPA were dominant in terms of frequency and concentration in the sediment samples, likely a consequence of the high use of glyphosate-based herbicides across European countries and their relatively high persistence in sediments (Silva et al., 2023). Previous studies reported high use but higher Kow compounds such as chlorpyrifos, cypermethrin and diazinon as top frequent and top concentration compounds in sediments (Ccanccapa et al., 2016; Masiá et al., 2013, 2015; Peris et al., 2022; Tóth et al., 2022). In our study we only detected traces of chlorpyrifos, and of its metabolites, likely a consequence of its ban in the EU in January 2020. Our cypermethrin figures were also much lower than those reported by for instance Peris et al. (2022), in the Ebro River Delta. The difference is likely related to their lower LOD for this compound and their focus on rice cultivation where the cypermethrin is used extensively (see more below on possible causes for country/region variations). Diazinon was not tested in our study as it is non-approved for use as PPP since 2007.

Due to the widespread use of glyphosate-based herbicides, we cannot inform much on the source of contamination, if from farms connected/in the vicinity to water bodies, or from more distant farms within the watershed, where this type of herbicides may have been used. It is also important to acknowledge that pesticide use in urban areas can also contribute to the pesticide findings in the waterbodies, and that application information is rather often not enough to fully explain sediment findings. The hydromorphological characteristics of the water body for instance can promote either conservation/accumulation either degradation of pesticides. Pesticide and sediment characteristics, as already touched upon in this paper, are also key factors. Indeed, it is known that some pesticide residues have stronger and faster adsorption to sediments, and their desorption is much less effective and incomplete even after a long equilibration time. Sediment particle size and organic matter content are known to play a crucial role in the sorption-desorption dynamics (Gao et al., 1998). Such dynamics translate into a double role of sediments: sink and source of contaminants. Our study is closely linked with the sink/accumulation aspect. Anthropogenic activities and natural events can lead to the re-suspension of contaminated sediments, and remobilization of contaminants to the liquid phase, and sediment-dwelling organisms can accumulate such contaminants and introduce them into the aquatic food chain. The results of this study have also illustrated that persistent residues (legacy and or of current use; $DT50_{WS} > 100$ days) were the most common compounds in the sediments. This is even though the sediment samples were collected in the middle of the growing season. DDT residues, for instance, banned in Europe decades ago (Grieco et al., 2000), were still frequently found in the sediment samples. These compounds, and respective background levels, should be accounted for in risk assessments, and for the definition of environmental quality standards. It should be also recognized that some pesticides, currently approved in the EU market, and of moderate to high persistency like AMPA and azoxystrobin, can accumulate in sediments over time, potentially leading to concerningly high concentrations (Kjær et al., 2011; Grunewald et al., 2001). The identification of specific pesticides exhibiting prolonged persistence in sediment samples is of paramount importance. For targeted pesticide regulatory measures, to evaluate persistence models accuracy, etc. Our results highlight the presence of certain chemical compounds with high half-lives and/or concentrations, such as AMPA, glyphosate, DDT, fludioxonil, azoxystrobin, fluopicolide, boscalid, and prosulfocarb. This persistence may be attributed to the chemical properties of the pesticides, such as their molecular structure and hydrophobicity, making them less available for microbial degradation (Borggaard and Gimsing, 2008; Sviridov et al.,

2015). Other relevant factors include sediment composition, organic matter content, and the composition of the local microbial communities. Our study underscores the need to explore these factors in greater detail to unravel the complex interactions governing pesticide fate in sediments. Additionally, the impact of land use practices on sediment contamination status requires further investigation. Indeed, the presence of (certain) pesticides in sediments, can indicate the absence of effective integrated pest management strategies, and weak or inadequate regulations regarding pesticide use and/or water bodies protection measures (Stenberg, 2017; McRobert et al., 2003; Van den Berg et al., 2020).

This dominance of widespread use and legacy compounds (glyphosate/AMPA/DDT/DDE) was also visible in our mixture root analyses, which seems not to be the best approach to identify associations between crop type and mixtures in sediments. Nevertheless, overall, pesticide contamination was found to be country-dependent, with Czech sediments presenting the highest number of pesticide residues and the highest concentrations among sampled countries. The same pattern was observed in soils, also covered by the SPRINT campaign. Soils were collected approximately at the same time as sediments, in nearby sometimes connected agricultural fields (Knuth et al., 2024). The higher numbers and concentrations of pesticide residues in sediments of a certain country are most likely a result of more intensive and/or diverse agricultural practices in the sampled region, but also of sediment characteristics. Czech Republic samples contain the highest organic matter content among all samples which could explain at least in part the higher pesticide figures. Water analyses (from the waterbodies where sediment was collected, and sampled at the same time as sediments) show the highest total pesticide concentration in Dutch, Portuguese and French samples. Glyphosate was the compound with the highest median concentration in water, followed by 2,4-D and MCPA. The discrepancy in water-sediment-top ranks is linked with pesticide properties and soil-water partition coefficients.

Finally, alongside its wide compound and spatial coverage, the sampling design has brought some limitations to the study. The main ones are the limited number of samples and the fact we had only one sampling time. Inter- and intra-annual variations are possible due to variations in pesticide use, and seasonal natural fluctuations (e.g. rainfall regime, river flow). In this study, the relation between the residues found in the sediments and their source is not completely understood due to a lack of data, and the complexity of the problem. For future works, it is recommended to do an inventory of all farms in the watersheds, including information on the farming system, type of crop, and pesticide residues used, but also a connectivity study, exploring links and discontinuity points between the farms and the studied water bodies. Last but not least, in-situ and ex-situ impacts (re-use of dredge sediments) risk assessment, out of the scope of this paper, should be further explored. This is especially relevant for the establishment/ evaluation of sediment quality guidelines and pesticide/dredge sediment life cycle assessments.

5. Conclusions

Our sampling design, covering eight European countries, highlighted the ubiquitous presence of pesticide residues in waterbed sediments. The number, type, and concentrations of the pesticides varied among countries, highlighting the need for further explanatory driving factors and comprehensive monitoring programs. Some patterns and common challenges were still identified. Glyphosate and its main metabolite AMPA were the two most frequently detected compounds and the ones with the highest concentrations in the sediment samples. These together with DDTs were included in most of the mixtures found. Indeed, almost all sediment samples presented a mix of currently used and banned compounds, stressing challenges around diffuse pollution and pesticide legacy effects. Understanding the dynamics and impacts of (mixtures of) pesticide residues is essential for the protection and conservation of

aquatic ecosystems, but also to achieve the soil, air and water zero pollution vision of the European Union by 2050. By studying pesticide residues in sediments, we can also uncover valuable insights into their potential "chemical time bomb" aspect, highlighting the need to explore the acute and long-lasting effects of pesticide use on aquatic systems, and ultimately the need for more sustainable and environmentally conscious agricultural practices.

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CRediT authorship contribution statement

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Declaration of competing interest

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

Data availability

Data will be made available on request.

Acknowledgments

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2024.119754.

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