CHAPTER 2

TEMPORAL VARIATION IN PESTICIDE CONCENTRATIONS AND FRESHWATER MACROFAUNA DIVERSITY IN DITCHES OF THE FLOWER GROWING AREA OF THE NETHERLANDS

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Introduction

The Netherlands is known worldwide for flower production. In 2011, the total export of horticultural products from the Netherlands (including seeds, ornamentals, plants, vegetables, fruits, nuts, spices, vegetables and fruits) reached 20.9 billion Euro (Centraal Bureau voor de Statistiek, 2012). The export of ornamentals (that includes flowers and flower bulbs), and other plants in 2011 accounted for 8.1 billion Euro (Centraal Bureau voor de Statistiek, 2012) and was one of the largest in the world. To ensure good quality of floricultural products, pesticides and fertilizers are applied in flower fields to enrich the soil, and control pests and weeds. The Netherlands is also a water-rich country covered with ditches and canals. These water systems possess a relatively high aquatic biodiversity. Intensive agriculture in close proximity to these open water systems creates difficulties in the control and management of surface water quality.

The national-level monitoring of pesticide levels in surface waters is conducted by water management organizations. The collected data is further evaluated in the so-called Pesticide Atlas (see http://www.bestrijdingsmiddelenatlas.nl/) (De Snoo et al., 2006). This tool visualizes the locations where pesticide concentrations were measured and allows comparison of actual pesticide concentrations with various water quality standards, analysis of time trends, and relating pesticide data to land use types.

The pesticide levels in surface waters often exceed the Maximum Permissible Concentration (MPC) in ditches located in the flower growing area of the Netherlands (province South Holland) (Vijver et al., 2008; De Snoo & Vijver, 2012). However, as an overall trend, the surface water quality in the Netherlands with respect to pesticides tends to increase, even though at many sites high pesticide concentrations are still found (Vijver et al., 2008). Whether this overall improvement of the water quality is reflected in the actual performance of aquatic biota in ditches, has not yet been investigated.

In the current study, we aimed to analyze the long-term trends in pesticide concentrations (over the period 1975 - 2010) and aquatic macrofauna diversity (over the period 1983 - 2010) in ditches of the flower growing area of the Netherlands. The research questions were: 1) did water quality in the flower growing area of the Netherlands improve over the previous decades with respect to pesticide residues in surface water? 2) did aquatic macrofauna diversity in ditches increase over time? To address the research questions, we analyzed a database obtained from the Water Board Rijnland. The database included concentrations of pesticides and macrofauna diversity data collected in the flower growing area. We hypothesized that pesticide levels decreased over the previous decades because chemicals with more specific modes of action have been produced by the chemical industry, so that pesticides can be applied at lower concentrations. In addition, less persistent chemicals have been produced in the last decades. We also hypothesized that macrofauna diversity increased over the previous decades, as a result of the improved water quality (reduced pesticide concentrations in surface waters) and all policy measures undertaken to control and reduce pollution of surface waters.

Materials and methods

Description of database

A database containing pesticide concentrations and macrofauna diversity in ditches of the flower growing area of the Netherlands (province Southern Holland) was obtained from the Water Board "Hoogheemraadschap Rijnland" (Leiden, the Netherlands). The area monitored by the Water Board lies mostly on sandy soil, with a smaller patch of light clay and peat, according to the soil maps presented in "Grondsoortenkaart" 2006 - Simplified Soil Map of the Netherlands (Wageningen UR – Alterra, 2006). The majority of sampling sites where pesticides were measured are located in the flower growing area (characterized by sandy and light clay soil types). In the current study, pesticide data only from the flower growing area was analyzed. Sampling sites where macrofauna data was collected are located in the flower growing area, peat and nature reserve areas (characterized by sandy soil type). In the current study, macrofauna data collected in the flower bulb growing area and the nature reserve was analyzed.

Pesticides data

Pesticide concentrations were measured at 92 locations in the research area over the period 1975 - 2010. The overall number of pesticides (including pesticides and their degradation products) analyzed was 109 and the total number of pesticide measurements done was 33560. Table 1 contains the list of pesticides analyzed, their EC50 values (derived in a 48 hour test with *D. magna*, endpoint immobility), time periods at which pesticides were measured in water samples, and the total number of measurements done for each pesticide.

Table 1. List of pesticide measured in water samples, with corresponding EC50 values (based on immobility test with *D. magna* 48 h) and reference to EC50 values.

Name of the compound	EC50, µg/L	Reference	Measurement years	Number of measurements
1.2 dichloropropane	55900	OECD SIDS (2003)	1987-1992	57
2.4 dichlfeenazijnzuur	*N*[4]	*N*	1988-1995	63
2.meth4chlfenazijnzuur	*N*	*N*	1996-2002	133
2.meth4chlfenboterzuur	*N*	*N*	2003-2006	124
2.meth4chlfenpropionzuur	*N*	*N*	1991-2004	131
2-nitrophenol	210[5]	EPA (1980)	1998	2
3-methyl-4-nitrophenol	12000	OECD SIDS (1994)	1997-1998	10
aldicarb	420	PPDB	1990-2010	264
aldicarb-sulfon	250	Reference 1[7]	1999-2010	261
aldicarb-sulfoxide	800	Reference 1[7]	1999-2002	78
adrin	28	PPDB	1980-2005	417

Table 1. List of pesticide measured in water samples, with corresponding EC50 values (based on immobility test with *D. magna* 48 h) and reference to EC50 values. (*Continued*)

Name of the compound	EC50, µg/L	Reference	Measurement years	Number of measurements
aminomethylphosphonic acid	691000	Traas & Smit (2003)	2001-2005	192
atrazine	85000	PPDB	1990-2010	187
bentazone	64000	PPDB	1994-2004	166
Benzothiazole	19	Reference 2[8]	1997	2
bitertanol	4460	PPDB	2002-2010	79
butocarboxim	3200	PPDB	1999-2002	78
butocarboximsulfoxide	*N*	*N*	1999-2002	78
captafol	3340	PPDB	1999	12
captan	7100	PPDB	1987-1999	105
carbaryl	6.4	PPDB	1999-2002	78
carbendazim	150	PPDB	1987-2010	1990
carbofuran	9,4	PPDB	1997-2010	342
chlorbromuron	5800	PPDB	2003-2010	58
chlorfenvinphos	0.25	PPDB	1995-2010	293
chlorpropham	2600	PPDB	1992-2010	297
chlorothalonil	84	PPDB	1994-2010	93
chloridazon	132000	PPDB	1990-2010	1595
chloroxuron	2950	PPDB	2001-2010	61
cyanazine	49000	PPDB	1998	1
DDD[1]	9	PPDB	1980-2005	383
DDE[2]	1	PPDB	1980-2005	384
DDT[3]	5	PPDB	1980-2005	384
o.p'-DDD	9	PPDB	2001-2005	17
p.p'-DDD	9	PPDB	2001-2005	17
o.p'-DDE	1	PPDB	1983-2005	29
p.p'-DDE	1	PPDB	2001-2005	18
o.p'-DDT	5	PPDB	2001-2005	18
p.p'-DDT	5	PPDB	2001-2005	18
desethylatrazine	5100[6]	Ralston-Hooper et al. (2009)	1995	1
dichlobenil	6200	PPDB	1980-2004	462
1.3-dichloropropene	3600	PPDB	1991-1992	16
dichlorvos	0.19	PPDB	1990-2010	236
dieldrin	250	PPDB	1980-2005	416
diethyltoluamide	75000	PPDB	2001-2010	198
difenoconazole	770	PPDB	2001-2002	2
dimethoate	2000	PPDB	1987-2010	119

Table 1. List of pesticide measured in water samples, with corresponding EC50 values (based on immobility test with *D. magna* 48 h) and reference to EC50 values. (*Continued*)

Name of the compound	EC50, µg/L	Reference	Measurement years	Number of measurements
dinoseb	240	PPDB	1990-1997	5
diuron	5700	PPDB	1999-2010	1370
dinitrosol	1100	PPDB	1990-2010	76
alpha-endosulfan	440	PPDB	1980-2005	340
endrin	4.2	PPDB	1980-2005	417
ethiofencarb	220	PPDB	1999-2010	132
ethofumesate	14000	PPDB	2001-2010	166
ethylenethiourea	21600	PPDB	1992-1993	21
fenamiphos	1.9	PPDB	1999-2010	56
fluazinam	220	PPDB	1998-1999	13
flutolanil	6800	PPDB	1997-2010	1668
folpet	680	PPDB	1999	12
furalaxyl	39000	PPDB	1999-2004	16
glyphosate	40000	PPDB	2001-2005	192
heptachlor	42	PPDB	1980-2005	350
heptachlor epoxide	240	PPDB	1980-2005	350
hexachlorobenzene	500	PPDB	1975-2005	552
alpha-hexachlorocyclohexane	1000	IPCS INCHEM (1991)	1975-2005	491
beta-hexachlorocyclohexane	500	IPCS INCHEM (1991)	1975-2005	491
gamma-hexachlorocyclohexane	1600	PPDB	1975-2005	560
HTI	*N*	*N*	1996-1997	28
imazalil	3500	PPDB	1999-2010	185
imidacloprid	85000	PPDB	1999-2010	1051
iprodione	660	PPDB	1997-2010	183
isoproturon	580	PPDB	2001-2010	335
lenacil	8400	PPDB	1997-2010	55
linuron	310	PPDB	1998-2010	234
metalaxyl	28000	PPDB	1999-2008	52
metamitron	5700	PPDB	2004-2010	1076
methabenzthiazuron	30600	PPDB	2002-2007	3
methiocarb	8	PPDB	1999-2010	261
methiocarb methoxy sulfone	180000	PPDB	1999-2010	261
methomyl	7.6	PPDB	1999-2010	261
methyl isothiocyanate	76	PPDB	1990-1998	73
metolachlor	23500	PPDB	1997-2008	2
metoxuron	215600	PPDB	1997-2010	314

Table 1. List of pesticide measured in water samples, with corresponding EC50 values (based on immobility test with *D. magna* 48 h) and reference to EC50 values. (*Continued*)

Name of the compound	EC50, µg/L	Reference	Measurement years	Number of measurements
oxamyl	319	PPDB	1997-2010	263
2,3',4,4',5-pentachlorobiphenyl	*N*	*N*	1980-1987	45
parathion-ethyl	2,5	PPDB	2001-2008	101
parathion-methyl	7.3	PPDB	1987-2010	230
pencycuron	300	PPDB	1999-2005	103
permethrin	0.6	PPDB	1999	1
pirimicarb	17	PPDB	1995-2010	199
pirimiphos-methyl	0.21	PPDB	1997-2010	292
prochloraz	4300	PPDB	1992-2008	342
procymidone	1800	PPDB	1994-2008	297
propham	23000	PPDB	1997	3
propachlor	7800	PPDB	1997-2002	16
propoxur	150	PPDB	1992-2010	325
propyzamide	5600	PPDB	2001-2010	81
prosulfocarb	510	PPDB	1999-2004	110
pyrimethanil	2900	PPDB	1999-2002	9
simazine	1100	PPDB	1990-2010	359
tebuconazole	2790	PPDB	2004-2008	7
terbuthylazine	21200	PPDB	1997-2010	68
thiram	11	PPDB	1990-1992	16
tolclofos-methyl	48000	PPDB	1987-2010	384
toluenesulfonamide	210000	OECD SIDS (2002)	1997-2004	2
tolylfluanid	190	PPDB	2001-2004	101
tri-allate	91	PPDB	2001	1
trichostatin	*N*	*N*	1998	1
vinclozolin	3650	PPDB	1987-2010	238

[1] DDD - dichlorodiphenyldichloroethane
[2] DDE - dichlorodiphenyldichloroethylene
[3] DDT - dichlorodiphenyltrichloroethane
[4] *N* - EC50 was not found
[5] EC50 value derived in 24 h test with daphnids
[6] EC50 value derived in 96 hour test with *Hyalella Azteca*, age below 7 days, test conditions: freshwater, temperature 25 ºC
[7] Ministerie van Landbouw, Natuurbeheer en Visserij (1998)
[8] Ministry of the Environment (Environmental Health Department, Environmental Risk Assessment Office)

Because different pesticides were measured in water samples during the period 1975 - 2010, the annual variation in concentrations of individual pesticides over this time period could not be analyzed. To analyze the change in pesticide levels in surface waters over the period 1975 - 2010, toxic units (TU) were calculated for each sample. TU were calculated as follows:

$$\sum_{i=1}^{n} TUi = \frac{\mathrm{Ci}}{EC50, D.magna},$$

Where, TU_i is the toxic unit of the pesticide *i, Ci* - is the concentration (µg/L) of the pesticide *i*; and EC50 - the corresponding Effect Concentration (48 hours) of *D. magna* exposed to substance *i* (µg/L). When the concentration of a pesticide was below the limit of detection, half of the detection limit was used in the analysis. We used EC50 values because those values could be made available for 101 out of 109 compounds. We know that NOEC (No Observed Effect Concentration) for *D. magna* derived in a 21 day test is a more sensitive parameter for TU calculation, but only a limited NOEC data was found in literature (59 out of 109 compounds). Logarithms of toxic units (base 10) for each sample were then plotted versus time. The total number of compounds analyzed each year, the number of compounds analyzed per sample, and the total number of samplings per year were also calculated and plotted versus time. The relationship between the toxic units and the number of pesticides measured per sample was analyzed by means of regression analysis. Additionally, to account for the different number of pesticides measured over the years 1975 - 2010, TUs were normalized by the number of pesticides measured per sample.

As a next step, sampling locations at which log TU exceeded zero (meaning that the concentration of at least one pesticide was higher than the EC50 value and therefore provided a potential risk to aquatic biota) were identified and analyzed separately. Pesticides contributing mostly to TU exceedances were identified, and concentrations of these individual pesticides were plotted versus time. In addition, pesticides mostly measured in samples (measured more than 250 times), measured until (and including) the year 2010, with concentrations exceeding detection limits in at least 15% of the measurements, were identified. Concentrations of these pesticides were plotted versus time and analyzed with linear regression. The authorization dates of these pesticides were retrieved and added to the graphs.

Macrofauna data

The macrofauna dataset covered the period 1983 - 2010. The total number of macrofauna samples collected over time was 84: 74 samples were collected in the ditches of the flower bulb area and 10 samples were collected in the nature reserve. We calculated the Shannon diversity (H) index for macrofauna collected in two areas according to the formula:

$$H' = \sum_{i=0}^{n} pi \times \ln(pi),$$

where p*i* is the proportion of species *i* relative to the total number of species (p_i). The relationship between Shannon diversity index and time was analyzed with linear regression, separately for macrofauna collected in flower bulb growing area and nature reserve. The difference in Shannon diversity indices between the flower growing area and the nature reserve was analyzed with the t-test assuming the equal variance.

Most of the macrofauna and pesticide data were collected not consistently over time and sampling site. For this reason, causal relationships between pesticide levels in ditches and macrofauna diversity could not be analyzed.

Non-metric Multi-Dimensional Scaling (MDS) based on Bray-Curtis similarity matrix was applied to macrofauna species data to visualize similarities in macrofauna species composition between the different years. All macrofauna samples were divided in five groups according to the sampling periods: 1975 - 1979, 1980 - 1985, 1986 - 1990, 1991 - 1999, 2000 - 2010 (these time intervals corresponded to the main periods of TU change over time). The difference in macrofauna species composition between the groups of samples was tested by the analysis of similarity (ANOSIM) test. Before the analysis, macrofauna data were log(x+1) transformed. Multivariate analysis was performed in PRIMER Software (Clarke & Gorley, 2006).

Results and discussion

Annual variation in pesticide concentrations

The total number of pesticides analyzed in water samples per year increased over time and reached a maximum of 60 - 80 in 2002 - 2005, compared to 4 compounds measured in 1975 - 1979 (Figure 1A). The number of pesticide samples analyzed per year increased simultaneously with the number of compounds analyzed (Figure 1B): from 5 - 50 compounds analyzed in 1974 - 1998 until 200 - 250 analyzed in 2000 - 2006. This means that in 2000 - 2006 the probability to detect high pesticide concentrations in water was higher than in 1974 – 1998, due to the higher measurement frequency. The number of pesticides measured per sample also tended to increase over time with a maximum observed in 2000 - 2004. However, during the years 2000 - 2010 this number varied between 5 and 50 (Figure 1C).

Figure 1D shows toxic units plotted versus time. The lowest TUs were found in 1975 - 1980. Nevertheless, this statement should be considered with care because in 1975 - 1979 only four pesticides (hexachlorobenzene, alphahexachlorocyclohexane, beta hexachlorocyclohexane and gamma-hexachlorocyclohexane) were analyzed in water samples (Figure 1D). In 2006 – 2010, the TU largely remained below zero. This means that pesticide concentrations did not exceed the EC50 for *D. magna*.

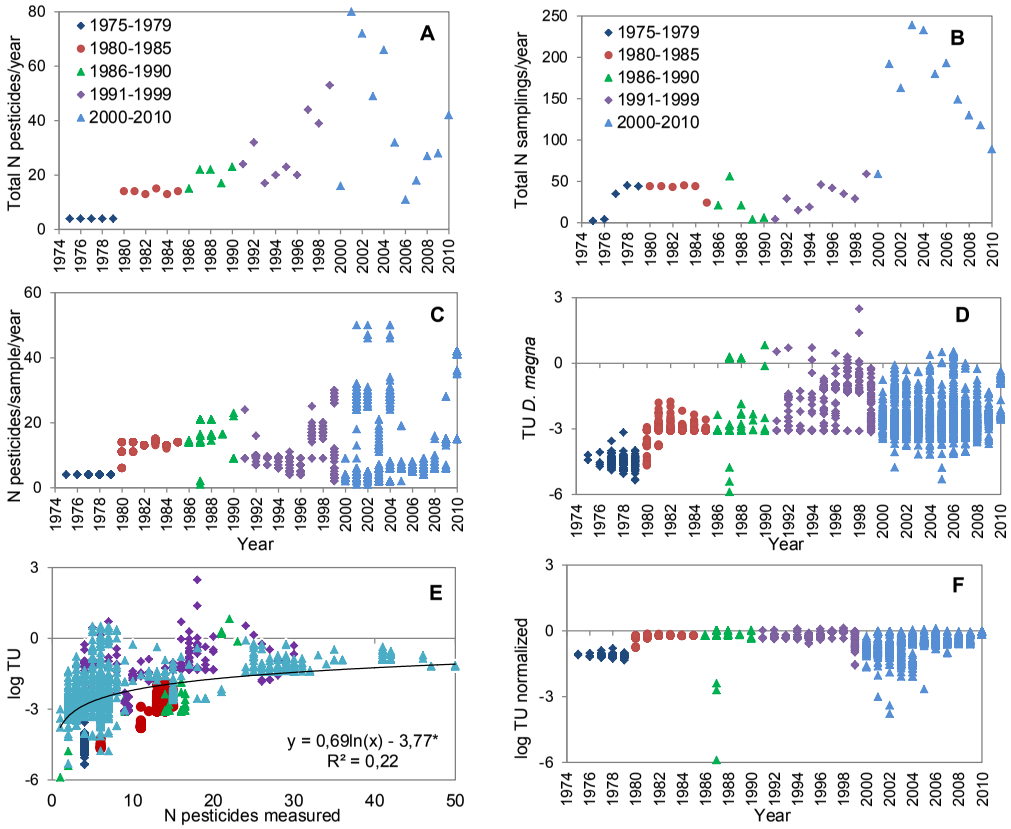

Figure 1. Total number (N) of pesticides measured in water samples per year (A), total number of samplings per year (B), number of pesticides measured in one sample per year (C), Toxic Units (D) plotted versus time, regression analysis between toxic units and the number of pesticides measured per sample (E), Toxic Units normalized by the number of pesticides measured per sample plotted versus time (F) * statistically significant at $p<0.05$

TU values in 1980 - 1987 were two log units higher than in 1975 - 1980. Starting from the year 1980, highly toxic pesticides (to *D. magna*) were measured in water samples, along with other less toxic compounds. Such toxic pesticides included DDT and its degradation products (DDE and DDD), eldrin, pirimicarb, dichlorvos (starting from year 1990), chlorfenvinphos (starting from 1995), pirimiphos-methyl and carbofuran (starting from 1997), permethrin, fenamiphos, carbaryl and methomyl (Table 1). The presence of these compounds in water samples starting from the year 1980 has led to a significant increase in TU values in the years 1980 - 1981 (Figure 1D). However, even though EC50 values of these compounds are very low (vary in the range 0.19 µg/L – 9.4 µg/L), their individual concentrations in surface waters rarely exceeded the EC50 values. The highest values of TU were found at a number of locations in 1987 - 1997 and 2003 - 2007. Pesticides that contributed mostly to the high TU (exceeding 1) were pirimiphos-methyl, carbendazim (in years 1987 - 2007), dichlorvos and chlorfenvinphos (in years 1994 - 1996). Among these pesticides, pirimiphos-methyl was one of the pesticides found at high concentrations in surface waters on the national scale in years 2003 - 2004, and was mainly linked to land use types of floriculture and greenhouse production (Vijver et al, 2008).

Most of the TU exceedings in the current study were observed in the periods 1987 - 1997 and 2003 - 2007. When pesticides contributing to TU exceedances were analyzed on an individual compound basis, it was found that concentrations of pirimiphos-methyl and carbendazim in surface waters of the study area revealed a significant negative trend over time (Figure 2). Concentrations of dichlorvos and chlorfenvinfos were not plotted versus time because most of their concentrations (up to 94%) remained below the limit of detection.

Concentrations of the other frequently measured pesticides revealed a significant declining trend over time (for instance, tolclophos-methyl, imidacloprid, chloridazon, isoproturon, chlorpropham, diuron, simazine, metoxuron). Concentration of prochloraz did not change significantly over time. Even though exceedances of EC50 values for several pesticides were found, the TU at most of the sampling sites remained below 1: out of 2505 sampling sites at which pesticides were measured, the TU exceeded 1 at 80 sites, what makes 3.2% of all locations.

As can be seen in Figure 1D, TU increased simultaneously with the number of pesticides analyzed per sample. When the number of pesticides measured per sample was plotted versus TU, a significant positive trend was found (Figure 1E), suggesting a dependence of TU on the number of compounds measured per sample. Figure 1F shows normalized TU plotted versus time. The normalized TU remained stable between 1974 and 1998, followed by a decrease in 1998 - 2010 with lowest values observed in 2000 - 2004 (Figure 1F).

Annual patterns in macrofauna diversity

Figure 3 shows the Shannon index of macrofauna diversity plotted versus time. The difference in Shannon diversity indices between the two areas was not statistically

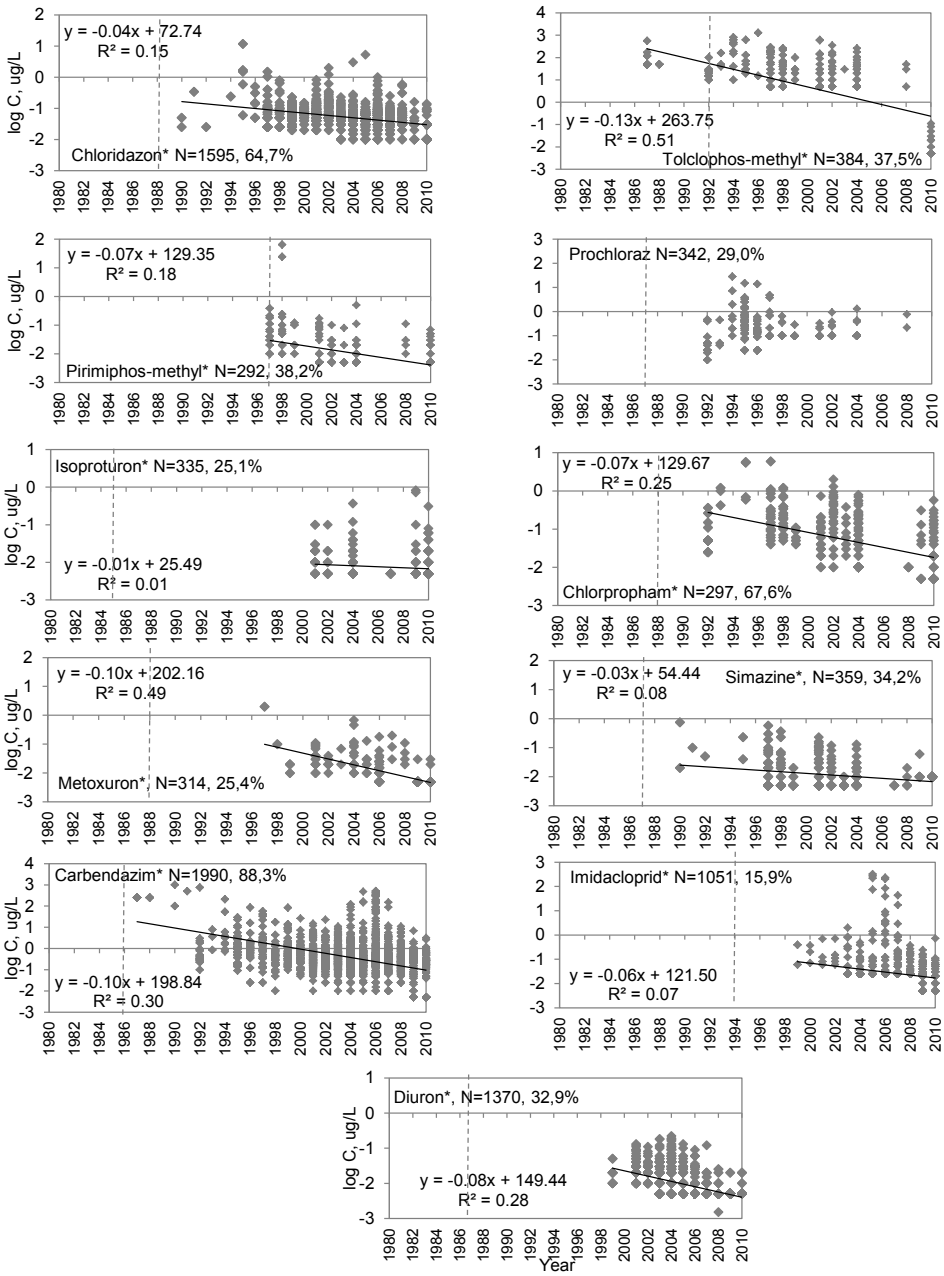

Figure 2. Concentrations of most frequently measured pesticides (logarithms with base 10, log C) plotted versus time, number of measurements done for each pesticide (N), and the percentage of measurements in which pesticide concentration exceeded the limit of detection (%). The dashed line corresponds to pesticide authorization date. *the regression line and equation are shown in case when relationship between pesticide concentration and time was statistically significant, as identified by regression analysis ($p<0.05$)

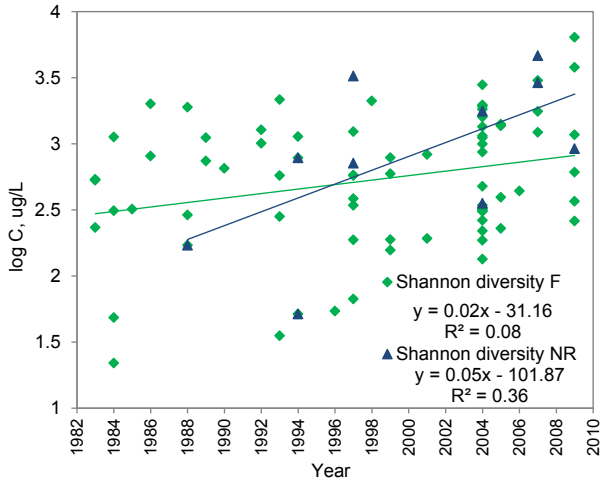

Figure 3. Shannon index of macrofauna diversity in ditches next to flower fields (green points) and nature reserve (blue points) plotted versus time. Regression lines and equations are shown. F = sampling sites in the flower growing area (p = 0.017), NR = sampling sites in the nature reserve (p = 0.066)

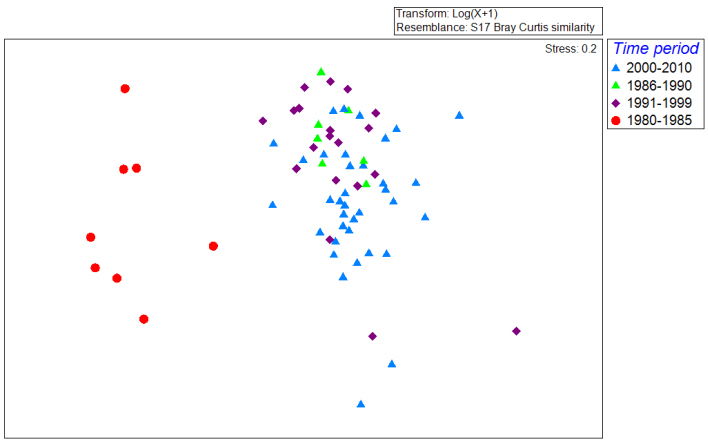

Figure 4. Non-metric multidimensional scaling of the macrofauna species composition Different colors correspond to different sampling periods. Summary of ANOSIM analysis testing the differences in macrofauna community composition between the sampling periods is given in Table 2.

Table 2. Results of ANOSIM analysis testing the differences in macrofauna community composition between sampling periods

Groups tested	R statistic	Significance level %
2000-2010 vs 1986-1990	0.078	23.8
2000-2010 vs 1991-1999	0.204	0.2*
2000-2010 vs 1980-1985	0.896	0.1*
1986-1990 vs 1991-1999	-0.076	72.0
1986-1990 vs 1980-1985	0.852	0.2*
1991-1999 vs 1980-1985	0.821	0.1*

*statistically significant at $p<0.05$

significant, as identified by the t-test. The Shannon index of macrofauna diversity in both the flower growing area and the nature reserve tended to increase over time ($p=0.017$ and $p=0.066$ respectively).

An MDS plot of macrofauna species composition is presented in Figure 4. Samples collected in 1980 - 1985 formed a distinct cluster, suggesting dissimilar macrofauna species composition in these years compared to other years (Figure 4). The possible explanation is that macrofauna in 1980 - 1985 was characterized by lower species diversity compared to the following years (Figure 3). ANOSIM analysis revealed significant difference between macrofauna species composition in 1980 - 1985 and that in later years (Table 2). Similarly, macrofauna community composition in 1991 - 1999 was significantly different from that in 2000 - 2010.

Conclusions

In the current study, we could not quantitatively link pesticide levels in surface waters and macrofauna diversity because pesticide and macrofauna data were not collected consistently over time and sampling site. Hence, it was not possible to perform a correlative analysis between pesticides levels in ditches and macrofauna diversity. To infer causal relationships between pesticide levels and the performance of aquatic biota, further field research is needed.

Addressing the two research questions, our results are the following. 1) Pesticide levels in ditches of the flower growing area changed over the previous decades. Toxic Units normalized by the number of pesticides measured per sample remained stable between 1974 and 1998, followed by decrease in 2000 – 2010, with the minimum values observed in years 2000 - 2004. Concentrations of the most frequently measured pesticides (like tolclophos-methyl, imidacloprid, chloridazon, isoproturon, chlorpropham, diuron, simazine, metoxuron) decreased over time confirming our starting hypothesis, while concentrations of

other pesticides (like prochloraz) remained stable over time. Carbendazim and pirimiphos-methyl contributed mostly to the exceedances of toxic units in recent years. 2) Macrofauna diversity in ditches of the flower growing area and watersheds of nature reserve increased over time. Macrofauna species composition in 1983 - 1985 was significantly different from that in the later years.

Acknowledgments

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