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This appendix contains translations to English of the summary, conclusions and captions of figures, tables and appendices for the following report:

Resultat från miljöövervakningen av bekämpningsmedel (växtskyddsmedel). Långtidsöversikt och trender 2002-2012 för ytvatten och sediment

Results from the national monitoring program of pesticides. Long-term overview and trends for surface water and sediments, 2002-2012.

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Contents

1 Summary

Pesticide analyses have been conducted by the Swedish University of Agricultural Sciences since 2002, within the framework of the national environmental monitoring programme (Agricultural Soils) managed by the Swedish Environmental Protection Agency. The analyses include collection of surface water samples and sediment for determination of pesticide residues in four streams draining small model catchments representative of intensely cultivated areas within the counties of Västergötland, Östergötland, Halland, and Skåne, the dominant agricultural regions of Sweden. Samples are also collected from two rivers, Skivarpsån and Vege å, in Skåne.

This report provides an in-depth description of the model catchments in terms of growing conditions (particularly climate and soil type), dominant crops, and pesticide use. Further, we present a compilation of analytical data on pesticides in surface water and sediment for the period 2002-2012, including a detailed description of detection frequency, surface water concentrations of commonly used substances, and emerging trends during the period. Appendices include detailed information on weather conditions in the four catchments, the scope of the analyses, and a summary of detection frequency and concentration per substance and area.

The catchment sizes vary between 770 ha (Västergötland) and 1630 ha (Halland), of which approx. 90% is arable. The soils within the four catchments are representative of Swedish agricultural soils in general, ranging from sandy soils in the catchment in Halland to heavy clays in the Västergötland catchment. The catchments in Halland and Skåne have greater mean annual precipitation (775 mm and 730 mm, respectively, for 2002-2012) and higher mean annual temperature (~8°C), while the catchments in Västergötland and Östergötland are drier (650 mm and 580 mm, respectively) and slightly colder (~7°C). For all catchments, this represents an increase in mean annual temperature of nearly 1°C compared to the 30-year norm (1961-1990) and all catchments, except Halland, also exhibit an increase in mean annual precipitation by around 100 mm. This increased rainfall compared with the 30-year norm mainly occurs in summer, with an average increase for May-October of 60-90 mm.

There were no major changes in the crops grown in the model catchments during 2002-2012, apart from a decreased area of fallow in all catchments except Skåne and an increased area of maize in the Halland catchment (total 5% in 2012). The dominant crops in each catchment are as follows: Västergötland – cereal (grown on ~75% of arable land), oilseeds, and peas; Östergötland – cereal (~65%), oilseeds, potatoes, forage ley, and peas; Halland – cereal (~50%), forage leys, potatoes, sugar beet, and peas; and Skåne – cereal (~65%), sugar beet, oilseeds and peas.

The proportion of arable land treated with some form of pesticide increased during 2002-2012 in the Västergötland and Östergötland catchments, from ~80% in the beginning of the period to ~90% in later years. This was mainly due to decreasing area under fallow and forage ley in these catchments. In the Halland catchment, around 20% of the arable land was under fallow

and ley during 2002-2012.This is a higher proportion than in the other catchments, and correspondingly a smaller proportion of the arable land was treated with pesticides (80%). In the Skåne catchment, the pesticide-treated proportion of arable area was around 95% throughout the entire period. Herbicides were used principally on the entire pesticide-treated acreage in all four catchments, but the application of fungicides and insecticides showed greater variability within and between the catchments.

The total amount of active pesticide ingredients applied within each catchment, calculated per unit treated area (kg/ha) was twice as high in the southern catchments, Skåne and Halland, as in Västergötland and Östergötland. This is a consequence of differences in climate and choice of crops between the regions. Overall, despite variations between years, the total use decreased by around 0.25 kg/ha in all catchments from 2002 to 2012.

Within each catchment, the choice and relative distribution of crops within the arable area and the use of pesticides (calculated as amount of active ingredient applied per unit treated area and proportion of treated area) can be considered comparable to those in the respective county, provided the intensity of cultivation within the catchment is taken into account. Thus, the catchments can be considered representative of intensive agricultural areas for each county. Likewise, the spread of soil types found across the model catchments is reasonably representative of arable land in Sweden.

In each model catchment, surface water samples were taken with automatic samplers (time-integrated weekly samples, with sub-samples every 80 minutes), while the two rivers were sampled manually with a bottle on a pole (momentary grab samples). The majority of samples were collected during the summer months, *i.e.* the period during which pesticide use is most intense. This resulted in between 20-30 samples per model catchment and year, and nine samples per river and year. Additionally, in Skåne another automatic surface water sampler provided 12-42 flow-proportional samples per year. Flow-proportional means that an increase in water flow corresponds to more frequent sampling. Sediment sampling was carried out once a year (in September) in all streams in the catchments and in the rivers.

The number of substances analysed per sample of surface water increased from 76 in 2002 to 131 in 2012. During the period 2002-2012 the analytical methods progressively improved, with *e.g.,* lower detection limits and more substances included in analyses from 2009 onwards. The substances targeted in analyses represent the majority of substances used by farmers in the model catchments. In 2012, between 78% and 99% of the total amount (kg) of pesticides used in the catchments were included in the analyses.

In total 140 substances were included in the analyses of surface water during the entire or part of the period 2002-2012. Of these, 10% were detected often (in >50% of samples), while 50% of the substances were detected seldom or never (in <2% of samples).

The concentrations of pesticides in surface water generally displayed no clear trends in 2002-2012, and the average total concentration for an individual sample during the entire period

was around 1.0 µg/L. This is partly because a few substances make up the majority of the total concentration and their concentrations remained relatively constant during the period.

On average, 13 substances were detected per sample in 2002-2008 and 22 substances per sample in 2009-2012. Samples from the Skåne catchment and the rivers had more substances than the other catchments. The substances encountered most frequently in 2002-2012 were the herbicides bentazone (100% of all samples), glyphosate (89%), isoproturon (76%), MCPA (75%), and metazachlor (65%). These substances were also the most commonly used in terms of amount and/or treated area during the period, which partly explains why they were detected in most samples. However, this was not the case for bentazone, which was not extensively used. The concentrations and detection frequencies of these substances showed no clear trends during the period.

The proportion of surface water samples with concentrations of one or more pesticides exceeding its respective environmental quality standard (EQS) remained relatively constant in the period 2002-2012, with the limit exceeded in around 45% of samples. Diflufenican, a commonly occurring herbicide in water samples (50% of all samples), was the substance that most frequently exceeded its EQS (25% of all samples), although the frequency of exceedance varied greatly between catchments (2% in Östergötland, 68% in Skivarpsån). Other substances detected in concentrations over the EQS were the fungicide picoxystrobin (14% of all samples) and the herbicides isoproturon (7%) and MCPA (6%).

Some substances that were banned before or at the start of the monitoring period (year 2002), for example atrazine, terbuthylazine, their degradation products, and BAM (degradation product of diclobenil), are persistent and can still be encountered in surface water. For terbuthylazine and its degradation products DETA, however, the concentrations and frequency of EQS exceedance both decreased during the period. Also in sediments, a number of long banned substances, *e.g.,* DDE, occurred relatively frequently. All substances detected in sediment samples have a high capacity to bind to organic material and/or clay minerals. Glyphosate was the substance detected at highest concentrations and was one of two substances, the other being esfenvalerate, found in all catchments. Considerably fewer substances were detected in the sediment samples than in the surface water samples. The highest number of substances was found in sediment from the Skåne catchment, where the frequency of detection also was highest.

In general, it was difficult to discern any clear trends for individual pesticides, since 11 years (2002-2012) is a relatively short period for many substances due to variability in use and weather conditions between years.

Despite some differences between the model catchments regarding climate, soil type, and crop distribution, our data shows that the total concentration of pesticides in surface water did not differ substantially between the model catchments This was consistent for all types of pesticides (herbicides, fungicides and insecticides). In addition, the total concentration measured in the rivers did not differ substantially from those measured in the model

catchments, despite differences in sampling method, proportion of arable land, size of catchment area, and discharge.

Since the existing EQS values for different pesticides have not been updated for several years and are only preliminary for some substances, the exceedance amount and frequency patterns could shift if new studies were conducted that led to the guideline values being adjusted, upwards or downwards.

2 Conclusions

Weather data from the environmental monitoring of pesticides in four agricultural model catchments in Sweden reveal an increase in summer precipitation of 60-90 mm (May-October) during the period 2002-2012 compared with the national 30-year norm (1961-1990). A wetter climate can increase the risk of pesticide leaching, emphasizing the importance of continuous, long-term monitoring of climate impacts within environmental monitoring programs.

According to our analyses, the soils in the four model catchments included in the monitoring program are representative of Swedish arable soils. The wide range of soil textures found in the catchments differs in their susceptibility to leaching, from sands to clays.

The crops grown within the model catchments are relatively representative for their respective county, except they include less ley, *i.e.* cropping is generally more intense within the model catchments than in the county in general. No major changes were found regarding the crops grown during the period, except for the area of fallow decreasing slightly towards the end of the period. This corresponded to a simultaneous increase in the proportion of arable land being treated with pesticides in the catchments.

The amounts of active substances applied, calculated as kg per unit area, showed a weakly declining trend in all four model catchments during the period 2002-2012, with a generally higher level of pesticide use in the Southern catchments in Halland and Skåne than in the catchments in Västergötland and Östergötland.

Total use of fungicides and insecticides varied between years, whereas the use of herbicides was more consistent. However the substances used varied, as new products were introduced and others disappeared. This requires flexibility in the analysis programme, although the results show that the majority of the substances used during the period were included in the analyses.

The concentrations of pesticides in surface water generally displayed no clear trends in 2002-2012, with the average total concentration for an individual sample during the entire period remaining around 1.0 µg/L. This was partly because a few substances with a relatively constant concentration over the years contributed most to the total concentration. They include the herbicides bentazone, glyphosate and MCPA.

Herbicides are the most commonly used group of pesticides and this is reflected in them being most frequently detected in surface water. They were also detected more frequently in elevated concentrations in all catchments and rivers included in the environmental monitoring program.

Analytical advancements made it possible to detect lower concentrations from 2009 onward than in the beginning of the monitoring period. As a consequence, some substances that had been difficult to detect at environmentally relevant levels, *i.e.* below the EQS, could be

monitored more accurately in later years. As a consequence, some trends may be challenging to interpret during this period, because an increase in *e.g.,* number of detected substances or detection frequency may be caused by the improved ability to detect substances at lower concentrations and not an actual increase in occurrences.

No trend was seen in the proportion of surface water samples with concentrations of pesticides exceeding their EQS values during the period 2002-2012. The substances that most commonly exceeded their EQS values were mainly herbicides, although most herbicides never exceeded their EQS, because they generally exhibit relatively low toxicity to aquatic organisms and thus have relatively high EQS values.

The substances that most frequently exceeded their EQS in the period 2002-2012 were the herbicides diflufenican, isoproturon, MCPA, metribuzin, metazachlor, and terbuthylazine. In addition, the insecticides imidacloprid and tiacloprid and the fungicide picoxystrobin were detected relatively often in concentrations exceeding the EQS. These three substances were approved for use after the monitoring programme started in 2002 and were only analysed in the later part of the period.

In sediment samples analysed 2003-2012, considerably fewer pesticides were found than in surface water samples. The highest concentrations detected were of glyphosate, although some long banned substances, *e.g.,* DDE, occurred relatively frequently. All substances detected in sediment samples have a high capacity to bind to organic material and/or clay minerals.

In general, it is difficult to discern any clear trends for individual pesticides, since 11 years (2002-2012) is a relatively short period due to variability in use and weather conditions between years.

Despite some differences between the model catchments regarding climate, soil type, and crop distribution, our data shows that the total concentration of pesticides in surface water did not differ substantially between the model catchments This was consistent for all types of pesticides (herbicides, fungicides and insecticides). In addition, the total concentration measured in the rivers did not differ substantially from those measured in the model catchments, despite differences in sampling method, proportion of arable land, size of catchment area, and discharge.

Since the existing EQS values for different pesticides have not been updated for several years and are only preliminary for some substances, the exceedance amount and frequency patterns could shift if new studies were conducted that led to the guideline values being adjusted, upwards or downwards.

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Figure 65. Detection frequency for the most commonly occurring substances in sediment samples during 2003-2012. Only substances with a detection frequency above 10% are included. Page 79

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Appendix 1.

Precipitation, stream hydrographs, and temperature in the model catchments 2002-2012, as well as river hydrographs for Skivarpsån and Vege å. The 30-year norms (1961-1990) for precipitation and temperature are shown for comparison and hydrographs are presented in relation to the average for 2002-2012.

Figure B1 – 1. Monthly precipitation in model catchment O18 (Västergötland) during 2002-2012 as percent of monthly average for the 30-year norm (1961-1990). Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Figure B1 – 2. Monthly average temperature for the model catchment O18 (Västergötland) compared to the 30-year norm.

Figure B1 – 3. Monthly drainage in model catchment O18 (Västergötland) during 2002-2012. Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Table B1 - 1. Precipitation data for the model catchment in Västergötland (O 18).

Figure B1 – 4. Monthly precipitation in model catchment E 21(Östergötland) during 2002-2012 as percent of monthly average for the 30-year norm (1961-1990). Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Figure B1 – 5. Monthly average temperature for the model catchment E 21 (Östergötland) compared to the 30-year norm.

Figure B1 – 6. Monthly drainage in model catchment E 21(Östergötland) during 2002-2012. Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Table B1 - 2. Precipitation data for the model catchment in Östergötland (E 21).

Figure B1 – 7. Monthly precipitation in model catchment N 34(Halland) during 2002-2012 as percent of monthly average for the 30-year norm (1961-1990). Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Figur B1 – 8. Monthly average temperature for the model catchment N 34 (Halland) compared to the 30-year norm.

Figure B1 – 9. Monthly drainage in model catchment N 34 (Halland) during 2002-2012. Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Table B1 – 3. Precipitation data for the model catchment in Halland (N 34)

Figure B1 – 10. Monthly precipitation in model catchment M 42 (Skåne) during 2002-2012 as percent of monthly average for the 30-year norm (1961-1990). Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Figure B1 – 11. Monthly average temperature for the model catchment M 42 (Skåne) compared to the 30-year norm.

Figure B1 – 12. Monthly drainage in model catchment M 42 (Skåne) during 2002-2012. Grey shading denote May-October, *i.e.* the ordinary sampling period for surface water.

Table B1 – 4. Precipitation data for the model catchment in Skåne (M 42).

Figure B1 – 13. Hydrograph (l/s) for Skivarpsån; average flow per month 2002-2012 compared to monthly average for the same period. Grey shading denote the ordinary sampling period for surface water, May-October.

Figure B1 – 14. Hydrograph (l/s) for Vege å; average flow per month 2002-2012 compared to monthly average for the same period. Grey shading denote the ordinary sampling period for surface water, May-October.

Årsnederbörd – cumulative precipitation (in mm) for the entire year.

Nedb. maj-okt – cumulative precipitation (in mm) from May 1 to 31 October.

 # dagar kum. nb ≤10 – number of consecutive days with a maximum of 10 mm cumulative precipitation. This parameter represents the length of a drought period and is calculated for the period between May 1 and October 31.

Max kum. nb 10 dagar – Maximum cumulative precipitation (in mm) during a 10 day period. This is calculated for the period between May 1 and October 31.

Max dygn-nb – Maximum precipitation (in mm) during a 24 hour period between May 1 and October 31.

 # dagar ≥10 – Number of days (not necessarily consecutive) with a precipitation of 10 mm or more during the period May 1 to October 31.

Appendix 2a.

Summary of analyzed substances during 2002-2012 and the year they were added to or removed from the analyses of surface water samples. Symbols indicate that the substance was included in the analyses for surface water from **streams (□)** and/or **rivers (X)** that year.

Appendix 2b.

Summary of analyzed substances and the year they were added or removed from the analyses for **sediment samples**, 2002-2012. Circles (○) indicate that the substance was included in the analyses that year.

Appendix 3.

Evolution of detection limits during 2002-2012 (µg/l, median values) in surface water samples from streams (ordinary sampling period) and rivers. Light red numberindicate that the detection limit is up to 10 times higher than the EQ (RV; µg/L). Dark red numbers indicate a detection limit more than 10 times higher than the EQ. Unless otherwise noted the EQ value is the standard Swedish EQ (Kemikalieinspektionen, 2014).

Substance types: H= herbicide; F= fungicide; I= insecticide; B= biocide; TV= growth regulator

- = EQ value does not exist
* = EQ according to the environmental quality standard (AA-MKN) for terrestrial water according to EU directive 2013/39/EU. Maximum concentration for preventing acute toxicity (MAC-MKN) is normally higher than this value.
# = refers to the total concentration of all isomers.
[a] = Preliminary EQ according to Andersson et al., 2009.

[b] = Preliminary EQ according to Andersson & Kreuger 2011.
[c] = Calculations assume that the EQ is the same as for the original substance n (Asp & Kreuger, 2005).
[d] = Preliminary EQ based on data from Agritox (2013).

Appendix 4.

Summary of the ordinary sampling period, 2002-2012, in **Västergötland** (O 18); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 5.

Summary of the ordinary sampling period, 2002-2012, in **Östergötland** (E 21); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 6.

Summary of the ordinary sampling period, 2002-2012, in **Halland** (N 34); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 7.

Summary of the **winter sampling**, 2002-2012, in **Halland** (N 34); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average

concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 8.

Summary of the ordinary sampling period, 2002-2012, in **Skåne** (M 42); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 9.

Summary of the **winter sampling**, 2002-2012, in **Skåne** (M 42); substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 10a.

Yearly summary of **flow-proportional** sampling in comparison to the time-integrated samples in Skåne (M 42), 2006-2012 (no flow-proportional samples from 2008); ”Max kvot” represents the highest detected ratio (largest difference) per substance between the concentration in a flow-proportional sample and the time-integrated sample (weekly average concentration, VMH) for the same week (*i.e.* the flow-proportional sample was taken during the week represented by that VMH).

Appendix 10b.

Summary of **flow-proportional** sampling for the entire period 2006-2012 (not including 2008) in comparison to the time-integrated sampling in Skåne (M 42). Max kvot” represents the highest detected ratio (largest difference) per substance between the concentration in a flow-proportional sample and the time-integrated sample (weekly average concentration, VMH) for the same week (*i.e.* the flow-proportional sample was taken during the week represented by that VMH). For all samples the maximum concentrations in the flow-proportional and time-integrated samples are provided for the ordinary sampling period, May-November) irregardless of the sampling time. The table also lists the years that each substance has been included in the analyses and the EQ (RV) for the substances. Concentrations marked in bold denote that the EQ was exceeded for that sample.

Appendix 11.

Summary of the ordinary sampling period, 2002-2012, for **Skivarpsån**; substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 12.

Summary of the ordinary sampling period, 2002-2012, for **Vege å**; substances included in analyses, first and last year that the substance was included (see appendix 2 for details), application within the model catchment (the most commonly used substances is denoted by 1), total number of samples that were collected, proportion of samples with detects, number of detects as percent of the number of samples, maximum concentration measured, and percentile values for the weekly average concentration (VMH) as µg/L, and current EQ (RV) and number of detects above the EQ as percent of total number of samples. Exceedance of the EQ was calculated differently for samples analyzed between 2002-2008 and those analyzed between 2009-2012, see **section 3.7** for explanation of the calculations.

Appendix 13.

Substances analyzed in **sediment samples** from the four model catchments and the two rivers 2003-2012. Ones ample per area were collected every year in September, *i.e.* each substance was analyzed in at most 10 samples. Summary of the years that the substance was included and the detection frequency (in %) and maximum concentrations (µg per kg DW) for each sampling point. Substances that were only analyzed one year were excluded from this summary table.