Title: Modeling global distribution of agricultural insecticides in surface waters

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Abstract: Agricultural insecticides constitute a major driver of animal biodiversity loss in freshwater ecosystems. However, the global extent of their effects and the spatial extent of exposure remain largely unknown. We applied a spatially explicit model to estimate the potential for agricultural insecticide runoff into streams. Water bodies within 40% of the global land surface were at risk of insecticide runoff. We separated the influence of natural factors under human control determining insecticide runoff. In the northern hemisphere, insecticide runoff presented a latitudinal gradient mainly driven by insecticide application rate, in the southern hemisphere, a combination of daily rainfall intensity, terrain slope, agricultural intensity and insecticide application rate determined the process. The model predicted the upper limit of observed insecticide exposure measured in water bodies (n=80) in five different countries reasonably well. The study provides a global map of hotspots for insecticide contamination guiding future freshwater management and conservation efforts.

Key words: Insecticide runoff, Exposure models, Risk assessment, Global map

Capsule: We provide the first global map on insecticide runoff to surface water predicting that water bodies in 40% of global land surface may be at risk of adverse effects.

Highlights:

- first global map on insecticide runoff through modelling
- model predicts upper limit of insecticide exposure when compared to field data
- water bodies in 40% of global land surface may be at risk of adverse effects
- insecticide application rate, terrain slope and rainfall main drivers of exposure

Introduction

An estimated 2.3 × 10^6 tons of pesticides are annually applied to agricultural land at the global scale (Grube et al., 2011) to maintain high levels of production. This amount is equivalent to an annual application of 0.15 kg of pesticide per hectare of the total land surface of the earth. The environmental effects caused by these substances are fundamentally different from those of other classes of chemicals, as pesticides are intentionally designed and released into the environment to eradicate pests and weeds. Consequently, pesticides are a major threat to terrestrial (Barmaz et al., 2010; Boatman et al., 2007; Newton, 1995) and aquatic biodiversity (Liess & von der Ohe, 2005; Relyea, 2005; Schäfer et al., 2012; Verro et al., 2009), although the global extent of their effects remains largely unknown. Similarly, the relative importance of main factors driving the exposure at a global scale needs further research. Studies performed at the local and regional scale have demonstrated that agricultural practices (Brown et al., 2008; Tang et al., 2012) and landscape features (Schriever et al., 2007) determine the magnitude of exposure. Large-scale screening approaches, such as spatially explicit models based on Geographic Information Systems (GIS), allow for rapid and cost-effective exposure estimations (Schriever et al., 2007). The identification of areas of concern can trigger regional monitoring programs and, if necessary, exposure mitigation measures.

Given that insecticides are designed to control pest insect populations, they have the potential to impair aquatic invertebrate populations, which play a major role in aquatic ecosystem structure and functioning (Fleeger et al., 2003; Ippolito et al., 2012; Liess & Beketov, 2011; Liess & von der Ohe, 2005; Schäfer et al., 2012). Such estimation of the current state of insecticide contamination in freshwater bodies is pivotal for a targeted conservation and restoration of these ecosystems in order to achieve a satisfactory ecological quality. The need for a global scale perspective is at present particularly essential because farmers in many developing countries are changing from traditional subsistence farming to market-oriented intensive-crop farming

(Satapornvanit et al., 2004). Moreover, global climate change is proposed to result in a significant increase in the global insecticide application on crops, especially in industrialized countries (Boxall et al., 2009; Kattwinkel et al., 2011).

Several field studies documented that most insecticides can enter surface water bodies via surface runoff triggered by heavy rain episodes (Van der Werf, 1996; Wauchope, 1978). In this study we used existing raster maps (FAO & IIASA, 2006; Batjes, 2006) and spatial databases (NOAA; FAO) as input data for a spatially explicit model (Kattwinkel et al., 2011; Schriever & Liess, 2007; Schriever et al., 2007) estimating the surface Runoff Potential (RP) following strong rainfall events. This RP model (Eq. 1) used to estimate insecticide input into streams, was split into two parts to separate environmental factors (e.g. daily rainfall intensity, soil characteristics, slope of the terrain etc.), from human-controlled variables (due to agricultural practices) that determine insecticide runoff. Subsequently, the predictions of the model were compared to monitored peak flow measurements of insecticide contamination in streams of four different biogeographic regions.

Materials and Methods:

Model

We used a generic indicator (RP; runoff potential) that distinguishes stream sites based on key characteristics of the environment around the stream to assess the potential for insecticide runoff inputs (Kattwinkel et al., 2011; Schriever & Liess, 2007; Schriever et al., 2007). The RP is based on a mathematical model (Eq. 1) that estimates the amount (gLOAD) of a generic substance that was applied in the near-stream environment and that may reach the stream in response to a single rainfall event.

$$gLOAD_i = A_i \cdot D \cdot \frac{1}{1 + \frac{K_{OC} \cdot OC_i}{100}} \cdots \quad \text{(Eq. 1)}$$

The model equation is built on nine parameters. $\boldsymbol{i}$: grid cell index; $\boldsymbol{A_i}$: area [ha] of the stream corridor; $\boldsymbol{D}$: country-specific insecticide application rate [$g \cdot ha^{-1}$]; $\boldsymbol{K_{OC}}$: soil organic carbon-water partitioning coefficient [adim]; $\boldsymbol{OC_i}$: organic carbon content [%]; $\boldsymbol{s_i}$: slope [%]; $\boldsymbol{f(s_i)}$: influence of slope on runoff; $\boldsymbol{P_i}$: daily rainfall intensity [mm], $\boldsymbol{T_i}$: texture; $\boldsymbol{f(P_i,T_i)}$: volume of surface runoff due to precipitation [mm]; $\boldsymbol{p_i}$: proportion of croplands in cell i; $\boldsymbol{I}$: average plant interception [%].

All spatial calculations were performed with ArcGis 10 (ESRI, Redlands, CA, USA). The grid cell size used in the study was 5 × 5 arc-minutes. As a result, gLOAD reflected the mean generic exposure of a stream section that would be located in a cell and had the same environmental characteristics as the grid cell. The RP of an individual grid cell was derived as the log10-transformed gLOAD and was subsequently classified into five order-of-magnitude categories (RP values as class boundaries: -3; -2; -1; 0).

In addition, the model was split into two submodels: the first included all environmental variables affecting the RP (daily rainfall intensity, soil characteristics, and slope); the second incorporated those variables that are under human control (insecticide application rate, crop interception, fraction of land used for growing crops). By analogy with the classical risk equation, the first submodel was used to construct a vulnerability map, with vulnerability taken as the natural degree of susceptibility to runoff in each grid cell. The second submodel was used to derive the hazard associated with the human management of insecticides in agriculture. The vulnerability and hazard maps show completely different ranges of variation; hence, the class boundaries for these maps were assigned ex-post according to the distribution function of the values (quintiles).

Area of the stream corridor (A_i) - No real stream courses were considered in the present study. In accordance with previous studies (Schriever & Liess, 2007; Schriever et al., 2007), for each grid cell we considered a generic stream segment with one bifurcation. The near-stream environment was set to an area of 45 ha, which was derived from a two-sided 100-m stream corridor that extended 1500 m upstream from the site (the bifurcation was placed midway in the upstream corridor).

Insecticide application rate (D) - Country-based data on the rate of insecticide application were retrieved from the FAOSTAT database (FAO). All available data for each country referring to the years 2000-2010 were considered. Issues reported in metadata (e.g. data refer to imports only or data expressed in formulated products) were evaluated case by case and unreliable data were discarded. A country was considered only if at least two acceptable data points were present for the abovementioned period. The arithmetic mean of the insecticide application rate was calculated for each country, unless the specific coefficient of variation was > 100%. Then, if a clear trend was observed, only the latest value was considered (most representative of the present situation), else the country data was omitted. For those 84 out of 165 countries without respective data, the insecticide application rate was estimated. The estimation was done using a linear model with the predictors average accumulated temperature (see below), the fraction of insecticide high-consuming crops, and the GDP. The average accumulated temperature was included as predictor since a strong correlation between temperature and the rate of insecticide application has been identified in another study (Kattwinkel et al., 2011) for European countries.

We calculated the average accumulated temperature for each country using a map produced by FAO & IIASA (33) ($T_{mean} > 0$ °C), then weighting the value of each grid cell in the country by the corresponding proportion p_i of croplands (taken from FAO) to account for potential differences in temperature between crop and non-crop areas. The insecticide application rates may vary strongly among crops. Therefore, crops were divided into two classes on the basis of their typical insecticide amount requirements using the U.S. National Pesticide Use Database (Gianessi & Reigner, 2006). Insecticide high-consuming crops included all kinds of fruits, nuts and olives; insecticide low-consuming crops included all remaining crops (mostly cereals, vegetables, herbs and fiber crops). Typical insecticide application rates differed significantly between the two groups ($p < 0.0001$, Mann-Whitney and Kolmogorov-Smirnov tests). The fraction of insecticide high-consuming crops in each country was estimated on the basis of the information retrieved from FAOSTAT database (FAO) for the years 2000-2010. Finally, the insecticide application rate is also determined by the economic situation in a country. Therefore, we also included the country-specific GDP. GDP values (expressed in 2005 US dollars) per capita per country per year were retrieved from the Economic

Research Service's International Macroeconomic Data Set (ERC). Median GDP values were calculated for all countries over the period 2000-2010. The regression model was calculated with the software [R] using all the log-transformed available data at the country level. All country-based rates of insecticide application were normalized by dividing them by the application rate for Germany to enable comparability to those used in previous studies (Kattwinkel et al., 2011; Schriver & Liess, 2007). A detailed description of the procedure to derive country-specific insecticide application rate is reported in SI-I

K_{OC} - The soil organic carbon-water partitioning coefficient is substance specific. We considered all insecticides with respect to the application rate, therefore a substance-specific value could not be assigned. The K_{OC} value was set to 100 in accordance with (Kattwinkel et al., 2011; Schriever & Liess, 2007). This assumption is rather worst case since most insecticides (>75% of used compounds, according to our estimation) have K_{OC} values higher than 100, which would result in lower RP. However, in the absence of detailed per country information that would allow to compute an average K_{OC} per country, the assignment of a different value for the global K_{OC} would not change the relative scale obtained for RP.

Daily Rainfall Intensity (P_i) - RP is determined by single rainfall events. We calculated the median value of the monthly maximum precipitation for each of the 76,687 stations (over the entire available period) collected in the NOAA - GHCN daily database (NOAA). We then assigned to each station the maximum value within the productive season. Productive seasons were assessed using two maps included in the Food Insecurity, Poverty and Environment Global GIS Database (FGGD) (FAO & IIASA, 2006). First, we considered the areas in which a lack of water is the principal constraint on agriculture, according to the "Hierarchical distribution of severe environmental constraints map". In those areas, no time period limitations were established because it is likely that the rainiest month is the period of maximum productivity. In all other areas, the length of the growing period was assigned on the basis of the corresponding FGGD map (FAO &

IIASA, 2006), while the temporal collocation of the growing season was differentiated between the northern and southern hemispheres.

With this methodology, we assigned one value to each of the GHCN stations for which the coordinates were known. Hence, the points were interpolated over the global land surface. Several (ordinary kriging) semivariogram models were tested: the choice of the final interpolation model aimed at minimizing (i) the root-mean-squared prediction error and (ii) the difference between the latter and the average estimated prediction standard error. The best results in terms of prediction (60%, $p<0.0001$, F-test) and uncertainty estimation were obtained with a Stable semivariogram model.

The function *f(T,P)* proposed by the OECD (OECD, 1998) (Eq. 2) is based on empirical values and it is valid in the range of 6-100 mm. Therefore, all values of daily rainfall intensity below 6 mm or above 100 mm were set to 6 and 100 mm, respectively (accounting for 1% of total grid cells).

$$f(T,P)=\begin{cases}-5.86\cdot10^{-6}\cdot P_i^3+2.63\cdot10^{-3}\cdot P_i^2-1.14\cdot10^{-2}\cdot P_i-1.64\cdot10^{-2} & \textit{if } T_i=\textit{sand}\\ -9.04\cdot10^{-6}\cdot P_i^3+4.04\cdot10^{-3}\cdot P_i^2-4.16\cdot10^{-3}\cdot P_i-6.11\cdot10^{-2} & \textit{if } T_i=\textit{loam}\end{cases} \quad \text{(Eq. 2)}$$

A detailed description of the procedure to derive daily rainfall intensity is reported in SI-II

Plant interception (I) - Plant interception is plant- and growth-phase specific. Nevertheless, agricultural patterns (the growth phases during which plants are treated) are too complex and spatially heterogeneous to be represented in a global assessment. Also, a reliable representation of crop distribution patterns at global scale is not available. In the European Generic guidance for FOCUS surface water scenarios (FOCUS, 2012), plant interception values corresponding to four phenological stages are reported for 21 different crops. Insecticide are likely not to be applied during the first stage, when interception is zero for all crops. The mean and the median of the of the remaining values (n=63, 3 stages x 21 crops) is 0.5. Hence, a fixed plant interception value of 50% was applied to each grid cell.

Soil variables (T_i, OC_i) -The values of the soil variables were retrieved from ISRIC-WISE (Batjes, 2006). The organic carbon content of the first soil layer (0–20 cm) was considered. In the

ISRIC-WISE map, organic carbon is expressed in six classes. In our study, each cell was assigned the mean value of its class rounded to the nearest integer. Texture was derived from the database attached to the ISRIC-WISE map. The percentage content of clay, silt, and sand in the first soil layer (0–20 cm) was used to classify the texture as sandy or loamy. Criteria for the classification were retrieved from Finnern et al. (2005).

Proportion of cropland in each grid cell (p_i) - The occurrence of cropland was taken from the FGGD project (FOA & IIASA, 2006).

Slope (s_i) -The average slope was taken from the FGGD project (FOA & IIASA, 2006). In the relevant FGGD map, slope was expressed in terms of seven classes. In our study, each cell was assigned the mean value of its class rounded to the nearest integer. The function determining the influence of slope on runoff according to OECD (1998) is Eq. 3

$$f(s_i) = \begin{cases} 0.001423 \cdot s_i^2 + 0.02153 \cdot s_i, & \text{if } s_i \leq 20\% \\ 1, & \text{if } s_i > 20\% \end{cases} \qquad \text{(Eq. 3)}$$

Sensitivity analysis

A sensitivity analysis was carried out to analyze the influence of changes in each model input variable on RP scores. Model sensitivity was assessed by varying one parameter at a time in the ranges +/-25% and +/- 50%, while all other parameters were kept constant at their mean values (daily rainfall intensity = 28.9 mm; organic carbon = 2.17%; slope = 12.8%; fraction of cropland in each cell = 0.25, calculated disregarding cells for which this fraction was zero; insecticide application rate = 0.34, calculated as median among countries) or at specified values (K_{OC} =100; plant interception = 50%) .

Uncertainty analysis

An uncertainty analysis was performed to take into account, how estimated RP values could change in response to the uncertainty of the input parameters. Uncertainty ranges were estimated for each parameter in each grid cell. Values of one parameter at a time were set to the extremes of the corresponding uncertainty range (lower and upper limit) and RP values were recalculated. We evaluated changes in the RP classification of grid cells with respect to the original estimation.

Due to different data types in the input data, different approaches were used to describe the uncertainty ranges. For the insecticide application rate (*D*), 95% prediction limits of the linear model used for prediction (see above) were used as upper and lower extremes of the uncertainty range for each country. For daily rainfall intensity (P_i), a prediction standard error map associated with the kriging interpolation was calculated. Extremes of the uncertainty range for each grid cell were calculated by adding to (upper limit) and subtracting (lower limit) from the predicted value twice the prediction standard error. Extremes of plant interception (*I*) were set everywhere to 0-90% in accordance with (FOCUS, 2012). No accuracy assessment was available for the map used to estimate the proportion of cropland (p_i) in each grid cell (FAO & IIASA, 2006). However, this map was originally derived from the Global Land Cover 2000 (EC-JRC, 2003), whose accuracy has been assessed (Mayaux et al., 2006). We considered the higher error between the User's accuracy and the Producer's accuracy for the category "Cultivated and managed areas" and we used this value (27%) to estimate the extremes of the uncertainty range for each grid cell. Slope (s_i) and organic carbon (OC_i) were derived from maps expressed in terms of classes (values intervals). The extremes of these classes were used as the limits of the uncertainty ranges. To assess the uncertainty due to soil texture (T_i), the model was applied assuming that the soil was loamy and sandy everywhere. Detailed results of the uncertainty analysis are reported in SI-III

Quantile regression

The predictions of the model were compared to actual measurements of insecticide contamination in water bodies from previous field studies covering Germany (Liess & von der Ohe, 2005), France and Finland (Schäfer et al., 2007), Denmark (Rasmussen et al., 2011) and Australia (Schäfer et al., 2011). The sampling campaigns were designed to catch runoff-induced concentrations of the insecticides most widely used in the respective areas.

Measured insecticide concentrations were expressed as Toxic Units (TU), i.e. benchmarked by a standard test organism (*Daphnia magna* in this case) to allow for comparison of different substances. The maximum TUs per site (Liess & von der Ohe, 2005; Schäfer et al., 2013) were matched with the corresponding RP values estimated for the same sites. Further information on the studies and the raw TU data are provided in (Schäfer et al., 2013).

Quantile regression fits a continuous function through the local (with respect to the independent variable) value of the quantile of a dependent variable to account for variation in the quantile with the independent variable (Cade et al., 1999; Koenker & Bassett, 1978). We fitted linear, exponential and logarithmic models to the data. For each model both (i) a τ-specific version of the corrected Akaike Information Criterion (Koenker, 2013), (AICc(τ)) and (ii) the Akaike weights (w_i) (the relative likelihood of a model, given a data set and a set of models) were calculated.

The difference between the model AICc(τ) and the minimum AICc(τ) was used to choose the best-fitting model (Δi = AICc(τ) - min AICc(τ)), considering that the model with the lowest AICc(τ) generally provides the better description of the data. Values of $\Delta i \geq 2$ are suggested as a threshold to exclude alternative models; values of $\Delta i < 2$ indicate substantial support for the alternative model (Burnham & Anderson, 2002; Johnson & Omland, 2004). Last, we determined the best model form across the 80th, 85th, 90th percentiles by averaging w_i for each model from all three percentiles model selection analyses. To test and compare the goodness of the different models, the selection process was done for a modified dataset with all x values shifted to the positive range. The functional form selected with w_i was recalculated on the original dataset and was tested for statistical significance in terms of the probability that the slope and intercept were zero (Scharf et al., 1998).

Results and Discussion

The predicted magnitude of insecticide runoff driven by environmental variables was visualized in a vulnerability map (Fig. 1). The spatial pattern of vulnerability is determined by geomorphology and climate, particularly the slope of the terrain and the maximum intensity of rainfall during the crop growing season.

FIGURE 1

Subsequently, we produced a hazard map (Fig. 2) based on the anthropogenic factors that influence runoff such as the rate of insecticide application, and the share of land used for agriculture.

FIGURE 2

We combined the vulnerability and hazard maps into a map of predicted insecticide runoff potential (RP) (Fig. 3) and we show that more than 40% of the global land area was at risk of generating insecticide surface runoff to streams. Remarkably, RP values for more than 40% of that fraction (18% of the global land area) were classified as high to very high. This result receives support from a previous model-based study that identified pesticides as important pollutants of streams and rivers globally (Vörösmarty et al., 2010).

FIGURE 3

High hazard was sufficient but not necessary to yield high RP values: more than 90% of the grid cells characterized by a high (4^{th} quintile) or very high (5^{th} quintile) hazard resulted in RP classes high or very high, but half of cells in the highest RP class did not originate from high or very high hazard values. By contrast, high vulnerability was usually a prerequisite for very high RP values (86% of cells in the highest RP class presented high or very high vulnerability), but did not always translate into high RP classes.

The RP map indicated the presence of a north-south gradient in the northern countries, with increasing RP towards the south throughout Eurasia. The same trend was observed from North to Central America, except for the Midwestern United States. This tendency generally reflects the insecticide application rate. The insecticide application rate depends strongly on average temperature due to the dependency of several insect pests on the number of degree days (Herms, 2004). In addition, temperature is a limiting factor for the cultivation of high insecticide-consuming crops (e.g. fruits, nuts, olives and ornamentals). Hence, climate change will likely increase the proportional area with high and very high RP in the future, especially in the northern hemisphere (Kattwinkel et al., 2011). By contrast, the RP values in the southern hemisphere showed no or only slight changes in association with latitude. This probably reflects that the insecticide application rate in these countries is constrained by socio-economic factors. Growing economies in several parts of the southern hemisphere may soon overcome this constraint and attempt increasing their food production e.g. through increasing insecticide application rates, which in turn may increase proportions of land with high or very high RP in the future.

The relationship between measured insecticide concentrations (converted to Toxic Units (TU) to enable comparison of potential ecological effects) in streams of five different countries (France, Germany, Finland, Denmark, and Australia) and RP predictions was positive and linear for all the considered percentiles in the quantile regression (80^{th}, 85^{th}, and 90^{th}) (See SI-IV for details). In ecological studies, 90^{th}-percentile regressions have been used to estimate limiting-factor relationships; i.e. 90% of the observations are below the fitted line (Konrad et al., 2008). Since our sample size was relatively small (n= 80), we modeled 3 extreme percentiles (80^{th}, 85^{th}, 90^{th}) for a more robust analysis. The linear model consistently yielded the best goodness of fit with Δ_i always greater than 10 and an averaged w_i of 0.997 (Table SI-IV 1). The positive slope of the three quantile regression models indicated that the upper quantiles of the maximum logTU increased with RP (See SI-IV for details). Overall, RP represents a potential - not an exact prediction of the pollution level - and should be interpreted as a limiting factor that is valuable to identify potential hotspots of insecticide runoff. Due to the scale used in our study, it was not feasible to consider mitigation measures (vegetated buffer strips, unsprayed areas, artificial wetlands, etc.) implemented at local scale. Therefore, our predictions may overestimate the exposure in regions where such measures are applied. Note that measured insecticide data originated from four regions of Europe and Australia and are not representative for the whole world. Nevertheless, they originated from regions with considerable differences in terms of landscape and agricultural practices.

Several field studies reveal that the abundance of macroinvertebrates sensitive to pesticide pollution decreases with increasing insecticide pollution measured during runoff episodes (Liess & von der Ohe, 2005; Schäfer et al., 2007). Importantly, RP predictions for 10 km^2 grid cells have been shown to provide reasonable predictions of abundances of sensitive macroinvertebrate species from random pulls of streams in Germany, and grid cells with predicted high or very high RP revealed reduced abundances of sensitive macroinvertebrate species in 55% and 90% of the streams, respectively (Schriever & Liess, 2007). In the light of these results, we find it worrying that we predict 18% of the global land surface having high or very high RP. Moreover, several studies confirmed that undisturbed upstream sections, such as forest patches, increase recovery of macroinvertebrate communities from pesticide pollution measured during storm flow (Schäfer et al., 2007 and reference therein). For 60% of the area with high and very high RP values in the present study, forests cover less than 20% of the surface area. Therefore, increased community recovery potential facilitated by upstream forested stream sections is not expected to be high in the grid cells characterized by predicted high to very high RP.

The sharp contrasts in some parts of the hazard map reflect national borders because: (i) insecticide application rate data were averaged on a country basis and (ii) the collection, analysis, and distribution of those data often vary among countries. To our knowledge, only the database maintained by the Food and Agriculture Organization of the United Nations (FAO) holds data on insecticide usage for most countries of the world. The insecticide rate gradient spans more than three orders of magnitude. Thus, the resolution is too low to account for regional differences. In addition, the insecticide use had to be estimated for 84 of the 165 countries included in the analysis (linear regression model, $R^2 = 0.55$, $p < 0.001$ for full model, see methods for details), because data were not available or not reliable. To our knowledge, only one other study has estimated the rate of pesticide application for countries without data (Esty et al., 2005). However, that study did not distinguish between insecticides, herbicides and fungicides, which is essential for the present study.

The sensitivity analysis revealed that the RP model was most sensitive to changes in slope. In addition, the model showed a high degree of sensitivity to changes in daily rainfall intensity values, whereas the organic carbon content exhibited smaller influence. Generally, a variation of +/- 25% in the input parameters caused an average change in RP by |0.08| to |0.18|. A variation of +/- 50% in the input parameters would result in an average change in RP by |0.18| to |0.41|. Note that the interval between the extremes of the same RP class is |1.00| (except for the lowest and highest class, which have no lower and upper limit, respectively). Detailed results of the sensitivity analysis are reported in SI-III

Insecticide application rates represent the greatest source of uncertainty in the final outcome. Oscillation within the estimated prediction interval (see material and methods for details) for this input parameter can cause a RP class shift for 97.6% of grid cells, with 24.4% shifting by +/- 2 classes. When plant interception is set to the most extreme values (0-90%), 91.6% of the grid cells change to an upper or lower neighboring RP class. However, such values are unlikely to be widespread over the entire productive season, therefore this assessment largely overestimates the actual uncertainty due to this parameter. Uncertainty of daily rainfall intensity led to 61% of potentially misclassified grid cells. The risk of misclassification due to uncertainty of the remaining parameters was much lower (See SI-III for details), though for slope, shifting by up to 4 classes was possible in some rare cases.

To conclude, our analysis identifies regions where insecticides may represent a major threat to the aquatic biodiversity (Liess & von der Ohe, 2005; Vörösmarty et al., 2010). These regions should be scrutinized for the actual exposure. To date the majority of studies dealing with insecticide contamination of streams have been conducted on a small scale, mostly involving single catchments (Verro et al., 2002). Only recently have empirical and modeling studies started to extend the focus to larger areas, such as regions (Liess & von der Ohe, 2005), countries (Huber et al., 2000), or continents (Kattwinkel et al., 2011, Schriever & Liess, 2007; Schriever et al., 2007). This is the first attempt to assess insecticide exposure at a global scale. The separation of "vulnerability" (influential environmental factors; Fig. 1) and "hazard" (anthropogenic factors; Fig. 2) can be used to identify appropriate strategies for managing the use of insecticides in agricultural activities. Whether to mitigate hazard by making changes in the cropping system, or vulnerability by introducing modifications in the local environment should be decided on a case by case basis, if an actual risk at the local or regional scale has been identified.

The maps available in this study can increase awareness of citizens and regulators in areas where the ecological effects of insecticides are likely to be significant. In addition, they could prompt national or international authorities to foster targeted local investigations. In fact, environmental management needs to be operatively performed at regional and local scales, but investment policies can be addressed at continental or even global scales by international agencies and authorities (e.g., FAO, UNEP, and OECD).

References

Barmaz S, Potts SG, Vighi M (2010) A novel method for assessing risks to pollinators from plant protection products using honeybees as a model species. Ecotoxicology 19:1347–59.

Batjes NH (2006) ISRIC–WISE derived soil properties on a 5 by 5 arc–minutes global grid (version 1.0), ISRIC Report 2006/2.

Boatman N, Parry H, Bishop J, Cuthbertson A (2007) in Biodiversity Under Threat., eds Hester R, Harrison R (RSC Publishing), pp 1–32.

Boxall ABA et al. (2009) Impacts of Climate Change on Indirect Human Exposure to Pathogens and Chemicals from Agriculture. Environ Health Persp 117:508-514.

Brown DL, Giles DK, Oliver MN, Klassen P (2008) Targeted spray technology to reduce pesticide in runoff from dormant orchards. Crop Prot 27:545–552.

Burnham KP, Anderson D (2002) Model Selection and Multi-Model Inference (Springer-Verlag, New York). 2nd Ed.

Cade BS, Terrell JW, Schroeder RL (1999) Estimating effects of limiting factors with regression quantiles. Ecology 80:311–323.

Esty D et al. (2005) The 2005 Environmental Sustainability Index: Benchmarking National Environmental Stewardship. Yale Center for Environmental Law and Policy.

European Commission, Joint Research Centre (2003) Global Land Cover 2000 database.

Finnern H, Grottenthaler W, Kühn D, Pälchen W. (2005) Bodenkundliche Kartieranleitung. Schweizerbart'scheVerlagsbuchhandlung (Hannover, Germany).

Fleeger JW, Carman KR, Nisbet RM (2003) Indirect effects of contaminants in aquatic ecosystems. Sci Total Environ 317:207–33.

FOCUS (2012) Generic guidance for FOCUS surface water Scenarios (version 1.2)

Food and Agricultural Organisation (FAO), United Nations, FAOSTAT online database, http://faostat.fao.org/ (Accessed 02/2011).

Food and Agricultural Organisation (FAO) & International Institute for Applied Systems Analysis (IIASA) (2000) Map of global accumulated temperatures (T_{mean}> 0 °C).

Food and Agricultural Organisation (FAO) & International Institute for Applied Systems Analysis (IIASA) (2006) Mapping biophysical factors that influence agricultural production and rural vulnerability, by H. van Velthuizen et al., Environment and Natural Resources Series No. 11 (Rome, Italy).

Gianessi L, Reigner N (2006) Pesticide use in U.S. crop production: 2002 Insecticides and other pesticides. (CropLife Foundation, Washington, D.C).

Grube A, Donaldson D, Kiely T, Wu L (2011) Pesticides Industry Sales and Usage - 2006 and 2007 Market Estimates (Washington, DC).

Herms DA (2004) Using degree-days and plant phenology to predict pest activity. In IPM Integrated Pest Management) of Midwest Landscapes, eds Krischik V, Davidson J (Minnesota Agricultural Experiment Station), pp. 49-59.

Huber A, Bach M, Frede HG (2000) Pollution of surface waters with pesticides in Germany: modeling non–point source inputs. Agric Ecosyst Environ 80:191–204.

Ippolito A, Carolli M, Varolo E, Villa S, Vighi M (2012) Evaluating pesticide effects on freshwater invertebrate communities in alpine environment: a model ecosystem experiment. Ecotoxicology 21: 2051-2067.

Johnson JB, Omland KS (2004) Model selection in ecology and evolution. Trends EcolEvol 19:101–108.

Kattwinkel M, Kühne J-V, Foit K, Liess M (2011) Climate change, agricultural insecticide exposure, and risk for freshwater communities. Ecol Appl 21:2068–2081.

Koenker R (2013) quantreg: Quantile Regression. R package version 4.98. http://CRAN.R-project.org/package=quantreg.

Koenker R, Bassett G (1978) Regression Quantiles. Econometrica 46:33–50.

Konrad CP, Brasher AMD, May JT (2008) Assessing stream flow characteristics as limiting factors on benthic invertebrate assemblages in streams across the western United States. Freshw Biol 53: 1983–1998.

Liess M, Beketov MA (2011) Traits and stress: keys to identify community effects of low levels of toxicants in test systems. Ecotoxicology 20:1328–1340.

Liess M, Von Der Ohe PC (2005) Analyzing effects of pesticides on invertebrate communities in streams. Environ Toxicol Chem 24:954–965.

Mayaux P et al. (2006) Validation of the Global Land Cover 2000 Map. IEEE Trans Geosci Remote Sens 44: 1728-1739.

National Oceanic and Atmospheric Administration (NOAA), Daily Global Historical Climatology Network (GHCN Daily), ftp://ftp.ncdc.noaa.gov/pub/data/ghcn/daily/ (Accessed 02/2011).

Newton I (1995) The contribution of some recent research on birds to ecological understanding. J Anim Ecol 64:675–696.

Organisation for Economic Cooperation and Development (OECD) (1998) Report of Phase 1 of the Aquatic Risk Indicators Project. (Paris, France).

Rasmussen JJ, Baattrup-Pedersen A, Wiberg-Larsen P, McKnight US, Kronvang B (2011) Buffer strip width and agricultural pesticide contamination in Danish lowland streams: Implications for stream and riparian management. Ecol Eng 37:1990–1997.

Relyea RA (2005) The impact of insecticides and herbicides on the biodiversity and productivity of aquatic communities. Ecol Appl 15:618–627.

Satapornvanit K, et al. (2004) Risks of pesticide use in aquatic ecosystems adjacent to mixed vegetable and monocrop fruit growing areas in Thailand. Austral J Ecotox 10:85–95.

Scharf F, Juanes F, Sutherland M, Charf FRSS, Uanes FRJ (1998) Inferring ecological relationships from the edges of scatter diagrams: comparison of regression techniques. Ecology 79:448–460.

Schriever CA, Liess M (2007) Mapping ecological risk of agricultural pesticide runoff. Sci Total Environ 384:264–279.

Schriever C, von Der Ohe PC, Liess M (2007) Estimating pesticide runoff in small streams. Chemosphere 68:2161–71.

Schäfer RB et al. (2007) Effects of pesticides on community structure and ecosystem functions in agricultural streams of three biogeographical regions in Europe. Sci Total Environ 382:272–285.

Schäfer RB et al. (2011) Effects of Pesticides Monitored with Three Sampling Methods in 24 Sites on Macroinvertebrates and Microorganisms. Environ Sci Technol 45:1665–1672.

Schäfer RB et al. (2012) Thresholds for the effects of pesticides on invertebrate communities and leaf breakdown in stream ecosystems. Environ Sci Technol 46:5134−5142.

Schäfer RB et al. (2013) How to characterize chemical exposure to predict ecologic effects on aquatic communities? Environ Sci Technol 47:7996-8004.

Tang X, Zhu B, Katou H (2012) A review of rapid transport of pesticides from sloping farmland to surface waters: Processes and mitigation strategies. J Environ Sci 24:351–361.

U.S. Department of Agriculture, Economic Research Centre (ERC). Real Historical Gross Domestic Product (GDP) and Growth Rates of GDP for Baseline Countries/Regions (in billions of 2005 dollars). International Macroeconomic Data Set. Online database http://www.ers.usda.gov/Data/Macroeconomics/ (Accessed 04/2011).

Van der Werf HMG (1996) Assessing the impact of pesticides on the environment. Agric. Ecosyst Environ 60:81–96.

Verro R et al. (2002) GIS-based system for surface water risk assessment of agricultural chemicals. 1. Methodological approach. Environ Sci Technol 36:1532–8.

Verro R, Finizio A, Otto S, Vighi M (2009) Predicting Pesticide Environmental Risk in Intensive Agricultural Areas. I: Screening Level Risk Assessment of Individual Chemicals in Surface Waters. Environ Sci Technol 43:522–529.

Vörösmarty CJ et al. (2010) Global threats to human water security and river biodiversity. Nature 467:555–561.

Wauchope RD (1978) The pesticide content of surface water draining from agricultural fields – a review. J Environ Qua 7:459–72.

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Figure legends

Fig. 1. Global insecticide runoff vulnerability map.

The map shows the potential magnitude of insecticide runoff regardless of the actual agricultural activity and incorporates all natural variables included in the model to calculate RP. Class boundaries have been assigned ex-post according to the distribution of the values (see Materials and Methods).

Fig. 2. Global insecticide runoff hazard map.

The map considers all variables of the RP model that are under human control and are connected to agricultural activities. Class boundaries have been assigned ex-post according to the distribution of the values (see Materials and Methods). Grey areas indicate the absence of any relevant agricultural activity.

Fig. 3. Global insecticide RP map.

The map shows the spatial distribution of potential insecticide runoff to stream ecosystems. According to this estimation, the surface waters in 43% of the total global land area are potentially subject to insecticide load as a consequence of current agricultural practices. The class boundaries (-3;-2;-1;0) are the same as those used in previous studies (Kattwinkel et al., 2011). Grey areas indicate the absence of any relevant agricultural activity.

Figure 1

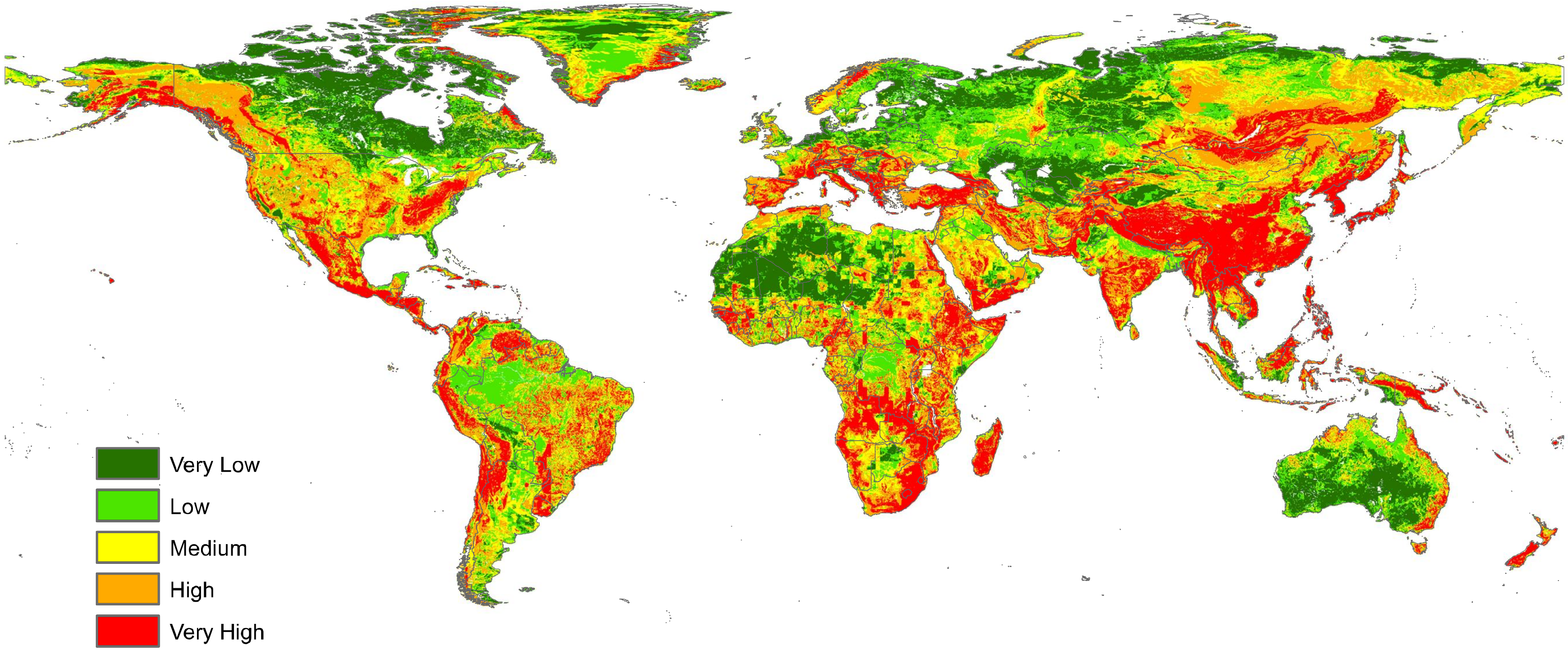

Figure 2
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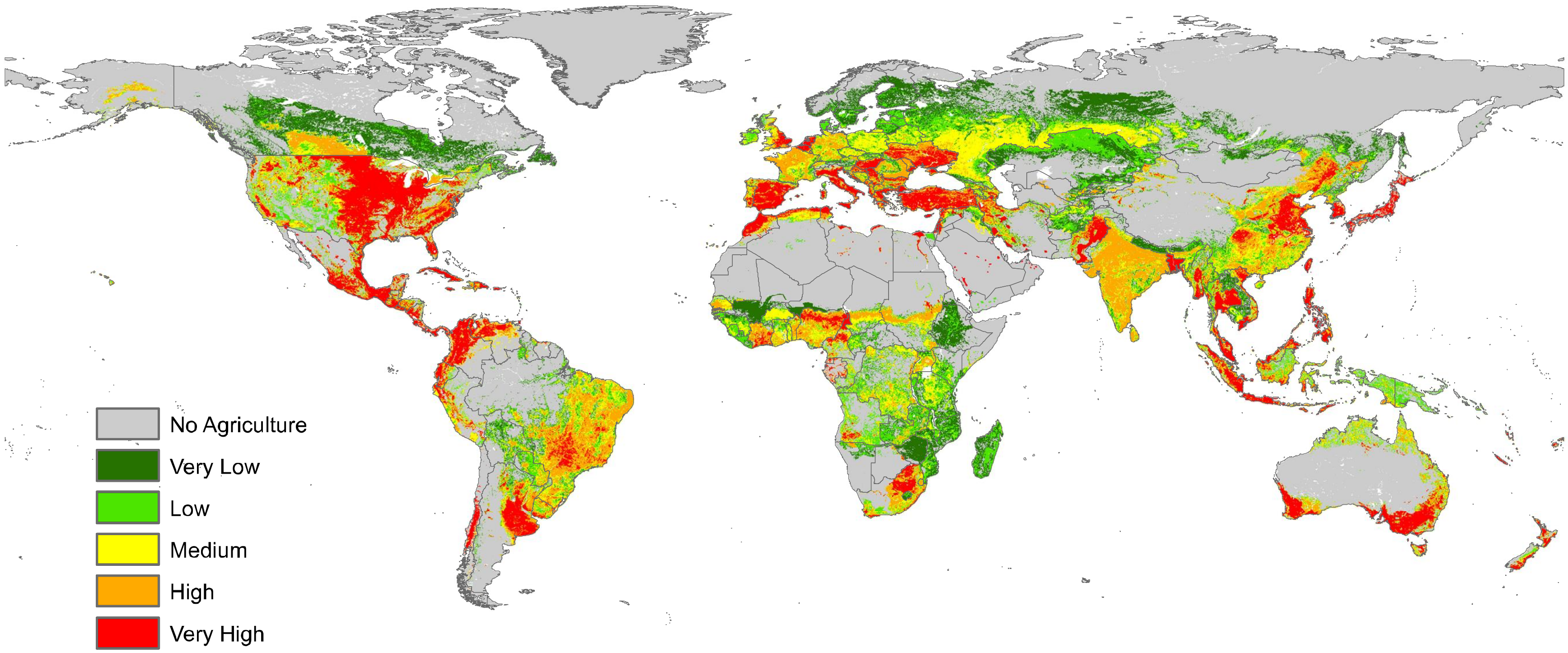
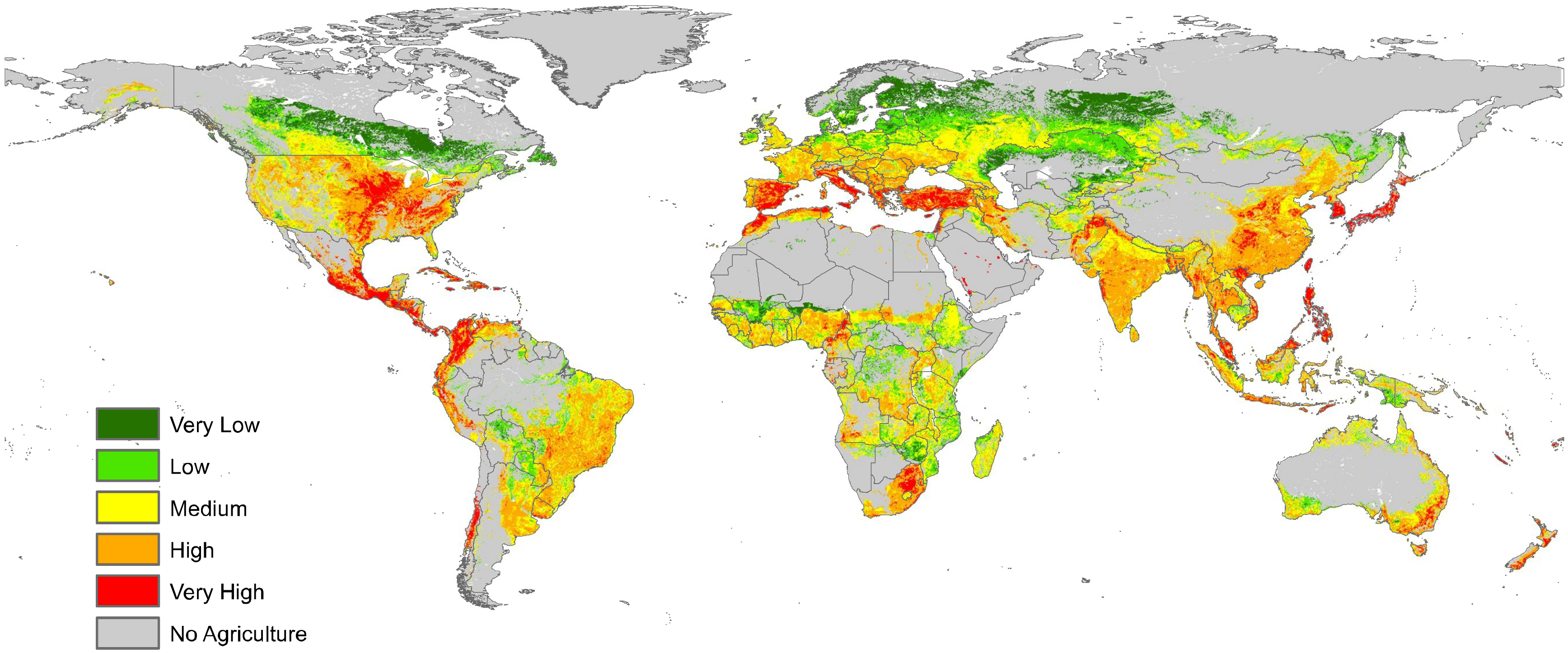


Figure 3



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