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RESEARCH ARTICLE

Pesticide authorization in the EU—environment unprotected?

Sebastian Stehle[1] **· Ralf Schulz**[1]

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Abstract Pesticides constitute an integral part of high-intensity European agriculture. Prior to their authorization, a highly elaborated environmental risk assessment is mandatory according to EU pesticide legislation, i.e., Regulation (EC) No. 1107/2009. However, no field data-based evaluation of the risk assessment outcome, i.e., the regulatory acceptable concentrations (RACs), and therefore of the overall protectiveness of EU pesticide regulations exists. We conducted here a comprehensive meta-analysis using peer-reviewed literature on agricultural insecticide concentrations in EU surface waters and evaluated associated risks using the RACs derived from official European pesticide registration documents. As a result, 44.7 % of the 1566 cases of measured insecticide concentrations (MICs) in EU surface waters exceeded their respective RACs. It follows that current EU pesticide regulations do not protect the aquatic environment and that insecticides threaten aquatic biodiversity. RAC exceedances were significantly higher for insecticides authorized using conservative tier-I RACs and for more recently developed insecticide classes, i.e., pyrethroids. In addition, we identified higher risks, e.g., for smaller surface waters that are specifically considered in the regulatory risk assessment schemes. We illustrate the shortcomings of the EU regulatory risk assessment using two case studies that contextualize the respective risk assessment outcomes to field exposure. Overall, our meta-analysis challenges the field relevance and protectiveness of the regulatory environmental risk assessment conducted for pesticide authorization in the EU and indicates that critical revisions of related pesticide regulations and effective mitigation measures are urgently needed to substantially reduce the environmental risks arising from agricultural insecticide use.

Keywords Pesticide · Surface water · Europe · Risk assessment · Regulation (EC) No. 1107/2009 · Regulatory acceptable concentration · Meta-analysis

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✉ Sebastian Stehle
stehle@uni-landau.de

[1] Institute for Environmental Sciences, University Koblenz-Landau, Fortstrasse 7, 76829 Landau, Germany

Introduction

Agricultural areas cover 40 % (174.1 million hectares) of the total land area of the EU-28, and two thirds (65.8 %) of these farmlands are used for the cultivation of arable and permanent crops (Eurostat 2013). In 2013, pesticides with an approximate input value of 11 billion Euros were applied to European arable lands (European Commission 2014). The widespread and intentional release of these highly biologically active substances poses threats to non-target aquatic and terrestrial ecosystems across the EU. Surface waters are especially at risk as systems that are likely to receive agricultural non-point source inputs due to their often close proximities to arable lands (Stehle and Schulz 2015; Davies et al. 2008; Schulz 2004). We focus here on insecticides, as this particularly toxic group of pesticides exhibits a high toxicity potential for aquatic organisms that are crucial for ecosystem structure and functions (Schulz 2004; Schäfer et al. 2012; US EPA 2014).

Stehle and Schulz (2015) showed that insecticides threaten aquatic biodiversity on a global scale, but did they not specify results, e.g., for the highly regulated EU. Although large-scale

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investigations of insecticide exposure of EU surface waters are lacking (Stehle and Schulz 2015), a recent study (Malaj et al. 2014) using governmental monitoring data and standard toxicity data derived from a single species laboratory test showed that, out of various organic pollutants, insecticides particularly jeopardize the integrity of EU freshwater ecosystems. In addition, several additional small-scale field studies conducted in the EU reported that pesticide exposure produced adverse effects on the aquatic ecosystem structure and function (e.g., Bereswill et al. 2013; Schäfer et al. 2012; Beketov et al. 2013; Schulz 2004). However, no scientific study exists that has evaluated pesticide, or specifically insecticide, field concentrations in EU agricultural surface waters using the regulatory acceptable concentrations (RACs) defined by the environmental risk assessment conducted for official EU authorization. The present study thus particularly addresses for the first time the essential question of whether the fundamental assumption of this pre-authorization EU pesticide risk assessment, i.e., that RACs are not exceeded in the field, is indeed met.

The new EU Regulation (EC) No. 1107/2009 (European Commission 2009a), which replaced Directive 91/414/EEC (European Commission 1991), together with the recently updated guidance document on tiered risk assessment for plant protection products (EFSA 2013, taking effect 01. January 2015), form the basis of the environmental regulatory risk assessment, which is mandatory for the authorization of active substances in the EU. Generally, the EU regulatory risk assessment is based on a single active ingredient toxicity assessment concept, and it follows a tiered approach, in which higher tiers are less conservative but more complex and are meant to be more realistic than lower tiers (EFSA 2013). Tier I of the aquatic effect assessment consists of acute and chronic laboratory toxicity tests using standard test organisms and the application of large assessment factors (AFs, i.e., 100 for acute and 10 for chronic toxicity tests) for RAC derivation (see EFSA (2013) for details). In cases in which there is an unacceptable risk indicated in this first tier, higher tier studies, such as species sensitivity distributions and aquatic micro-/mesocosm tests, are performed to derive an RAC, which is considered more realistic and less conservative (EFSA 2013). In particular, micro-/mesocosm studies are often conducted for the refined risk assessment of insecticides (Table 1). The AFs applied to these higher-tier studies are substantially lower than tier-I AFs and are set on a case-by-case basis (see EFSA (2013) for details). RAC comparisons with the predicted environmental concentrations (PECs) derived from exposure modeling (see FOCUS (2001) and EFSA (2013) for details) thus indicate either an acceptable risk for aquatic ecosystems or the need for a specific application prescription (e.g., no-spray buffer zones close to surface waters) that becomes part of the registration procedure as legally binding label amendments for the farmer. Overall, the EU Regulation (EC) No. 1107/2009 claims that a high level of environmental protection is required (e.g., in article 1 and 4.3). In detail, this directive states that "no unacceptable effects on the environment" shall result from pesticide use and particularly refers in this context to biodiversity (European Commission 2009a). In addition to these general protection goals, Nienstedt et al. (2012) and EFSA (2010) defined specific protection goals for main groups of aquatic organisms (algae, aquatic plants, aquatic invertebrates, aquatic vertebrates, aquatic microbes) covering ecosystem services potentially affected by pesticides. In general, to maintain ecosystem services and thus to adhere to these specific protection goals, aquatic taxa need to be protected at the population level (see Nienstedt et al. (2012) and EFSA (2010) for details).

Therefore, after a pesticide is authorized and in use, field concentrations exceeding their RACs must not occur in order not to compromise pre-authorization risk assessment outcome and to adhere to the general and specific protection goals outlined in EU pesticide legislation (EFSA 2010, 2013; European Commission 2009a; Nienstedt et al. 2012). Based on a meta-analysis of field studies conducted by Beketov et al. (2013), Stehle and Schulz (2015) argued that aquatic biodiversity is reduced by 29 % at agricultural stream sites with insecticide concentrations only slightly (i.e., a factor of 1.12) above regulatory threshold levels relative to uncontaminated sites. It follows that insecticide concentrations exceeding their RAC in the field in fact lead to unacceptable effects on aquatic biodiversity. Consequently, the extent of RAC exceedances in EU surface waters reveals two important details: (i) the actual protectiveness and effectiveness of pre-authorization regulatory risk assessment schemes and thus EU pesticide legislations and (ii) the significance of insecticide exposure as a threat to aquatic biodiversity in EU surface waters. However, despite extensive decades-long pesticide application in European agricultural areas, this information has never been analyzed on a European scale. Such an analysis is urgently needed considering that a recent study (Knäbel et al. 2012) revealed substantial failures in the European regulatory pesticide exposure assessment due to insecticide surface water concentrations. However, this study did not provide any information on the relationship between insecticide surface water concentrations and the RACs.

Therefore, the present study had the following three objectives:

i. To evaluate the overall protectiveness of the official EU regulatory pesticide risk assessment conducted for pesticide authorization using the agriculturally related insecticide exposure of EU surface waters and RACs;
ii. To contextualize the different risk assessment tiers and the associated protection levels with the insecticide risks in the field and to validate the field relevance of the EU regulatory risk assessment by considering, e.g., different

Table 1 The insecticides included in the meta-analysis, their final regulatory acceptable concentrations for water (RAC_{SW}) and sediments (RAC_{SED}), their respective tiers (higher tiers denote microcosm/mesocosm studies) of the RAC_{SW} setting and their approval status under Regulation (EC) No. 1107/2009 (DG SANCO 2014)

Insecticide	Class	Status under Reg. (EC) No. 1107/2009	RAC_{SW} (μg/L)	EU risk assessment tier of final RAC_{SW} setting (tier-I RAC_{SW}[a] in μg/L)	RAC_{SED} (μg/kg)
Endosulfan	OC	Not approved	1.3[b]	Higher tier (0.02)	0.026[c]
Azinphos-methyl	OP	Not approved	0.32[d]	Higher tier (0.011)	–
Chlorpyrifos	OP	Approved	0.1[d]	Higher tier (0.001)	1.1[c]
Diazinon	OP	Not approved	2.4[d]	Higher tier (0.0041)	0.95[c]
Malathion	OP	Approved	1.25[d]	Higher tier (0.0072)	0.9[c]
Parathion-ethyl	OP	Not approved	0.024[d]	Tier I	0.13[c]
Parathion-methyl	OP	Not approved	0.073[d]	Tier I	0.96[c]
Carbofuran	Carb	Not approved	0.0205[d]	Tier I	–
Acrinathrin	Pyr	Approved	0.0087[d]	Higher tier (0.00022)	–
Bifenthrin	Pyr	Approved	0.005[d]	Higher tier (0.001)	–
Cyfluthrin	Pyr	Approved	0.0068[d]	Tier I	–
β-cyfluthrin	Pyr	Approved	0.00068[d]	Tier I	–
Cypermethrin	Pyr	Approved	0.025[d]	Higher tier (0.003)	1.8[e]
α-cypermethrin	Pyr	Approved	0.015[d]	Higher tier (0.003)	1.8[e]
Deltamethrin	Pyr	Approved	0.0032[d]	Higher tier (0.0026)	1.3[c]
Esfenvalerate	Pyr	Approved	0.01[d]	Higher tier (0.001)	0.41738[f]
Fenvalerate	Pyr	Not approved	0.0022[b]	Tier I	0.88[f]
λ-cyhalothrin	Pyr	Approved	0.0021[d]	Tier I	10.5[d]
Permethrin	Pyr	Not approved	0.025[b]	Tier I	0.87[c]
Acetamiprid	Neo	Approved	0.5[d]	Tier I	–
Imidacloprid	Neo	Approved	0.3[d]	Higher tier (0.552)	–
Thiacloprid	Neo	Approved	1.57[d]	Higher tier (252)	–
Thiamethoxam	Neo	Approved	2.8[d]	Tier I	–

See Stehle and Schulz (2015) for further details on RAC_{SW} and RAC_{SED} derivation. "–" denotes that no sediment concentrations were reported for this insecticide in the literature; sediment refers to sediment and suspended particle concentrations

OC organochlorine, *OP* organophosphate, *Carb* carbamate, *Pyr* pyrethroid, *Neo* neonicotinoid

[a] RAC_{SW} set at the tier I level of the regulatory risk assessment for insecticides, which, however, did not pass at tier 1, meaning that a higher tier RAC_{SW} was used for final authorization

[b] BBA (2001)

[c] Crommentuijn et al. (2000)

[d] EFSA (2014); DG SANCO (2014)

[e] US EPA (2012)

[f] RAC_{SED} derived by the application of the modified EPA method according to Crommentuijn et al. (2000) and Akerblom et al. (2008)

types of water bodies and the pesticide mixture toxicity; and

iii. To relate the ecotoxicological significance of insecticide surface water exposure to those of other pesticide groups, to analyze the aquatic risks for different insecticide classes, specifically those of EU Water Framework Directive (WFD) priority substances.

The present study thus denotes an important extension of the work of Stehle and Schulz (2015) as it is the first to report insecticide RAC exceedance frequencies particularly for EU surface waters and, among others, as it subsequently contextualizes insecticide field exposure to the pre-authorization regulatory risk assessment schemes and EU pesticide legislations.

Materials and methods

Dataset on the insecticide exposure of EU surface waters

We extracted all scientific studies (n=165, published between 1972 and 2012) reporting measured insecticide concentrations (MICs, i.e., the concentrations actually detected and quantified) resulting from the agricultural non-point source pollution of surface waters for the 28 EU member states from the global insecticide exposure dataset provided in Stehle and Schulz (2015; see this publication for detailed information on the entire literature review process, selection criteria and information retrieval). The dataset evaluated here thus represents an exhaustive compilation of Europe-wide insecticide surface water concentrations. In addition to the insecticide concentrations in the water (μg/L), sediment or suspended particles (μg/kg), the scientific studies provided information on the sampling location (including the distinction between freshwater and estuarine waters and the hydrology of surface water bodies), the catchment size, the sampling interval, the sampling date, the limit of quantification (LOQ), and the quantity and concentrations of additional pesticides present in a given sample. Further on, we classified the certainty that the MIC resulted from an agricultural non-point source entry.

In total, our analysis comprised MICs of 23 insecticide compounds, and 15 of these 23 insecticides are currently authorized for agricultural uses in the EU under the new pesticide Regulation (EC) No. 1107/2009 (Table 1); the other eight compounds that are currently not authorized had, however, formerly been authorized for agricultural uses in the EU. We classified these compounds for further analyses into four (organochlorines, organophosphates and carbamates (named "organophosphates"), pyrethroids, neonicotinoids) generations of insecticide classes (see Table 1) based on their ecotoxicological mode of action (Yu 2008) and the time period of their market introduction (Denholm et al. 2002).

Compilation of European RACs

The derivation and application of the RACs were as follows (see Stehle and Schulz (2015) for further details): The RAC_{SW} (Table 1) were used to evaluate the measured insecticide concentrations in the water phase (MIC_{SW}). The RAC_{SW} were derived from official European pesticide registration documents (EFSA 2014; DG SANCO 2014) and denote the final acute tier-I or higher-tier ecotoxicity endpoints, including the AF determined within the regulatory aquatic risk assessment of their respective insecticide compounds. As no official European pesticide registration documents were available for the insecticides endosulfan, fenvalerate, and permethrin, we used the toxicity endpoints and associated AFs provided by the German Federal Office of Consumer Protection and Food Safety (BVL) for their respective RAC_{SW} (BBA 2001). Further details, EU risk assessment tiers of the final RAC_{SW} setting, and references for RACs are specified in Table 1.

RAC_{SED} (Table 1) are not determined by default for all pesticide compounds within the official EU regulatory risk assessment procedure (EFSA 2013; DG SANCO 2002); this threshold level was thus only available from EU risk assessment documents for the insecticide lambda-cyhalothrin. To overcome this limitation, we applied the RAC_{SED} derived from the regulatory US EPA pesticide ecological risk assessment (US EPA 2012; available for cypermethrin and cypermethrin-alpha) or, in cases in which no official EU or US RAC_{SED} was available, maximum permissible concentrations (referred to here also as RAC_{SED}; Crommentuijn et al. 2000) to insecticide sediment concentrations (MIC_{SED}).

An evaluation of the EU regulatory risk assessment using MICs

We evaluated the protectiveness and field relevance of the pre-authorization EU pesticide regulatory risk assessment and underlying EU pesticide legislations and guidance documents in the following contexts:

First, we assessed the overall protectiveness of the regulatory EU pesticide risk assessment and the ecological significance of insecticide exposure by comparing all MICs to the respective EU-level RACs for the approval of active substances.

Second, we evaluated the insecticide exposure of the water bodies specifically considered in the EU pesticide regulatory risk assessment, i.e., the small edge-of-field freshwater bodies in close proximity to agricultural fields (European Commission 2009a; EFSA 2013; FOCUS 2001). We therefore restricted the evaluation of our dataset to MICs reported for water bodies with catchment sizes of up to 1 km^2 (i.e., the water body size used in the regulatory FOCUS exposure assessment (FOCUS 2001; EFSA 2013)) and to MICs reported for water bodies with catchment sizes of up to 10 km^2 in order to be less restrictive about the specific focus of the EU regulatory risk assessment and to include surface waters that are still typical for agricultural landscapes (Davies et al. 2008) but are not particularly addressed under the EU WFD. We further distinguished between different types of surface waters, i.e., we evaluated the MICs separately for freshwater and estuarine water bodies. As the regulatory pesticide risk assessment and the resulting RACs are valid only for MICs caused by agricultural non-point source pollution (European Commission 2009a; EFSA 2013), we further restricted our dataset to the MICs definitively attributable to this source using information, e.g., on land use, insecticide application schemes, and the routes of entry provided in the scientific studies (see Stehle and Schulz (2015) for detailed classification criteria). This strict classification procedure enabled us to attribute a specific insecticide concentration to agricultural non-point source pollution with high confidence and to subsequently analyze those exposure incidences separately. It is important to note that all these restricted analyses led to even worse outcomes, i.e., even higher RAC exceedance rates; therefore, the evaluation of the EU pesticide risk assessment using the entire dataset indicates less risk than is actually present.

Third, from the official EU pesticide registration documents, we determined whether the final RAC_{SW} used for the authorization of a given active substance was derived from the first tier of the regulatory risk assessment, or, in cases in which tier I was not passed, by conducting higher-tier effect assessment studies. In the latter case, we also extracted the associated tier-I RAC_{SW} from the respective registration documents (Table 1). Given this information, we evaluated the MIC_{SW} separately for (i) the compounds finally regulated by tier-I risk assessment and (ii) the compounds regulated using higher-tier RACs. In addition, we applied in an additional assessment respective tier-I RAC_{SW} to all MIC_{SW}, i.e., also to the insecticides that were in fact authorized using higher-tier risk assessments.

Finally, we separately evaluated the RAC exceedance frequencies for the different insecticide substance classes (i.e., organochlorines, organophosphates, pyrethroids, neonicotinoids) and for the different pesticide groups (i.e., herbicides, fungicides, insecticides). Regarding the latter, we evaluated the differences in pesticide water-phase concentration levels, tier-I RAC_{SW} values, as determined by the official EU pesticide risk assessment of a given pesticide compound, and the respective concentration to tier-I RAC_{SW} ratios for fungicides, herbicides, and insecticides using all the samples analyzed for multiple pesticide exposure. In detail, we extracted from samples containing pesticide mixtures in addition to insecticide concentrations, the concentrations of all further pesticide compounds detected.

It is worth mentioning that the pesticide registration in the EU is based on a two-stage registration system, with an initial assessment of active substances at the EU level (which is considered in this study) and the subsequent registration of plant protection products containing approved active substances by member states. However, member states can only authorize the use of plant protection products after an active substance has passed the EU regulatory risk assessment and has been added to the list of approved active substances eligible for agricultural uses in the EU.

Water Framework Directive: an assessment of priority substances

The EU WFD 2000/60/EEC (European Commission 2000) uses a retrospective risk assessment approach by comparing chemical monitoring data with environmental quality standards (EQSs) for EU-wide priority substances. We assessed the MIC_{SW} of the three compounds listed as priority substances by the WFD (i.e., endosulfan, chlorpyrifos, and cypermethrin (including isomers)) (European Commission 2013), as detected in the water bodies considered in this directive (i.e., catchment sizes > 10 km^2), by using their respective maximum acceptable concentration EQS values (MAC-EQSs) for inland surface waters. The MAC-EQSs, which should not be exceeded by a single concentration in the aquatic ecosystem of concern, are as follows: 0.01 μg/L for endosulfan, 0.1 μg/L for chlorpyrifos, and 0.0006 μg/L for cypermethrin (including isomers) (European Commission 2013).

An evaluation of pesticide mixture toxicity

To evaluate the ecotoxicological significance of mixture toxicity for EU surface waters, we compared all water-phase pesticide concentrations quantified in a given sample containing multiple pesticides (n=516 out of the total of 1140 samples analyzed) to the respective tier-I threshold levels (i.e., ecotoxicity values including AFs) for the three taxonomic groups (i.e., fishes, invertebrates, primary producers) considered in the EU regulatory risk assessment. We calculated tier-I threshold levels by dividing the lowest acute LC_{50} or EC_{50} values (compiled from PPDB (2013) and official EU pesticide registration documents (EFSA 2014)) for fish, *Daphnia*, green alga, an additional arthropod species (for substances with an insecticidal mode of action), and macrophytes (for substances with a herbicidal mode of action) by their respective AFs (i.e., 100 in the case of fish, *Daphnia*, and arthropods and 10 in the case of primary producers; EFSA 2013).

The mixture toxicity was calculated separately for each respective taxonomic group by summing up the concentration to tier-I threshold level ratios for all the pesticides detected in a surface water sample to obtain the risk quotient of the mixture (RQ_{mix}) for a given taxonomic group:

$$\mathrm{RQmix} = \sum_{i=1}^{n} \frac{\mathrm{MPCi}}{\mathrm{TLi}}$$

where MPCi is the measured pesticide concentration of the compound i quantified in a given sample; TLi is the acute tier-I threshold level for a given taxonomic group of the pesticide i; and with $RQ_{mix}<1$ indicating an acceptable risk for a specific taxonomic group.

We used this approach as its modified version (which uses modeled exposure data instead of MPC), and the underlying principle of concentration addition (Kortenkamp et al. 2009) is proposed by the EU Commission for the regulatory risk assessment of pesticide mixture toxicities for individual taxonomic groups (EFSA 2013). Moreover, this approach is generally considered as broadly applicable for pesticide mixture toxicity evaluations (Deneer 2000; Cedergreen et al. 2008).

Linear model analysis

We conducted a hierarchical linear model analysis to quantify the influence of different drivers on the outcome variable logarithmic MIC_{SW} to RAC_{SW} ratio. The following independent variables were entered in the analysis using a complete-case approach (Pigott 2009): (i) the log sampling interval, (ii) the log catchment size, (iii) the sampling date, and the dummy-coded categorical variables for (iv) EU risk assessment tiers of the RAC_{SW} setting (tier I ("0") vs. higher tier ("1")), (v) status under Regulation (EC) No. 1107/2009 (approved ("0") vs. not approved ("1")), and (vi) insecticide substance classes (organochlorines ("0"), organophosphates and carbamates ("1"), pyrethroids ("2")). We excluded the neonicotinoid substance class as only a total of 33 EU surface water concentrations were documented in the peer-reviewed literature, and this number was further reduced to only six MIC_{SW} available for complete-case linear model analysis.

Automatic and manual model building were used to identify independent variables and potential interactions with the highest explanatory power for the response variable logarithmic MIC_{SW} to RAC_{SW} ratio and best-fit models (see Stehle and Schulz (2015) for further details). Model checking included heteroskedasticity, the normal distribution of residuals and the influence of single observations using residual-leverage plots and Cook's distance. All computations were done with the open source software R (version 2.15.2 for Mac OS X 10.6.8).

Results

The insecticide exposure of EU surface waters: an evaluation of the regulatory risk assessment

Overall, 44.7 % (n=700 cases) of the 1566 MICs reported for EU surface waters exceeded their respective RACs. In particular, 37.1 % of the 1352 MIC_{SW} exceeded their RAC_{SW} up to a factor of 125,750, and 93 % of the 214 MIC_{SED} exceeded their RAC_{SED} up to a factor of 31,154 (Fig. 1). Information on the MICs for the 23 insecticide compounds was available for 385 sites located in 16 of the 28 EU member states (Fig. S1 in Supplementary Material), with most MICs originating from southern EU countries: Greece (n=487), Spain (n=415), and Italy (n=152). Additional summary statistics for the EU dataset are displayed in Table 2.

The temporal analyses of the insecticide exposure data (Fig. S2; Fig. S3; Table S1) indicates that risks did not decrease over time; this is in accordance with the results of the linear model analysis (Table 3), which predicted significant increases in MIC_{SW} to RAC_{SW} ratios over time when considering the influences of covariates. In total, 546 (38 %) of all

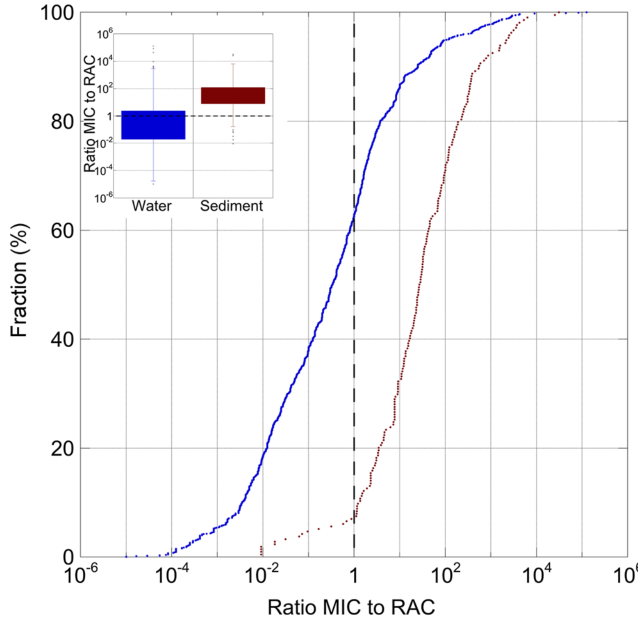

Fig. 1 Distribution curves for MICs relative to their respective RACs. *Blue* represents the MIC_{SW} relative to substance-specific RAC_{SW} (n=1352) and *brown* represents MIC_{SED} relative to substance-specific RAC_{SED} (n=214). The *inlet* shows the overall variation of the MIC to RAC ratios for water and sediment concentrations

MICs were detected after the year 2000, with 40.5 % of these exposure incidences exceeded the respective RAC (Table S1).

Approximately 90 % of all MICs (n=126) exceeded their RACs in small edge-of-field surface waters with catchment sizes of up to 1 km^2, as well as >75 % of all MICs (n=273) in the case of water bodies with catchment sizes of up to 10 km^2 (Table S2). The linear model analysis, which predicted significantly higher MIC_{SW} to RAC_{SW} ratios for smaller surface waters (Table 3), supports these results. In addition, RAC exceedance frequencies for freshwater systems (45.4 %, n=1430) were higher compared with those derived for estuarine surface waters (37.5 %, n=136) (Table S2). The restriction to exposure incidences (n=581), which could be linked with high confidence to agricultural non-point source entries, resulted in RAC exceedance frequencies of 60.9 % (Table S3).

The risk assessment of the three WFD priority substances (i.e., chlorpyrifos, endosulfan, cypermethrin (including isomers)) included in our meta-analysis showed that 57.5 % of their MIC_{SW} (n=146) exceeded their respective MAC-EQS values. All cypermethrin concentrations (n=29), as well as 73.3 % of the endosulfan (n=60) and 19.3 % of the chlorpyrifos concentrations (n=57), exceeded their respective MAC-EQS values.

The risk assessment tiers of RAC_{SW} determination and aquatic risks in the field

Ten of the 23 insecticide compounds considered here gained authorization for agricultural uses in the EU by passing tier I

Table 2 Summary statistics (the number of measured insecticide concentrations (MICs)) for important parameters of the EU insecticide exposure dataset

Parameter[a]	Minimum	25th percentile	Median	75th percentile	Maximum
Sampling date (n=1447)	1969	1989	1996	2004	2010
Catchment size (km^2, n=1320)	0.02	15	800	3315	180,000
Sampling interval[b] (days, n=1192/1054)	0.0416/0.0416	14/12	30/30	60/60	180/180
RAC exceedances per country (%, n=1566)		Belgium (n=26) 3.9 Bulgaria (n=1) 100 Cyprus (n=3) 66.7 Denmark (n=7) 100 France (n=46) 76.1 Germany (n=138) 83.3 Greece (n=487) 35.2 Hungary (n=3) 0 Italy (n=152) 54.6 Netherlands (n=60) 33.3 Poland (n=33) 27.3 Portugal (n=94) 21.3 Romania (n=5) 0 Spain (n=415) 33.5 Sweden (n=17) 94.1 UK (n=79) 78.5			
Hydrology (n=1419)		Lotic surface waters: 1211 (85.3 %); lentic surface waters: 208 (14.7 %)			
Type of surface water (n=1566)		Freshwater systems: 1430 (91.3 %); estuarine waters: 136 (8.7 %)			
Source (n=1566)		Non-point source[c]: 1222 (78 %); rainfall-induced runoff: 159 (10.2 %); rice field effluents: 81 (5.2 %); spray drift: 41 (2.6 %); aerial application: 27 (1.7 %); irrigation-induced runoff: 18 (1.1 %); drainage: 18 (1.1 %)			
Insecticide classes (n=1566)		Organochlorine insecticide: 143; organophosphorus insecticides: 1224; pyrethroids: 143; neonicotinoids: 33			

[a] There are fewer MICs for some parameters due to missing information in studies

[b] The first value is for all (water and sediment) MICs, and the second value is for MIC_{SW} only

[c] The non-point source denotes that the exact diffuse pollution source was not specified

of the regulatory environmental risk assessment for aquatic organisms, whereas 13 compounds were approved using higher risk assessment tiers (i.e., RAC_{SW} derivation using microcosms or mesocosms) (Table 1). The tier-I RAC_{SW} levels of the 10 insecticides (median 0.02225 μg/L) are noticeably lower than the RAC_{SW} levels of the 13 compounds (median 0.1 μg/L) derived through higher-tier risk assessment (Fig. 2). However, the median toxicity towards tier-I standard test organisms is approximately one order of magnitude higher (i.e., lower RAC_{SW} values) for the latter 13 compounds. Furthermore, the higher-tier RAC_{SW} of these 13 compounds are approximately 1.5 orders of magnitude higher than their associated tier-I RAC_{SW} levels (median: 0.003 μg/L; Fig. 2).

The MIC_{SW} of the 10 compounds that were approved using tier-I RAC_{SW} led to significantly (Table 3) higher RAC_{SW} exceedances (64.9 %; n=576) compared with those of the 13 insecticides that were approved using higher-tier RAC_{SW} (16.4 %; n=776; Table 4). However, if we only consider the tier-I RAC_{SW} for all 23 insecticide compounds in the assessment of MIC_{SW}, 71.4 % (n=1352) of the MIC_{SW} exceeded the RAC_{SW} (Table 4).

The risk assessment for different insecticide substance classes and pesticide groups

The MIC_{SW} of pyrethroids (n=108) led to the highest percentage of RAC_{SW} exceedances (70.4 %; see also Table 3 for a comparison of insecticide classes in the linear model analysis), followed by the MIC_{SW} of organophosphorus insecticides (37.5 %; n=1100) and neonicotinoids (24.2 %; n=33); in contrast, only 3.6 % of the MIC_{SW} (n=111) reported for the organochlorine insecticide endosulfan exceeded the RAC_{SW} (Fig. S4). Insecticide sediment exposure led to >90 % RAC_{SED} exceedance frequencies for all substance classes (organochlorine insecticides (n=32) 100 %; organophosphorus insecticides (n=124): 90.3 %; pyrethroids (n=58) 94.8 % RAC_{SED} exceedance frequencies), except for neonicotinoids, for which no MIC_{SED} was reported in the scientific literature.

We detected higher absolute field concentrations for fungicides (median 0.96 μg/L) compared with those of herbicides (median 0.063 μg/L) and insecticides (median 0.034 μg/L) (Fig. 3a) in the samples containing multiple pesticides (n= 516). However, the risk assessment for these pesticide groups showed higher tier-I RAC_{SW} exceedance frequencies for

Table 3 The results of linear model analyses predicting logarithmic MIC_{SW} to RAC_{SW} ratios (R^2=0.612; adjusted R^2=0.609; p<0.001; n=942)

	Estimate	*t* value	*p* value
Intercept	−22.270	−2.992	0.00285
Catchment size	−0.262	−9.076	<0.001
Sampling interval	−0.274	−6.566	<0.001
Sampling date	0.012	3.160	0.00163
SC (OP)	0.108	0.474	0.6355
SC (Pyr)	1.349	5.267	<0.001
RA tier (higher tier)	−2.017	−28.142	<0.001

The substance class (SC) (reference category: organochlorine insecticides) and risk assessment tier of RAC_{SW} derivation (RA tier) (reference category: tier I) were entered as dummy-coded variables, and the catchment size and sampling interval were entered as log-transformed variables. The same main effects analysis was also performed using the organophosphates/carbamates insecticide substance class as the reference category for calculating the significance level of pyrethroids vs. organophosphates/carbamates (B=1.241; t value=9.646; p<0.001). The insecticide substance class neonicotinoid was excluded due to the small number of cases (n=6) available for statistical analysis. The categorical variable authorization status under Regulation (EC) No. 1107/2009 did not show significant explanatory power for the outcome variable

OP organophosphates/carbamates, *Pyr* pyrethroids

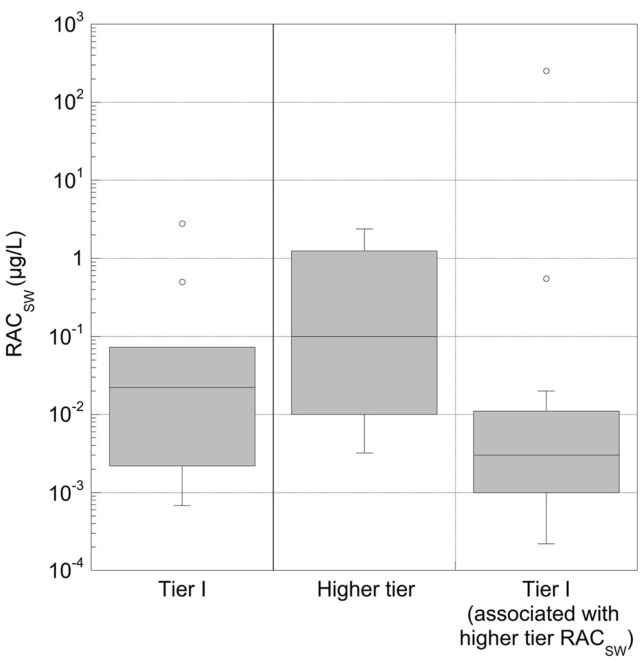

Fig. 2 A comparison of the RAC_{SW} levels derived from the different tiers of the official EU pesticide risk assessment (n (insecticides) tier-I risk assessment: 10, median RAC_{SW}=0.02225 µg/L; n (insecticides) higher-tier risk assessment: 13, median RAC_{SW}=0.1 µg/L). The tier-I RAC_{SW} associated with higher-tier RAC_{SW} (n (insecticides): 13, median RAC_{SW}=0.003 µg/L) denote RAC_{SW} derived from the first tier risk assessment for insecticides, which were finally approved using higher-tier studies (microcosms/mesocosms, see Table 1)

insecticides (53.1 %) compared with those of fungicides (31 %) and herbicides (3.8 %); in addition, the insecticide median concentration to tier-I RAC_{SW} ratio (1.25) is approximately one and two orders of magnitude higher compared with those of fungicides (0.13) and herbicides (0.019) (Fig. 3c).

Risk assessment for pesticide mixtures in EU surface waters

Overall, 135 different pesticides (66 insecticides; 42 herbicides; 27 fungicides) were detected in the 608 samples analyzed in total (i.e., water and sediment samples) for pesticide mixture occurrence in EU surface waters. Mixtures of pesticides occurred in 90 % (n=462 cases) of the insecticide water-phase samples with information on additional pesticides (n=516 out of the total of 1140 samples analyzed); these samples contained up to 13 pesticide compounds (Table S4). The results for sediment samples were comparable, i.e., 87 % of all samples with information on additional compounds (n=92) contained up to 11 pesticides.

The RQ_{mix} of the water-phase samples containing multiple pesticides (n=462) indicated the highest risks for invertebrates, as 82.7 % of these samples showed RQ_{mix} exceedances for this taxonomic group of up to a factor of 1,840,805 (Fig. 4). In relation to fish, 39.6 % of the samples had a RQ_{mix} >1 up to a factor of 18,377, whereas only 8.2 % of the water-phase samples led to a RQ_{mix} >1 for algae/macrophytes, with 829 as the highest RQ_{mix}.

Discussion

The insecticide exposure of EU surface waters: the protectiveness of EU pesticide legislation

Our meta-analysis shows that approximately 45 % of all MICs at >215 sites (i.e., >55 % of all (n=385) sites with MIC data) across the EU exceeded their respective RACs (Fig. 1; Fig. S1). It follows that insecticides are an important threat to European freshwater biodiversity, as insecticide levels > RACs lead to severe biodiversity reductions (Stehle and Schulz 2015). This conclusion is in line with smaller-scale field studies reporting pesticide-induced adverse effects on ecosystem function and aquatic biodiversity in small agricultural surface waters (e.g., Schäfer et al. 2012; Berenzen et al. 2005; Bereswill et al. 2013) and a study conducted on organic pollutants in larger EU surface waters (Malaj et al. 2014). However, for the first time, the present meta-analysis uses empirical evidence based on scientific data and official RACs to illustrate the extent of the risk for European aquatic ecosystems. In terms of regulatory implications, the risk assessment findings presented here question the fulfillment of

Table 4 An evaluation of MIC_{SW} as a function of the regulatory risk assessment tiers of the RAC_{SW} setting

	No. (%) of MIC_{SW} below RAC_{SW}	No. (%) of MIC_{SW} above RAC_{SW}
An evaluation of MIC_{SW} based on the final RAC_{SW} used for the authorization of compounds		
Insecticides with a tier-I RAC_{SW} (n=10; 576 MIC_{SW})	202 (35.1)	374 (64.9)
Insecticides with a higher-tier RAC_{SW} (n=13; 776 MIC_{SW})	649 (83.6)	127 (16.4)
An evaluation of MIC_{SW} based on tier-I RAC_{SW} for the 23 insecticide compounds		
Insecticides authorized by a higher-tier RAC_{SW} (n=13; 776 MIC_{SW})	184 (23.7)	592 (76.3)
All MIC_{SW} (n=23; 1352 MIC_{SW})	386 (28.6)	966 (71.4)

the general protection goals outlined in Regulation (EC) No. 1107/2009 and of the specific protection goals defined by Nienstedt et al. (2012) and the EFSA PPR Panel (EFSA 2010) based on the ecosystem service concept for the regulatory risk assessment of pesticides in the EU. Regarding the latter, Nienstedt et al. (2012) argued that the protection of ecosystem services for the fulfillment of the specific protection goals requires the protection of biodiversity in agricultural landscapes; our data, however, indicate clear biodiversity impairments (see also Stehle and Schulz 2015) in agricultural surface waters due to insecticide exposure. Importantly, not only the endpoints of the regulatory effect assessment (i.e., RAC) are exceeded in the field but also those of the regulatory exposure assessment (i.e., PEC; Knäbel et al. 2012; Knäbel et al. 2014); it must therefore be concluded that the current pre-authorization regulatory risk assessment schemes including associated risk mitigation obligations (i.e., pesticide application prescriptions) and underlying EU pesticide regulations, do not protect the aquatic environment. In addition, the insecticide field exposure data presented here do not provide a final conclusion on the reasons for RAC exceedances in the field, i.e., the failure of the prospective regulatory exposure and risk assessment or of farmers' adherence to regulatory risk mitigation obligations such as no-spray buffers; however, Knäbel et al. (2012) suggest both factors' contributions to insecticide risks for EU surface waters.

In addition to its overall protectiveness, our data also challenge the field relevance and focus of the EU pesticide regulatory risk assessment. Interestingly, we found the highest RAC exceedances for the MICs detected in small edge-of-field water bodies (Table S2) and for those definitively resulting from agricultural non-point source entries (Table S3). Although this finding can be explained (Schulz 2004; Stehle et al. 2013), one would nevertheless expect lower risks in surface waters and for exposure sources that are the specific focus of the aquatic regulatory risk assessment. On the contrary, surface waters not specifically targeted by regulatory risk assessment schemes, such as estuarine ecosystems (EFSA 2013), are also heavily affected by insecticide pollution; 37.5 % of the MICs exceeded their RACs, even though estuaries often are not located in close proximity to agricultural areas and non-contaminated seawater dilutes insecticide exposure (Steen et al. 1999).

There are additional issues that alert us to severe problems. First, approximately 90 % of the MIC_{SW} assessed here were measured using single or fixed-interval sampling strategies, which considerably underestimate actual insecticide exposure levels (Stehle et al. 2013; see also the result of the linear model

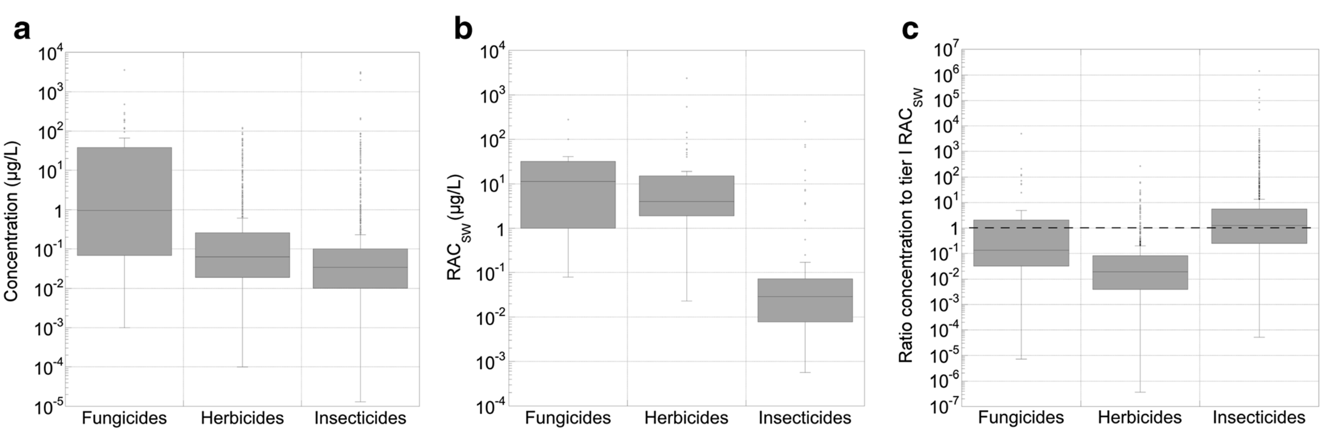

Fig. 3 Boxplots of the water-phase concentrations detected in EU surface waters (**a**), the regulatory acceptable concentrations (RAC_{SW}) derived from tier I of the European pesticide risk assessment (**b**) and related field concentration to tier-I RAC_{SW} ratios (**c**, *dashed line* indicates the RAC_{SW}) for the different pesticide groups. The comparison is based on fungicide (n=87; 23 compounds), herbicide (n=852; 36 compounds), and insecticide (n=1408; 59 compounds) water-phase concentrations detected in the 516 samples analyzed for the occurrence of multiple pesticide exposure

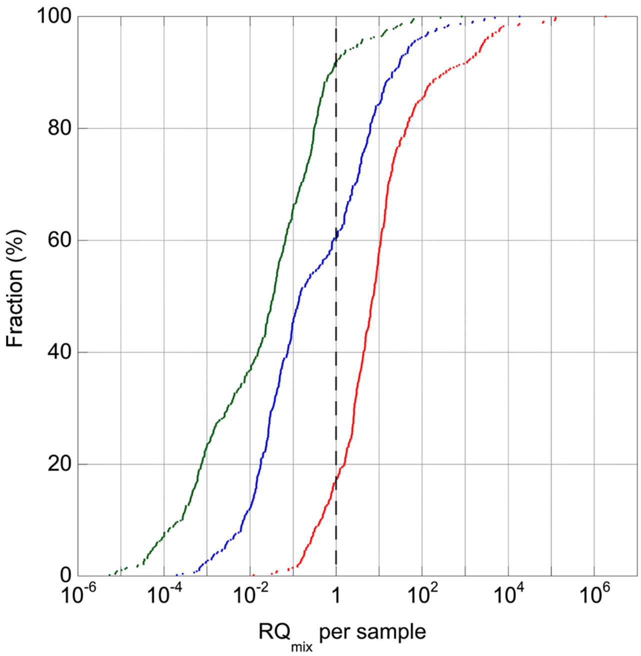

Fig. 4 Pesticide mixture toxicities detected in the water phase of EU surface water samples (n=462), expressed as risk quotients (RQ_{mix}) for algae/macrophytes (*green*), fishes (*blue*), and invertebrates (*red*). A RQ_{mix} >1 indicates a risk for the respective taxonomic group

analysis (Table 3), which indicates higher RAC_{SW} exceedances for shorter sampling intervals). Second, no scientific knowledge on insecticide surface water exposure exists for large parts (i.e., approximately 80 %) of European high-intensity agricultural areas (Fig. S1), which indicates that future monitoring studies are needed to further quantify risks across the EU; this research is of even more importance because climate change is expected to lead to increasing insecticide application in EU agriculture (Kattwinkel et al. 2011). Third, our meta-analysis shows that pesticides occur as mixtures in 90 % of the samples analyzed for multiple compounds (Table S4), with nearly 40 % of these samples containing more than 5 and up to 13 pesticides per sample. Importantly, most of the studies analyzed surface water samples for selected pesticide compounds only; thereby most likely they potentially missed compounds that were additionally present (see also Moschet et al. (2014) on this topic). However, these findings on pesticide mixture occurrences in the field challenge the protectiveness of the RAC, which is defined for single active ingredients only (EFSA 2013) and thus not covering potential combined or even synergistic effects (e.g., Denton et al. 2003; Belden and Lydy 2006). Fourth, only a marginal difference in RAC exceedances exists between the 15 insecticide compounds currently authorized in the EU and the eight compounds that are no longer approved (Table S5). This finding is supported by our linear model analysis, which could not detect a significant explanatory power for the differentiation of authorized and non-authorized compounds (Table 3). We therefore conclude that the cancelation of the authorization of obsolete active ingredients under Directive 91/414/EEC and Regulation (EC) No. 1107/2009 did not reduce insecticides' acute risks for surface waters; this claim, again, challenges the overall effectiveness of EU pesticide legislations. Within this context, we identified even higher MIC_{SW} to RAC_{SW} ratios after the enforcement of the Directive 91/414/EEC in 1993 (Fig. S2; Fig. S3) and, as opposed to the global MIC data presented by Stehle and Schulz (2015), for more recent sampling dates independent of the influence of covariates, such as the increased detection of more toxic pyrethroids in recent years (Table 3). Moreover, 40.5 % of all MICs detected since the year 2000 exceeded respective RACs (Table S1), which challenges the general perception of decreasing environmental risks (see, e.g., Lamberth et al. 2013; Devine and Furlong 2007) due to the market introduction of newer insecticide compounds and the enforcement of more stringent environmental regulations. However, other reasons not concerning aquatic organisms (e.g., high mammalian and avian toxicities of organophosphates) presumably led to the withdrawal of hazardous pesticide compounds under Directive 91/414/EEC, so that the overall environmental risks might nonetheless be reduced over time (Cross and Edward-Jones 2011).

Overall, our data and those of Knäbel et al. (2012, 2014) indicate that a critical reconsideration of the entire EU pesticide regulatory risk assessment approach including enforcement of mandatory risk mitigation obligations is imperatively needed; these findings must be seriously considered in future revisions of EU pesticide regulations. In addition, effective risk mitigation measures (e.g., Reichenberger et al. 2007; Stehle et al. 2011) have to be implemented and enforced, inter alia within National Action Plans, as requested by EU Directive 2009/128/EC (Sustainable Use Directive for Plant Protection Products (European Commission 2009b)). EU agricultural policies and subsidies should also be critically reconsidered, as they currently foster agricultural intensification and agrochemical use (Pe`er et al. 2014).

In relation to the WFD, the scientific exposure data presented here confirm recent findings based on governmental data (Malaj et al. 2014), which showed that insecticide pollution is a significant stressor in large EU surface waters. This confirmation, however, is a crucial finding, as the characterization of the chemical status of a large proportion of water bodies is still deficient due to lacking (European Environment Agency 2012) and often inappropriate (Stehle et al. 2013) governmental monitoring. Furthermore, our meta-analysis identified substantially higher RAC_{SW} exceedance frequencies (32.6 %; n=763) in large EU surface waters for the 20 non-priority substances included in our meta-analysis compared with those of the three priority substances (15.8 %; n=146). This finding challenges the WFD priority substance selection criteria (see also Von der Ohe et al. (2011) and Schäfer et al. (2011)) that currently disregard the high ecotoxicity potential of modern insecticides. Real-world exposure data and actual ecological risks in the field should trigger the future identification and prioritization of WFD priority substances.

The protectiveness of the regulatory risk assessment tiers

The MIC_{SW} of the compounds authorized using a higher-tier risk assessment show considerably lower RAC_{SW} exceedances (Table 4). This finding is in line with the general principles underlying the pre-authorization regulatory risk assessment, i.e., the outcomes of higher risk assessment tiers are less conservative compared with those of lower tiers (EFSA 2013), which consequentially leads to less frequent exceedances of these higher-tier RACs in the field. Most importantly, tier-I RACs are derived based on the ecological threshold option (ETO), which accepts only negligible effects, whereas the derivation of higher-tier RACs based on micro-/mesocosm studies generally accepts (temporary) clear population level effects (i.e., RACs derived based on the ecological recovery option, ERO-RACs; see EFSA (2013) for details). However, it is thought-provoking that such liberal higher-tier RACs drive the final regulatory risk assessment specifically of extremely toxic insecticide compounds. These insecticides have a substantially higher intrinsic ecotoxicity potential towards aquatic (standard test) organisms compared with those of the compounds authorized using tier-I RAC_{SW} (Fig. 2; Table S6). It follows that the most toxic insecticides are authorized using least conservative RACs, i.e., those based on ERO. Considering this high toxicity potential and that these liberal higher-tier RACs are set with hardly any margin of safety, they should never be exceeded in the field to prevent unacceptable adverse effects. Our data (Table 4), however, clearly disprove this assumption.

There are two more critical issues that have to be considered in this context. First, higher-tier RAC_{SW} are considerably less conservative compared with tier-I RAC_{SW} levels (Fig. 2) due to the substantial reduction of AFs (up to two orders of magnitude); however, this reduction in conservatisms is not justified by actually lower ecotoxicity potentials (Table S6). Although this AF reduction is often reasoned by the higher complexities and ecological realism of the higher-tier microcosm/mesocosms studies (EFSA 2013), the inherent limitations of these artificial model ecosystem studies (see, for example, Crane and Giddings (2004) and references therein) jeopardize the protectiveness of higher-tier RAC_{SW} for real-world situations in the field, especially in cases in which an AF of one was employed (Table S6); these limitations and the resulting uncertainties are therefore not covered by the regulatory pesticide risk assessment. In addition, the occurrence of pesticide mixtures, consecutive exposure events, and confounding factors (e.g., hydraulic stress, exposure to nutrients) in the field further challenge the protectiveness of higher-tier RAC_{SW} set with low AFs. However, according to EFSA (2013), an AF of one is not used anymore since commencement of this guideline. Nevertheless, higher-tier RAC_{SW} should, in consistency with tier-I RACs, generally be derived using the ETO and thus without already allowing for clear population level effects (sensu ERO-RACs).

Second, recent field studies (Schäfer et al. 2012; Beketov et al. 2013; Peters et al. 2013) reported pesticide-induced adverse effects at concentrations even well below (i.e., 1/10 to 1/100) conservative tier-I RAC_{SW}. In addition, based on statistical analyses, Luttik et al. (2011) argued that the AFs of 100 used for tier-I RAC_{SW} derivation may not adequately cover interspecies sensitivity variation. These findings provide evidence that even the conservative RAC_{SW} are potentially not protective in the field. An even worse protection level may thus be expected for the even less conservative higher-tier ERO-RAC_{SW}, although they have been established under conditions that are considered more realistic.

Overall, we conclude that in addition to cases with RAC exceedances, the occurrence of unacceptable adverse effects in the field can potentially not be excluded for the 35 % of MIC_{SW} that comply with conservative tier-I ERO-RAC_{SW} and are even more likely for the 83.6 % of MIC_{SW} that comply with higher-tier ERO-RAC_{SW} (Table 4). Our findings on the lack of the protectiveness of higher-tier RACs for insecticide compounds are in line with a recent study on aquatic ecosystems and fungicides (Zubrod et al. 2015), which also claimed that the higher-tier regulatory EU risk assessment does not provide an adequate level of protection. EFSA (2013) acknowledges these regulatory risk assessment shortcomings by admitting that the RAC_{SW} may not be protective for all cases occurring in the field; the effects not covered by the prospective risk assessment, the combined effects between pesticides and environmental stressors, the exposure to multiple pesticides, and the repeated exposure due to serial pesticide application are potential reasons for these uncertainties. As a consequence, EFSA (2013) postulates further strengthening the link between the RACs and real-world field situations, e.g., by conducting appropriate field studies that clearly link pesticide exposure to related effects. We believe our study addresses one aspect of this issue. However, further targeted studies are urgently needed.

Risk assessment for pesticide groups and insecticide classes: a proposal for a new hazard-based cut-off criterion

The comparison of pesticide risks shows that insecticides particularly threaten EU surface waters (Fig. 3c). This finding is explained by the substantially higher ecotoxicity potential of insecticides. The median insecticide tier-I RAC_{SW} (0.029 μg/L) is more than two orders of magnitude lower compared with those of herbicides (4 μg/L) and fungicides (11.3 μg/L; Fig. 3b). This high ecotoxicity potential of insecticides overcompensates the absolutely higher field concentrations of fungicides and herbicides (Fig. 3a), which result from higher application rates and physicochemical properties (e.g., large DT_{50} values, high water solubilities), which foster surface water exposure (Stehle et al. 2011, 2013). Our findings support those of Stehle et al. (2011), who also reported lower concentrations and higher ecotoxicological risks for insecticides compared with those of herbicides and fungicides at the inlet and outlet of vegetated treatment systems. The high ecotoxicity of insecticides, particularly for aquatic invertebrates (Devine and Furlong 2007), together with the overall high sensitivity of this group of organisms to pesticide exposure (US EPA 2014), is also a major reason that aquatic invertebrates are at risk to the largest extent when exposed to multiple pesticides (Fig. 4).

Overall, our results provide strong evidence that regulatory risk assessment and risk management for insecticides particularly needs reconsideration; targeted and more protective risk assessment concepts, stricter decision criteria, and mandatory risk mitigation obligations should be defined specifically for the authorization procedures of insecticides. However, it is important to note that field data-based meta-analyses are also needed for herbicides and fungicides to thoroughly evaluate the protectiveness and field relevance of the EU regulatory risk assessment for these pesticide groups. For example, the standard test organisms currently used in the aquatic effect assessment of pesticides are potentially unsuitable for adequately assessing fungicide effects in the field (Zubrod et al. 2015; Maltby et al. 2009).

Excluding neonicotinoids, for which a valid conclusion is hindered due to insufficient data, the development and authorization of newer insecticide classes led to an increase in acute environmental risks for surface waters (Fig. S4; Table 3), with the pyrethroids outpacing the other insecticide classes due to their extremely high toxicities for non-target organisms (Spurlock and Lee 2008) and their fast mode of action (Schulz and Liess 2000; Forbes and Cold 2005; Solomon et al. 2001). Pyrethroids' acute toxicities for fishes and invertebrates are several orders higher than those of other pesticides (Table S7), which substantially increases the ecotoxicological risks for aquatic ecosystems. Balderacchi and Trevisan (2010) showed that authorized pesticides are generally less toxic, less hydrophilic, and more rapidly degraded than non-authorized pesticide compounds; this finding, however, does not account for pyrethroids (Table S7). We therefore propose considering a new hazard-based cut-off criterion, very Toxic, fast Mode of Action (vTfMoA), in the regulatory risk assessment of pesticides. This criterion could complement the hazard-based cut-off criteria introduced by the new Regulation (EC) No. 1107/2009, which aim to enhance human and environmental health protection (Table S8). However, the exact classification schemes for the vTfMoA criterion still have to be defined, e.g., by using acute toxicity thresholds and time-to-event analyses (Newman and McCloskey 1996). The implementation of the vTfMoA criterion could substantially reduce the environmental risks caused by extremely toxic and rapidly acting pesticides, such as pyrethroids, which, despite their high acute risk potentials and related RAC exceedances, are still predominantly authorized in the EU (Table 1). However, another fact to consider here is that the introduction of additional hazard-based cut-off criteria potentially decreases the anticipated number of active ingredients to be (re-)authorized and therefore available for crop protection in Europe (ECPA 2006); this fact should not be ignored considering the increasing resistance of target pests (Denholm et al. 2002).

Until now, only very limited field data are available for neonicotinoids (Fig. S4), which, in addition to their entirely different mode of action, selectivity, plant systemicity, persistence, and resulting delayed effects (Jeschke and Nauen 2008; Tennekes and Sanchez-Bayo 2011; Sanchez-Bayo 2014), hinders a thorough assessment of their acute risks for EU surface waters. However, numerous recent studies reporting the substantial ecological effects of neonicotinoids in aquatic and terrestrial ecosystems (e.g., van Dijk et al. 2013; Hallmann et al. 2014; Chagnon et al. 2015; Goulson 2013) strongly indicate that further research is needed on the ecological consequences of neonicotinoid use. In this context, it is important to note that insecticide use patterns in the EU have changed substantially over the past few decades, with the discontinuation of many organochlorine and organophosphate insecticides and recent increases in pyrethroid and neonicotinoid use (together, with an insecticide market share of approximately 40 % in 2008; Jeschke et al. 2010). Future monitoring studies should therefore particularly focus on contemporary insecticide classes and newly introduced insecticide compounds; the sampling strategy must be suitable for the compounds of concern (Stehle et al. 2013) and must be conducted by independent organizations.

Case studies: EU regulatory risk assessments for bifenthrin and imidacloprid

This study uses the EU authorizations of the insecticides bifenthrin (EFSA 2011) and imidacloprid (EFSA 2008) to illustrate the lack of field relevance and margins of safety in the current EU regulatory risk assessment schemes. The predicted aquatic exposure concentration of bifenthrin was calculated to be 0.0049 μg/L, using FOCUS step-4 PEC_{SW} (incorporating 20 m no-spray buffer and 80 % runoff reduction) and thus making use of essentially all exposure mitigating assumptions that the FOCUS model provides. The effect assessment for aquatic organisms for this compound defined based on a higher-tier mesocosms study with a no observed ecologically adverse effect concentration (NOEAEC) of 0.015 μg/L and an AF set to 3, a RAC_{SW} of 0.005 μg/L. Overall, the final higher-tier regulatory risk assessment for bifenthin indicated an acceptable aquatic risk, as the final RAC_{SW} of 0.005 μg/L is higher than the final PEC_{SW} of 0.0049 μg/L. In essence, the active substance bifenthrin was authorized in the EU using the highest and therefore least conservative tiers in both the exposure and effect assessment, with a difference of 0.0001 μg/L (or 0.1 ng/L) between the PEC_{SW} and the higher-tier RAC_{SW}. Although this procedure appears formally correct according to legal requirements, it immediately becomes evident that, from a scientific point of view, it cannot be ensured that this small margin of safety is protective considering multifaceted field conditions. It must be concluded that the field relevance, as well as the margin of safety, of such an aquatic risk assessment is considerably questionable. Within this context, it is worth noting that all bifenthrin concentrations detected in EU surface waters (n=8) exceeded both the PEC_{SW} and the RAC_{SW}, which suggests that unacceptable effects occur in the field and further challenges the protectiveness of the current regulatory risk assessment approach for real-world situations.

The case study for the neonicotinoid insecticide imidacloprid reveals further regulatory risk assessment uncertainties. The EU authorization of this compound was based on FOCUS step-4 PEC_{SW} (incorporating 95 % spray drift reduction and 90 % runoff reduction) ranging between 0.152 and 0.429 μg/L subject to crop and FOCUS scenarios (see EFSA (2008) for details). The higher-tier RAC_{SW} of 0.3 μg/L was based on a mesocosm study with a NOEC of 0.6 μg/L and an AF of 2. It follows that the risk assessment already forecasts surface water concentrations potentially to exceed the RAC_{SW} under certain conditions. EFSA (2008) thus admits: "Overall it is concluded that a high risk for aquatic organisms is indicated for the representative uses in orchards and tomatoes requiring substantial risk mitigation measures to reduce spray drift and runoff. "Imidacloprid surface water concentrations (n=21) were reported for six countries across the EU, with concentrations reaching up to >200 μg/L (Mohr et al. 2012; Starner and Goh 2012) and 28.6 % of all MIC_{SW} exceeding the RAC_{SW}; these findings, again, challenge the overall protectiveness of the pre-authorization regulatory risk assessment in the EU.

Conclusion and recommendations for risk assessment amendments

For the first time, we evaluated the protectiveness and field relevance of the regulatory EU pesticide risk assessment on a continental scale. As a result, our meta-analysis shows that MICs frequently exceed the RACs set for the authorization of active substances at the EU level. This finding reveals the critical failures of the EU pesticide regulations and the substantial and widespread ecological risks for the aquatic biodiversity. Moreover, even compliance, especially with higher-tier RACs, may not provide sufficient protection for aquatic ecosystems. The lack of consideration of pesticide mixtures and significantly increasing risks due to the market introduction of newer insecticide compounds poses further challenges to the overall protectiveness of EU pesticide legislation; the latter are also important for the future selection of WFD priority substances. Overall, we conclude that the European pre-authorization regulatory risk assessment for insecticides (and pesticides in general) must be substantially improved in terms of field relevance and environmental protectiveness. We therefore propose the following five risk assessment amendments:

(i) The conservatism of the regulatory exposure assessment must be increased, e.g., by only considering step 1 PECs or by applying safety factors to step 3 and 4 PECs (see also Knäbel et al. (2012) and Knäbel et al. (2014) for further information); in addition, the scope of the exposure assessment must be extended to larger surface waters and estuarine systems. Alternatively, the entire FOCUS exposure assessment approach must be completely revised and the protectiveness of the revised approach must be validated independently using field data.

(ii) The uncertainties of the overall pre-authorization risk assessment must be substantially reduced, and its protectiveness must be increased; in particular, a critical reconsideration of the ecotoxicity endpoints (including magnitude and duration of effects considered acceptable for ERO-RACs) and AF used in higher-tier risk assessment for the RAC derivation and authorization of highly toxic compounds must thoroughly be addressed. In addition, mixture toxicity must be considered in the prospective regulatory risk assessment, and the implementation of additional hazard-based cut-off criteria, e.g., for extremely toxic compounds, should be considered. Incidences of unacceptable adverse effects at concentrations below the RACs in the field should be excluded with high confidence.

(iii) The overall link between the regulatory risk assessment and the actual situation in the field must be considerably strengthened, and findings from field studies on pesticide exposure and effects must be used for a retrospective validation of the current EU regulatory risk assessment, particularly for its future development. The fundamental rationale of the risk assessment, i.e., to protect aquatic biocenoses in the field, not in the computer or any sort of artificial test system, must be the driver for all future risk assessment revisions.

(iv) Effective risk management measures (e.g., large non-cropped buffer zones) should be mandatory for all pesticide approvals.

(v) An obligatory validation of the risk assessment through targeted chemical and biological post-authorization monitoring programs must be implemented for compounds of concern to ensure that their application does not lead to unacceptable effects in the field.

In addition to these risk assessment amendments, farmers' knowledge about appropriate pesticide use and environmental awareness must also substantially be improved through obligatory professional training, and adherence to risk mitigation obligations (i.e., application prescriptions) should be monitored. Above all, the reliance of EU agriculture on pesticides should be critically reconsidered and replaced by more environmental friendly alternatives, such as truly integrated pest management and organic farming, wherever possible.

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Conflict of interest The authors declare that they have no competing interests.

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