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Thinking big! Landscape-scale evaluation of pesticide pollution and ecological risks in a protected mediterranean wetland

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ABSTRACT

Conventional prospective Environmental Risk Assessments (ERAs) often fail to account for the complex spatial-temporal dynamics of pesticide transport and exposure, limiting their ecological relevance. In this study, we developed a spatially explicit modelling approach to assess pesticide exposure and risks in the Albufera Natural Park (ANP), a protected Mediterranean coastal wetland heavily influenced by intensive rice cultivation. The model chain integrates hydrological simulations (erahumed), pesticide dynamics in rice paddies (RiceWQ), and hydrodynamic transport (Delft3D), allowing for a comprehensive evaluation of pesticide emissions from rice paddies to drainage channels, and to the Albufera Lake. Surface water concentrations were calculated for 8 pesticides applied to three rice varieties over a period of 10 years. Ecotoxicological risks were calculated for single compounds and for pesticide mixtures using acute and chronic Species Sensitivity Distributions. The model accurately reproduced hydrological dynamics and pesticide concentrations, with the fungicide Azoxystrobin showing the best agreement with monitoring data. Through mass balance analysis, we quantified substantial pesticide inputs into the ANP and showed that their dispersion and degradation were driven by application timing, hydrological flows, and pesticide specific properties. Acute pesticide risks were identified in the rice fields, with individual pesticide PAFs (Potentially Affected Fraction) reaching up to 12 % of species for Azoxystrobin, and mixture pesticide PAFs exceeding 22 %. In the Albufera lake, the maximum acute risk reached 7 %. Chronic risks calculated based on 21-day time weighted average concentrations were driven by Azoxystrobin and Difenoconazole in late summer, with additional contributions from Penoxsulam and MCPA in early summer. Mixtures of up to five substances exceeding the chronic 5 % PAF were identified, resulting in potential risks for 40 % of species in the Albufera Lake. This study highlights the importance of considering full pesticide application schemes and landscape-scale models for assessing pesticide risks to non-target aquatic organisms in protected aquatic ecosystems.

1. Introduction

The production and use of chemicals have grown exponentially over the last 50 years, with the number of chemicals being introduced far exceeding the regulatory capacity of governments (Landrigan et al., 2018; UNEP, 2019; van der Vegt et al., 2022). To regulate pesticide use and mitigate its environmental risks, jurisdictions worldwide rely on Ecological Risk Assessment (ERA) frameworks. These typically involve a prospective evaluation of pesticide active ingredients and formulated products before market registration, as well as retrospective assessments through post-market monitoring (van der Vegt et al., 2022; Tarazona et al., 2024). Prospective ERA are typically conducted on edge-of-field water bodies following a substance-by-substance basis, which does not adequately reflect the complexities of real-world exposure scenarios, where multiple pesticides are applied within one or multiple crop cycles in heterogeneous agroecological landscapes (Brühl & Zaller, 2019; Topping et al., 2020; Morrissey et al., 2023; Tarazona et al., 2024). In this context, spatial exposure modelling has emerged as a powerful tool in ERA, particularly for evaluating the spatial-temporal distribution and impacts of pesticide mixtures on distant, interconnected habitats

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(Topping & Lagisz, 2012; Topping et al., 2013; Focks et al., 2014).

The Albufera Natural Park (ANP), Valencia (Spain), is a Mediterranean coastal wetland which encompasses an extensive area of rice paddies and a complex network of irrigation and drainage ditches that are connected to the Albufera Lake – the largest freshwater lake in Spain. Historically, the Albufera Lake functioned as a brackish water estuary and was directly connected to the sea. However, human intervention drastically altered its hydrological regime: the natural inlets were closed to facilitate rice cultivation, effectively transforming the system into a freshwater lake dependent on irrigation inflows and seasonal management (Soria et al., 2021a). Over the past 50 years, the ecological functioning of the Albufera has undergone profound changes due to the combined effects of multiple stressors (Amador et al., 2024a). Agricultural intensification, hydrological alterations, urban expansion, and nutrient enrichment have significantly impacted the structure and functioning of this ecosystem (Martín et al., 2020; Amador et al., 2024a). These changes have resulted in increased eutrophication, loss of submerged vegetation, and shifts in aquatic community structure, raising concerns about the long-term resilience of the ecosystem (Romo et al., 2005; Martín et al., 2020; Amador et al., 2024a).

Despite its eutrophication status, the ANP is subjected to intensive pesticide use during the rice growing season, including herbicides targeting competing weeds, insecticides against aphids, and fungicides to control rice blast (Martínez-Megías et al., 2023). Several studies have monitored the occurrence of pesticides in the ANP, showing significant risks formed by complex agricultural mixtures to non-target aquatic organisms (Calvo et al., 2021; Soriano et al., 2023; Martínez-Megías et al., 2024). Nevertheless, most of the studies consist of isolated monitoring events at different sampling points and thus do not allow the characterization of complete spatio-temporal dynamics of pesticide pollution, leading to inaccurate estimations of environmental exposure and risks (van den Brink et al., 2018). Understanding the complexity of pesticide exposure scenarios is key for the identification of hazardous pesticide mixtures and for performing cumulative ecological risk assessments in vulnerable ecosystems such as Mediterranean coastal wetlands.

Therefore, the aim of this study was to implement a spatial modelling approach to enhance our understanding of the distribution and risks of pesticides used in rice cultivation on the ecological integrity of a Mediterranean coastal wetland. Specifically, the objectives of this study were (1) to assess how pesticides spread across the ANP using a rice paddy model and a hydrodynamic model, (2) to conduct a mass balance to evaluate their fate and transport across environmental compartments, (3) and to quantify the ecological risks posed by individual compounds and their mixtures at the landscape-scale. Finally, we discuss how the modelling approach implemented here can be used for setting specific protection goals and for formulating sustainable agricultural strategies, which can be directly applied to other Mediterranean wetlands.

2. Materials and methods

2.1. Study area and pesticide applications

The ANP presents a heterogeneous landscape where rice cultivation is the dominant land use, as it occupies approximately 130 km^2 of the park's total area (209 km^2). Despite a generally consistent agricultural schedule, management practices vary across space and time mainly due to differences in water availability and farming strategies. To capture this complexity, the rice farming area was divided into 552 clusters according to Martínez Megías et al. (2023). Each cluster represents a set of rice paddies with the same hydrological and agricultural regime, which are influenced by the rice variety planted on each of them. Three dominant rice varieties were considered in this study: J. Sendra, which accounts for 80 % of the production area; Bomba, covering 10 % and primarily cultivated in the surroundings of the Albufera Lake; and Clearfield, also comprising 10 %, predominantly planted in the northern region of the ANP. The rice varieties were randomly assigned to the rice production clusters until they reached the target coverage within the assigned rice farming area (Fig. 1).

Each rice variety followed a specific pesticide application scheme (Fig. 1), which included one insecticide (Acetamiprid) and five herbicides (Bentazone, Cycloxydim, Cyhalofop-butyl, MCPA, and Penoxsulam) applied by agricultural truck, and two fungicides (Azoxystrobin and Difenoconazole), applied by helicopter. Differences in drainage frequency, pesticide combinations, and application timings reflected the specific requirements of the J. Sendra, Bomba, and Clearfield varieties (Fig. 1). By integrating this level of detail into crop variety, pesticide use, and hydrological management across spatial units, the modelling framework was able to reflect the inherent landscape complexity and management-driven variability of the study area.

2.2. Hydrological modelling

The spatial and temporal variability of rice cultivation, combined with its substantial water requirements, makes the Albufera a highly complex hydrological system. Water flows into the system through irrigation channels, moves through rice fields, and ultimately reaches the lake, with sluice gates controlling the lake's outflow to the sea (Fig. 1). To represent this system, we developed the hydrological component of the *erahumed* model, which operates within a sequential framework. The model begins by calculating the lake's water balance, using continuous outflow data provided by the Júcar River Basin Authority (CHJ, 2024) as a baseline. Total water inflows were then estimated, assuming that changes in lake depth result from the combined effects of inflows, outflows, precipitation and evaporation. Precipitation and evaporation data were obtained from the Benifaió meteorological station, with records provided by the Agroclimatic Information Service for Irrigation (SIAR, 2024)

To capture the spatial variability of water inputs, the total lake inflows were distributed across the 26 primary irrigation ditches, following Soria et al. (2021b). Water inflows and outflows for each rice paddy cluster were assigned to the nearest irrigation ditch. Once all clusters were linked to specific ditches, the total water inflow to the ANP was distributed among the ditches based on the cumulative surface area of the clusters they irrigate. Subsequently, water inflow and outflow at the cluster level were simulated. This was done under the assumption that the total outflow from the rice paddies matches the combined total inflow from the ditches and lake, and that the water levels in the rice paddies align as closely as possible with the water depths and drainage periods outlined in Fig. 1. Further details on the governing equations of the hydrological model and its assumptions are provided in the Supporting Information (Text S1). The output of the *erahumed* model consisted of daily time series for daily meteorological conditions for the study area, and daily water inflow, outflows and water volume for each rice cluster.

2.3. Pesticide exposure modelling in rice paddies

The autoRiceWQ model (Fuentes-Edfuf & Martínez-Megías, 2022), a Python implementation of the RiceWQ model (Williams et al., 1999), was used to simulate pesticide exposure at the rice cluster scale. The RiceWQ is a pesticide fate model used for the determination of pesticide concentrations in rice production systems, including potential contamination of surface water and groundwater. The model considers several environmental fate and dissipation processes, including volatilization, degradation, adsorption, leaching and runoff. Simulations were conducted for each of the eight pesticides, based on the application schemes outlined in Fig. 1, as well as meteorological and hydrological data generated by the *erahumed* model for each cluster (Text S1). The simulations also incorporated scenario parameters and the physico-chemical properties of the pesticides, as detailed in Tables S1 and S2, respectively. The autoRiceWQ simulations covered the period from 2013 to 2023 and

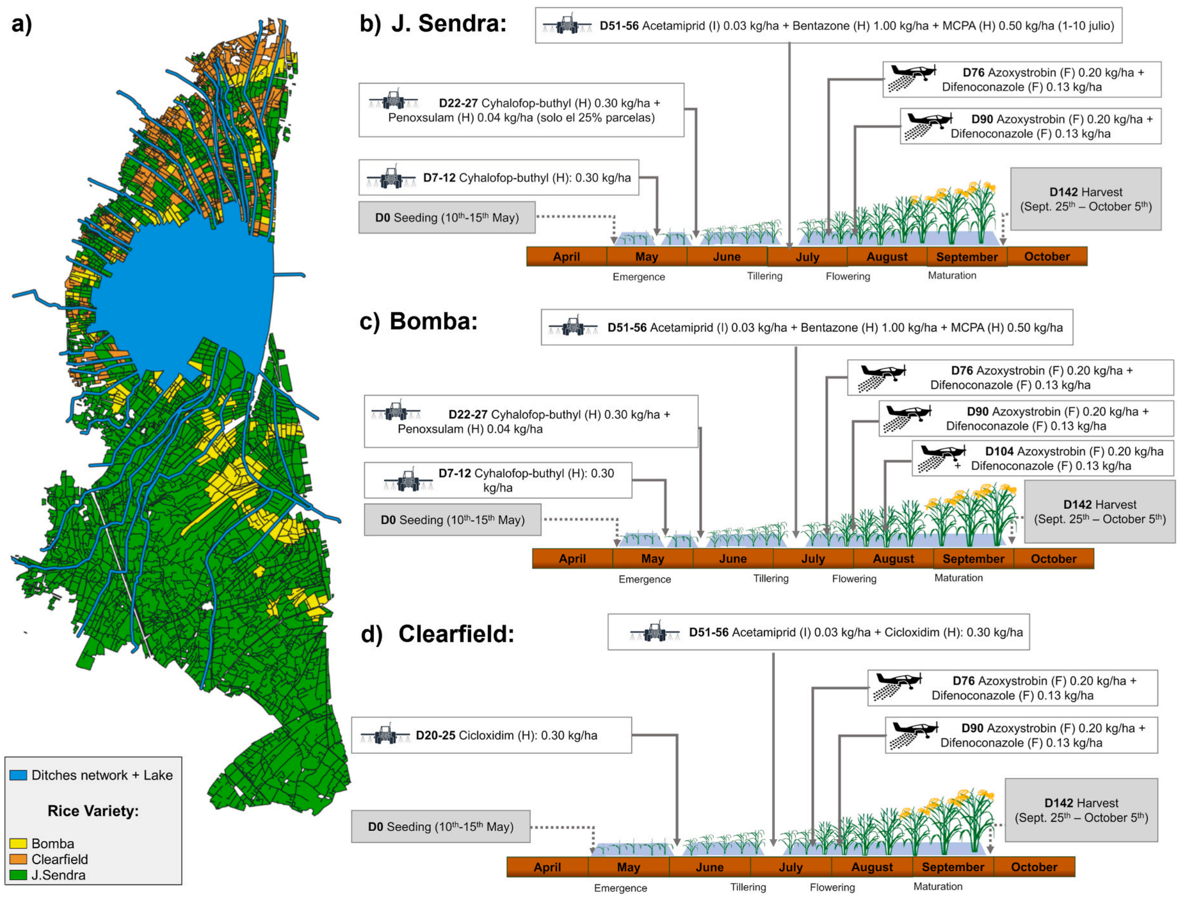

Fig. 1. Map showing the spatial configuration of the Albufera Natural Park (ANP) and the distribution of the three rice varieties (a). Pesticide application scheme and hydrological regime of the three different rice varieties cultivated in the ANP: J. Sendra (b), Bomba (c) and Clearfield (d).

produced daily time series of pesticide concentrations in the water column and in the water discharge point of each rice cluster.

2.4. Pesticide exposure modelling in down-stream water bodies

A hydrodynamic simulation was conducted to model the transport of pesticide residues from rice paddies to the Albufera Lake through the ditch network. This was done using the Delft3D Flow model (Deltares, 2025), which solves equations for fluid motion, continuity, and mass transport, allowing for a detailed representation of water and contaminant dynamics over space and time. A hydrodynamic grid was designed as a flexible mesh, where cell sizes ranged between 50 and 200 m, depending on the resolution required for the representation of the terrain morphology. Next, a hydrodynamic component was defined based on the ANP characteristics. This incorporated the drainage network of the 26 main ditches (Fig. 1), the lake's bathymetry, and its sea outlets, which were implemented as open boundaries with discharge data sourced from the Júcar River Basin Authority (CHJ, 2024). Each ditch was assumed to have a original uniform water depth of 1 m, with discharge points representing the rice fields. Each ditch included an initial point for water inflow, followed by additional discharge points corresponding to the different rice fields. These points accounted for both water volume and pesticide concentrations based on the RiceWQ model outcomes. Delft3D Flow model simulations were run in two-year blocks to manage its computational load, covering the full 10-year period from 2013 to 2023, using a time step of 1 min. The model was run using the parameter settings described in Table S3 and using the meteorological data sourced from the Benifaió station (SIAR, 2024). The model simulations generated 1-min time series of pesticide concentrations for the grid cells representing the ditches and the Albufera Lake.

2.5. Landscape-scale exposure assessment

A landscape-scale assessment of pesticide exposure concentrations was done integrating the outcomes of the RiceWQ model (for rice clusters) and the Delft3D model (for ditches and Albufera Lake). These outputs were georeferenced by joining model data to rice cluster and grid shapefiles and creating pesticide exposure concentration maps with R (R Core Team, 2024) using the *ggplot2* package (Wickham, 2016). Spatio-temporal exposure dynamics were represented for pesticide exposure concentrations in each time step, which allowed the identification of pesticide peaks in each grid cell for the calculation of acute risks, and the calculation of 21-day average exposure concentrations for the assessment of chronic risks.

Violin plots were also created using *ggplot2* (Wickham, 2016) to compare the maximum pesticide exposure distribution across habitats within the ANP. These were built with the 100 highest exposure concentrations—separately for exposure peak and 21-day average concentrations—extracted from each model run and habitat type (i.e., rice cluster, drainage ditches and the Albufera lake), ensuring a maximum of one observation per spatial unit (i.e., grid cells for ditches and the lake, clusters for rice fields). In addition, for the day of maximum exposure concentrations in the Albufera Lake, both acute and chronic exposure maps were created, allowing for the evaluation of conservative spatial patterns of pesticide exposure across different compounds and habitats.

2.6. Pesticide exposure model validation

The reliability of the model approach was assessed through a dual validation process, focusing on hydrology and pesticide exposure concentrations. As for the system's hydrology, simulated changes in lake water depth were compared with observed data from a single gauging station, using the depth measurements provided by CHJ (2024) as benchmark. The agreement between modelled and observed lake water depth series was evaluated using the root mean square error (RMSE) and the normalized root mean square error (NRMSE). The coefficient of determination (R^2) was also reported, as it remains a widely used performance metric in hydrological models (e.g. Arnold et al., 2012), despite its sensitivity to autocorrelated time series. This analysis confirmed the model's ability to replicate the lake's water depth dynamics, a critical factor for simulating water flow and pesticide exposure at the landscape-scale.

The validation of pesticide exposure concentrations focused on three pesticides: Bentazone (herbicide), Acetamiprid (insecticide) and Azoxystrobin (fungicide), for which field monitoring data was available from two sources: the long-term monitoring programme executed by the regional government in the Albufera Lake (iZONASH, 2024), and a dedicated pesticide sampling campaign conducted in 2022, which included the assessment of pesticide exposure concentrations in three sampling points of the Albufera Lake (see Text S2). Modelled mean pesticide exposure concentrations in the lake were calculated and compared with field measurements using graphical analyses and statistical metrics (RMSE and R^2) in R (R Core Team, 2024). Finally, we conducted an ad hoc sensitivity analysis focused on the role of key environmental drivers (i.e., temperature, precipitation, and water inflow) in shaping maximum pesticide concentrations across rice fields, irrigation ditches, and the lake. Further details on the methods used for this sensitivity analysis are provided in Text S3.

2.7. Pesticide mass balance

A mass balance was conducted to evaluate the fate of the applied pesticides and their losses along the hydrological pathway. This analysis involved assessing annual pesticide loads into the ANP (based on application rates), pesticide discharge from the rice paddies into the ditches (using outcomes from the RiceWQ model), pesticide loads into Albufera Lake (by defining discharge points from the irrigation ditches into the lake within the Delft3D model grid), and pesticide export to the sea (based on modelled concentrations and corresponding water discharge rates).

2.8. Ecological risk assessment

Acute and chronic risks for individual substances, as well as the cumulative risk of their mixtures, were calculated across the entire study period (2013–2023). To do so, we sourced aquatic toxicity data for primary producers, invertebrates, and vertebrates for the pesticides studied here from the ECOTOXicology Knowledgebase of the U.S. Environmental Protection Agency (US EPA, 2024). These data were subsequently screened and categorized as described in the Supporting Information (Text S4) based on Rico et al. (2021). Species Sensitivity Distributions (SSDs; Posthuma et al., 2001) for acute and chronic risks were then generated by fitting a log-normal distribution to acute and chronic toxicity data, respectively (Table S4). The resulting SSD curves, along with individual toxicity data points for acute and chronic risk characterization are presented in Figs. S4 and S5, respectively.

Ecological risks for each compound individually were determined by calculating the Potentially Affected Fraction (PAF) of species affected by the pesticide exposure concentration. Acute PAF values, which generally relate to aquatic organism mortality or immobilisation, were determined on the basis of the peak exposure concentration and the acute SSD parameters (Table S4), while chronic PAF values, which relate to effects on growth, development, or reproduction, were obtained using the 21d Time Weighted Average (TWA) concentration and the chronic SSD parameters (Table S4), for each minimum spatial unit (rice clusters and grid cells). Risks were considered to be unacceptable when the exposure concentration resulted in a PAF larger than 5 %, so that less than 95 % of species would be protected. This is consistent with widely accepted ecological protection goals and established practices in SSD-based risk assessments (Posthuma et al., 2001).

To assess ecological risks of pesticide mixtures, we calculated the PAF of species affected by the pesticide mixture in each minimum spatial unit following the msPAF approach (multi-substance PAF; De Zwart, & Posthuma; 2005). This was done by grouping pesticides according to their toxic Toxic Mode of Action (TMoA), following the classifications established by the resistance action committee's databases for fungicides, herbicides and insecticides (FRAC, 2024; HRAC, 2024; IRAC, 2024), and applying the Concentration Addition model for pesticides within the same TMoA and the Independent Action model for different TMoAs. The methods used for the calculation of the PAF and msPAF calculations are described in detail in Rico et al. (2021) and Martínez-Megías et al. (2024).

Similarly to the exposure assessment, we created violin plots showing the differences in acute and chronic PAF and msPAF values across habitats. In addition, maps for PAF and msPAF were also created for the 10-year simulation period. Then, for each habitat, the location with the highest number of pesticides exceeding the 5 % risk threshold was selected, and the evolution of risk over time was visualised using heatmaps for individual substances and the pesticide mixture.

3. Results and discussion

3.1. Model performance and validation

The model results indicate a good agreement between the modelled and observed water depth levels of the Albufera Lake. The small deviations (i.e., overestimations during the flooding period after the rice growing season and slight underestimations elsewhere) coincide with periods of large water inflow and are likely due to the assumed 1 m depth in drainage ditches in the absence of bathymetric data. It is important to note that the winter flooding phase is less relevant from a toxicological perspective, as the most critical periods of pesticide exposure take place during spring and summer. Nevertheless, with an RMSE of 0.04 m, an NRMSE of 5 %, and an R^2 of 70 %, the model accurately captured temporal variations in water depth (Fig. 2).

Regarding pesticide exposure, model performance varied across compounds. The RMSE was used to verify that the modelled concentrations were within the observed order of magnitude, while the NRMSE was employed to facilitate comparisons between compounds, given its normalisation by the range of observed concentrations. The fungicide Azoxystrobin showed the best agreement between simulated and observed concentrations, with an RMSE of 0.07 μg/L and an NRMSE of 13 %. The herbicide Bentazone exhibited the highest absolute discrepancies (RMSE of 2.7 μg/L), although the relative error (NRMSE) remained similar at 13 %, reflecting the broader range of observed concentrations. In contrast, Acetamiprid presented a low RMSE (0.07 μg/L), but a higher NRMSE of 25 % (Fig. 2). This elevated NRMSE reflects the small range of observed concentrations rather than poor model performance, as the actual deviations remain minimal.

Sensitivity analyses revealed that temperature generally reduced maximum exposure in paddies and ditches for all compounds except Azoxystrobin, likely due to its August application under high evapotranspiration conditions (Fig. S1). In contrast to the negligible effects observed for Azoxystrobin and Bentazone, rainfall led to a notable increase in acetamiprid concentrations (Fig. S2). In ditches, maximum exposure of all three compounds increased with rainfall, while in the lake, only azoxystrobin showed a marked increase, probably due to enhanced wash off and outflow from paddies following application (Fig. S2). Inflow increased maximum exposure concentrations in ditches by promoting export from paddies, but reduced it in the lake through stronger dilution (Fig. S3).

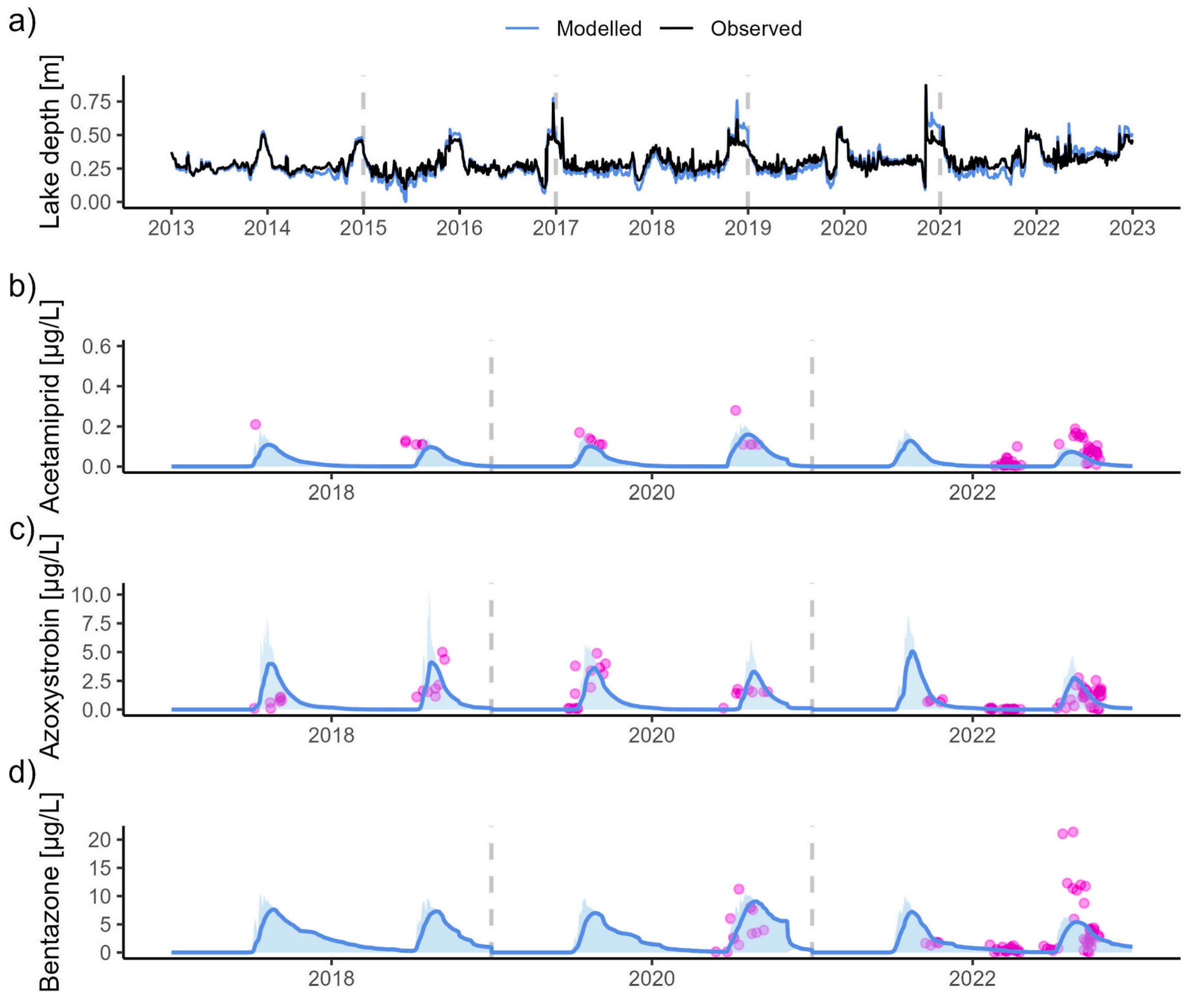

Fig. 2. Comparison of modelled and observed water depth of the Albufera Lake (a). Comparison of modelled (line) and measured (dots) pesticide concentrations in the Albufera Lake: Acetamiprid (b), Azoxystrobin (c), Bentazone (d). The line represents the median of modelled pesticide concentrations across grid cells, while the shaded area shows the range between the 5th and 95th percentiles, reflecting spatial variability in concentrations within the lake.

3.2. Pesticide exposure assessment

Azoxystrobin reached the highest water concentrations within the rice paddies, with maximum values around 100 µg/L (Fig. 3a). This is particularly noteworthy given that its application rate (0.2 kg/ha) was significantly lower than that of other pesticides like Bentazone (1.0 kg/ha) or MCPA (0.5 kg/ha; see Table S2). However, Azoxystrobin is applied over flooded fields by helicopter, which facilitates direct transfer into the water column. High concentrations (60 µg/L) were also calculated for MCPA in the rice paddies, mainly due to its low affinity for sediments, as reflected by its low water/sediment partition coefficient (Fig. 3a; Table S2). Bentazone, although applied at twice the rate of MCPA, exhibits stronger sorption to sediments, which limited its overall concentration in the water column, with peak values around 50 µg/L. For the rest of compounds, maximum concentrations in the water column of the rice paddies ranged from approximately 1 µg/L for Acetamiprid to 15 µg/L for Cycloxidim, with intermediate values of 12.5 µg/L and 3 µg/L for Difenoconazole and Penoxsulam.

Maximum concentrations for Azoxystrobin, Bentazone and MCPA in drainage ditches were comparable, with values of 30 µg/L. For the rest of compounds, concentrations were generally lower, ranging from 0.5 µg/L for Acetamiprid to 7 µg/L for Cycloxydim, with Difenoconazole and Penoxsulam reaching 6 µg/L and 1.75 µg/L, respectively.

The highest concentrations detected in the Albufera lake were those of the herbicide Bentazone, with peaks reaching nearly 20 µg/L—levels that have been observed experimentally within the lake (Fig. 2). MCPA and Azoxystrobin showed maximum concentrations in the lake close to 12 µg/L. Maximum lake concentrations for Difenoconazole, Penoxsulam, Cycloxidim, and Acetamiprid ranged between 0.3 and 1.3 µg/L, indicating comparatively lower exposure levels relative to the other compounds (Fig. 3).

The 21-day chronic concentrations for the different compounds generally followed similar patterns to those observed for the peak concentrations. However, the absolute values were strongly influenced by compound-specific physico-chemical properties. For instance, while Bentazone maintained relatively high 21-day TWA concentrations in ditches (around 25 µg/L), MCPA (21 µg/L) and Azoxystrobin (16 µg/L) showed a more pronounced decline over time (Fig. 3).

The concentrations predicted in this study are in line with values reported in previous monitoring efforts. For example, MCPA has been detected at levels around 10 µg/L in agricultural catchments and Mediterranean wetlands (Morton et al., 2021; Barbieri et al., 2021). Bentazone concentrations exceeding 20 µg/L have been documented both in the ANP and in the Ebro Delta (Wildlife Service, 2017; Barbieri et al., 2021), while Azoxystrobin has been reported at values approaching 30 µg/L in agricultural drainage waters and around 5 µg/L in the Albufera Lake (Berenzen et al., 2005; Wildlife Service, 2017). Similarly, Acetamiprid has been measured at slightly higher concentrations than those estimated by the model, with field observations reaching 0.25 µg/L in the ANP and Ebro Delta compared to the modelled maximum of 0.2 µg/L

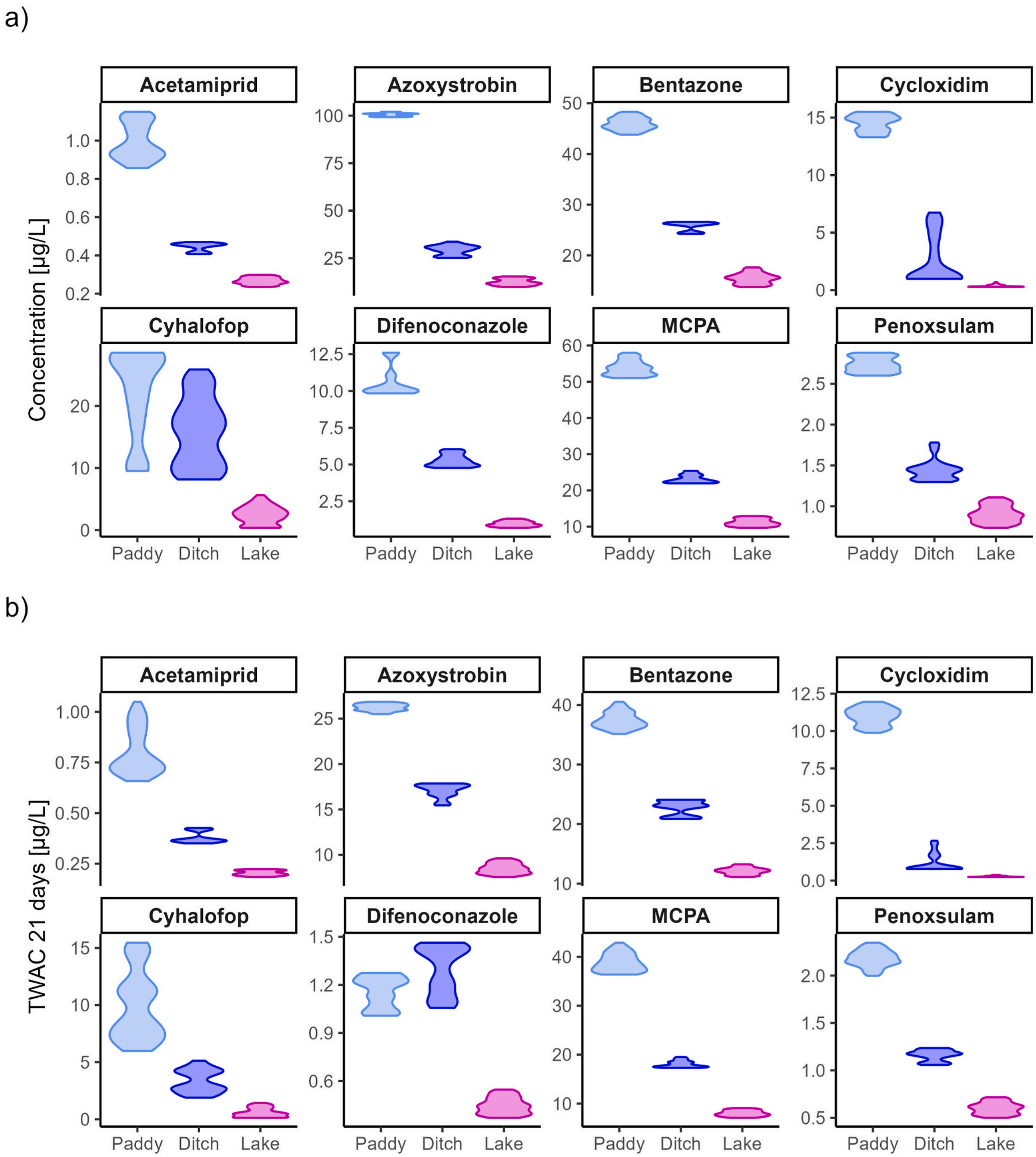

Fig. 3. Acute (a) and chronic (b) pesticide exposure concentrations across habitat types (μg/L). Violin plots represent the interquartile range (25th to 75th percentile). Distributions represent the dataset constructed from the 100 highest simulated concentrations per run, allowing a maximum of one observation per spatial unit (i.e., paddy cluster, ditches, or lake cells).

(Wildlife Service, 2017; Barbieri et al., 2021).

The spatial patterns of acute and chronic pesticide exposure across the Albufera Lake are shown in the Supporting Information (Figs. S6 and S7). Herbicides exhibited the most pronounced spatial contrasts, largely driven by the coexistence of three distinct rice varieties in the northern region, each associated with different herbicide formulations. Each compound displayed peaks in acute and chronic concentrations at different moments, with chronic exposure peaks typically occurring a few weeks later, reflecting the compound-specific degradation and transport processes (Figs. S6 and S7). Variety-specific pesticide applications, combined with runoff events and the degradation and transport properties of individual compounds, emerged as the main factors shaping the spatial and temporal distribution of exposure in the ANP. Overall, the spatial-temporal patterns in exposure observed in this study are broadly consistent with previous research. For instance, Soriano et al. (2024a, 2024b) and Calvo et al. (2021) showed higher pesticide loads in the southern part of the ANP, while the northern area is considered to be more exposed to down-the-drain chemicals from the Valencia city and peri-urban regions (Soriano et al., 2023). Sources of uncertainty in our local exposure predictions could be related to the simplifications in hydrological dynamics, such as the use of average flow rates and fixed boundary conditions; assumptions around pesticide application schedules and timing; the use of generalised degradation rates rather than site-specific data; and the lack of representation of farmer decision-making. All together, these uncertainties may have affected the accuracy of local predictions and could be addressed in follow up studies.

A noteworthy pattern observed in the pesticide exposure simulations relates to pesticide transport times. Compounds applied to (semi-)dried fields (i.e., herbicides and insecticides) tended to remain for longer periods in the paddies, reaching the drainage ditches in an average of two days, and up to three days in the case of Penoxsulam. In contrast, compounds applied over flooded fields were almost immediately transferred to the ditches. Regardless of the compound, the time between detection in the ditches and arrival to the lake was consistently short (approximately one day) reflecting the high hydrological connectivity of the system once drainage begins. These results not only raise questions about the compliance of pesticide applications with the protection goals of the ANP, but also highlight how quickly pesticides can travel.

The exposure modelling results highlight the need to account for both compound-specific properties and landscape-scale processes when evaluating pesticide exposure in agricultural wetlands, as environmental concentrations emerge from the interplay between chemical behaviour and spatial-temporal factors such as hydrological connectivity, meteorology or application timing (Focks et al., 2014). The use of a spatially explicit modelling approach allowed us to capture and quantify the spatial variability in pesticide exposure across the landscape, revealing how distinct habitats within the same system experience markedly different exposure dynamics, an essential consideration for understanding chemical pressures in complex landscapes like the ANP. Moreover, it resonates with EFSA's vision for the future of environmental risk assessment, as it represents a clear shift away from traditional edge-of-the-field assessments (EFSA, 2016). Thus, the modelling approach presented here provides a scalable and policy-relevant framework for supporting more ecologically grounded decision-making.

3.3. Pesticide mass balance

Based on the application schemes considered here, we estimated that approximately 35 tonnes of pesticides are yearly applied to the ANP (Table 1). Bentazone accounted for the highest pesticide load (with 11.6 tonnes/year), followed by Cyhalofop-buthyl (7.0), MCPA (5.8), Azoxystrobin (5.5), and Difenoconazole (3.5). In contrast, Cycloxidim, Penoxsulam and Acetamiprid were introduced in lower quantities, with 0.8, 0.5 and 0.4 tones/year, respectively (Table 1). We estimate that, on average, more than 2 ± 0.6 (mean ± standard deviation) tonnes of pesticides are discharged annually from rice paddies to ditches, accounting for roughly 6 % of the total applied amount. Bentazone and Azoxystrobin exhibited the highest pesticide emissions from the rice paddies, with 0.68 ± 0.11 and 0.64 ± 0.13 tonnes/year, respectively, which correspond to 6 % and 12 % of the total mass applied. Cyhalofop-buthyl and Difenoconazole, the two compounds with the highest sediment affinity, were mostly adsorbed and biodegraded within the rice paddies, showing the lowest emission rates. Our mass balance shows that the Albufera Lake receives a total amount of 1.5 ± 0.46 tonnes of pesticide active ingredients yearly, which roughly corresponds to 4 % of the total pesticide mass applied to the ANP. Such value, however, does not take into account pesticide transformation products and metabolites generated in the rice paddies and diches.

Most compounds followed a consistent spatial pattern, with southern ditches emerging as dominant pesticide contributors to the lake (Fig. S8). Cycloxidim was the main exception, as it is only applied in the northern part of the ANP. Our results also indicate that 0.58 ± 0.19 tonnes of pesticide active ingredients are yearly discharged into the Mediterranean Sea, with Bentazone, Azoxystrobin, and MCPA accounting for 95 % of the total discharge. From the spatial analysis, it can be concluded that 45 % is discharged by the Gola del Pujol outlet, and 20 % and 35 % by the Gola del Perelló and the Gola del Perellonet outlets, respectively.

To our knowledge, this is the first study tracking the distribution of agricultural pesticides across an entire agricultural landscape in a protected Mediterranean wetland. This achievement is particularly relevant as it provides a quantitative framework to assess the retention, transformation, and export of contaminants at the landscape-scale. Furthermore, it also demonstrates the critical role of mediterranean coastal wetlands in acting as a sink for upstream agrochemical contaminants. This function is well illustrated by the ANP, where the lake and its ditch system prevent nearly three-quarters (73 %) of the pesticide emissions from rice paddies from reaching the marine environment. However, relying solely on their natural retention capacity, risks compromising other vital ecosystem services, such as biodiversity conservation or water quality (Berg et al., 2017; See next section). In this context, the implementation of intermediate treatment systems, such as constructed wetlands to reduce pesticide loads into the Albufera Lake emerges as a promising solution, given their proven success both within and beyond the ANP (Vymazal & Březinová, 2015; Carabal et al., 2024; Martínez-Megías et al., 2024).

3.4. Ecological risk assessment

Ecological risks were assessed by means of the PAF approach for single compounds. Acute risks for single compounds were only identified in the rice paddies, with Azoxystrobin, Acetamiprid, Penoxsulam and Difenoconazole PAFs reaching 12 %, 7 %, 6 % and 6 %, respectively (Fig. S4A). Regarding chronic risks, Azoxystrobin remained the most prominent compound in the rice paddies, with a maximum PAF of 32 %, followed by Cyhalofop-buthyl (30 %), MCPA (23 %), Penoxsulam (20 %), Difenoconazole (14 %), Acetamiprid (13 %), Cycloxydim (11 %) and Bentazone (9 %). In drainage ditches, the figures were similar, with Azoxystrobin showing the highest chronic risk (25 %), followed by Cyhalofop-buthyl (18 %), Difenoconazole (16 %), MCPA (12 %), Penoxsulam (10 %), Acetamiprid (7 %), and Bentazone (4 %). In the lake, only Azoxystrobin (18 %), Difenoconazole (10 %), MCPA (9 %), Cyhalofop-buthyl (9 %), Penoxsulam (8 %), and Acetamiprid (5 %) exceeded the 5 % threshold (Fig. 4 and S10). As illustrated in Fig. S5, the

Table 1
Mass balance for pesticides applied to rice paddies in the Albufera Natural Park. Values represent the total amount applied, the amount emitted from rice paddies to drainage ditches, the inputs from drainage ditches into the Albufera Lake, and from the lake to the Mediterranean Sea. Mean values ± standard deviations are based on the 10-year simulation period (2013–2023).

Pesticide	Total applied to the ANP	Rice paddy emissions		Inputs to the Albufera lake		Inputs to the Mediterranean sea	
	(kg/year)	(kg/year)	%	(kg/year)	%	(kg/year)	%
Acetamiprid	389	14.2 ± 5.52	3.66 %	10.32 ± 3.11	2.65 %	4.01 ± 1.65	1.03 %
Azoxystrobin	5460	637 ± 112	11.7 %	365 ± 88.6	6.69 %	155 ± 43.6	2.84 %
Bentazone	11685	680 ± 145	5.82 %	795 ± 173	6.80 %	305 ± 78.7	2.61 %
Cycloxidim	779	32.5 ± 14.1	4.17 %	7.49 ± 3.19	0.96 %	2.00 ± 0.79	0.26 %
Cyhalofop	7011	70.3 ± 149	1.00 %	47.0 ± 124	0.67 %	10.2 ± 22.6	0.15 %
Difenoconazole	3549	69.0 ± 15.9	1.94 %	11.3 ± 4.46	0.32 %	5.59 ± 3.43	0.16 %
MCPA	5843	546 ± 128	9.35 %	237 ± 55.4	4.06 %	91.4 ± 37.3	1.56 %
Penoxsulam	467	42.9 ± 7.47	9.18 %	20.1 ± 5.18	4.32 %	7.28 ± 2.23	1.56 %
Total	**35183**	**2093 ± 578**	**5.95 %**	**1494 ± 457**	**4.25 %**	**580 ± 190**	**1.65 %**

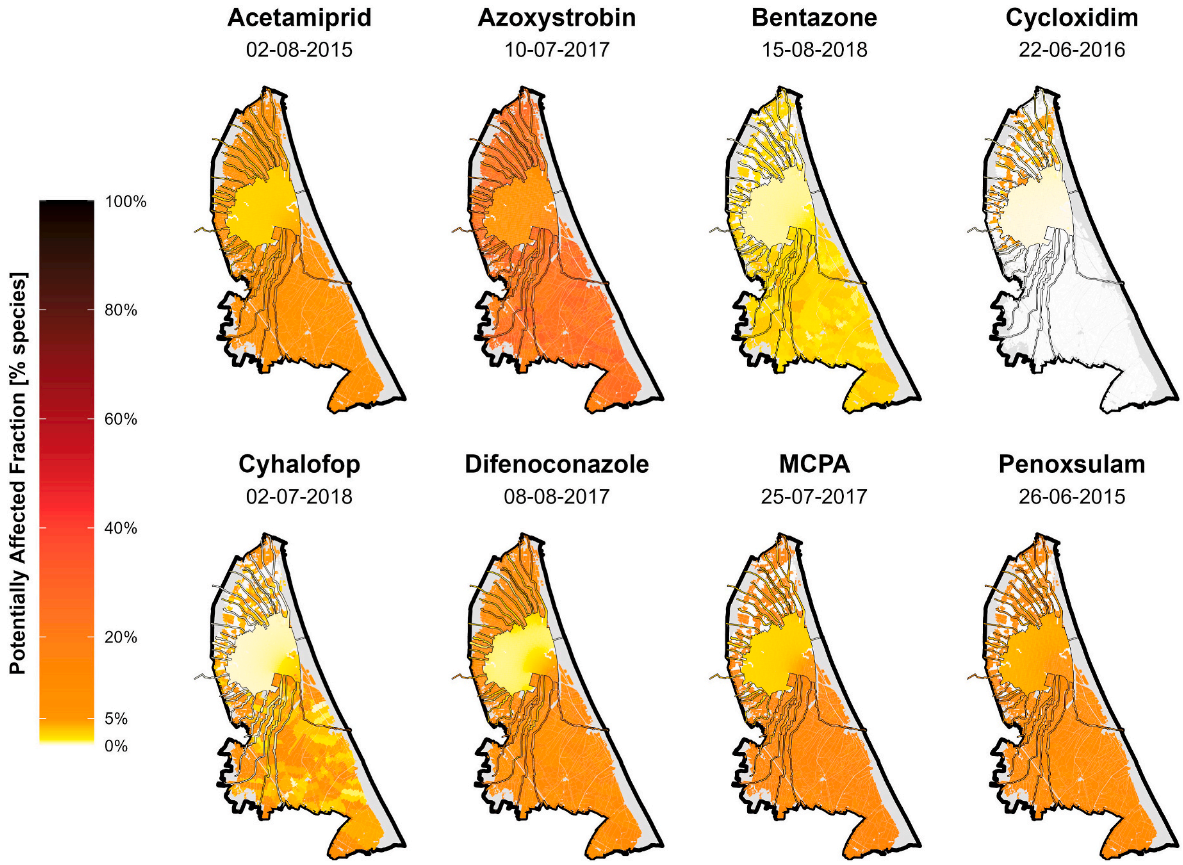

Fig. 4. Spatial distribution of chronic risks (measured as the percentage of potentially affected of species, PAF) for each compound on the day of peak exposure in the Albufera Lake across the 10-year simulation period. PAF was derived by applying the corresponding chronic Species Sensitivity Distributions (SSDs) to pesticide concentrations calculated for rice paddies (using the RiceWQ model), irrigation ditches, and the Albufera lake (using the Delft3D model).

fact that six pesticides exceed the 5 % chronic risk threshold beyond the agricultural areas is particularly concerning; not only because they include herbicides, fungicides, and an insecticide with distinct modes of action, but also because some of them (notably Azoxystrobin, Difenoconazole, and MCPA) exhibit chronic toxicity across all major biological groups (primary producers, invertebrates, and vertebrates) at environmental concentrations.

Mixture toxicity at the landscape-scale was assessed using the msPAF approach based on concentration addition and independent action depending on the toxic mode of action of the assessed compounds. The msPAF for acute exposure reached 25 % of species affected in paddies, 22 % in ditches (with substantial variation linked to episodic runoff), and 7 % in the Albufera Lake (Fig. S9B). The chronic msPAF indicated 55 %, 45 %, and 32 % of species being affected in rice paddies, ditches, and the lake, respectively (Fig. S10B). The southern section of the lake and its adjacent ditches—particularly the one connecting to the Gola del Perelló—stand out as the areas with higher acute risks posed by pesticide mixtures (Fig. 4 and S11), which is echoed in the chronic risk map (Fig. 5). This spatial pattern primarily reflects the larger area of rice fields drained by those ditches and is a direct consequence of the current hydrological management strategy, which seeks to route a substantial portion of irrigation return flows through the lake to enhance flushing capacity and mitigate eutrophication (Soria et al., 2021; Amador et al., 2024a). This finding is also consistent with previous monitoring efforts that identified the same areas as hotspots of pesticide pollution (Calvo et al., 2021; Soriano et al., 2024a; Soriano et al., 2024b). Notably, this southern sector also experienced a massive die-off of macrophytes in late July 2016 (Wildlife Service, 2017), shortly after reaching one of the highest recorded levels of coverage in the last 50 years, a situation that raises questions about the potential role of pesticide toxic pressure in triggering such ecological collapse. Moreover, Amador et al. (2024a) demonstrated that, although toxic pressure in the Albufera remains relatively stable throughout the year, pesticide-driven toxicity in terms of msPAF reaches its highest levels in August.

The dynamics of pesticide mixture toxicity are represented in Fig. 6 for three areas of the ANP representing the three different habitats. Acute risks tend to occur at specific time points and are mostly confined to rice fields and drainage ditches. Due to their short duration, complex mixtures are rare under acute exposure conditions, and only Acetamiprid and Azoxystrobin coincided with PAFs exceeding 5 %. Chronic risks reveal more frequent and persistent mixtures exceeding the threshold, particularly in rice paddies and ditches (Fig. 6). In rice paddies, there are distinct episodes where up to five substances exceed the 5 % PAF threshold simultaneously—patterns not observed in ditches or the lake (Fig. 6). Chronic toxic risk also exhibits a marked temporal pattern, in which contaminants with different modes of action and toxicity succeed one another over time, resulting in prolonged exposure windows and sustained ecological pressure. In the early stages of the rice growing season, herbicides such as MCPA, Penoxsulam, and Cyhalofop-butyl dominate the mixture toxic profile, particularly in rice paddies and in the southern sector of the ANP (Figs. 5 and 6). As the season progresses into mid-July, Acetamiprid enters the system, increasing the toxicological pressure. Chronic toxic pressure peaks late, in mid-August, fuelled by fungicide toxicity (Azoxystrobin and Difenoconazole) and the build-up due to earlier applied compounds, leading to complex mixtures of up to four substances in the lake. It is important to note that some of these substances have been shown to cause ecological impacts on ANP's functional and structural parameters in mesocosm experiments at the exposure levels identified here (Amador et al., 2024b; Grillo-Ávila et al., 2025), thereby supporting the theoretical risks established through

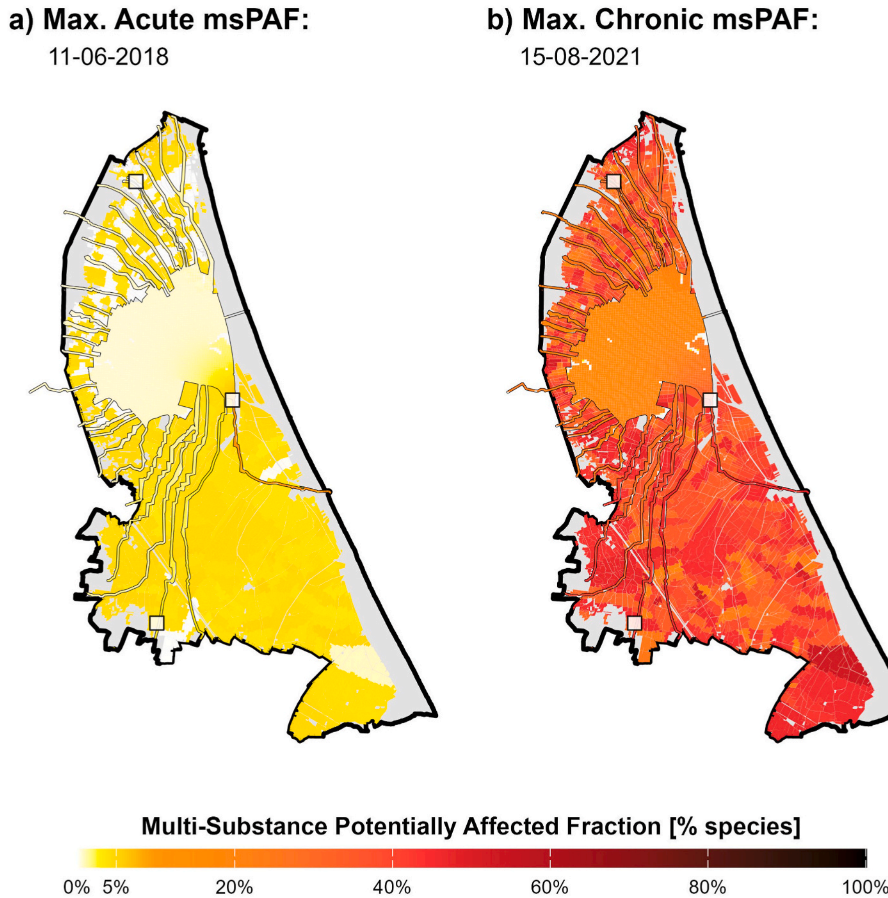

Fig. 5. Spatial distribution of acute (a) and chronic (b) pesticide mixture toxicity, expressed as the multi-Substance Pontentially Affected Fraction (msPAF), on the day of maximum risks in the Albufera Lake over the 10-year simulation period. Risk estimates were derived assuming concentration addition for compounds sharing the same toxic mode of action. White squares indicate the locations selected to represent the temporal dynamics of pesticide mixtures within each habitat type—specifically, from up to down: a paddy cluster, a lake point, and a ditch. The temporal dynamics of the calculated PAF and msPAF at these locations are illustrated in Fig. 6.

SSDs. These theoretical risks are reflected in Fig. S4 and Fig. S5, where the plotted SSDs reveal a wide diversity of potentially affected organisms. This suggests that various elements of the ecosystem's structure and function are likely to be altered, potentially disturbing the integrity of trophic interactions and key ecological processes (Relyea, 2009; Pereira et al., 2018).

Furthermore, although the modelling framework employed here assumed additivity, interactions between multiple compounds may lead to synergistic outcomes on single species endpoints, as have been shown for MCPA and Bentazone (Nielsen & Dahllöf, 2007) or among azole fungicides and azoxystrobin (Rösch et al., 2017). Due to the lack of reliable data on non-additive effects across species, we could not incorporate them into the msPAF calculations. However, as noted by EFSA (2019), concentration addition remains the default assumption in such cases, given its conservative and general applicability.

The results of this study provide an in-depth understanding of the ecological risks associated with rice cultivation in a protected Mediterranean wetland, while underlining the value of spatial modelling efforts and their alignment with the theoretical frameworks and emerging priorities in ERA outlined by EFSA (EFSA, 2016) and multiple authors (Garcia-Alonso & Raybould, 2014; Topping et al., 2020; Morrissey et al., 2023; Tarazona et al., 2024). This modelling framework not only advances scientific understanding of pesticide risks but also provides actionable insights for environmental management. It supports the definition of Specific Protection Goals (SPGs), trough the definition of spatial and temporal boundaries of species protection (EFSA, 2016). By identifying vulnerable areas and critical exposure windows, spatial risk assessments can inform practical decisions such as implementing buffer zones, prioritising monitoring efforts, or temporarily restricting pesticide use during periods of heightened ecological vulnerability (Nienstedt et al., 2012; Tarazona et al., 2024). Moreover, it helps anticipate cascading ecological effects; for instance, herbicide-driven suppression of green algae could promote cyanobacterial blooms (Lürling & Roessink, 2006), while pesticide mixtures affecting large Cladocera might compromise the resilience of clear-water states (Rumschlag et al., 2020). Ultimately, incorporating such tools into regulatory and conservation frameworks can help ensure that agricultural practices remain compatible with the long-term ecological integrity of multifunctional landscapes like the ANP.

CRediT authorship contribution statement

Pablo Amador: Writing – original draft, Methodology, Investigation, Conceptualization. **Valerio Gherardi:** Writing – review & editing, Methodology, Investigation, Conceptualization. **Yasser Fuentes-Edfuf:** Writing – review & editing, Methodology, Investigation. **Claudia Martínez-Megías:** Writing – review & editing, Methodology, Investigation. **Andreu Rico:** Writing – review & editing, Supervision, Methodology, Investigation, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

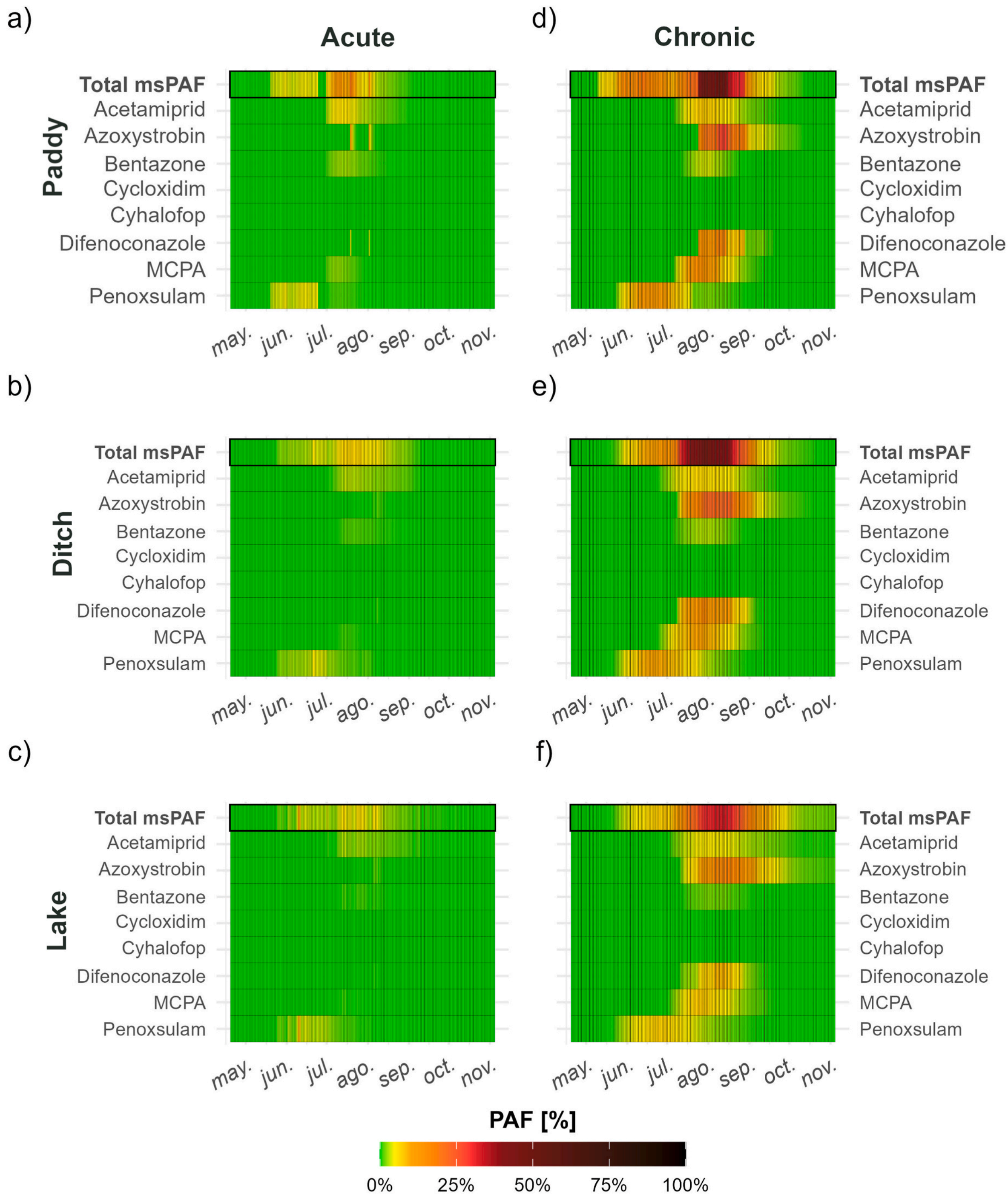

Fig. 6. Heatmaps showing the temporal dynamics of individual PAFs and msPAF for the representative locations marked in Fig. 5. Panels a) and d) display acute and chronic risks, respectively, for a rice paddy; b) and e) for a drainage ditch; and c) and f) for the Albufera Lake.

the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2025.126918.

Data availability

Data will be made available on request.

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