Environmental Science & Technology

Article

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# Regulatory FOCUS Surface Water Models Fail to Predict Insecticide Concentrations in the Field

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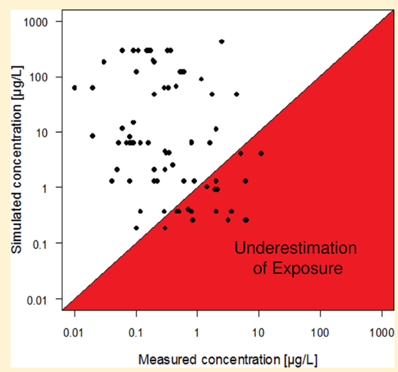
*Supporting Information*

**ABSTRACT:** The FOrum for the Co-ordination of pesticide fate models and their USe (FOCUS) exposure models are used to predict the frequency and magnitude of pesticide surface water concentrations within the European regulatory risk assessment. The predictions are based on realistic worst-case assumptions that result in predicted environmental concentrations (PEC). Here, we compared for the first time a larger data set of 122 measured field concentrations (MFC) of agricultural insecticides extracted from 22 field studies to respective PECs by using FOCUS steps 1–4. While FOCUS step 1 and 2 PECs generally overpredicted the MFCs, 23% of step 3 and 31% of step 4 standard PECs were exceeded by surface water MFCs, which questions the protectiveness of the FOCUS exposure assessment. Using realistic input parameters, step 3 simulations underpredicted MFCs in surface water and sediment by 43% and 78%, respectively, which indicate that a higher degree of realism even reduces the protectiveness of model results. The ratios between PEC and MFC in surface water were significantly lower for pyrethroids than for organophosphorus or organochlorine insecticides, which suggests that the FOCUS predictions are less protective for hydrophobic insecticides. In conclusion, the FOCUS modeling approach is not protective for insecticide concentrations in the field.




## INTRODUCTION

The application of pesticides to agricultural areas can result in transport to adjacent nontarget environments. In particular, surface water systems are likely to receive agricultural pesticide input.[1] When insecticides enter aquatic environments, they may pose a substantial threat to the ecological integrity of surface water systems, as they are highly toxic to a wide range of aquatic organisms such as invertebrates and many fish species.[2,3] In the European Union (E.U.), the registration procedure (i.e., E.U. Directive 1107/2009)[4] for the authorization of new pesticides consists of an effect assessment, which is based on a variety of toxicity tests, and an exposure assessment, which relies on modeling, as usually no field data are available.[5] The FOrum for the Co-ordination of pesticide fate models and their USe (FOCUS) modeling approach[6] is used in the European Union to determine the predicted environmental concentration (PEC) in surface water and sediment and is intended to reflect the exposure levels of specific pesticide compounds under (realistic) worst-case conditions.

FOCUS step 1 is based on very simple assumptions and scenarios and accounts for extreme worst-case pesticide loading[6] without considering specific additional characteristics such as pesticide application time, crop type, or climate. Within step 2, sequential application patterns and pesticide degradation are taken into account in concert with regional or site-specific parameters such as crop interception and runoff. A static ditch with a water depth of 30 cm and a sediment layer of 5 cm is considered to be the model water body for both steps 1 and 2. In step 3, the FOCUS concept uses 10 realistic worst-case scenarios, which are assumed to cover approximately 33% of the total agricultural area in the European Union.[6] In addition, site-specific environmental parameters such as soil type, slope, climatic conditions (e.g., temperature and precipitation), and three different water bodies (i.e., pond, ditch, and stream) that are typical for each of the 10 scenarios are included. The step 3 exposure assessment uses mechanistic models to consider pesticide leaching via drainage,[7] surface runoff,[8] and spray drift as well as fate and transport processes in the respective water bodies.[9] FOCUS step 4 includes mitigation options with different levels of complexity[10] such as no-spray buffer zones or vegetated filter strips. The PECs in FOCUS step 1 and 2 play a minor role in the regulatory risk assessment of insecticides in the European Union. Of the 29 insecticides listed on Annex I of E.U. Directive 1107/2009[4] (for which the European Food Safety Authority risk assessment was publicly available), the risk estimation for 24 insecticide compounds was based on the FOCUS step 3 (four compounds) and step 4 (20 compounds) PEC calculations. The FOCUS surface water working group claims that the highest PEC in surface water ($PEC_{sw}$) estimates from the 10 scenarios would represent at least the 90th percentile (worst-case) for surface water exposures.[6]

Received: November 16, 2011
Revised: July 9, 2012
Accepted: July 17, 2012
Published: July 17, 2012

To the best of our knowledge, no study has compared a large number of pesticide field exposure data across a wide range of situations with the PECs derived from the complete FOCUS modeling approach. Hence, this study aimed to evaluate the PEC predictions by using measured pesticide concentrations from field studies.

In detail, we tested the following four hypotheses using 122 insecticide concentrations extracted from field monitoring studies:

(1) A maximum of 10% of the FOCUS step 3 PECs underestimate the measured field concentrations (MFCs).
(2) The calculated FOCUS PECs exhibit a high correlation with the measured insecticide concentrations in water and sediment. The degree of correlation improves from step 1 to step 4.
(3) The predictive capability of the FOCUS exposure model is similar across all insecticide substance classes.
(4) The PECs that resulted from step 3 simulations with realistic input data are lower than those resulting from FOCUS step 3 standard simulations, and the correlation of PECs with respective MFCs increased relative to FOCUS step 3 standard calculations.

## MATERIALS AND METHODS

**Selection of Insecticide Field Studies.** The field studies ($n$ = 22; Table S1, Supporting Information) that reported insecticide concentrations in lotic surface water resulting from agricultural non-point-source pollution (i.e., spray drift, edge-of-field runoff, drainage) were selected from the studies listed in a review by Schulz[11] and from multiple literature databases (i.e., ISI Web of Science, BIOSIS Previews, CAB Abstracts). Only studies containing detailed information on site-specific parameters regarding climate, landscape characteristics, agricultural pesticide use, crop conditions, and entry routes were selected for the simulation of PECs in the surface water and the sediment. Five organophosphorus insecticides that are important in terms of global application rates,[12] and the organochlorine insecticide endosulfan were included in this study. Furthermore, 10 pyrethroid compounds were considered in this analysis, as pyrethroids are one of the most important types of modern insecticides.[13,14] The selected substances are shown in Table S2 (Supporting Information). If multiple insecticide concentrations were reported in a publication, only the peak concentrations that originated from different entry events were classified as separate events; therefore, multiple concentrations in one publication can be regarded as independent. In these investigations, insecticide concentrations were measured in three European countries (France, Germany, and Italy), the United States, South Africa, and Argentina between 1995 and 2007. The respective water bodies were comparably small; that is, their catchment sizes ranged from 0.1 to 700 $km^2$. The majority of water bodies (approximately 90%) had catchment areas <190 $km^2$.

**Overview of FOCUS Model Calculations.** All of the insecticide concentrations that were extracted from field studies (as described above) were compared to PEC calculations using the tiered FOCUS surface water approach in accordance with E.U. Directive 1107/2009. In detail, the PEC values were derived from FOCUS step 1, 2, 3, and 4 calculations (see Table 1 and FOCUS surface water report[6]), which were also calculated within the exposure assessment in the regulatory pesticide registration. These PEC calculations were designated as FOCUS standard calculations.

**Table 1. Overview of FOCUS Tiers and Their Characteristics Used for Comparison with Actual Field Data**[a]

| FOCUS tier | models used | characteristic | adaptations made |
|---|---|---|---|
| step 1 standard | FOCUS steps 1 and 2 | extreme worst-case | no |
| step 2 standard | FOCUS steps 1 and 2 | worst-case | no |
| step 3 standard | SWASH[b] | realistic worst-case | no |
| step 3 realistic[c] | SWASH[b] | realistic | yes |
| step 4 standard | SWAN | realistic worst-case | no |

[a]See text for details. [b]Includes PRZM, MACRO, and TOXSWA. [c]FOCUS calculations using appropriate step 3 scenarios and information from actual insecticide monitoring field studies.

In addition to the standard FOCUS simulations, step 3 calculations were also performed by adapting the model input data to the actual field conditions; these calculations were designated as FOCUS realistic calculations. FOCUS steps 1–2 model version 1.1[6] was used to calculate the tier 1 and tier 2 PECs in surface water and sediment. FOCUS step 3 standard and realistic simulations were performed by use of MACRO version 4.3b,[7] PRZM version 3.2.1b,[8] and TOXSWA version 2.1.3.[9] These different models are integrated into the SWASH shell version 3.1.2.[6] Step 4 calculations were made with SWAN version 1.1.3[15] by taking the mitigation options into account (see below for details on all calculations).

**Input Parameters for PEC Calculations Using FOCUS.** *FOCUS Standard Scenarios (Steps 1–4).* FOCUS exposure calculations rely on several input parameters related to pesticides, applications, crop type, climate, and landscape.[6] The input parameters for the steps and their sources are provided in Table 2. If no crop type was specified in a field study, the crops commonly grown in the specific study region and for which the use of the particular insecticide was permitted were selected. If several crops were cultivated in an agricultural area where a field study was conducted, then multiple FOCUS PEC calculations were performed with all possible crop and scenario combinations. Therefore, a total of approximately 250 step 1 and 2 FOCUS calculations, as well as approximately 1200 step 3 and 4 calculations, were conducted, and the maximum PECs ($n$ = 122 cases) were subsequently compared to the actual insecticide concentrations that have been measured in the field (see Data Analysis section for details). If the field concentrations were measured at specific time periods after distinct entry events (28 out of 122 cases), then to account for degradation and downstream losses, this aspect were also considered instead of simply using the maximum PECs for comparison with MFCs.

*FOCUS Step 3 Realistic Calculations.* FOCUS step 3 realistic calculations were performed by use of all available realistic field study information regarding insecticide use patterns, climatic conditions, landscape, and water body characteristics (see also Table S4, Supporting Information). If the reported field conditions differed from the FOCUS scenario assumptions, then the standard parameters and scenario conditions of the FOCUS model were adjusted.

Table 2. Description and Source of FOCUS Input Parameters

| category | relevant FOCUS step | parameter | source[a] |
|---|---|---|---|
| physicochemical insecticide properties[b] | steps 1−3 | $K_{OC}$, $DT_{50}$, water solubility, etc. | Footprint Pesticide Property Database[16] according to FOCUS[6] or from E.U. registration documents[17] |
| application data[c] | steps 1−4 | application rate,[d] number and interval of applications, application timing | E.U. registration documents according to GAP;[17] U.S. RED documents[18] |
| scenario | step 2 | northern or southern Europe | selected according to field study information |
| scenario | step 3 | D1−D6, R1−R4[e] | selected according to field study information |
| cultivated crops | steps 1−3 | maize, cereals, fruit crops, etc. | selected according to field study information |
| water body | step 3 | ditch, stream | selected according to field study information |
| mitigation option[c] | step 4 | no-spray buffer zone, vegetated filter strip | GAP from E.U. registration documents;[17] U.S. RED documents[18] |

[a]GAP, good agricultural practice; RED, registration eligibility decision. [b]All insecticide parameters used for FOCUS modeling are given in Table S2 (Supporting Information). [c]For field studies conducted in the European Union, information was taken from E.U. registration documents or producer product labeling. For field studies conducted elsewhere and for European studies where no other source was available, information was taken from U.S. RED documents. [d]Application rates used are given in Table S3 (Supporting Information). [e]D1−D6 are the standard drainage scenarios implemented in FOCUS step 3 for different locations in Europe, and R1−R4 are the standard runoff scenarios.

For the field studies that reported surface runoff as an insecticide exposure pathway, insecticide application was simulated as a granular application to exclude spray drift as an entry route for the PEC calculations. The study information on insecticide application patterns were included in FOCUS calculations via the application definition section from the FOCUS SWASH program, which selects the application dates within a user-defined application window.[6] The exact application dates were included in the MACRO, PRZM, and TOXSWA input files if the time interval between the application date and the precipitation events was clearly identifiable in the field monitoring studies.

The landscape and water body characteristics (e.g., field size, slope, distance between field and water body), which affect the drainage or runoff inputs, were included in the PRZM and MACRO input files after the project definition in SWASH. Furthermore, user-defined water bodies were included in the SWASH database for simulation of the insecticide fate and transport in TOXSWA. To this end, the individual hydraulic characteristics of the respective water bodies were extracted from the publications. The available temperature or precipitation data (as reported in field studies) were included in the PRZM or MACRO climatic input files and were considered in TOXSWA by defining new site-specific scenarios. Details on FOCUS step 3 realistic calculation adaptations are shown in Table S4 (Supporting Information). Changes in the application scenario (e.g., application rate; $n = 5$) were applied to 19 of the 22 field study simulations. More realistic climate data were available for 13 studies. Characteristics from the water body (e.g., water body width) and the surrounding agricultural areas (e.g., field size) were used to realistically simulate the insecticide concentration levels from 11 field studies.

The final PEC comparisons (resulting from FOCUS step 3 realistic calculations with MFCs) were also based on the maximum PEC values. However, in accordance with the PEC comparisons that resulted from the standard calculations, the actual field concentrations (measured at a specific date after a relevant entry event) were also compared to step 3 realistic PECs, which were calculated for these specific data (surface water $n = 15$; sediment $n = 13$) to account for the degradation process, the insecticide fate, and the downstream loss.

**Data Analysis.** Generally, the maximum PECs were compared with the respective MFCs if multiple PECs were available from the FOCUS calculations for one event, that is, when different crops or scenarios were regarded as potentially relevant for the respective field conditions. The effects of different insecticide substance classes (i.e., organophosphate, organochlorine, pyrethroid) and water body size [i.e., water body width <1 m, water body width >1 m (up to ca. 4.5 m), or unknown water body size; Supporting Information] on the ratio of simulated to measured concentration (PEC divided by MFC) were analyzed by two separate single-factor analysis of variance (ANOVA) tests followed by Tukey's honestly significant differences (HSD) post hoc test for pairwise multiple comparisons. The numerical data (PEC/MFC ratio) used in the ANOVA tests were transformed (ln [$x$]) prior to the statistical analysis to satisfy the assumption of normally distributed residuals and homogeneity of variance. All statistical analyses and graphics were made with the open-source software package R (www.r-project.org), version 2.11.1.[19]

## RESULTS

**Comparison between MFC and PEC from FOCUS Standard Scenarios (Steps 1−4).** Figure 1 shows the relationship between PECs and MFCs in surface water ($n = 77$) and sediment ($n = 45$). A comparison of FOCUS step 1 PECs and MFCs showed that the sediment and water MFCs were generally overpredicted up to 32 000 times (median: $10^2$) (Figure 1a, Table 3). Only 4% of the simulated water concentrations underestimated the real concentrations. All of the PEC in sediment ($PEC_{sed}$) estimates from FOCUS step 1 were higher than the measured sediment concentrations. In FOCUS step 2 assessments, most of the $PEC_{sw}$ and $PEC_{sed}$ values were higher than the field concentrations (Figure 1b, Table 3). However, 13% of the sediment and 14% of the water predictions underestimated the respective MFCs up to 15 times, while the median concentrations showed a general overestimation of 13 and 35 times in sediment and water, respectively (Figure 1b, Table 3). Most (77%) of the simulated water concentrations resulting from step 3 were greater than the concentrations detected in the field, with a median PEC to MFC factor greater than 10. However, 23% of the $PEC_{sw}$ values underestimated the insecticide field concentrations (by more than 10 times in 4% of cases) in water. In addition, 42% of all simulated FOCUS step 3 sediment concentrations underestimated the MFC in sediment ($MFC_{sed}$) (Table 3), while the median values coincided comparably well (Figure 1c). For step 4 calculations, approximately a third (i.e., 31%) of the simulated water concentrations underestimated the field concentrations and 6.5% were underestimated by more than 10 times. In

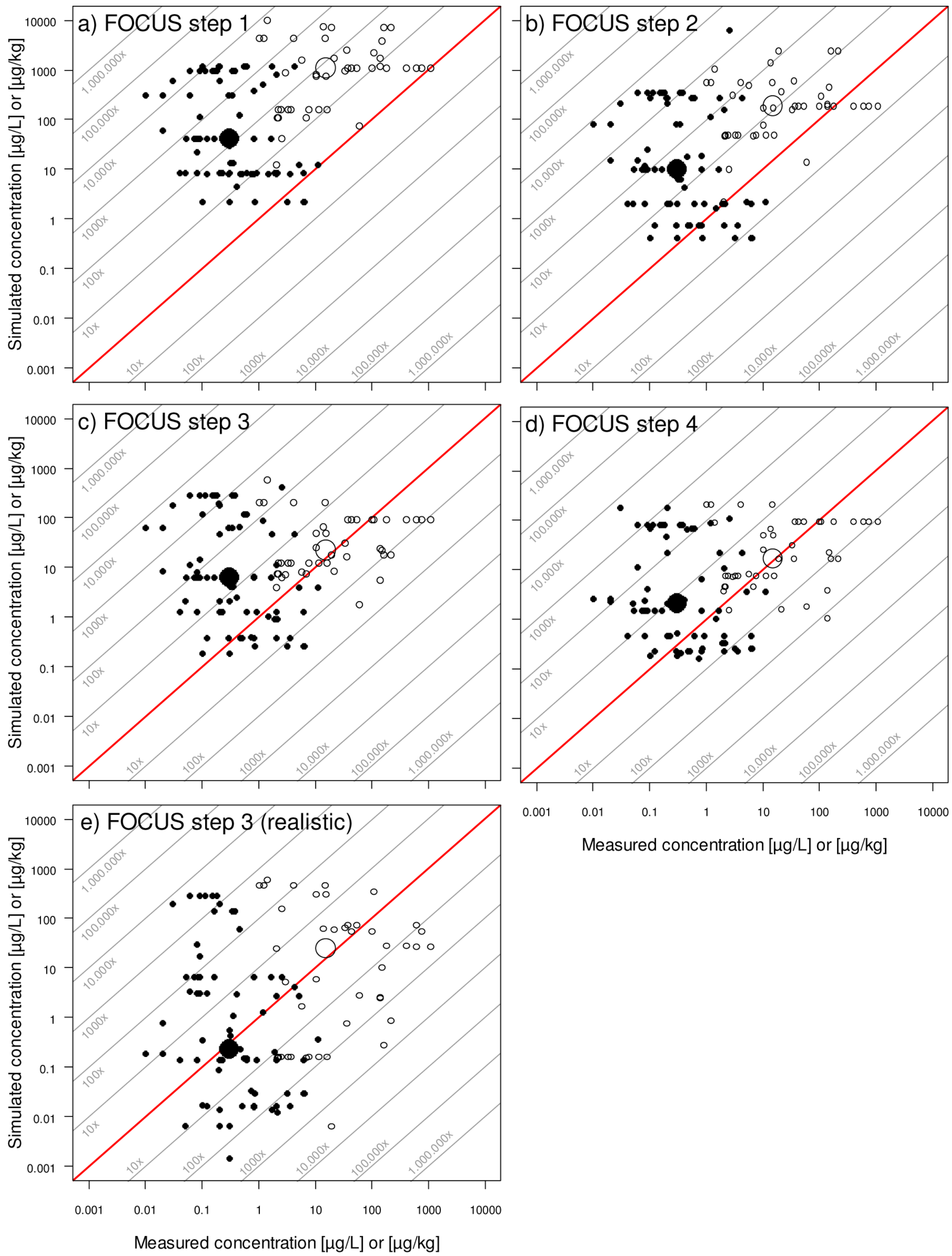



**Figure 1.** Relationship between simulated and measured insecticide concentrations for FOCUS standard scenarios and FOCUS step 3 realistic calculations using information from field studies. (●) Water ($n$ = 77); (○) sediment ($n$ = 45); larger circles, overall medians. The 45° line denotes identity between PEC and MFC. The gray lines indicate over- and underestimation by orders of magnitude. The simulated concentrations are displayed on the $y$-axis so that the MFC overestimations are plotted above the 45° line.

addition, 49% of all $PEC_{sed}$ values calculated by step 4 underestimated the $MFC_{sed}$ values by up to 130 times (Figure 1d, Table 3).

We also found that the underestimation rate for MFC in surface water ($MFC_{sw}$) is even higher (33.3% of FOCUS step 3 $PEC_{sw}$ exceedances instead of 23% for complete data; Table 3) when the evaluated data set is restricted to only the E.U. data. In addition, Figure 1 clearly shows that there is no obvious relationship between the simulated and measured insecticide concentrations for all of the FOCUS steps.

Furthermore, our analysis showed that, in all of the FOCUS standard steps, the pyrethroids ($n$ = 17) had significantly lower

Table 3. Over- and Underprediction and Adequately Matching Data for FOCUS Standard and Realistic Simulations[a]

| FOCUS tiers | surface water | | sediment | |
|---|---|---|---|---|
| | overprediction (PEC > MFC), % | underprediction (PEC < MFC), % | overprediction (PEC > MFC), % | underprediction (PEC < MFC), % |
| step 1 | 96 | 4 | 100 | 0 |
| step 2 | 86 | 14 | 87 | 13 |
| step 3 | 77 | 23 | 58 | 42 |
| step 3 (realistic)[b] | 57 | 43 | 22 | 78 |
| step 4 | 69 | 31 | 51 | 49 |

[a]PEC, predicted environmental concentration; MFC, measured field concentration. [b]FOCUS calculations using appropriate step 3 scenarios and information from insecticide field studies.

ratios of PEC to MFC and thus higher levels of real-world data underestimation than the organophosphates ($n = 55$) and the organochlorines ($n = 5$) (organochlorine−pyrethroid step 4, $p = 0.02$; all other $p < 0.001$, see Supporting Information, Figure S1); however, there was no significant difference between organochlorines and organophosphates.

**Comparison between MFC and PEC from FOCUS Step 3 Realistic.** A considerable proportion of all calculated FOCUS step 3 realistic PECs underestimated MFCs for water (43%) and sediment (78%) (Table 3, Figure 1e). Approximately 26% of $PEC_{sw}$ and 51% of $PEC_{sed}$ values were more than 10 times lower in FOCUS step 3 realistic simulations than the MFCs (Figure 1e). In addition, 12% of $PEC_{sw}$ and 7% of $PEC_{sed}$ cases exceeded the MFCs by more than 100 times (Figure 1e).

## ■ DISCUSSION

**Protectiveness of FOCUS Predictions.** Generally, the degree of conservatism should decrease from FOCUS steps 1 to 4, which is in agreement with our results comparing PEC and MFC values. Consequently, the percentage of insecticide MFCs in surface water that exceed the PECs increased from 4% for step 1 to 31% for step 4 (Figure 1, Table 3).

FOCUS[6] states that uncertainty will always prevail "to some degree in environmental risk assessment"; however, the use of the FOCUS scenarios as part of the E.U. registration process "provides a mechanism for assessing pesticide PECs in surface water and sediment with an acceptable degree of uncertainty". For several reasons, our study results question whether the degree of uncertainty of the regulatory exposure model outcomes generated by the FOCUS is acceptable. First, 23% of $PEC_{sw}$ and 42% of $PEC_{sed}$ values that resulted from step 3 calculations underpredicted the corresponding MFCs. This rejects our first hypothesis, which states that a maximum of 10% of the calculated FOCUS step 3 PECs would underestimate the field data. The ≤10% exceedance value had also been hypothesized as a quality threshold by the FOCUS working group.[6] A similar situation holds true for FOCUS step 4 results, as almost a third of the $PEC_{sw}$ values underpredicted the insecticide MFCs (Table 3). This result is remarkable when it is considered that FOCUS step 4 is the most realistic standard tier available in European regulatory exposure modeling and is almost exclusively used in risk assessment for insecticides currently registered in the European Union. In addition, this is the first study demonstrating that the field concentrations of insecticides can even exceed the FOCUS step 1 (surface water) and 2 (surface water and sediment) PECs (Figure 1, Table 3).

Overall, these results indicate that the FOCUS modeling approach is not reliable in predicting insecticide concentrations when compared to real-world surface water situations. This result also means that unacceptable ecological effects could arise from agricultural insecticide uses, which are not assessed by the regulatory risk assessment.[20] The fact that we are not aware of this situation is somewhat surprising, given Hendley's[21] claim in 2003 that monitoring initiatives should be performed and used explicitly for exposure model validation within a so-called "moditoring" approach.

In addition to model inaccuracies (see below), the underestimation of insecticide field concentrations by the respective PECs might also be attributed to farmers' malpractice during the insecticide application, for example, nonadherence to no-spray buffer zones. However, this malpractice would not explain the 23% underestimation of $MFC_{sw}$ values resulting from FOCUS step 3 calculations, as this step does not include any pesticide application restrictions for farmers (e.g., no-spray buffer zones). Consequently, this suggests that a theoretical maximum of only 8% (i.e., the difference between 23% and 31% underestimations of MFCs in steps 3 and 4, respectively) of cases in which FOCUS step 4 $PEC_{sw}$ underestimates the $MFC_{sw}$ could be attributed to farmers' malpractice. Nevertheless, it is possible that farmers do not adhere to the required application rates.

Generally, it can be argued that the FOCUS modeling approach is valid only for the pesticide registration process in the European Union and respective European agricultural settings, so that an evaluation of the FOCUS PECs should include only the insecticide concentrations measured within the European Union. However, comparison of step 3 $PEC_{sw}$ to $MFC_{sw}$ values (derived only from E.U. studies) showed that eight of 24 (33.3%) field concentrations were underestimated, which is higher than the average of 23% derived from the global data (Figure 1, Table 3). This result clearly indicates that the FOCUS models also failed to predict protective insecticide field concentrations for conditions in Europe.

**Quality of FOCUS Predictions.** Predicted concentrations should be protective,[6] and the exposure model simulations should also provide some degree of realistic estimates of field concentrations, that is, there should ideally be a close relationship between measured and predicted concentrations. However, the results of our study show that the number of matches between predicted and measured concentrations was generally low. If a difference between predicted and measured concentrations of ±10% is regarded as adequate,[6,22] up to 6% of step 2 PECs in surface water and 9% of step 3 PECs in sediment matched the MFCs. In total, 97.7% of the step 3 PECs were not within ±10% of the MFCs. Even when the performance threshold is set at ±30% to account for uncertainties in the field measurements, 92.6% of step 3 PECs did not match the MFCs in surface water and sediment. In contrast to our second hypothesis, Figure 1 shows that there is no positive relationship in terms of a statistically significant slope >0 in a linear regression model between predicted and measured concentrations. For all FOCUS steps, the data points form a rectangular cluster that spans several orders of magnitude on both axes. The complete absence of a relationship between the predicted and measured data highlights the importance of the inherent FOCUS model restrictions. This model weakness appears instrumental in the results of our study and needs to be urgently addressed rather than focusing only on the problems associated with farming practices (discussed above). In addition, the relationship did not improve from FOCUS step 1 to step 4 (Figure 1).

The low quality of FOCUS predictions, as described here, is in contrast to the results derived from a series of test runs using FOCUS step 3 models and scenarios in which generally good (although not quantified) agreement between predicted and measured concentrations in drainflow and runoff had been declared by the FOCUS group.[6] However, only a few field studies have compared predicted environmental concentrations and measured field data. For example, Padovani and Capri[23] showed that the PECs derived from TOXSWA 1.2 (used in Dutch registration process) accurately predicted the measured pesticide concentrations that resulted from spray drift input, as measured and predicted concentrations were below the detection limit in almost all cases. Furthermore, Singh and Jones[24] demonstrated that the U.S. Environmental Protection Agency (EPA) PRZM model provides a "reasonable estimate" of the edge-of-field chemical runoff ($n$ = 17), as the simulated data were within an order of magnitude of the measured data. In addition, Jackson et al.[25] compared predicted pesticide concentrations ($n$ = 40) in drinking water reservoirs (calculated with the U.S. EPA's pesticide exposure models, FIRST and PRZM/EXAMS) with the monitoring data and found a general overestimation of field concentrations by several orders of magnitude. Until now, no studies have compared the actual field concentrations of pesticides in more than 40 cases. Hence, the study presented here with $n$ = 122 from 22 different field studies is the largest study to evaluate the FOCUS approach.

Although FOCUS steps 3 and 4 are regarded as sophisticated, higher-tier pesticide exposure calculations, several inherent restrictions and model weaknesses exist, which may partly explain the PEC calculation inaccuracies. First, the pesticide exposure resulting from the upstream fields is integrated in a too-simplified manner, as a constant additional pesticide loading of 20% irrespective of upstream conditions is assumed. A further important model weakness is the absence of hourly weather data. The standard daily runoff fluxes calculated by PRZM are translated to hourly data by assuming a peak runoff rate of 2 mm/h. For example a 16 mm daily runoff event translates into an eight hour runoff loading of 2 mm/h,[6] although it is possible that during heavy rainfall events, a large proportion of the 16 mm daily runoff occurs in a considerably shorter time period. This pragmatic translation may not realistically reflect the peak pesticide concentrations,[26] particularly during extreme rainfall events.[27] Moreover, the fact that surface water exposure caused by runoff and drainage entries cannot be simulated simultaneously by the FOCUS model is unrealistic and may result in an underestimation of the field concentrations.[28] Even if the $PEC_{sed}$ generally plays a minor role in regulatory risk assessment due to the lack of adequate sediment toxicity data for benthic organisms, note that the simplified assumptions underlying FOCUS $PEC_{sed}$ calculations (i.e., identical sediment layer properties across all step 3 scenarios) could result in high uncertainties.

**Evaluation of Factors Influencing PEC to MFC Ratios.** Our results show that for low (i.e., < 0.5 $\mu$g/L) water-phase insecticide field concentrations, the PECs are higher than the MFCs in all of the FOCUS standard steps (Figure 1a–d). The reason for this dicrepancy is that most of the $MFC_{sw}$ were compared to initial $PEC_{sw}$ values, although the latter most likely did not represent the peak concentrations that are commonly detected when an event-triggered sampling is used.[20,27] This explanation is corroborated by Figure 2, which shows that 50%

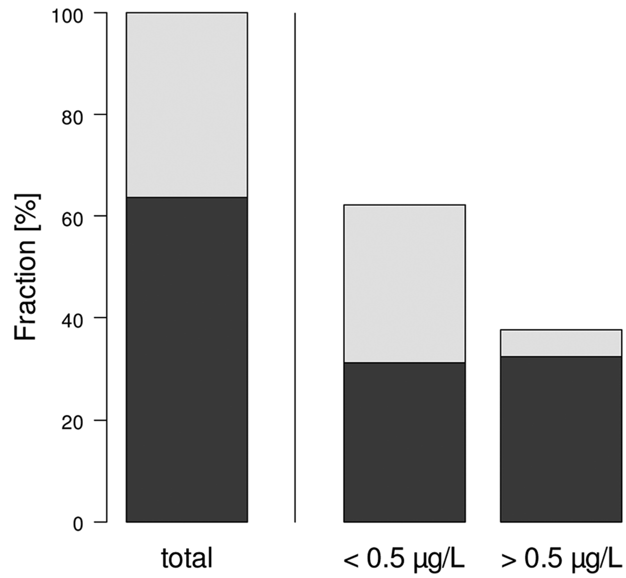



**Figure 2.** Fraction of insecticide surface water concentrations that were detected by event-related sampling (black bars) for all ($n$ = 77) concentrations as well as for concentrations <0.5 $\mu$g/L ($n$ = 48) and >0.5 $\mu$g/L ($n$ = 29). Gray bars denote non-event-related sampling. See also Supporting Information for sampling details of individual field studies.

of the 48 $MFC_{sw}$ values below 0.5 $\mu$g/L were obtained via an event-related sampling approach, which was the case for 86% of the 29 insecticide concentrations >0.5 $\mu$g/L. This result suggests that the field concentrations represented by the $MFC_{sw}$ values used here are still lower than the concentrations present in the field. Consequently, the degree to which the FOCUS PECs underestimate the MFCs could be considerably higher. When only the field concentrations that were measured by event-triggered sampling are analyzed, a much higher number (i.e., 18 of 49 $MFC_{sw}$; 37%) of field concentrations were underestimated by step 3 $PEC_{sw}$.

Our third hypothesis (that the predictive capability of the FOCUS modeling approach is similar for all substance classes) was not confirmed by our analysis. Compared to organochlorines and organophosphates, highly toxic pyrethroids had significantly lower ratios of $PEC_{sw}$ to $MFC_{sw}$ for all FOCUS standard steps. Although the sample size for this analysis is rather low for organochlorines, this suggests that the FOCUS model particularly underpredicted the pyrethroid MFCs in surface water. This result is remarkable, as over the past decades pyrethroids have become increasingly important agricultural insecticides.[29] Generally, synthetic pyrethroids are highly hydrophobic and characterized by low water solubility and have high organic carbon–water partitioning coefficient ($K_{OC}$) values, which lead to a rapid and strong sorption to soil and sediment in the environment.[30,31] Luo and Zhang[32] stated that

PRZM is known to inadequately predict the pesticide transport associated with soil erosion. This assertion may explain the underestimation of insecticide PECs arising from runoff entries for strongly sorbing pyrethroids, as the pesticides associated with eroded soils are removed only from the uppermost soil compartment.[33] In addition, Jones and Mangels[34] list several PRZM deficiencies (e.g., overestimation of downward movement, underestimation of pesticide persistence in soil) that could also lead to the underestimation of field concentrations.

As the $K_{OC}$ value is a key input parameter for exposure modeling, in addition to data used that were published in the Footprint Pesticide Property Database,[16] we recalculated the PECs with the $K_{OC}$ values from EPISUITE 4.1[35] for the substances for which $K_{OC}$ values showed a large variance. However, we detected only small differences in the amount of surface water underestimation in step 1 (from 4% to 5%) and for the sediment in step 3 (from 42% to 44%). In addition, we found no significant differences between the PEC/MFC ratios calculated with $K_{OC}$ values from EPISUITE 4.1 and the Footprint Pesticide Property Database (Supporting Information). Furthermore, we recalculated the PEC values for bifenthrin and fenvalerate using the degradation half times ($DT_{50}$) for soil from the American Crop Protection Association (ARS) Pesticide Properties Database,[36] which differed considerably from the Footprint[16] values; however, we found no change in the amount of overestimation or underestimation for the FOCUS realistic calculations (Supporting Information). Therefore, we concluded that the model outcome for the investigated substances was insensitive to the range of reported $K_{OC}$ values and half-lives. Note that the experimental $K_{OC}$ values for strongly hydrophobic substances might be biased toward having particularly low values due to the lack of complete phase separation during the experimental setup. A detailed sensitivity analysis is needed to clarify the general influence of physicochemical substance properties on model outcomes.

**Evaluation of FOCUS Realistic Simulations.** The FOCUS surface water working group notes that the scenarios used in the E.U. pesticide registration process "do not mimic specific fields, and nor are they necessarily representative of the agriculture at the location or the Member State after which they are named. (...) crops or situations have been adjusted with the intention of making the scenario more appropriate to represent a realistic worst-case for a wider area".[6] To overcome this generalizing nature of the FOCUS standard scenarios, we performed realistic FOCUS step 3 calculations using all site and insecticide use characteristics available from scientific field studies. The results of FOCUS step 3 realistic calculations showed that, with these adaptations, 43% (instead of 23%) of calculated $PEC_{sw}$ and $PEC_{sed}$ values underestimated the MFCs (Figure 1, Table 3). The substitution of worst-case assumptions by real-world data in step 3 realistic calculations explains the generally lower PECs compared to step 3 standard calculations (Figure 1). Realistic step 3 PEC values underestimate the MFCs to a larger extent, despite the use of more realistic (i.e., lower) application rates, which suggests that the emission rates are not a likely cause of our overall study results.

Overall, our fourth hypothesis (stating that the PECs resulting from step 3 realistic calculations are lower than the PECs resulting from step 3 standard calculations) was confirmed by our results. More importantly, the relationship between the PEC and the MFC did not improve by using more realistic entry data for step 3. Again, this result indicates that the FOCUS modeling approach is most likely due to an inappropriate mechanistic representation of the relevant processes not capable of predicting the actual field exposure levels.

In conclusion, our study clearly revealed the need for further targeted modification and calibration of the central processes of the FOCUS exposure models. It appears that further testing is necessary to investigate factors that may potentially influence the model outcomes and to reassess the adequacy of the model input variables (see Blenkinsop et al.[37] for development of alternative FOCUS climate scenarios). Beyond that, our data provide evidence to recommend a further safety or assessment factor for the exposure side of pesticide risk assessment to address the current uncertainties, unless it is clearly demonstrated with sufficient probability that all of the possible field exposures are covered by regulatory models. If we continue to use the current FOCUS scenarios to assess the exposure of insecticides, then FOCUS step 1 data should be used, or FOCUS step 3 or step 4 results must be accompanied by a general safety factor of about 10 to consider the claims originally made when the FOCUS models were implemented.

## ■ ASSOCIATED CONTENT

### Ⓢ Supporting Information

Additional text, four tables, and two figures with information on selected field studies and modeling input parameters. This material is available free of charge via the Internet at http://pubs.acs.org.

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### Notes

The authors declare no competing financial interest.

## ■ ACKNOWLEDGMENTS

This study has been funded by the German Society for the Advancement of Sciences (DFG SCHU 2271/6-1). We thank four anonymous reviewers and the editor for valuable comments on earlier versions of the manuscript.

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