Atrazine and dichlobenil residues in Uppsala soil? A study of soil cores

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by

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Abstract

Pesticides residues of atrazine and dichlorobenzamide (BAM) have been found in ground and drinking water in Uppsala, Sweden during the last years. To examine where the pesticides can possibly leach down to the ground water and to look at the distribution and levels of these pesticides in soil two locations were chosen, Uppsala university hospital and Eklundshof, known to be contaminated with atrazine and BAM. At each location two soil cores were collected to a depth of 4 and 5 m respectively, and pesticide concentrations were determined by GC-MS. BAM was detected in all soil samples at Uppsala university hospital, while dichlobenil was only found in one of the topsoil samples from Uppsla university hospital. No other pesticide residues were found. The levels of pesticides were rather low and whether these locations contaminate the ground and drinking water is difficult to determine. A comparison between data from the soil samples and a mathematical model called MACRO did not match very well, mainly because of uncertainties with regard to half-life and the amounts of pesticides applied.

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1. Aim

The aim of the study was to examine the distribution of atrazine and dichlobenil residues in soil profiles to a depth of 5 m, in order to examine whether these pesticides could be leached into the groundwater of the Uppsala esker, and to determine the levels of these pesticides throughout the profiles. A second task was to model the movement and levels of atrazine and dichlobenil residues in a mathematical program called MACRO (Jarvis, 1994) and to compare the results from MACRO and the levels of pesticides in the soil profiles.

2. Introduction

The use of pesticides in Sweden is relatively small compared to other countries and the government is working for even lower use of pesticides, yet pesticide residues are found in all kinds of water; streams, surface water and groundwater (Hessel, Kreuger & Ulén, 1997). Pesticide residues in drinking-water is not acceptable according to Swedish drinking water regulation (National Food Administration, 1993) and at pesticide levels higher than 0.1 µg/l for individual pesticides action must be taken. This is based on the EC recommendation that the level of 0.1 µg/l for individual pesticides in drinking water shall not be exceeded (EC, 1998).

Atrazine is one of the most commonly detected herbicides in surface water, groundwater and drinking water (Hessel et al. 1997). 2,6-Dichlorobenzamide (BAM), which is a metabolite of dichlobenil, is also commonly found in groundwater and drinking water. BAM has been the most frequently found pesticide residue in water between 1985 and 1999 (Ulén & Kreuger, 2000).

According to Wallman (2000), traces of BAM was first observed in 1993 in the drinking water of the Department of Environmental Assessment in Ultuna, Uppsala and analyses of the groundwater around Uppsala between 1997-1999 show traces of atrazine and dichlobenil residues, some exceeding 0.1 µg/l (Appendix 1).

Degradation of these pesticide residues in topsoil is generally slow, and water soluble, persistent species can be found deep down in soil layers and at great distances from the actual application site. This was one of the reasons why atrazine and dichlobenil were banned in 1989 by the Swedish National Chemical Inspectorate, (Hessel et al. 1997). Yet they are still detected in water (Wallman 2000).

Atrazine and dichlobenil were ingredients in herbicide preparations that were mostly used on graveled areas like paths, courtyards, industrial sites and railway lines. One of the most common preparations with atrazine and dichlobenil was Totex strö, another common preparation that only contained dichlobenil was Casoron. Other preparations containing atrazine and/or dichlobenil were Prefix, Primal and Silver. No preparations containing these substances is approved in Sweden and the last product in use was banned in 1990 (Swedish National Chemicals Inspectorate, 2001).

Dichlobenil is a cellulose biosynthesis inhibitor, inhibits dividing meristems, germination of seeds and damages rhizomes and it is selective because it is bound to the top 5-10 cm of the soil (Tomlin, 1997). Atrazine inhibits photosynthetic electron transport. It is adsorbed through

the roots, translocated via the xylem, and accumulates in the apical meristems and leaves (Tomlin, 1997).

2.1 Fate of dichlobenil and atrazine in soils

Because of the relatively high volatility of dichlobenil, loss of dichlobenil from the soil surface can be rapid especially under circumstances of strong wind. Low temperature and heavy rain decrease evaporation, and this is the case also if the pesticide is cultivated into the soil, and if the pesticide is spread as granules (Torstensson, 1984).

The movement of dichlobenil with water through soil profiles is extremely slow due to its low water solubility and adsorption on soil constituents. Adsorption of dichlobenil to soils is dependent on the content of organic matter. In pure sand and on clay adsorption is rather low (PEMP, 2000). The adsorption potential can be described by the partitioning coefficient for the substance equilibrium between water and octanol, K_{ow}. The log K_{ow} value for dichlobenil is 2.7 (Tomlin, 1997).

In laboratory experiments with eight different soil types, the half-life of dichlobenil varied between 1.5 and 12 months. In soils dichlobenil is gradually broken down microbiologically to 2,6-dichlorobenzamide (BAM) (Figure 1), the rate depending on the soil type, water content and temperature (PEMP, 2000). Dichlobenil can also be rapidly hydrolysed to BAM by strong alkalis (Tomlin, 1997). The degradation of dichlobenil does not follow first-order degradation kinetics; with small amounts of dichlobenil in soils the degradation becomes very slow and dichlobenil residues can remain for a very long time in soils (Torstensson, 1984).

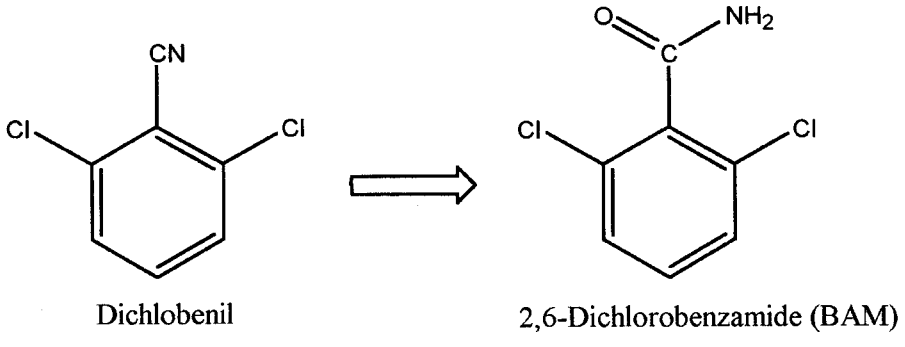


Figure 1. Hydrolysation or microbiologically degradation of diclobenil (forming BAM).

BAM, being more water-soluble than dichlobenil (log K_{ow} of BAM is 1.25) may leach in soil profiles, and has a very long half-life >660 days (Swedish National Chemicals Inspectorate, 2001).

Atrazine is relatively stabile in neutral soils, has a log K_{ow} of 2.5 (Tomlin, 1997), but is rapidly hydrolysed by first-order kinetics to hydroxyatrazine in strong acids and alkalis (Figure 2). Hydrolysis is affected by temperature, pH, organic matter content and soil moisture content. The rate increases as soil pH decreases and soil organic carbon increases and a wet soil will slow down degradation (Mersie & Seybold, 1996).

Microbial degradation of atrazine results in two metabolites (Figure 2); the major metabolite is desetyl atrazine (DEA), and desisopropyl atrazine (DIA) is formed to a lesser extent (Mersie & Seybold, 1996).

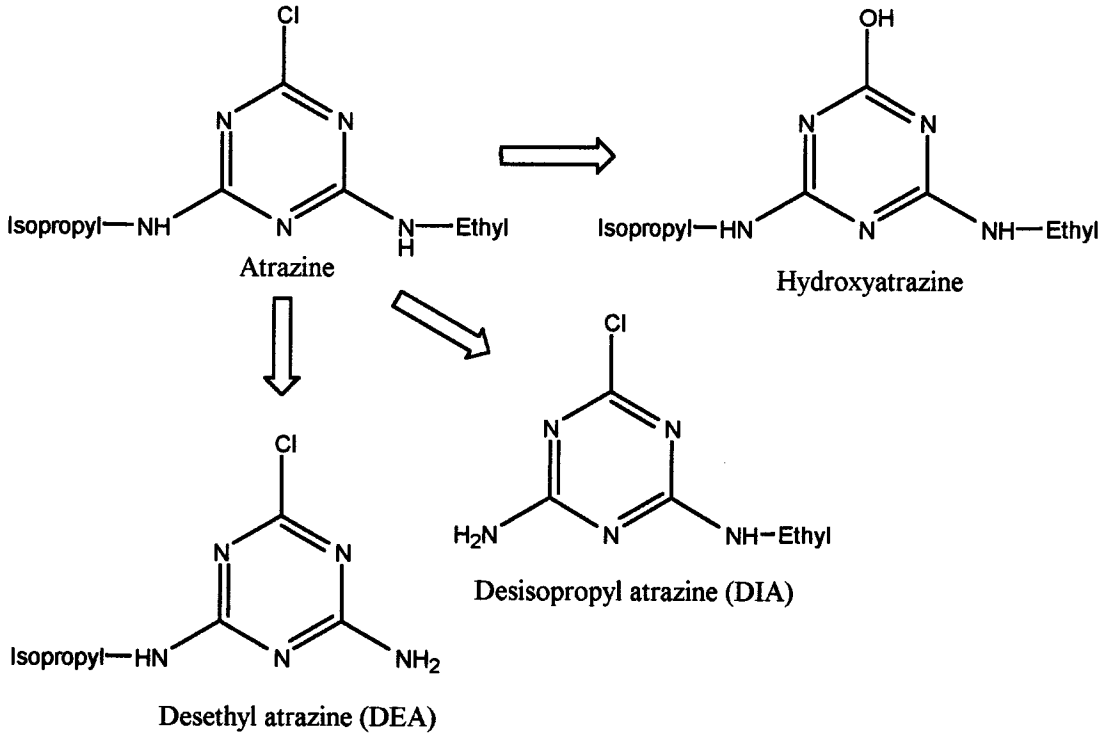


Figure 2. Hydrolysation of atrazine (forming hydroxyatrazine) or microbiologiclly degradation of atrazine (forming DIA and DEA).

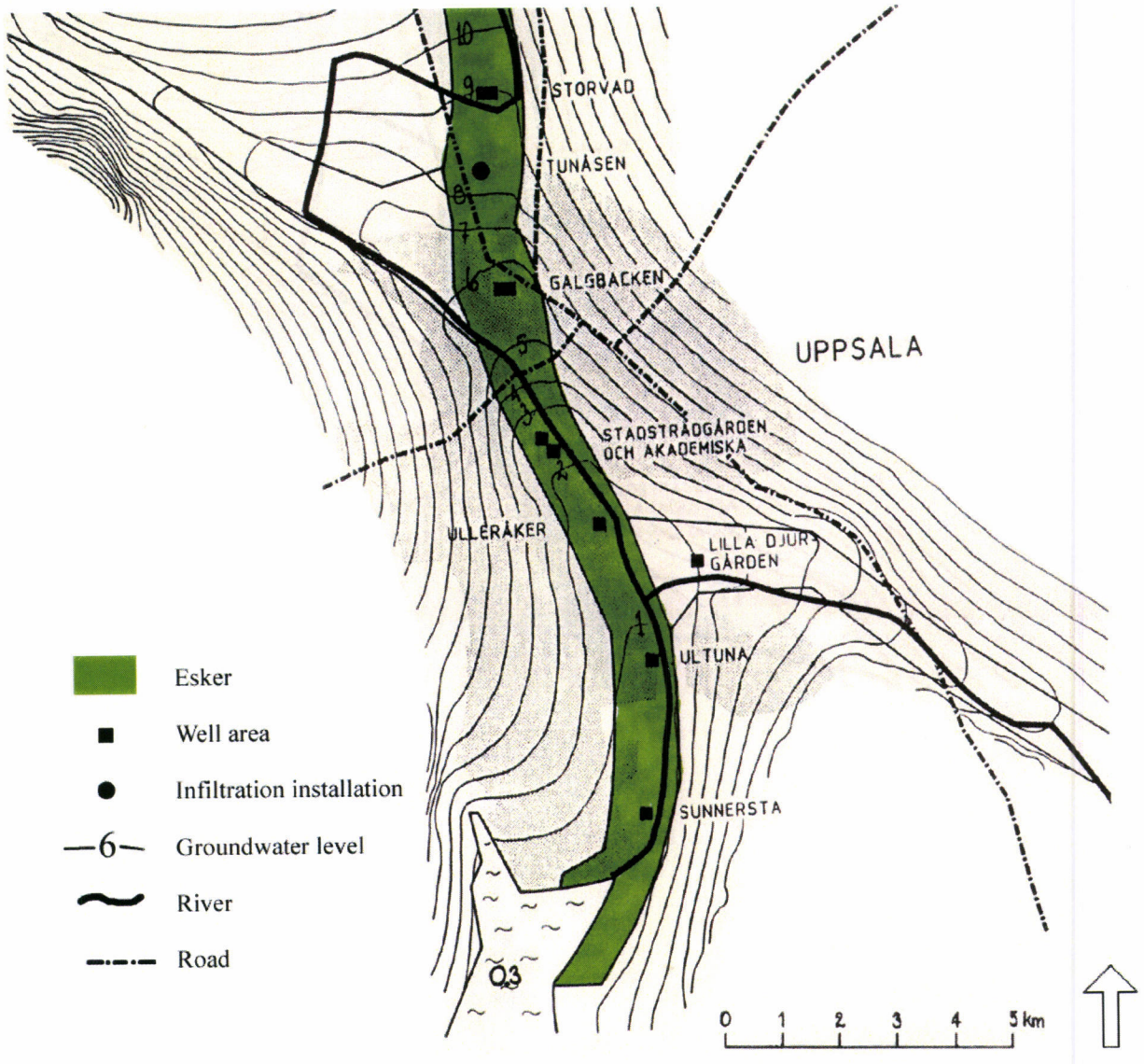
Hydroxyatrazine is usually formed to a greater extent than DEA and DIA, and hydroxyatrazine has a higher sorption and is more stable in soils than atrazine, DEA and DIA (Lerch, Thurman & Blanchard, 1999).

2.2 Formation and behavior of ground water around Uppsala

The esker that runs through Uppsala in a north to south direction is the most important ground water resource in Uppsala (Figure 3). The esker consists of sand, gravel and stone and is most extensive in the center of Uppala and to the south. In the centre of town the esker is partly over-stratified with thick clay layers. The clay layer is built up by glacial clay over-stratified by muddy post-glacial clay (Axelsson & Holmén, 1990).

The precipitation that falls on surface clay layers mostly run off to surface water but the precipitation that falls on the surface of the esker infiltrates. The local formation of ground water is built up by precipitation that infiltrates direct on the esker or in moraine around Uppsala and by artificial infiltration of surface water in the esker in order to ensure sufficient capacity for drinking water production. In general, the groundwater flow direction is from both the east and west towards the esker and in the esker, while in the esker the water is flowing to the south and eventually end ups in the lake Ekoln (Axelsson & Holmén, 1990).

The water supply of Uppsala is mainly based on groundwater uptake from the esker. There are eight large well areas in the central part of the esker where groundwater is taken out from the moraine layer (Figure 3) (Axelsson & Holmén, 1990).



Modified from Axelsson and Holmén, 1990.

Figure 3. The Uppsla esker trough Uppsala and the wells from north to south: Storvad, Galgbacken, Stadsträdgården, Uppsala university hospital, Ulleråker, Lilla Djurgården, Ultuna and Sunnersta.

2.3 Structure of this study

Soil cores were collected where Wallman found the highest levels of pesticides. Two soil cores at each location to a depth of 5 and 4 m respectively were collected at Uppsala university hospital and Eklundshof. The soil samples were analysed for water content, organic matter, pH and pesticide concentration.

Physical and chemical data from the soil cores from Uppsala university hospital were used in a mathematical program called MACRO to simulate the movement and levels of pesticides and to compare with the analysed soil samples.

3. Materials and Methods

3.1 Soil analysis

3.1.1 Sampling

Because the aim of the study was to examine if atrazine and dichlobenil residues are distributed and thus transported through the soil and if they possibly could have reached the subsoil water, two locations were chosen from Wallman (2000) (Appendix 2). Only one of Wallman´s locations (Eklundshof) was reported to contain atrazine and metabolites, and was selected (figure 4).

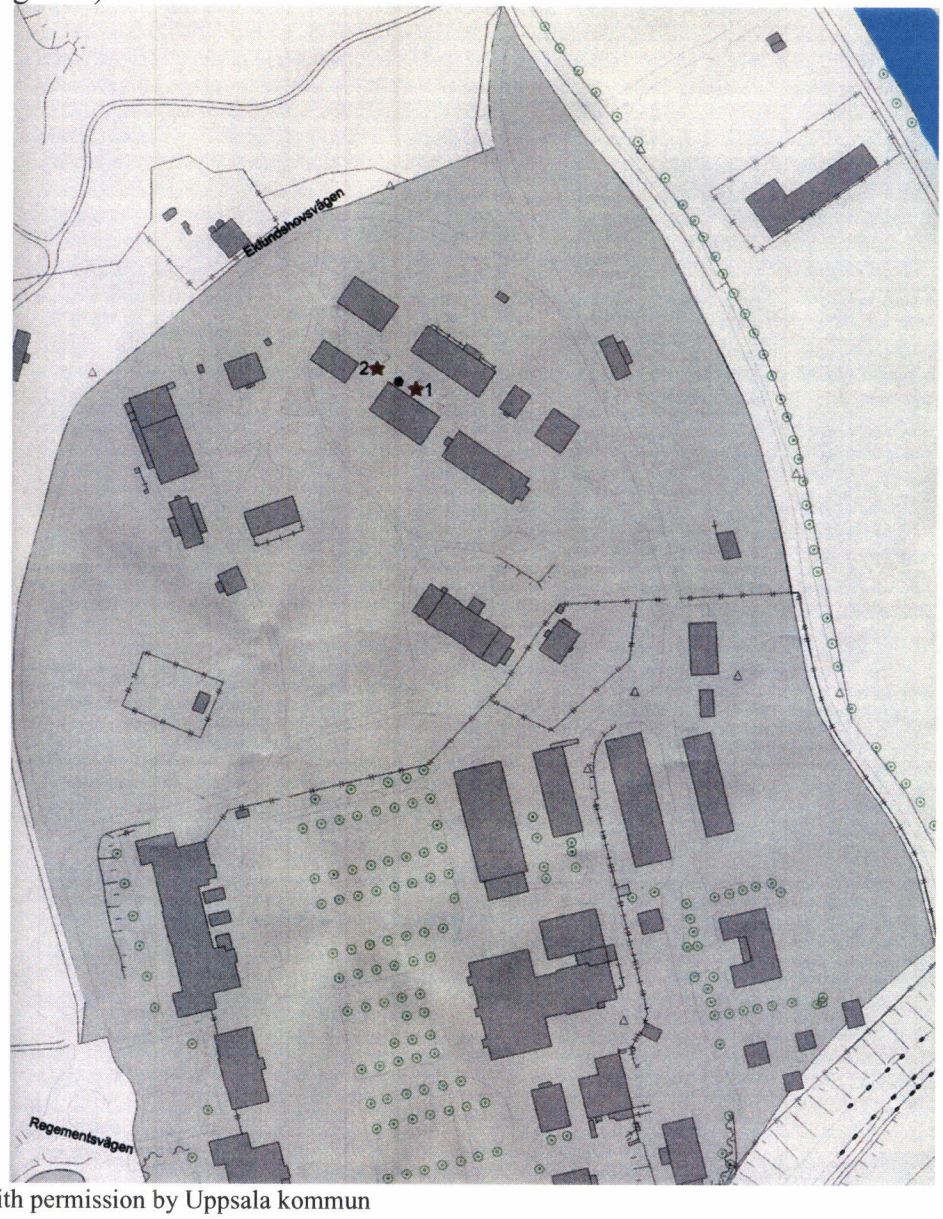


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Figure 4. Sampling locations of the soil cores from Eklundshof marked as stars and Wallman´s sampling location as a dot.

The other location selected (Uppsala university hospital) contained dichlobenil and BAM throughout the previously studied soil profile (15-100 cm) according to Wallman (2000) (figure 5).

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Figure 5. Sampling locations of the soil cores from Uppsala university hospital marked as stars and Wallman´s sampling location as a dot.

The plan was to take out three soil cores at each location (in order to account for the variability) to a depth of 5 m and to collect three continuous soil samples for every meter. It was not possible to take out a larger number of samples per meter because of restrictions with regard to the lowest amount of soil possible to analyse for the analytes of interest. The Geological Survey of Sweden carried out the coring using a derrick (∅=7 cm). The samples were collected on 2000-10-05. For every meter the drill was taken up and three soil core samples were collected and packed in plastic bags. Each soil sample weighed between 0.5 and 2.5 kg before drying. Because of cables and filling material in the ground it was impossible to find spots to drill three soil cores at each location, so only two soil cores could be drilled at each site. The soil cores were not taken out at exactly the same spots as in Wallman (2000)

because of cables, filling material and shrubs, but the samples were taken out only a few meters from the exact spots sampled in the earlier work. Furthermore, it was only possible to drill to a depth of 4 meter at Eklundshof because of the sorted material in the esker which become coarser with depth, and deeper than 4 m the texture was too coarse to be collected by the drill. The soil samples were stored at –18 °C until analysis.

3.1.2 Classification, SOM, water content and pH

The samples from Uppsala university hospital were very clayey (glacial clay over-stratified by post-glacial clay) and a simplified field method was used to decide the clay content for the soil samples. According to the method, a piece of soil with the right water content (not to sticky) is rolled between the palm of one hand and a piece of wood and the size of the resulting rolls is determined by the clay content (Linde, 1991). The clay pieces were rolled to 1 mm or less in diameter, which correspond to 40% or more of clay. In the soil samples between 3-5 m there were thin (1-2 cm) horizontal layers of pure sand, but the sand content was very low compared to the clay content in these soil samples. The topsoil samples were not possible to roll at all because of coarser material such as wood sticks and bark pieces.

Another field method was also used to decide the clay content for the samples of Uppsala university hospital. A piece of soil is rolled to the thickness and length of a pen and then bent to a circle (Linde, 1991). It was possible to bend the soil samples without any cracks or very small (except from the topsoil samples) which, in agreement with the method described above, indicates a clay content of 40 % or more in the soil samples.

Another method, where soil is poured through sieves with different wire netting size and then shaken for 15 minutes, was used to determine the soil texture for the soil samples of Eklundshof. This material is coarser compared to the samples of Uppsala university hospital, and it is sorted because it is a part of the esker. Soil samples were chosen from both profiles (four from each profile), they were weighed and dried at 105 °C for 24 hours, after that they were weighed again and passed through sieves with different sizes corresponding to the grain classification of the SGFs laboratory committee (Almén & Talme, 1975). After that each fraction was weighted and the distribution was calculated as a percentage. From this information the soil type could be determined (Almén & Talme, 1975).

To determine water content and LOI (loss of ignition) a sub sample (about 10 g) was taken out from each soil sample and put into crucibles. The samples were dried at 105 °C for 24 hours. All samples were weighed before and after the drying. The water content was calculated according to:

$$\text{Water content (\%)} = [(m_{\text{before drying}} - m_{\text{after drying}})/m_{\text{before drying}}] * 100 \qquad (m=\text{mass})$$

The dried soil was put in an oven again and the samples were combusted using a temperature program with a maximum temperature of 550 °C. All samples were weighted before and after the combustion. The LOI was calculated according to:

$$\text{LOI (\%)} = [(m_{\text{before ashing}} - m_{\text{after ashing}})/m_{\text{before ashing}}] * 100$$

To determine pH, 15 g of the soil samples were mixed with 75 ml of distillated water by rotation for 2 hours in a plastic cup. The soil samples were left to sediment for 24 hours and

after that water from each sample was collected in small bottles to make possible determination of pH (Meterlab PHM model 210 pH-meter).

3.1.3 Pesticide analysis

3.1.3.1 Preparation of soil samples and recovery tests

Two archived dried soils with no contamination of pesticides were used for the recovery tests. The soil samples were collected in the late 70's by the Swedish county agencies in a study of topsoil from the most important cultivated areas within Sweden (Ståhlberg, 1980). The archived soils were selected on the basis of soil texture to fit the soil samples of Uppsala university hospital and Eklundshof. Soil number 11 was collected in Norrbotten and had a clay content of 5-15 % and pH 6.4 (corresponding to the soil samples from Eklundshof) and soil number 28 was collected in Östergötland and had a clay content of 25-40 % and pH 7.6 (corresponding to the soil samples from Uppsala university hospital) (Ståhlberg, 1980).

A standard solution was prepared for the recovery tests. The relative concentration of the pesticides in the standard solution were: atrazine 2.45 mg/ml, desetyl atrazine 5.73 mg/ml, desisopropyl atrazine 16.83 mg/ml, dichlobenil 3.25 mg/ml and BAM 7.86 mg/ml.

To prepare the soil samples for the recovery test 16 g of archived soil were put in a mortar together with varying amount/concentrations of a diluted (ten times) standard solution. Ten to 200 µl were taken out from the diluted standard solution to the different recovery tests and 2 ml of Milli-Q water. The samples were covered with aluminum foil and left to stand for 3 hours and after that mixed with a spoon.

The recovery tests were carried out on three different occasions and with two different GC-MS instruments.

All soil samples from Eklundshof and Uppsala university hospital were shaken to get a homogeneous mix (the soil samples from Eklundhof had in some cases to be sieved, to a maximum particle size of 3 mm, before preparation). Fourteen grams were taken out from each soil sample and mixed with Hydromatix to remove water from the sample and after that ground in a mortar.

3.1.3.2 Extraction

An equivalent of 7 g of dry soil was taken out from each soil sample that had been mixed with Hydromatix, and the recovery test soil samples, and put into cellulose thimbles. An internal standard mixture was added to each sample (80 µl of a mixture containing etion 4.144 µg/ml and terbutylazine 1.905 µg/ml).

Acetone/dichlormetane 1:1 (v/v; 70 ml) was added and the extraction were carried out for three hours in a Soctex Avanti model 2050 Auto Extraction System. The cellulose thimble was immersed in the solvent for two hours and then rinsed with condensed solvent for one hour while held above the solvent surface.

The extract was filtered and dried with sodiumsulphate. Cyclohexane (5 ml) was added and the solvent was reduced by rotary evaporation to a volume of about 0.5 ml. The volume was

adjusted to 2 ml with cyclohexane/acetone 9:1. The extracts and standard solutions were studied by GC-MS.

3.2 GC-MS conditions

Most of the samples were analysed on an Agilent model 6890 gas chromatograph (GC) in conjunction with a model 5973 mass spectrometer (MS). The GC was equipped with a H5-5MS column with dimensions of 30 m x 0.25 mm i.d. and 0.25 µm film thickness, provided by Chrompack Sverige AB, Nacka, Sweden. The injector temperature was set by a temperature program to 65 °C for 0.02 min, increasing 700 °C/min to 280 °C were it was held for 1.5 min. The source temperature was 230 °C, the detector interface temperature was 300 °C, and the injection volume was 2 µl in the splitless mode. The oven temperature was set to 70 °C for 2 min, increasing 15 °C/min to 150 °C, then 3 °C/min to 200 °C and then 15 °C/min to 290 °C, where it was held for 2 min.

The rest of the samples were analysed on a Hewlett Packard model 5890 GC attached to a VG Masslab LTD model TRIO 1 MS. The GC was equipped with a CP-sil 5 CB column with dimensions of 60 m x 0.25 mm i.d. and 0.25 µm film thickness, provided by Chrompack Sverige AB, Nacka, Sweden. The injector temperature was 250 °C, the source temperature was 200 °C, the detector interface temperature was 290 °C and the injection volume was 2 µl in the splitless mode. Oven temperature was set to 90 °C for 1 min, increasing 30 °C/min to 210 °C and then 4 °C/min to 290 °C, where it was held for 10 min.

3.3 Simulations with MacroDB

Two simulation programs MacroDB (Jarvis, Hollis, Nicholls, Mayr & Evans, 1997) and MACRO (Jarvis, 1994) were used to model the movement of the different pesticides through a soil profile corresponding to that at the sampling location of Uppsala university hospital.

Physical and chemical data from the sampling location of Uppsala university hospital were used as input to the program MacroDB (the program can only simulate to a depth of 2.64 m) (table 1). Simulations were carried out in Macro over a period of 26 years. Only two kg per hectare of the active ingredient dichlobenil was simulated to be applied (to compensate for the high volatility) as granules on grass (7 cm high) on day 120 and 210 each year during the first 15 years (no application after that). The half-life was set to 30 days for dichlobenil in the topsoil and between 33-66 cm to 60 days and further down to 183 days. For BAM the half-life was set to 660, 330 and 220 days, in three different simulations.

Table 1. Parameters from the soil cores of Uppsala university hospital used in MacroDB simulation.

Properties	a -horizon	b -horizon	c-e -horizon
Thickness (cm)	33	33	198
Texture	silty clay	clay	clay
Bulk density (g/cm^3)	1.3	1.4	1.5
Organic carbon (%)	4.8	2.3	2.0
pH	7.5	7.5	8
Structure	moderate fine blocky	moderate fine blocky	strong fine blocky

4. Results

4.1 Soil analysis

4.1.1 Classification, SOM, water content and pH

The soils at both sampling locations were classified as mineral soils since the LOI was less than 20% (table 3). They were also classified as sorted material; glacial- and postglacial clay at Uppsala university hospital and Eklundshof is situated on the Uppsala esker.

The soil samples from Uppsala university hospital were classified as clay and the samples showed a strong and blocky structure. Soil samples from Eklundshof were classified as gravely sand in the upper part of the profiles and deeper down as sandy gravel (Jordartsbedömning, kompendium för kurs MV 4305, 2000), (table 2).

Table 2. The soil samples from Eklundshof shown as a percentage of the different texture classes. Characteristics are shown for one sample per meter.

Eklundshof	Depth (m)	Coarse gravel (≥6 mm) %	Fine gravel (6-2 mm) %	Coarse sand (2-0.6 mm) %	Medium sand (0.6-0.2 mm) %	Fine sand (0.2-0.06mm) %	Coarse silt (≤0.06) %
Profile 1	0.33-0.66	-	2.5	44.5	44	5	4
	1.33-1.66	-	9	43	29	11	8
	2.33-2.66	-	1.4	25.6	48	18	7
	3.33-3.66	27	25	14	17	12.5	4.5
Profile 1	0.00-0.33	0.5	10	35.5	41	7	6
	1.33-1.66	0.5	44.5	34.5	12	3	5.5
	2.33-2.66	8	44	29	12	3.5	3.5
	3.33-3.66	9	42	34	9	3	3

The topsoil samples from both Uppsala university hospital and Eklundshof differed from the subsoil samples. Because of human impact the topsoil samples had a different soil texture and also a higher LOI content.

Soil samples from Uppsala university hospital had pH values somewhere between fully 7 in the topsoil and fully 8 in the deeper subsoil samples, (table 3). Soil samples from Eklundshof had pH values between roughly 7 in the upper part of the profiles and 8 in the lower part, (table 3). This is rather neutral pH values, which is typical for this region.

The LOI was highest in the topsoil samples and immediately below, the highest value of Uppsala university hospital was 4.8% by dry weight and this location generally had higher LOI values compared to Eklundshof. Further down in the soil profiles the LOI values were lower, Uppsala university hospital about 2% and Eklundshof about 1%.

Table 3. The water content, LOI content and pH for the soil samples of Uppsala university hospital and Eklundshof.

Uppsala university hospital	Depth (m)	Water content (%)	LOI content (%)	pH
Profile 1	0.00-0.33	11.7	3.5	7.4
	0.33-0.66	10.0	2.6	7.5
	0.66-1.00	10.3	2.5	7.5
	1.00-1.33	11.8	2.5	7.3
	1.33-1.66	19.0	1.6	8.3
	1.66-2.00	20.1	1.9	8.4
	2.00-2.33	25.9	2.2	7.9
	2.33-2.66	29.8	2.8	8.2
	2.66-3.00	21.7	1.8	8.4
	3.00-3.33	23.2	2.1	8.5
	3.33-3.66	27.1	1.8	8.4
	3.66-4.00	21.7	1.5	8.3
	4.00-4.33	28.0	1.9	8.5
	4.33-4.66	27.6	1.8	8.4
	4.66-5.00	23.7	2.1	8.3
Profile 2	0.00-0.33	16.4	4.8	7.5
	0.33-0.66	17.8	2.3	7.4
	0.66-1.00	9.7	1.5	7.8
	1.00-1.33	27.9	2.0	8.3
	1.33-1.66	24.3	2.4	8.2
	1.66-2.00	21.1	1.6	8.2
	2.00-2.33	23.4	2.7	8.2
	2.33-2.66	23.6	1.7	8.6
	2.66-3.00	24.8	2.1	8.5
	3.00-3.33	23.5	1.5	8.5
	3.33-3.66	7.7	0.9	8.8
	3.66-4.00	24.2	2.3	8.4
	4.00-4.33	18.7	1.6	8.8
	4.33-4.66	21.4	1.9	8.6
	4.66-5.00	22.4	1.9	8.6

Eklundshof	Depth (m)	Water content (%)	LOI content (%)	pH
Profile 1	0.00-0.165	8.2	2.3	7.5
	0.165-0.33	5.2	1.4	7.4
	0.33-0.66	3.0	0.8	7.3
	0.66-1.00	3.0	0.7	7.2
	1.00-1.33	5.0	1.0	7.8
	1.33-1.66	4.4	0.8	7.7
	1.66-2.00	4.4	0.7	7.8
	2.00-2.33	5.0	0.8	7.9
	2.33-2.66	-	-	-
	2.66-3.00	8.0	0.8	7.6
	3.00-3.33	4.6	0.6	8.0
	3.33-3.66	-	-	-
	3.66-4.00	5.2	0.7	8.0
Profile 2	0.00-0.33	6.7	2.0	7.1
	0.33-0.66	3.7	1.2	6.9
	0.66-1.00	2.7	1.0	6.6
	1.00-1.33	3.1	1.0	6.9
	1.33-1.66	3.6	0.9	6.8
	1.66-2.00	3.4	1.0	6.9
	2.00-2.33	2.9	0.7	7.1
	2.33-2.66	-	-	-
	2.66-3.00	3.3	0.9	7.0
	3.00-3.33	-	-	-
	3.33-3.66	3.1	0.7	7.0
	3.66-4.00	-	-	-

- = Samples that were not tested.

4.1.2 Pesticide analysis

4.1.2.1 Recovery tests

The results from the GC model HP 5890 MS model TRIO 1 (the last three recovery tests in (table 4) show in general a lower recovery for the added pesticides compared to the results from the GC model Agilent 6890 MS model 5973. The recovery tests with the lower concentration of pesticides show in some case a recovery of more than 110%. Some individual recovery values were to low or to high to be acceptable (<75 % or >120%), but the mean and median values for all five pesticides are within the acceptable range and the total coefficient of variation for each analyte, considering all concentrations, is less than 10 %.

Table 4. Recovery test. Concentration of pesticides added to the archived soils, the extracted amount of pesticides and the calculated recovery of the pesticides.

Pesticide mixture* added (ml)	Substance	C_{added} (µg/kg)	$C_{extracted}$ (µg/kg)	Recovery Atrazine (%)	Recovery EIA (%)	Recovery DIA (%)	Recovery Dichlobenil (%)	Recovery BAM (%)
10	Atrazine	5.35	1.8	119				
	DEA	3.6	2.7		75			
	DIA	10.5	8.9			84		
	Dichlobenil	2.0	2.1				103	
	BAM	4.9	5.8					119
10	Atrazine	5.35	2.2	141				
	DEA	3.6	3.5		98			
	DIA	10.5	13.1			125		
	Dichlobenil	2.0	1.7				83	
	BAM	4.9	7.3					148
20	Atrazine	3.1	3.8	125				
	DEA	7.2	6.1		85			
	DIA	21.0	18.5			88		
	Dichlobenil	4.1	4.1				100	
	BAM	9.8	12.2					125
20	Atrazine	3.1	4.4	145				
	DEA	7.2	6.8		95			
	DIA	21.0	21.5			102		
	Dichlobenil	4.1	3.7				92	
	BAM	9.8	12.5					128
30	Atrazine	4.6	4.7	103				
	DEA	10.7	9.0		83			
	DIA	31.6	28.7			91		
	Dichlobenil	6.1	6.2				102	
	BAM	14.7	16.1					109
30	Atrazine	4.6	5.0	109				
	DEA	10.7	10.1		94			
	DIA	31.6	31.2			99		
	Dichlobenil	6.1	5.7				94	
	BAM	14.7	16.8					114
50	Atrazine	7.6	7.7	101				
	DEA	17.9	15.0		84			
	DIA	52.6	45.3			86		
	Dichlobenil	10.1	10.6				104	
	BAM	24.6	26.6					108
50	Atrazine	7.6	7.2	94				
	DEA	17.9	12.5		70			
	DIA	52.6	36.8			70		
	Dichlobenil	10.1	9.4				93	
	BAM	24.6	25.6					104
100	Atrazine	15.3	14.0	91				
	DEA	35.8	29.9		84			
	DIA	105.2	85.2			81		
	Dichlobenil	20.3	18.1				89	
	BAM	49.1	46.4					94

Table 4. (continued)

Pesticide mixture* added (ml)	Substance	C_{added} (µg/kg)	$C_{extracted}$ (µg/kg)	Recovery Atrazine (%)	Recovery EIA (%)	Recovery DIA (%)	Recovery Dichlobenil (%)	Recovery BAM (%)
100	Atrazine	15.3	14.6	95				
	DEA	35.8	29.0		81			
	DIA	105.2	87.2			83		
	Dichlobenil	20.3	17.5				86	
	BAM	49.1	48.4					98
100	Atrazine	15.3	15.6	102				
	DEA	35.8	30.2		84			
	DIA	105.2	92.1			88		
	Dichlobenil	20.3	19.4				95	
	BAM	49.1	51.4					105
50**	Atrazine	7.6	6.3	82				
	DEA	17.9	14.3		80			
	DIA	52.6	38.4			73		
	Dichlobenil	10.1	4.8				47	
	BAM	24.6	19.7					80
100**	Atrazine	15.3	12.6	83				
	DEA	35.8	26.6		74			
	DIA	105.2	71.7			68		
	Dichlobenil	20.3	15.3				75	
	BAM	49.1	35.6					72
200**	Atrazine	30.6	24.4	80				
	DEA	71.6	49.4		69			
	DIA	210.4	131.7			69		
	Dichlobenil	40.6	20.6				51	
	BAM	98.3	66.8					68
Mean				104	82	85	86	104
Median				101	82	85	92	106
Standard deviation				8.6	4.6	1.6	3.8	7
Cofficient of variation				8.3	5.9	1.9	4.4	6.7

* A standard solution according to 3.1.3.1

** Analysed ont he GC model HP 5890 MS model TRIO 1.

4.1.2.2 Soil samples

Only BAM and dichlobenil were traced in the soil samples from Uppsala university hospital and no pesticides at all were traced at Eklundshof (table 5). Dichlobenil was only found in one sample, the topsoil sample of profile 1. The highest concentration of BAM at each spot was found in the topsoil layer and the concentration tended to decrease with depth (Figure 6).

Table 5. Concentration of BAM and dichlobenil found in the soil samples at the two locations of Uppsala university hospital.

Location 1			Location 2		
Depth (m)	BAM (µg/kg d.w.)	Dichlobenil (µg/kg d.w.)	Depth (m)	BAM (µg/kg d.w.)	Dichlobenil (µg/kg d.w.)
0.00-0.33	28.3	5.4	0.00-0.33	13.1	-
0.33-0.66	23.3	-	0.33-0.66	2.5	-
0.66-1.00	25.1	-	0.66-1.00	7.6	-
1.00-1.33	10.0	-	1.00-1.33	3.5	-
1.33-1.66	4.8	-	1.33-1.66	11.8	-
1.66-2.00	12.8	-	1.66-2.00	6.5	-
2.00-2.33	12.4	-	2.00-2.33	15.2	-
2.33-2.66	22.4	-	2.33-2.66	4.7	-
2.66-3.00	14.0	-	2.66-3.00	5.0	-
3.00-3.33	9.4	-	3.00-3.33	6.3	-
3.33-3.66	10.3	-	3.33-3.66	3.1	-
3.66-4.00	5.8	-	3.66-4.00	4.6	-
4.00-4.33	3.6	-	4.00-4.33	2.3	-
4.33-4.66	2.6	-	4.33-4.66	0.9	-
4.66-5.00	7.9	-	4.66-5.00	1.8	-

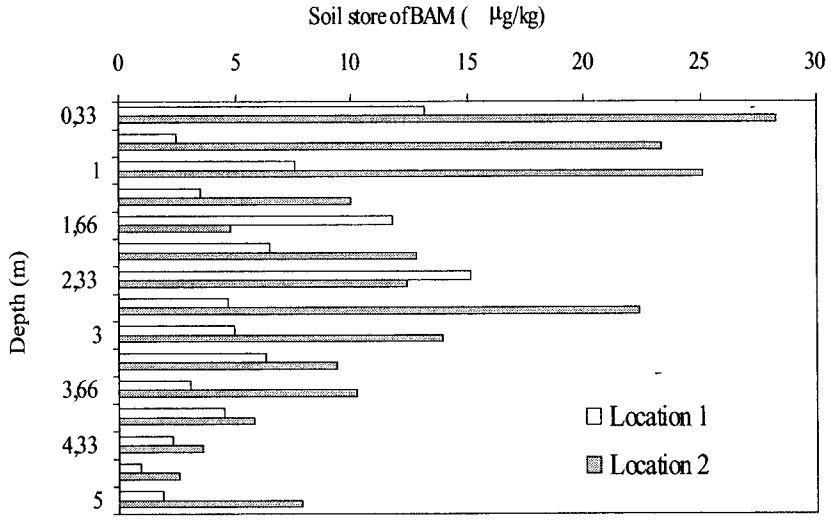

Figure 6. Concentration of BAM found in the soil samples at the two locations of Uppsala university hospital.

4.2 Simulations with MACRO

The simulation of dichlobenil gave the result that there were no pesticide residues left at all in the soil profile in the year 2000; in the simulations, the last residues disappeared in 1992/1993 (Appendix 3). The first simulation of BAM (with half-life 660 days) showed the highest pesticide residues of BAM, with the highest values ranging between 700-800 µg/kg d.w. at a depth of 1.3-2.0 m and almost no pesticide residues between 0-0.33 m depth in the year 2000 (Appendix 4). The second simulation (half-life 330 days) showed the highest value, around

225 µg/kg d.w. at a depth of 1.3-1.75 m and there were practically no pesticide residues left between 0-0.65 m depth in the year 2000 (Appendix 5). The third simulation (half-life 220 days) hardly showed any pesticide residues left between 0-0.65 m depth and the highest value was 70 µg/kg d.w. at a depth of 1.3-1.55 m depth in the year 2000 (Appendix 6).

5. Discussion

5.1 Soil analysis

It is obvious that pesticides have been used at Uppsala university hospital, and according to Wallman (2000) Casoron was used for weed control. The results from the soil samples support this as Casoron only contain dichlobenil and no atrazine.

BAM was found down to 5 m depth in clay at Uppsala university hospital, which supports the assumption that BAM can leach down to the groundwater. Why there were no traces of pesticides in the soil samples from Eklundshof might be explained by the soil texture, which provide a faster transport through the soil profile. Due to coarse texture and low water-holding capacity, and low sorption capacity pesticides are probably transported rather fast through the soil profile.

The result of the soil samples from Uppsala university hospital show lower concentrations compared to Wallman (2000), one soil sample of dichlobenil from the same area is one order of magnitude higher in Wallman's study than in the present one. The differences in the results between Wallman and this report might be explained by the fact that these soil samples were taken a few meters from the exact locations of Wallman and this indicates that the distribution of the pesticides in soils may be very variable.

5.2 Comparison between MACRO simulations and results from soil analysis

The differences between the MACRO simulations and the results from the soil profiles from Uppsala university hospital can be explained in several ways. Firstly, the actual supply of Casoron is very difficult to estimate because no one really knows the timings and the amounts of pesticide used. Furthermore the half-life is of great importance and the exact value depends on factors such as the soil type, organic matter content and temperature. Erroneous values here have large consequences for the Macro simulation results.

Whether the half-life of dichlobenil is based on laboratory- or field experiments is of great importance, for instance in a field experiment the volatilization is included but in a laboratory experiment it is not. MACRO is designed to use half-lives from laboratory experiments and can compensate for field temperature and moisture conditions, but the program can not account for volatilization of pesticides. In order to compensate for this the supply of the active ingredient dichlobenil was reduced which also can have considerable impact on the results.

Further, the program can only calculate with first order reaction kinetics, but the degradation of dichlobenil does not follow this assumption which gives a different degradation pattern in the soil profile compared to the soil profiles of Uppsala university hospital. The degradation rate constant of dichlobenil becomes smaller when the concentration of dichlobenil decreases.

In the MACRO simulations the greater part of the pesticide concentration is left at a depth of 1.3-1.5 m and no pesticides remain in the topsoil. The soil profiles from Uppsala university hospital are more difficult to interpret, but the BAM residues remaining in the topsoil do indicate a departure from first-order kinetics.

However, what does seem clear is that the initial estimate of half-life for BAM (660 days) is not appropriate for the Uppsala university hospital site, and that a much smaller half-life (220 days) was needed to give predictions of residues of the right order of magnitude.

Finally MACRO can only simulate to a depth of 2.64 m and is not exactly comparable with the soil samples from Uppsala university hospital taken to a depth of 5 m.

5.3 Conclusion

The detection of pesticide residues (BAM) down to 5 m depth at Uppsala university hospital suggests the possibility that the current groundwater contamination is a result of leakage from old deposits presently subject to transport through the soil.

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Appendix 1

Pesticide residues in the Uppsala ground water 1997-1999

The locations of the pumping stations are denoted in Figure 3

Location (from north to south)	Date	BAM (µg/l)	Atrazine (µg/l)	DEA (µg/l)	DIA (µg/l)
Galgbacken	990322*	n.d.	n.d.	n.d.	n.d.
	990417*	-	n.d.	n.d.	n.d.
Galgbacken	980115	n.d.	n.d.	n.d.	
Storvad	990322*	n.d.	0.01	n.d.	n.d.
	990427*	n.d.	0.01	n.d.	n.d.
Storvad	980115	n.d.	n.d.	n.d.	-
Uppsala univ. hospital 1	981028	n.d.	n.d.	n.d.	-
	990428*	-	0.08	0.09	0.02
	990531*	0.05	0.05	0.03	n.d.
	990324*	0.05	0.05	0.04	0.02
Uppsala univ. hospital 2	981028	0.06	0.05	n.d.	-
Uppsala univ. hospital (drinking water)	971230	n.d.	0.06	n.d.	-
Stadsträdgården S1	981028	n.d.	n.d.	n.d.	-
	980403	n.d.	n.d.	n.d.	-
Stadsträdgården S2	981028	n.d.	n.d.	n.d.	-
	980403	n.d.	n.d.	n.d.	-
Stadsträdgården S3	981028	0.01	0.05	n.d.	-
	980403	0.08	0.05	n.d.	-
	980114	n.d.	n.d.	n.d.	-
Stadsträdgården K1	990322*	0.03	0.03	0.02	n.d.
	990427*	n.d.	0.03	n.d.	n.d.
	990531*	nd	0.03	0.03	n.d.
Stadsträdgården K1	981028	0.08	n.d.	n.d.	-
	980403	n.d.	n.d.	n.d.	-
Ulleråker (drinking water)	971230	0.43	0.067	0.067	-
Ulleråker (drinking water)	981028	0.4	n.d.	n.d.	-
	990324*	0.5	0.07	0.07	0.01
	990428*	0.2	0.02	0.02	0.02
	990531*	0.3	0.05	0.05	n.d.
Ultuna (drinking water)	971230	0.24	n.d.	n.d.	-
Ultuna -alternative reservoir	971210*	0.2	0.01	0.02	n.d.
	981028	0.19	n.d.	n.d.	-
	990330*	0.2	0.02	0.02	0.02
	990429*	0.3	0.02	0.03	0.01
	990526*	0.2	0.02	0.02	n.d.
Ultuna -auxiliary reservoir	990330*	2.0	n.d.	0.02	n.d.
	990429*	2.0	0.009	0.02	0.03
	990526*	1.0	n.d.	n.d.	n.d.
Ultuna -Bigården	990330*	0.4	0.2	0.4	0.03
	990429*	0.3	0.2	0.4	0.02
	990531*	0.1	0.1	0.2	0.02

Appendix 1 continued

Location (from north to south)	Date	BAM (µg/l)	Atrazine (µg/l)	DEA (µg/l)	DIA (µg/l)
Ultuna main reservoir	971210*	0.2	0.02	0.03	n.d.
	971230	0.24	n.d.	n.d.	-
	980309*	0.3	0.1	0.1	n.d.
	980629*	0.4	0.06	0.06	n.d.
	981028	0.25	n.d.	n.d.	
	990330*	0.3	0.02	0.03	n.d.
	990429*	0.3	0.02	0.03	n.d.
	990526*	0.2	0.02	0.03	n.d.

n.d. = not detected
- = not analysed

The samples marked with an asterix (*) have been analysed by Miljölaboratoriet in Nyköping (data made available by Miljökontoret, Uppsala). The remaining samples have been analysed by the Department of Environmental Assessment /unpublished data).

The detection limit for BAM and atrazine were 0.05 µg/l at Miljölaboratoriet in Nyköping and 0.05 µg/l and 0.02 µg/l respectively at Department of Environmental Assessment.

Appendix 2

Locations selected from Wallman (2000).

Concentrations of dichlobenil, dichlorobenzamide (BAM), atrazine, desisopropyl atrazine (DEA) and desethyl atrazine (DIA) found in the soil samples from Uppsala university hospital (location C) and Eklundshof (location H). Data from Wallman (2000).

Location	Sample depth (cm)	$C_{dichlobenil}$ (µg/kg d.w.)	C_{BAM} (µg/kg d.w.)	$C_{atrazine}$ (µg/kg d.w.)	C_{DEA} (µg/kg d.w.)	C_{DIA} (µg/kg d.w.)
C:7	15-30	5.7	14	n.d.	n.d.	n.d.
C:7	85-100	3.9	199	n.d.	n.d.	n.d.
H:1	20-30	n.d.	35	80	81	81

n.d. = not detected

Appendix 3

The distribution of dichlobenil trough a soil profile over 26 years, as simulated by the Macro model using a half-life of 30 days in topsoil, 60 days at 33-66 cm depth, and 183 days at greater depth. For other parameters, se Section 3.3.

Year	Accumulated leakage (mg/m^2)	0-8 cm depth (µg/kg)	8-16 cm depth (µg/kg)	16-24 cm depth (µg/kg)	24-33 cm depth (µg/kg)	33-49 cm depth (µg/kg)	49-66 cm depth (µg/kg)	66-88 cm depth (µg/kg)	88-110 cm depth (µg/kg)	110-132 cm depth (µg/kg)	132-154 cm depth (µg/kg)	154-176 cm depth (µg/kg)	176-198 cm depth (µg/kg)	198-220 cm depth (µg/kg)	220-242 cm depth (µg/kg)	242-264 cm depth (µg/kg)
1975-12-17	0.0	701.0	61.6	10.1	1.9	0.6	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1976-12-17	0.0	922.7	98.0	16.9	3.5	1.1	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1977-12-17	0.0	784.0	101.3	21.6	7.4	2.1	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1978-12-17	0.0	878.9	105.2	25.7	7.9	1.7	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1979-12-17	0.0	853.7	111.3	36.3	17.6	6.0	2.0	0.5	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1980-12-17	0.0	836.5	114.8	34.4	12.6	4.9	1.5	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1981-12-17	0.0	718.9	93.9	24.9	8.0	3.3	0.9	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1982-12-17	0.0	846.7	112.1	31.8	12.3	4.3	1.1	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1983-12-17	0.0	808.9	82.1	16.2	3.5	2.4	0.7	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1984-12-17	0.0	936.9	90.9	18.3	4.8	2.0	0.6	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1985-12-17	0.0	927.4	85.4	10.5	1.2	0.8	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1986-12-17	0.0	857.6	68.0	6.0	0.5	0.3	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1987-12-17	0.0	950.0	56.6	8.7	4.2	2.2	0.6	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1988-12-17	0.0	756.5	127.6	37.4	14.4	4.6	1.3	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1989-12-17	0.0	714.6	67.4	12.6	2.9	1.8	0.5	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1990-12-17	0.0	68.2	19.0	5.4	1.9	1.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1991-12-17	0.0	4.9	3.1	1.3	0.5	0.4	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1992-12-17	0.0	0.4	0.4	0.2	0.1	0.2	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1993-12-17	0.0	0.0	0.0	0.0	0.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1994-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1995-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1996-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1997-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1998-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1999-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1920-12-17	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

Appendix 4

The distribution of 2,6-dichlorobenzamide (BAM) trough a soil profile over 26 years, as simulated by the Macro model using a half-life of 660 days. For other parameters, se Section 3.3.

Year	Accumulated leakage (mg/m^2)	0-8 cm depth (µg/kg)	8-16 cm depth (µg/kg)	16-24 cm depth (µg/kg)	24-33 cm depth (µg/kg)	33-49 cm depth (µg/kg)	49-66 cm depth (µg/kg)	66-88 cm depth (µg/kg)	88-110 cm depth (µg/kg)	110-132 cm depth (µg/kg)	132-154 cm depth (µg/kg)	154-176 cm depth (µg/kg)	176-198 cm depth (µg/kg)	198-220 cm depth (µg/kg)	220-242 cm depth (µg/kg)	242-264 cm depth (µg/kg)
1975-12-17	0.0	1813.3	717.0	258.7	70.5	12.8	4.7	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1976-12-17	0.0	2658.6	1717.8	909.7	411.0	89.0	30.1	5.7	1.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0
1977-12-17	0.0	2827.7	2797.6	1810.9	921.0	252.4	66.4	10.5	1.5	0.2	0.0	0.0	0.0	0.0	0.0	0.0
1978-12-17	0.0	3095.7	3331.9	2494.4	1494.5	430.3	139.0	26.9	3.0	0.3	0.0	0.0	0.0	0.0	0.0	0.0
1979-12-17	0.0	2659.2	3408.7	2996.0	2221.3	796.0	370.3	114.3	17.1	1.9	0.2	0.0	0.0	0.0	0.0	0.0
1980-12-17	0.0	2155.2	2914.5	2953.2	2672.0	1235.7	742.5	266.7	71.6	15.9	3.4	0.7	0.2	0.0	0.0	0.0
1981-12-17	0.0	2403.8	2742.7	2788.8	2699.3	1434.1	1052.2	454.9	154.9	44.6	12.3	3.2	0.8	0.2	0.0	0.0
1982-12-17	0.0	2232.1	3000.7	3005.4	2876.6	1603.4	1280.9	586.7	197.4	53.2	14.1	3.7	0.9	0.2	0.0	0.0
1983-12-17	0.0	3107.6	3066.7	2985.7	2845.6	1640.2	1396.9	696.9	279.9	86.3	24.3	6.6	1.7	0.4	0.1	0.0
1984-12-17	0.0	2974.1	3242.6	3051.6	2848.4	1688.5	1554.7	860.8	353.2	111.3	30.6	8.1	2.1	0.5	0.1	0.0
1985-12-17	0.0	3669.0	3716.5	3371.3	2952.8	1653.2	1513.0	927.1	405.8	125.1	34.9	9.4	2.5	0.6	0.1	0.0
1986-12-17	0.0	4346.1	4246.5	3604.1	2997.1	1697.4	1549.8	942.1	389.6	115.8	32.4	8.9	2.4	0.6	0.2	0.0
1987-12-17	0.0	3806.7	4387.1	3981.8	3232.2	1772.0	1697.6	1032.0	416.1	115.3	31.4	8.6	2.3	0.6	0.2	0.0
1988-12-17	0.0	1854.1	3400.4	3782.7	3585.3	2069.2	2034.2	1417.0	690.9	218.0	54.5	13.2	3.2	0.8	0.2	0.0
1989-12-17	0.0	3082.0	3397.0	3558.7	3481.2	2043.3	1933.0	1400.9	775.0	290.9	90.0	25.3	6.8	1.8	0.4	0.1
1990-12-17	0.0	1405.5	2839.3	3193.9	3274.1	2037.2	2127.0	1597.4	844.1	285.1	84.9	24.2	6.6	1.7	0.5	0.1
1991-12-17	0.0	250.2	1299.5	2061.4	2627.0	1867.2	2084.1	1761.0	1157.3	516.3	182.3	55.9	15.5	4.1	1.0	0.2
1992-12-17	0.1	76.5	598.1	1245.9	1877.6	1538.3	1904.0	1781.9	1315.1	693.6	290.1	103.3	32.8	9.6	2.6	0.6
1993-12-17	0.1	13.9	246.6	687.9	1195.9	1168.2	1704.8	1743.5	1424.1	837.3	385.2	144.3	46.7	13.9	3.9	0.9
1994-12-17	0.8	1.0	51.5	213.9	458.3	562.3	1091.6	1482.7	1537.4	1159.8	691.4	338.1	141.0	51.6	17.0	4.7
1995-12-17	3.3	0.1	10.8	64.1	175.5	225.3	568.3	1076.5	1389.7	1284.4	934.3	560.2	285.1	126.9	49.8	16.2
1996-12-17	9.1	0.0	2.5	25.9	86.5	127.2	347.1	831.8	1179.8	1178.3	956.7	655.1	385.2	199.2	92.2	34.6
1997-12-17	14.4	0.0	0.4	8.2	31.7	57.3	132.5	545.5	999.2	1137.8	1002.8	734.4	458.6	250.6	123.2	48.3
1998-12-17	27.8	0.0	0.0	1.3	5.8	23.6	12.0	250.3	671.0	961.2	1007.2	853.5	604.7	368.8	200.0	83.6
1999-12-17	54.4	0.0	0.0	0.4	0.1	14.9	1.3	132.1	461.7	749.5	878.9	835.3	665.4	457.7	282.9	134.9
1920-12-17	75.3	0.0	0.0	0.1	0.0	6.8	0.0	27.3	259.5	567.6	778.0	823.6	714.9	525.5	344.1	171.4

Appendix 5

The distribution of 2,6-dichlorobenzamide (BAM) trough a soil profile over 26 years, as simulated by the Macro model using a half-life of 330 days. For other parameters, se Section 3.3.

Year	Accumulated leakage (mg/m^2)	0-8 cm depth (µg/kg)	8-16 cm depth (µg/kg)	16-24 cm depth (µg/kg)	24-33 cm depth (µg/kg)	33-49 cm depth (µg/kg)	49-66 cm depth (µg/kg)	66-88 cm depth (µg/kg)	88-110 cm depth (µg/kg)	110-132 cm depth (µg/kg)	132-154 cm depth (µg/kg)	154-176 cm depth (µg/kg)	176-198 cm depth (µg/kg)	198-220 cm depth (µg/kg)	220-242 cm depth (µg/kg)	242-264 cm depth (µg/kg)
1975-12-17	0.0	1730.9	674.8	241.2	65.1	11.9	4.4	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1976-12-17	0.0	2498.7	1557.8	806.1	359.8	78.2	26.5	5.1	0.9	0.1	0.0	0.0	0.0	0.0	0.0	0.0
1977-12-17	0.0	2596.1	2448.2	1536.8	767.0	212.4	55.2	8.7	1.3	0.2	0.0	0.0	0.0	0.0	0.0	0.0
1978-12-17	0.0	2810.9	2830.4	2034.9	1184.0	340.7	109.4	21.3	2.4	0.3	0.0	0.0	0.0	0.0	0.0	0.0
1979-12-17	0.0	2429.3	2841.1	2377.7	1710.7	609.3	282.3	87.7	12.9	1.4	0.2	0.0	0.0	0.0	0.0	0.0
1980-12-17	0.0	1997.9	2431.0	2296.7	1971.2	898.8	534.4	191.6	51.0	11.3	2.4	0.5	0.1	0.0	0.0	0.0
1981-12-17	0.0	2223.4	2304.1	2143.7	1918.5	992.3	710.5	305.6	103.2	29.5	8.1	2.1	0.5	0.1	0.0	0.0
1982-12-17	0.0	2223.4	2304.1	2143.7	1918.5	992.3	710.5	305.6	103.2	29.5	8.1	2.1	0.5	0.1	0.0	0.0
1983-12-17	0.0	2852.2	2559.2	2263.1	1941.1	1055.0	857.0	419.4	165.0	50.2	14.0	3.8	1.0	0.2	0.1	0.0
1984-12-17	0.0	2723.4	2700.2	2302.8	1924.7	1065.1	914.9	492.0	196.3	60.7	16.4	4.3	1.1	0.3	0.1	0.0
1985-12-17	0.0	3341.9	3071.6	2507.3	1935.3	998.8	846.6	502.7	212.8	64.0	17.6	4.7	1.2	0.3	0.1	0.0
1986-12-17	0.0	3903.0	3426.2	2599.8	1890.9	977.0	817.4	481.7	192.0	55.4	15.2	4.1	1.1	0.3	0.1	0.0
1987-12-17	0.0	3416.0	3450.2	2810.0	2041.8	1018.0	864.2	498.3	192.2	51.5	13.7	3.7	1.0	0.3	0.1	0.0
1988-12-17	0.0	1703.5	2773.9	2815.0	2446.0	1276.4	1094.0	682.8	311.4	93.9	22.6	5.3	1.3	0.3	0.1	0.0
1989-12-17	0.0	2827.1	2731.2	2553.5	2260.1	1214.9	999.6	646.8	332.0	119.0	35.5	9.7	2.5	0.7	0.2	0.0
1990-12-17	0.0	1198.1	2216.6	2269.1	2109.5	1202.9	1092.2	719.9	346.2	109.7	31.4	8.7	2.3	0.6	0.2	0.0
1991-12-17	0.0	196.3	952.9	1425.8	1685.2	1112.2	1102.0	805.0	475.7	197.9	66.5	19.6	5.3	1.4	0.3	0.1
1992-12-17	0.0	54.0	401.4	801.4	1141.7	892.5	1001.8	812.8	532.1	259.8	102.8	35.1	10.8	3.1	0.8	0.2
1993-12-17	0.0	8.9	150.3	406.4	677.1	646.9	870.8	781.8	565.9	304.8	131.6	46.8	14.5	4.2	1.1	0.3
1994-12-17	0.2	0.6	28.7	116.5	240.2	297.5	554.6	676.8	628.1	433.2	240.8	111.4	44.4	15.7	5.0	1.3
1995-12-17	0.9	0.0	5.5	32.2	84.2	111.4	281.1	487.3	568.6	481.2	325.4	183.9	89.2	38.1	14.4	4.6
1996-12-17	2.6	0.0	1.2	11.8	37.4	57.5	161.6	359.0	461.5	423.6	320.9	207.6	116.4	57.9	25.9	9.5
1997-12-17	4.0	0.0	0.2	3.4	12.3	22.9	59.4	227.7	377.1	393.3	322.0	221.9	131.8	69.1	32.7	12.5
1998-12-17	7.4	0.0	0.0	0.5	2.1	8.1	5.3	102.7	247.5	325.5	316.7	251.6	168.6	98.1	51.1	20.7
1999-12-17	14.0	0.0	0.0	0.2	0.1	4.8	0.4	52.5	163.7	244.3	266.6	237.8	179.2	117.6	69.7	32.1
1920-12-17	18.9	0.0	0.0	0.0	0.0	2.0	0.0	11.6	89.5	178.3	226.8	224.7	184.0	128.6	80.5	38.7

Appendix 6

The distribution of 2,6-dichlorobenzamide (BAM) trough a soil profile over 26 years, as simulated by the Macro model using a half-life of 220 days. For other parameters, se Section 3.3.

Year	Accumulated leakage (mg/m^2)	0-8 cm depth (µg/kg)	8-16 cm depth (µg/kg)	16-24 cm depth (µg/kg)	24-33 cm depth (µg/kg)	33-49 cm depth (µg/kg)	49-66 cm depth (µg/kg)	66-88 cm depth (µg/kg)	88-110 cm depth (µg/kg)	110-132 cm depth (µg/kg)	132-154 cm depth (µg/kg)	154-176 cm depth (µg/kg)	176-198 cm depth (µg/kg)	198-220 cm depth (µg/kg)	220-242 cm depth (µg/kg)	242-264 cm depth (µg/kg)
1975-12-17	0.0	1652.5	635.0	224.8	60.1	11.0	4.1	0.2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1976-12-17	0.0	2351.4	1414.8	714.9	315.1	68.7	23.3	4.5	0.8	0.1	0.0	0.0	0.0	0.0	0.0	0.0
1977-12-17	0.0	2390.2	2150.6	1308.7	640.8	179.4	45.9	7.3	1.0	0.1	0.0	0.0	0.0	0.0	0.0	0.0
1978-12-17	0.0	2564.8	2421.2	1671.5	943.9	271.3	86.5	17.0	1.8	0.2	0.0	0.0	0.0	0.0	0.0	0.0
1979-12-17	0.0	2232.4	2393.0	1908.0	1332.8	471.6	217.5	68.0	9.8	1.1	0.1	0.0	0.0	0.0	0.0	0.0
1980-12-17	0.0	1859.5	2057.3	1816.0	1478.0	663.8	390.1	139.3	36.7	8.1	1.7	0.4	0.1	0.0	0.0	0.0
1981-12-17	0.0	2063.8	1963.8	1682.3	1394.2	700.8	488.3	208.3	69.6	19.8	5.4	1.4	0.3	0.1	0.0	0.0
1982-12-17	0.0	1907.7	2162.7	1835.6	1461.5	741.5	547.9	240.6	78.4	20.7	5.4	1.4	0.3	0.1	0.0	0.0
1983-12-17	0.0	2628.7	2164.7	1754.0	1365.4	700.2	540.6	257.8	98.8	29.6	8.2	2.2	0.6	0.1	0.0	0.0
1984-12-17	0.0	2505.7	2280.1	1778.2	1344.8	697.9	557.2	288.5	111.3	33.6	9.0	2.3	0.6	0.1	0.0	0.0
1985-12-17	0.0	3060.0	2574.7	1908.3	1311.0	627.0	491.4	280.2	114.1	33.4	9.0	2.3	0.6	0.2	0.0	0.0
1986-12-17	0.0	3527.2	2806.5	1920.4	1233.9	585.1	447.8	253.4	96.7	27.0	7.3	1.9	0.5	0.1	0.0	0.0
1987-12-17	0.0	3090.2	2759.6	2035.3	1344.7	616.8	462.4	248.5	90.8	23.4	6.1	1.6	0.4	0.1	0.0	0.0
1988-12-17	0.0	1572.9	2308.2	2157.6	1738.0	832.7	632.0	348.5	146.4	41.7	9.6	2.2	0.5	0.1	0.0	0.0
1989-12-17	0.0	2603.6	2242.0	1889.5	1525.5	761.6	555.4	318.4	149.5	50.6	14.5	3.8	1.0	0.2	0.1	0.0
1990-12-17	0.0	1026.4	1761.4	1661.5	1414.7	747.0	602.7	348.2	150.0	44.0	12.0	3.2	0.8	0.2	0.1	0.0
1991-12-17	0.0	154.9	708.3	1008.7	1117.3	691.8	622.1	398.3	210.3	80.7	25.5	7.2	1.9	0.5	0.1	0.0
1992-12-17	0.0	38.4	272.6	524.9	713.0	536.9	557.5	401.1	233.3	104.5	38.8	12.6	3.7	1.0	0.3	0.1
1993-12-17	0.0	5.7	92.6	243.7	391.5	368.9	466.6	377.8	244.7	120.2	48.2	16.1	4.8	1.3	0.3	0.1
1994-12-17	0.1	0.3	16.1	64.1	127.7	160.4	290.3	328.1	278.1	176.5	91.4	39.8	15.1	5.1	1.6	0.4
1995-12-17	0.3	0.0	2.8	16.4	40.8	55.9	141.1	230.8	249.6	196.1	123.8	65.9	30.3	12.4	4.5	1.4
1996-12-17	0.8	0.0	0.5	5.4	16.3	26.4	75.7	160.9	192.4	165.0	117.5	72.0	38.5	18.3	7.9	2.8
1997-12-17	1.2	0.0	0.1	1.5	4.7	9.4	26.2	97.3	150.3	146.5	112.6	73.4	41.5	20.8	9.5	3.5
1998-12-17	2.2	0.0	0.0	0.2	0.7	2.9	2.1	42.2	95.1	117.5	107.7	80.9	51.5	28.6	14.3	5.6
1999-12-17	3.9	0.0	0.0	0.1	0.0	1.6	0.1	20.5	59.9	84.3	87.0	73.6	52.8	33.1	18.8	8.4
1920-12-17	5.2	0.0	0.0	0.0	0.0	0.6	0.0	4.5	31.3	58.7	70.6	66.4	51.6	34.5	20.6	9.6