OCCURRENCE OF PESTICIDES IN SWEDISH WATER RESOURCES AGAINST A BACKGROUND OF NATIONAL RISK-REDUCTION PROGRAMMES - RESULTS FROM 20 YEARS OF MONITORING

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ABSTRACT

One important aim of the new EU thematic strategy on the sustainable use of pesticides is the protection of the aquatic environment. The intention is to reduce levels of harmful substances and the need for monitoring programmes is stressed, including the determination of concentrations of pesticide residues in water. Sweden has long-term experiences in pesticide monitoring programmes and also in the implementation of risk-reduction programmes for the use of pesticides. To assess the effects of political decisions, information on occurrence of pesticide residues in Swedish waters is collected and stored in a national database. Results from analyses of surface water and groundwater throughout Sweden now cover a 20-year period (1985-2005). The data show that the incidence of samples with a total pesticide concentration above 0.5 µg/L has decreased over the years, both from surface and ground waters. Especially in surface waters, having shorter pathways between the site of application and the receiving water body there has been a notable decrease in pesticide concentrations. This positive development is likely the result of some of the national risk-reduction measures introduced.

KEY WORDS: pesticides, monitoring, database, surface water, groundwater.

INTRODUCTION

In the mid-1990s, data on the occurrence of pesticide residues in groundwater and watercourses throughout Sweden were quite unknown. Various investigations had been conducted by local and regional authorities such as the Swedish National Food Administration, National County Boards and different Water Quality Associations, but there was no national monitoring programme at this time and no general overview of the situation. Therefore in 1996 a database was set up to store information on pesticide occurrence in surface water, groundwater and drinking water. The information now comprises a 20-year period (1985-2005), and contains information on more than 10 000 water samples covering all Swedish water districts and counties and 88% of municipalities. The database is administrated by the Division of Water Quality Management at the Swedish University of Agricultural Sciences, and is financed by the Swedish Environmental Protection Agency. The Division also manages the Swedish national monitoring programme, which started in 2002 in four small agricultural catchments. The programme includes investigations in different substrates such as groundwater, surface water and precipitation (Adielsson & Kreuger, 2007) and its aim is to quantify and chart variations of pesticides with time and space. It originated from a monitoring study set up in a small catchment in southern Sweden in 1990 to examine the loss of pesticides (Kreuger, 1998).

The objectives of the EU thematic strategy on the sustainable use of pesticides (COM (2006) 372 final) are to minimise the risks to health and environment from pesticides, by improving the handling of pesticides through education and enhancing the quality of application equipment, and to reduce the levels of harmful active substances in the environment. The document emphasises that determination of pesticide concentrations in the environment can be used to

verify model calculations, risk assessment predictions and how users act in compliance with restrictions and label instructions.

The overall objectives of this paper are to present a general overview of the occurrence of pesticides in Swedish surface and groundwater resources during the period 1985-2005. How is the pesticide situation in Swedish waters? Has it changed over the years? Have risk-reduction programmes had any impact on the occurrence and concentrations of pesticides in waters?

MATERIALS AND METHODS

The Swedish pesticide database was set up in 1996 in order to obtain a general overview of the occurrence of pesticides on a national level, i.e. the pesticides most frequently detected, their concentrations and their locations. Information dating back to 1985 has been collected, including data on type of water, sampling date and location, substances included in analysis, concentration levels detected, reporting limit for each substance and analytical laboratory. Every third year a larger update is conducted by sending out a questionnaire to Swedish counties, municipalities, water conservation associations and others. Between these major updates, information is also collected continuously on a smaller scale.

The results are collected from a multitude of different surveys and are therefore not homogeneous, i.e. analyses are not performed in the same way, do not include the same substances over time, and are not conducted for the same reasons. Some of the data are from random sampling; others are from more extensive monitoring programmes. There are large differences between surveys in number of sites, sampling intervals and the number of substances analysed. The description of the sampling sites varies, some have co-ordinates, some have actual property information, and most are identified by only a name. This can in some cases lead to difficulties in deciding whether a location has remained the same over the years.

This paper includes analytical results from 7 235 different samples accessible in the database (SQL/Access) in March 2007, mainly for groundwater and surface water. Some of the samples contain less information about the type of water investigated. For example, some are raw water used for municipal water supply where information on whether it is groundwater or surface water is lacking. Most of these ‘less-defined’ samples are from the earlier part of the 20-year-period included in this report. More recent raw water samples are sometimes mixtures of surface water and groundwater. Drinking water from private household wells has been assigned to the groundwater category, as has surface water infiltrated into groundwater bodies. This report does not include results on treated drinking water from municipal water supplies (today some 3 000 samples) since these results may be more dependent on the choice of water treatment than on the environmental state.

In this paper, findings below the limit of quantification (LOQ) but above the limit of detection (LOD), so-called traces, are included. The numbers of trace-values increased during the late 1990s, peaked in 2001 and thereafter decreased. The analytical laboratories have lowered the LOQ in subsequent years and hence more findings can be quantified at environmentally relevant concentrations. Results related to single substances only include the most frequently analysed substances, i.e. substances analysed more than 500 times.

The Swedish Chemicals Agency has derived water quality standards (guideline values) for approx. 100 different pesticides (KemI, 2004). The guideline values give the maximum concentration of each pesticide estimated not to cause any negative effects on aquatic organisms.

In the EU Drinking Water Directive (98/83/EC) and the new Groundwater Directive (2006/118/EC), the quality standard of groundwater and water used as drinking water has been set to 0.1 µg/L for individual pesticides and 0.5 µg/L for the sum of individual pesticides.

RESULTS

Of the 7 235 different samples included in this paper, 59% were groundwater and 38% surface water. A small proportion (3%) of the samples contains less information about the type of water investigated. A total of 305 different substances have been analysed over the years in surface and groundwater, 170 of them in more than 500 different samples.

The incidence of samples with a total pesticide concentration above 0.5 µg/L has decreased over the years (Figure 1). On the other hand, the percentage of samples with a total concentration below 0.1 µg/L has increased during the same period, reflecting refined analytical techniques.

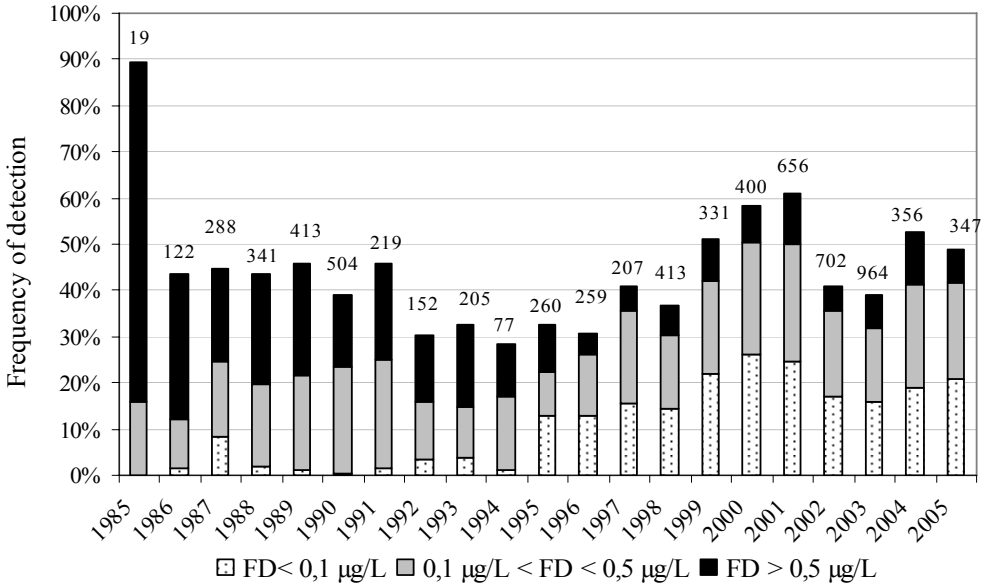

Figure 1: Frequency of detection (FD) for samples, with total pesticide concentration in different intervals. Figures above bars give the total number of samples each year.

GROUNDWATER

A total of 4272 samples collected originated from groundwater. The majority (67%) was groundwater samples intended for municipal water use and the remaining samples were mostly for private purposes (24%). The relationship between these two categories has changed somewhat over years, with more samples coming from municipalities in latter years. Overall, 1421 different sites have been investigated. The number of sites has generally increased over the years, with the number per year varying from 0 to 433 (Figure 2). The frequency of sites with at least one sample exceeding the water quality criteria has decreased.

Overall, 295 different substances have been analysed in groundwater, 136 of these in more than 500 samples. A total of 50 different substances have been detected in groundwater on one or more occasions and 36 of these have been analysed more frequently. The most commonly found substance is 2,6-dichlorobenzamide (sometimes abbreviated BAM), with a detection frequency of 40% during the 20-year period (Figure 3 and Table 1).

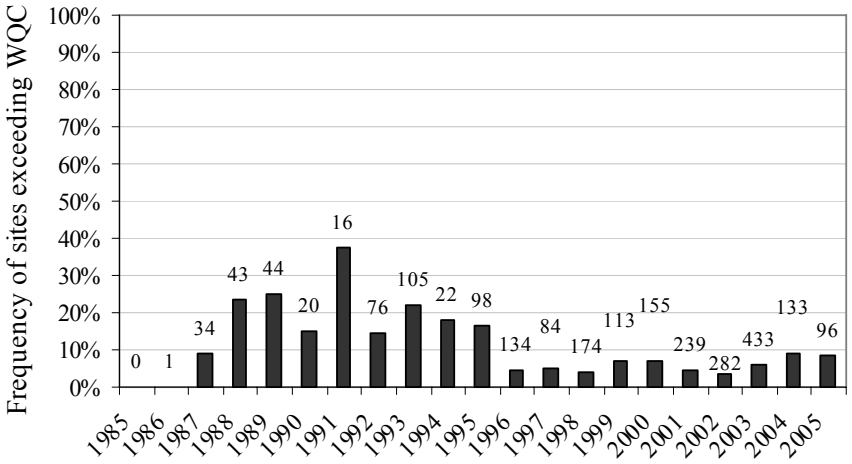

Figure 2: Frequency of groundwater sampling locations each year with one or more samples failing the drinking water quality criteria (WQC) of 0.5 µg/L (total pesticide concentration). Figures above bars give the total number of groundwater sites investigated each year.

This and two more of the top four most commonly found substances, atrazine and desethylatrazine, are residues of a former total weed-killer for non-agricultural purposes, named Totex Strö, which was banned in Sweden in 1989. This product was used in large quantities on gravel paths, industrial areas and embankments, places likely to leach and with low biological activity.

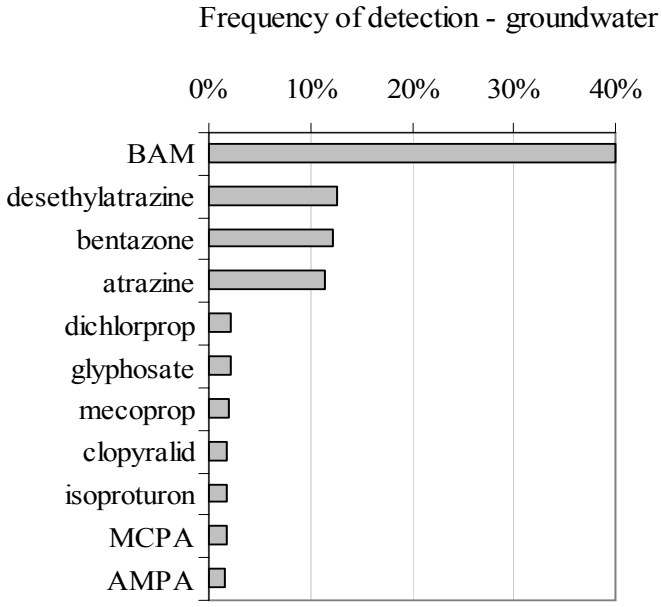

Figure 3: Frequency of detection for substances detected in more than 2% of the groundwater samples and analysed more than 500 times.

For single substances such as atrazine, desethylatrazine and bentazone, there has been no major change in findings of concentrations above 0.1 µg/L in groundwater between different time periods (Figure 4). The time intervals are set so that the number of samples is evenly distributed between the different periods. To minimise the influence of differences in LOD over the years, only findings above 0.1 µg/L are compared. For 2,6-dichlorbenzamide, comparisons between the time intervals are more difficult to interpret because of the differences in number of samples. All

four substances have been detected at levels over the quality standards in the Groundwater Directive on several occasions over the years (Table 1).

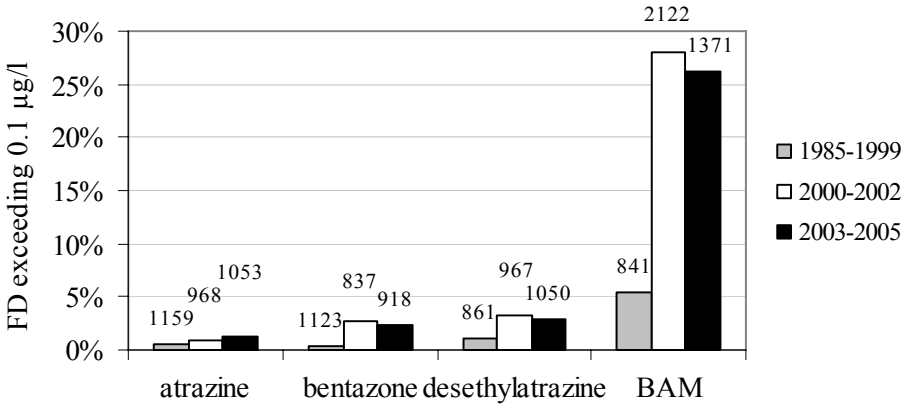

Figure 4: Frequency of detection (FD) for concentrations above 0.1 µg/L during three different time periods for the four most commonly detected substances in groundwater. Figures above bars give the total number of samples collected during each time interval.

Table 1: Substances detected in groundwater and surface water samples, 1985-2005. Percentage of samples with detections above 10% for groundwater and 15% for surface water

Substance	Number of samples	% samples with detections	Average reporting limit (µg/L)	Median detected conc. (µg/L)	Mean of detected conc. (µg/L)	Maximum conc. (µg/L)	Number of samples above quality standards (%)
Groundwater							
atrazine	3180	11%	0.05	0.05	0.37	14	135 (4%)
bentazone	2878	12%	0.05	0.09	1.9	280	174 (6%)
desethylatrazine	2878	13%	0.05	0.06	0.27	23	139 (5%)
BAM	3414	40%	0.01	0.13	0.24	5.4	844 (24%)
Surface water							
bentazone	2305	31%	0.05	0.1	0.48	80	1 (<0.1%)
BAM	1056	16%	0.05	<0.05	0.02	0.11	- *
dichlorprop	2438	18%	0.10	0.1	1.4	230	10 (0.5%)
MCPA	2442	27%	0.05	0.2	0.90	100	8 (<0.5%)
mecoprop	2440	17%	0.10	0.08	0.35	23	2 (<0.1%)

* - = Substance lacking a Swedish guideline value (WQS) for surface water quality.

SURFACE WATER

A total of 2725 samples collected originated from surface water. The number of samples investigating the environmental state of watercourses has decreased over the years. Samples

taken from water for municipal use now constitute a total of 22%, with this proportion increasing over the years at the expense of samples from environmental monitoring. A total of 509 different sites have been investigated, with the number per year varying from 8 to 103 (Figure 5). The number of sites investigated has remained fairly constant over the years, but the sites themselves have changed.

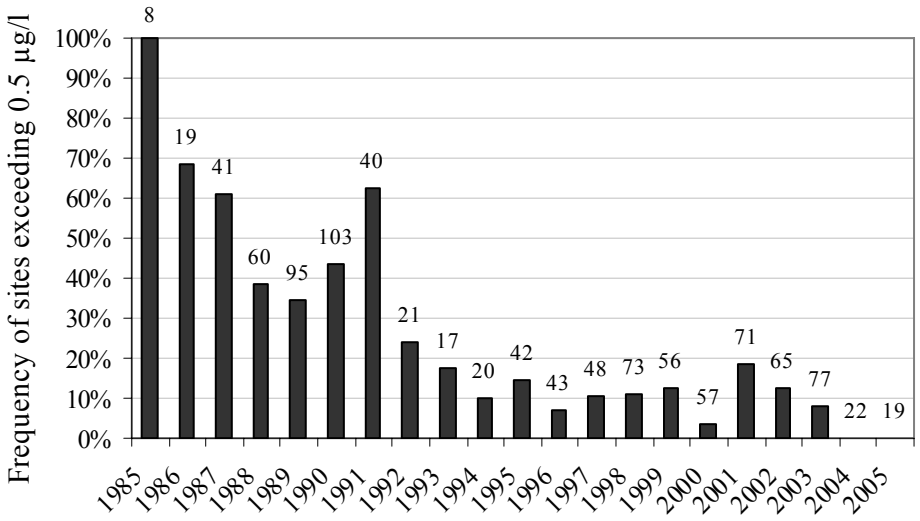

Figure 5: Frequency of surface water sites with one or more samples above 0.5 µg/L (total pesticide concentration). Figures above bars give the total number of surface water sites investigated each year.

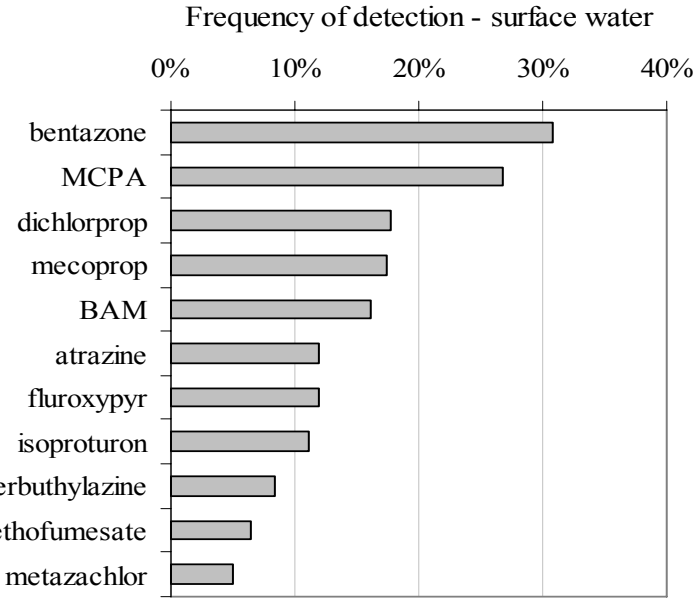

Figure 6: Frequency of detection for substances detected in more than 5% of surface water samples and analysed more than 500 times.

Overall, 268 different substances have been analysed in surface water, 103 of which have been found in more than 500 samples. A total of 68 different substances have been detected in surface water on one or more occasions and 47 of them have been analysed more frequently. The most commonly found substance has been bentazone, with a detection frequency of 31% (Figure 6 and Table 1). For the top four most commonly found substances in surface water (bentazone, dichlorprop, MCPA and mecoprop), there has been a substantial decrease in the frequency of

detection of concentrations over 0.1 µg/L (Figure 7). These substances have also occasionally (<1%) been found at concentration levels above their respective guidelines value (Table 1).

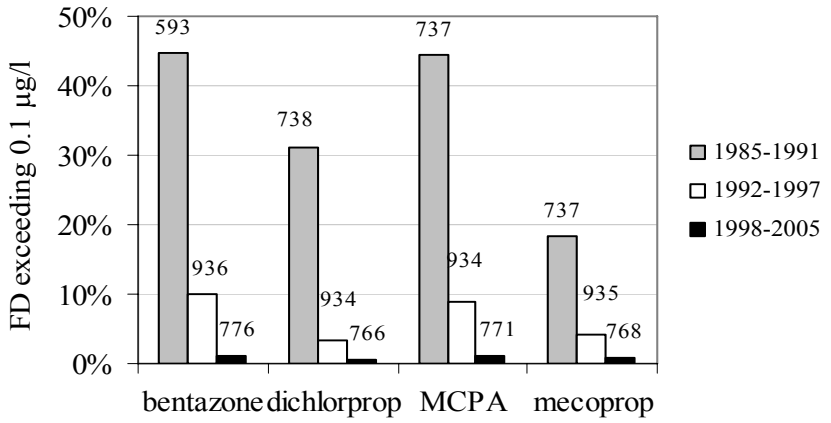

Figure 7: Frequency of detection (FD) for concentrations above 0.1 µg/L during three different time periods for the four most commonly detected substances in surface water. Figures above bars give the total number of samples collected during each time interval.

DISCUSSION

In Sweden, some measures have been taken to minimise the risk to health and environment during the 20-year period for which water quality data are currently available. Such measures include the Swedish national risk reduction programmes and an information campaign called Safe Pesticide Use that was launched in 1997. This campaign focused on safe filling and cleaning of agricultural sprayers and safety distances as regards surface runoff and wind drift. Furthermore, comparing the quantities of pesticides sold to agriculture and horticulture in 2005 with the average quantities sold during the early 1980s shows that there has been a 63% reduction in the 20-year period (KemI, 2006). All these efforts seem to have had an impact on the concentrations of pesticides in surface waters. There has been a substantial decrease in sites with a total pesticide concentration of 0.5 µg/L or more (Figure 5), and for the most commonly detected substances there has been a significant decrease in detection of concentrations of 0.1 µg/L or more over the years (Figure 7). However, for groundwater there has been no substantial reduction so far for the most commonly detected compounds at concentrations of 0.1 µg/L or more (Figure 4). The number of sites investigated has varied widely between years (Figure 2), making it difficult to interpret the differences in findings over the years. Nonetheless, the results demonstrates an overall longer time interval between the onset of risk mitigation measures (including banning of certain compounds) and a reduction in findings of pesticides in groundwater.

The Swedish EPA previously provided subsidies for monitoring in watercourses. This financial support was taken away in 1991 and the number of surveys decreased substantially. In the late 1990s, the number started to increase again. In 2003 the new regulations on drinking water (SLV, 2001) were implemented and the focus shifted from raw water to the treated water that reached the consumers. Consequently many municipalities now only investigate the drinking water and the monitoring in watercourses has decreased again. This leads to difficulties in following the occurrence of pesticides in the environment.

The top eleven substances detected (Figures 3 and 6) are all herbicides and five of these have been frequently found both in groundwater and surface water. Several of the most common substances in groundwater have been found at remarkably high concentrations (Table 1). The concentrations of these substances could be expected to be lower in groundwater than in surface water, since the groundwater has percolated through the soil and the pesticides are exposed to absorption and degradation. Concentrations at several micrograms per litre indicate that substances are being transported to the groundwater in an abnormal way, either through point sources such as spillage, accidents or leaking wells or through inappropriate use of substances, e.g. spraying on unsuitable sites.

CONCLUSIONS

Pesticides have been found on numerous occasions in Swedish waters during the 20-year period 1985-2005. Overall, the total incidence of concentrations above 0.5 µg/L has decreased. For surface water it is possible to discern a substantial reduction in pesticide concentrations, and also in the number of sites with high total detections of pesticides. There have been a number of national efforts made to reduce the levels of harmful substances in Swedish waters and it is likely that these have had an impact as regards surface waters. However, a similar improvement has not occurred in groundwater. The increase in the number of sites investigated makes it difficult to interpret the reduction in detections exceeding the water quality criteria. However the lack of decrease in detections of the most commonly occurring substances in groundwater may indicate that the efforts made to date are insufficient.

ACKNOWLEDGEMENTS

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