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Pesticides in rivers – Assessment of source apportionment in the context of WFD

ABSTRACT

The implementation of the Water Framework Directive (WFD) demands the identification of the significance of point and non-point sources of contaminants such as from pesticides. This paper introduces a first approach exemplarily for the situation in Germany. The model-approach DRIPS estimates spatially differentiated surface water inputs of pesticides via leaching, runoff and spray drift on a monthly basis. Model results, which are currently available for arable land (field crops), can be used directly for WFD requirements, need however actualization and further validating. For many river basins in Germany, a significant fraction of pesticide loads contaminating surface waters results from point source inputs of farmyards. As a first assessment criterion for point sources, the density of boom sprayers for field crops and blast sprayers for orchards, vineyards and hops could be taken into consideration. At present, groundwater inputs of licensed active ingredients are not considered to be significant on a larger scale in Germany.

Keywords: diffuse sources, pesticides, point source, water framework directive, water quality.

1. INTRODUCTION

With the Water Framework Directive (WFD) an approach comprising an inventory of water-courses, recognition of deficits and suggestion of measures aiming to improve water quality has been initiated for the EU. The "Indicative List of the Main Pollutants" (WFD Annex VIII) lists biocides and plant protection products prior to substances, which contribute to eutrophication. In the "List of Priority Substances (WFD Annex X)" the pesticides alachlor, atrazin, chlorpyrifos, diuron, alpha-endosulfan, isoproturon, simazin, and trifluralin are all except for alachlor classified as priority substances. Hence, pesticides as pollutants are treated with high priority in the WFD concept.

Recent concepts to implement the WFD in Germany largely focus on water contamination by nutrients (nitrogen and phosphorus). The water authorities of the federal states of Germany are currently working on harmonizing WFD implementation strategies. Parameters such as share of farmed land and farming intensity (nitrogen surplus in catchment) are in discussion to

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be established as criteria to assess significant nitrogen contamination from agriculture. Although the impact of pesticides on river quality has similar relevance as nutrient immissions, strategies to evaluate the various sources of pesticide inputs (synopsis ref. to Carter, 2000) are currently not yet launched in the WFD implementation.

This paper will give an overview of the current state of knowledge regarding pesticide inputs in surface waters from diffuse and point sources in Germany. Furthermore, several approaches based on experiences from Germany will be introduced here, as conceptual examples for identifying significant impacts on surface waters within the WFD. The authors intend to provide examples of how pesticide risk assessment could be conceptualized in the context of the WFD and hope to speed up the implementation of pesticide related measures into the WFD.

Annotation: The literature does not clearly differentiate between diffuse and point sources. In the WFD's terminology inputs from farmyards are generally treated as point sources whether the effluent enters the surface water directly or via a sewage plant (Carter, 2000).

2. MODELING DIFFUSE PESTICIDE INPUTS FOR GERMANY

The model DRIPS (Drainage, Runoff and spray drift Input of Pesticides in Surface waters) was developed as a first approach to estimate pesticide input into surface waters by diffuse sources on a yearly basis with a spatial resolution of 1 km² for the territory of Germany (Röpke, 2003; Röpke et al., 2004). As input parameters, the GIS-based model requires (i) a set of digital maps such as administrative boundaries, soils, annual precipitation, probability of rainstorm occurrence, CORINE land cover and stream network; (ii) physico-chemical properties of active ingredients (koc, dt50); (iii) the NEPTUN-database containing dosages and dates of pesticide applications on arable land (Roßberg et al., 2002). Within the model, the three routes of diffuse pesticide input (leaching, runoff, and spray drift) are estimated separately. Results presented in this paper are based on calculations for the top 59 active ingredients (a.i.) used in agriculture by volume of sales in 2000. NEPTUN data on orchard and vineyard applications were not yet at hand at the time this modelling exercise was conducted.

2.1 Surface runoff

Pesticide loss with surface water occurs event-specific. The estimate of the pesticide input from surface runoff consists of four components: First, the mean probability of the occurrence of a runoff-causing rainstorm is determined. The German Meteorological Service provides nationwide rainstorm-probability datasets for various rainstorm durations in a 7.5 x 7.5 km² grid. The mean time interval between the application and the intense precipitation results from the occurrence probability of the precipitation event and the dates of application. The runoff volume is determined using the method for calculating high-water flow developed by Lutz (1984). The mean concentration of active ingredients (dissolved phase only) in the runoff is



calculated according to the GLEAMS model (Mills and Leonard, 1984). According to this model approach, 15.0 t active ingredients per annum are estimated to enter surface waters via runoff for the territory of Germany from pesticide application on arable land in 2000. Fairly large inputs are computed for row crops, especially sugar beet, as the spatial distribution of 'hot spot'-regions in Germany indicates (Fig. 1a).

2.2 Drainage

The estimation of tile drainage input into surface waters is carried out in two steps (Huber et al., 2000). First, based on the leaching model PELMO (Version 2.01), for all combinations of site conditions and pesticide applications in Germany, the portion of an active ingredient leaching to a soil depth of 0.8 m is simulated. Second, the share of drained farmland is estimated for the respective area. Therefore, the input of an active ingredient via drainage as shown in Figure 1b in its spatial distribution can be calculated by multiplying the two values. Out of the 59 active ingredients considered, only six showed a noteworthy level of expected surface water input via drainage with an overall yearly sum of 185 kg a.i. The model does not consider the risk of pesticide input into groundwater.

2.3 Spray drift

The table values given by the German pesticide registration authority (Ganzelmeier et al., 1995; Rautmann et al., 2001) are used to calculate spray drift losses. For field crops a mean distance to the adjacent water body of 5 m during the application was assumed which corresponds to a spray drift loss rate of 0.57 % (90th-percentile) of the active ingredients applied. The drainage density, i.e. the frequency of surface waters per unit area, was computed on the basis of the digital surface water network (HAD, 2000). In total the model estimates for spray drift entries only amount to approx. 38 kg a.i. for field crops in 2000 (Fig. 1c). In contrast an earlier modeling exercise for the application year 1993 (Huber et al., 2000; Bach et al., 2001; BMU, 2001) showed, that spray drift inputs from vineyards (120 kg a.i.) and especially orchards (3100 kg a.i.) were by far larger. According to Huber et al. (2000) peaks were estimated for the region "Altes Land" (north of Hamburg) an intensively farmed fruit growing region in Germany. Here, the combination of adverse factors as the presence of a fairly narrow grid of ditches, tree rows very close to the ditches, high treatment frequency as well as spray drift prone application techniques (blast sprayers) lead to extremely high surface water inputs, as Bach et al. (2000) showed. As mentioned earlier, no pesticide application rates were available for orchards and vineyards for modelling inputs in the DRIPS reference year 2000.

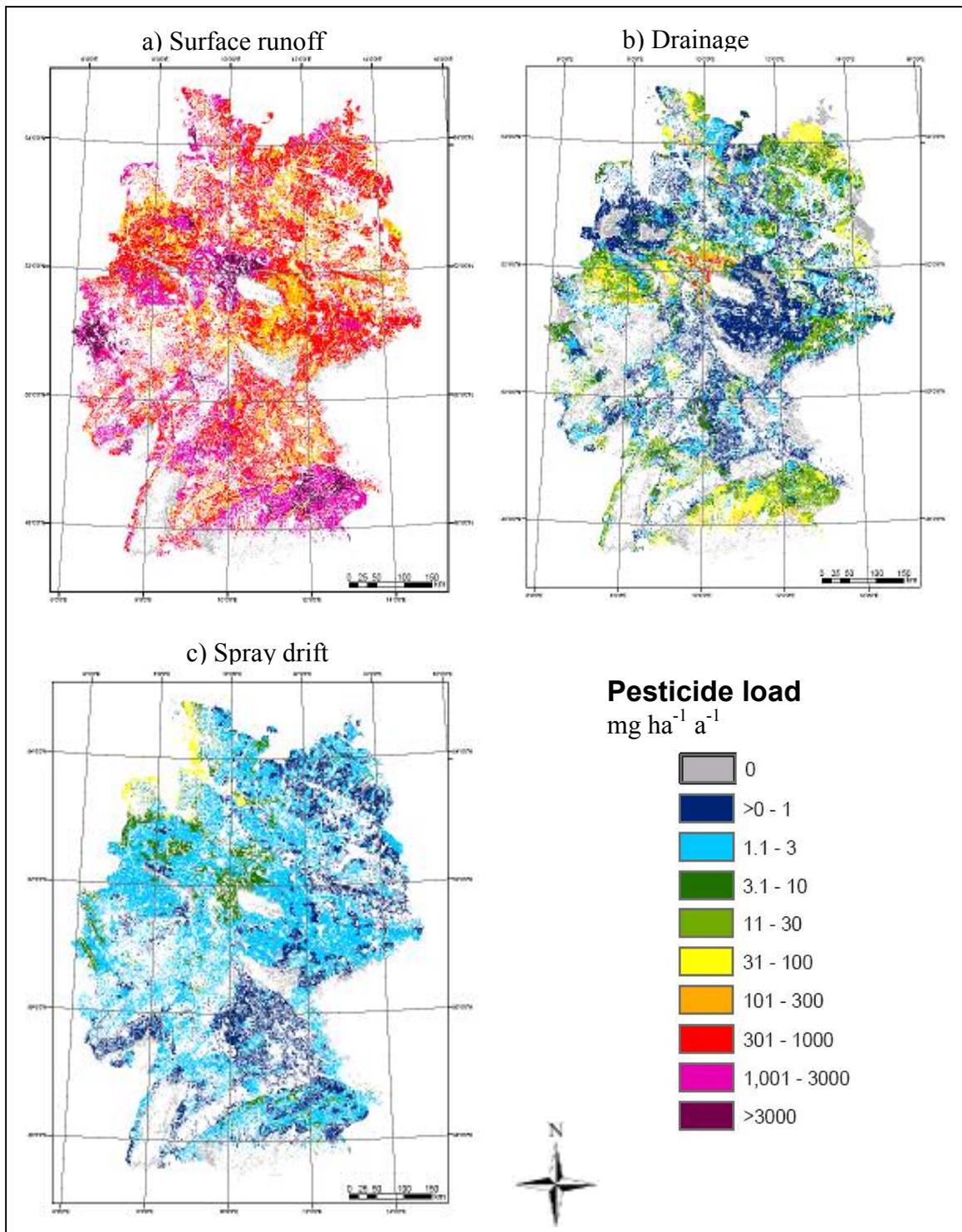


Fig. 1: Spatial distribution of diffuse pesticide losses from field crops (vineyards and orchards excluded) to surface waters in Germany via (a) surface runoff, (b) tile drains, and (c) spray drift according to model DRIPS (reference year 2000; Röpke, 2003; Röpke et al., 2004).



2.4 Magnitude of diffuse inputs from field crops

For the area of Germany a total input of 15.2 t a.i. was estimated with DRIPS as surface waters inputs from arable land as a sum of drainage, runoff, and spray drift for 2000 (Table 1). This value reacts however, very sensitive in relation to the input parameters. For example a variation of a substance's K_{oc} - and dt_{50} -value of $\pm 50\%$ will likewise leads to a variation of the modelled surface water inputs of 25 to 200 % of the initial value. The overall margin of error lies between a minimum of 3.75 t a.i. and a maximum of 30 t a.i., estimated by minimum-maximum spreading of the most sensitive parameters for the runoff input module.

Tab. 1: Estimated diffuse pesticide losses from field crops (in the sum of 59 substances used on arable land) into surface waters in Germany in 2000 according to model DRIPS (Röpke, 2003; Röpke et al., 2004).

Route	Surface water load (model DRIPS estimation) [kg a ⁻¹]	Fraction of total dosage ^a %
Surface runoff	14948	ca. 0.11 %
Drainage	185	ca. 0.0013 %
Spray drift	38	ca. 0.0003 %
Sum (59 a.i.)	15170	ca. 0.11 %

^a) Related to the total amount of 14053 t a.i. in the sum of 59 substances applied in field crops in Germany's agriculture in 2000 (acc. to NEPTUN data base, Roßberg et al., 2002).

Diffuse pesticide input varies to a great extend with different field crops (Table 2). The smallest losses of 0.1 g a.i./ha are calculated for rape seed followed by potatoes with 0.6 g a.i./ha. For grain and maize the modelled annual diffuse input reaches 1.3 and 1.7 g a.i./ha respectively. For sugar beet known to be very susceptible to runoff losses a five times higher input of 7.9 g a.i./ha is estimated. By comparison the figures of Huber et al. (2000) show similarly high specific inputs for orchards and vineyards of 5 and 6 g a.i./ha respectively. With the exception of orchards, runoff is the predominant pathway of entry for all forms of cropping.



Tab. 2: Modelled crop-specific diffuse pesticide losses^a into surface waters in Germany according to model DRIPS (Röpke, 2003; Röpke et al., 2004) and Huber et al. (2000).

Crop	Mean annual diffuse input ^a per hectare of cropping [g a.i. ha ⁻¹ a ⁻¹]	Major pathway of diffuse input
Grain ^b	ca. 1.3	runoff (drainage ^c)
Sugar beet ^b	ca. 7.9	runoff
Maize ^b	ca. 1.7	runoff
Potato ^b	ca. 0.6	runoff
Rape seed ^b	ca. 0.1	runoff
Field crops ^b (average)	ca. 1.5	runoff
Fruit culture ^{d,e}	ca. 5	spray drift
Viniculture ^d	ca. 6	runoff, spray drift

^a) Sum of 59 modelled active ingredients

^b) Calculated for year 2000 (ref. Röpke, 2003, Röpke et al. 2004)

^c) Tile drainage losses are important only in regions with heavy soils

^d) Calculated for year 1993 (ref. Bach et al., 2000, Huber et al., 2000)

^e) Excluding the pesticide losses from the region „Altes Land“ (northwest of Hamburg).

DRIPS results are always estimated individually for every active ingredient with available application NEPTUN data. In this paper results are presented in an aggregated manner since its scope is to present the capabilities of risk assessment modelling for surface waters in the WFD framework rather than to discuss the risk of individual substances. If the model is to be used for water quality assessment purposes, catchment specific estimations for individual substances are readily available.

2.5 Model validation

River loads from diffuse sources estimated by DRIPS cannot be directly compared to measured data for validation purposes. Firstly, this is due to the fact that only four gauging stations in Germany have a sampling frequency high enough to allow statistically valid calculation of annual pesticide loads (see chapter 3). Secondly, in larger river basins point source inputs from farmyards via the sewage system are in many cases a decisive source of pesticide pollution. Since DRIPS is designed to estimate diffuse source only, the model results tend to be lower than the measurements in the respective catchments.



The calculated diffuse inputs of active ingredients in surface waters were tested for soundness in an earlier study by comparing the model results with measured loads from 13 small to medium sized catchments (Fig. 2; see Huber, 2000, for details). Roughly half of the reference values were within the margins of 20 % to 500 % of the measured value. Considering the various restrictions, which impair the comparison such as the lack of model and/or parameter fitting, the authors find the agreement to be acceptable. River loads seem to be systematically underestimated for larger catchments due to the fact that the total load in these catchments is a composite of non-point and point source inputs with the latter one not being considered in the model.

3. POINT SOURCES

In recent years sewage plants were regarded as a relevant source of pesticide entry into surface waters. A number of research projects (overview: Bach et al., 2000) on pesticide loads from farmyards showed a great variance of the mean amount of pesticide effluent according to the number of farms connected to the observed municipal sewage system. Approximately 5 g a.i. was considered as the lower margin and 80 g a.i. as the upper margin per farm and measurement period. For a more precise survey of the specific pesticide input from farmyards, the sewage plant pesticide load needs to be related to the number of sprayers rather than to the number of farms connected to the sewage system. Unfortunately these accurate figures could not be acquired yet for these studies. Regarding the large margin of values, it does not seem very promising to calculate an average value such as load per farm or per sprayer respectively.

Furthermore, the relevance of farmyard inputs as a source of surface water contamination is verified by a number of measuring campaigns in smaller rivers, in which both, the diffuse and point source fraction of the total pesticide load, was estimated parallelly. Measurements of such kinds were taken during the mid nineties in five catchments (7 to 1900 km²) in Hessen (Germany) showed that wastewater treatment plant effluents contributed 65 % to 95 % to the total pesticide river load (Seel et al., 1996; Fischer et al., 1998; Frede et al., 1998; Müller et al., 2002).

To estimate point source inputs in larger catchments, all gauging stations in Germany were selected that measured pesticides loads at least 40 times per year either as grab or composite samples. Only four gauging stations met these criteria in Germany (Table 3). Diffuse pesticide inputs from arable land, which were estimated according to the model approach DRIPS, were deducted from the measured loads to calculate the fraction attributed to point sources (PS Perc. in Table 3). This fraction is not only attributed to farm yard effluents reaching the surface waters via sewage plants but also results from inputs of combine sewer overflows and pesticide production or formulation plants.



Tab. 3: River loads of active ingredients^a in four river basins in Germany, DRIPS modelled diffuse input^b and percentage of estimated contribution from point sources (reference year 2000).

Substance	River Rhein (Cologne) 141,000 km ²			River Main (Bischofsheim) 27,000 km ²			River Nidda (Praunheim) 1,900 km ²			River Ruhr (Westhofen) 1,900 km ²		
	Meas. ^c	Mod.	PS Perc.	Meas.	Mod.	PS Perc.	Meas.	Mod.	PS Perc.	Meas.	Mod.	PS Perc.
	[kg a ⁻¹]	[kg a ⁻¹]	%	[kg a ⁻¹]	[kg a ⁻¹]	%	[kg a ⁻¹]	[kg a ⁻¹]	%	[kg a ⁻¹]	[kg a ⁻¹]	%
Bentazon	106	151	#	70	54	23%	7	10	#	<DL	0.4	-
Chlortoluron	<DL	21	-	19	11	42%	4	0.3	92%	<DL	0.9	-
Isoproturon	2615	1163	56%	515	411	20%	35	72	#	7	27	#
MCPA	<DL	16	-	68	1	99%	4	0.2	95%	1	0.1	90%
Mecoprop-P	137	309	#	151	90	40%	13	0.6	95%	4	4	0%
Terbutylazin	<DL	84	-	53	42	21%	1	1.7	#	3	3	0%
2,4-D	497	2	100%	10	1	90%	<DL	0.1	-	1	0	100%

Meas.: measured river load (in 2000), Mod.: DRIPS modelled diffuse inputs.

PS Perc.: Estimated percentage from point sources = (Meas. – Mod.)/Meas. *100%.

: Modelled diffuse input exceeds measured river load.

<DL: River load below limit of detection.

- ^{a)} Selection of the substances which had been measured in all four river basin. Only active ingredients are accounted, which are registered in 2000 in Germany, used mainly in agriculture, and NEPTUN application data available (Roßberg et al., 2002).
- ^{b)} DRIPS calculated annual diffuse input of the respective a.i. into the surface waters of the river basin up to sampling station ignoring in-stream removal and degradation.
- ^{c)} Without diffuse source inputs into the river Rhine outside of Germany (ca. 28,000 km² catchment area in France and Switzerland).

In the investigated four catchments diffuse inputs account for up to 100 % of the total measured river load for some of the analysed substances. The results underline the relevance of point source inputs of pesticides also for larger river catchments. Nevertheless the modelled diffuse input exceeds the measured river load for a notable number of cases due to the fact that the DRIPS results as well as river pesticide load measurements are subject to large uncertainty.



4. IMPLEMENTATION INTO WFD

An inventory according to § 5 of the WFD comprises the identification of significant sources of surface water contamination of anthropogenic origin. The state of knowledge for pesticide contamination is by far patchier than for nutrient inputs. Approximately 60 gauging stations spread along the main rivers of Germany measure pesticide contamination. Since the measurements reflect inputs integrated over larger catchment areas no direct assumptions on specific sources (pathways) of contamination nor on ungauged sub-catchments can be made. In the case of diffuse nutrient contamination the evaluation will be based on simple criteria such as the fraction of arable land or livestock intensity which are demanded by the first step of the WFD. A similar approach can be outlined for the issue of the pesticide contamination of surface waters.

Diffuse Sources: One criterion might be the ‘estimated pesticide input according to the DRIPS model approach’, calculated with the procedures described above producing specific yearly inputs of active ingredients per hectare for field crops ($\text{g a.i. ha}^{-1}\text{a}^{-1}$) as results (figures for inputs from orchards and vineyards will be available for Germany in near future when NEPTUN data of pesticide dosages in viticulture and fruit growing are at hand). An alternative method is the use of the NEPTUN collection of regional pesticide application data for the respective crops (Roßberg et al., 2002). If coupled with a ranking system for assessing a substance’s specific risk of surface water input, spatially distributed risk indices could be derived indicating the river contamination in catchment areas. Ranking systems of such kind are listed in Reus et al. (1999).

Point sources: The few surveys conducted on this issue for Germany came up with relatively high entries from point sources for small-scale agricultural regions. Relating these quantitative values to other uninvestigated regions does not seem to be justified according to the present state of knowledge. The assumption that point source inputs are very closely related to the number of sprayers in a region seems to be reasonable. Pesticide inputs from point sources, such as from farmyards, occur due to spillages while filling or cleaning the sprayer and the disposal of excess tank-mixture on the yard. Contaminated water is washed off from the paved farmyard by rain or wash water entering either the sewage system or a river directly. The probability of input-scenarios of such kind is presumably directly proportional to the total number of sprayers within a river basin. Possible criteria are the density of sprayers in cropped areas and orchards per river basin. In Germany figures for these criteria are available on a state-level (Rautmann, 2002). According to the regional distribution of the sprayer density in Germany, it can be assumed that point source inputs are particularly a problem in the southern and southwestern states of Germany. The sprayer density is significantly lower in the northern and particularly the eastern states.

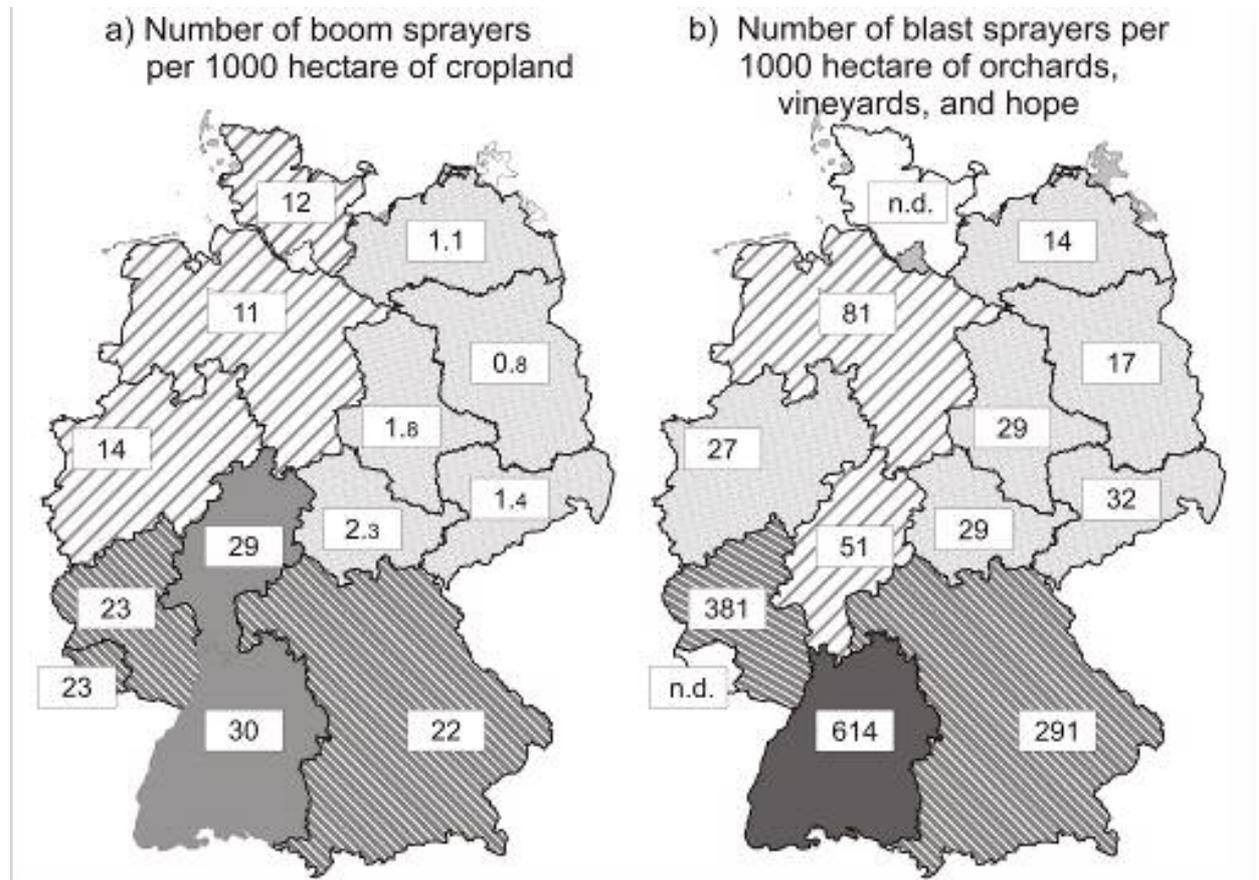


Fig. 2: Number of (a) field sprayers per hectare of arable land, (b) air blast sprayers per hectare vineyards, orchards and hops in Germany, year 2001 (Rautmann, 2002), n.d.: no data.

Ground water: Fairly large analytical datasets documenting groundwater contamination from pesticides are available for Germany. Substances were detected in roughly 231000 cases of sampled groundwater wells in between 1996 and 1998 (Domroese et al., 2001). Out of the 24 most commonly detected substances and their metabolites, 1.21 % of the samples exceeded the general threshold of $0.1 \mu\text{g L}^{-1}$ for drinking water. Only nine of these 24 active ingredients though were registered with the respective authorities for application at the time of sampling. Hence, for the currently registered substances only 0.25 % of the samples exceed the $0.1 \mu\text{g L}^{-1}$ threshold. In other words, only 217 cases were detected in a time span of three years. In regard to assessing the level of contamination in respect of the WFD, it is to conclude that no significant risk of groundwater contamination is to be expected if the currently registered substances are applied with Best Management Practice.



5. DISCUSSION

In the context of the WFD implementation the results presented in this paper are meant to provide a basis for discussion about the relevance of different sources of pesticide input into surface waters. Some restrictions have to be mentioned for evaluating the model results: Pesticide inputs with eroded soil particles were not considered in the model since no adequate soil erosion model of appropriate resolution is currently available for Germany. DRIPS model results are only to be treated as semi-quantitative indicators due to the fairly large margin of error. Results are mean estimates of the expected diffuse inputs of active ingredients – currently only from arable land - on a catchment level to be used for comparative statements such as: judging the hazard potential of different substances, pathways of entry, crops, agricultural areas, catchments etc.

The data of area- and crop-specific loads listed in Table 2 can only be compared with strong limitations due to the different reference years. Expected loads from field crop applications were modelled on the basis of the NEPTUN-survey for the year 2000, while loads from orchards and vineyards were assessed in an earlier study by Huber (2000) based on 1993 application data. Once again, DRIPS estimates for surface runoff, drainage and spray drift loads presented in this paper (Figure 1, Table 1, Table 2) stem from arable land applications only. With the NEPTUN data available for orchards and vineyards in the near future, model calculations will be conducted for the respective cultures. It is expected that the ongoing spread of modern drift reducing sprayer equipment in all agricultural branches (field crop farming, viticulture and fruit growing) will lead to a significant reduction of spray drift inputs into surface waters.

It is in the scope of the model DRIPS to calculate predicted environmental concentrations of pesticides in surface waters (PEC_{sw}) for a number of German river basins and sub-catchments from the load estimates (Röpke et al., 2004). The model DRIPS can assist the development of river management plans by estimating the possible exceeding of regulatory threshold concentration levels in river basins.

One of the ‘take home messages’ of this paper is, that according to our findings point sources (sewage of farmyards) have to be taken into account as a serious pathway of pesticide input into waters. For a certain number of substances and/or river basins in Germany point source inputs clearly dominate over non-point source input. It is astonishing, that only very few studies seem to have been published in any of the European states besides in Germany which clearly stresses the importance of point source inputs into water courses. Exceptions might be catchment monitoring studies conducted in the U.K., Sweden, and Switzerland suggesting that the inputs from farmyards might be more significant than previously recognized (Gerecke et al., 2002; Harris et al., 1991; Kreuger and Nilsson, 2001; Mason et al., 1999). Oakes et al. (1998) has been developed a decision tree procedure to identify short-term pollution events from point sources for river catchments.

In regard to the implementation of the WFD, the authors suggest to encourage the collection of empirical data on the relevance of the different surface water routes of input in a number of EU-member states. Up to day most water- and registration authorities are rather of the opinion, that the major sources of pesticide pollution stem from spray drift and probably to a



much lesser extend from surface runoff (inclusive soil erosion) and drainage inputs. However, in most of the cases there no scientific evidence is provided undermining this assumption. Currently, there is an evident lack of systematic field studies clearly identifying the pathways of entry responsible for measured concentrations. A separation of point and non-point inputs can only be achieved experimentally on the basis of a mass balance, if at least two of the three components (total river load, point source and non-point source immissions) can be sampled simultaneously (e.g. Müller et al., 2002).

The studies conducted in Germany indicated, that pesticide inputs from farmyard wastewater are of higher relevance if the following agricultural and sewer system traits are present in the respective region:

- high density of conventional field sprayers and blast sprayers (orchards, vineyards) within a river basin (e.g. more than ten sprayers per km² catchment area),
- filling and rinsing the sprayer equipment commonly occurs on paved surfaces, and
- these paved yards are connected to a stream directly or via the municipal sewage system (in Hessen as well as in other regions of Germany about 90 % of the farms are connected to a municipal sewage system).

It is expected, that point source inputs from farmyard wastewaters are also important in any country's river basins meeting the above conditions. Fortunately, point sources are rather easy to handle sources of emission when conceptualizing WFD river basins management plans. Abatement strategies focusing on enhanced advisory campaigns to increase the farmer's awareness are readily available and have already been applied successfully in Germany (Frede et al., 1998), U.K. (Mason et al., 1999), Sweden (Kreuger and Nilsson, 2001), and Switzerland (Gerecke et al., 2002).

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