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## POLLUTION AT AND BELOW SITES USED FOR MIXING AND LOADING OF PESTICIDES

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Sites used for mixing and loading of pesticides in sprayers and for washing tractors and sprayers may be point sources of pesticides. Pollution may be caused by accidental spills during filling, disposal of excess spray solution, rinsing of sprayer and tractor or from leaking nozzles on the sprayer.

Ground water sampled 2–4 m below sites used for mixing and loading has been analysed for 23 or 46 different pesticides and metabolites in two Danish counties (Storstrøm and Bornholm). Further, the surface pollution at sites used for mixing, loading and rinsing was determined by elution with water of soil sampled in the top 10 cm.

In all ground water samples pesticide pollution was determined to be above the European drinking water level ( $0.1 \mu\text{g L}^{-1}$ ). The highest concentrations and most pesticides were found below loading and mixing sites at machine pools, where the highest concentrations were the phenoxyacid herbicides dichlorprop ( $750 \mu\text{g L}^{-1}$ ) and 2,4-D ( $800 \mu\text{g L}^{-1}$ ). The herbicides bentazone, mecoprop and dinoseb were also found in relatively high concentrations ( $5\text{--}60 \mu\text{g L}^{-1}$ ).

The surface soil sampled at the top 0–10 cm at sites used for loading and washing sprayers at six farms was eluted with water. These analyses also showed that many different pesticides and relatively high concentrations could be leached out from the soil. Twenty-four different pesticides and metabolites were found, and though most concentrations were below  $10 \mu\text{g L}^{-1}$  about 10% of the water samples contained more than  $50 \mu\text{g L}^{-1}$ .

The results demonstrate that sites used for mixing, loading and washing can be seriously contaminated with pesticides even in ground water 2–4 m below the sites. This implies that ground water, nearby wells and well borings are at risk of pollution and indicates the need for better farm practice.

**Keywords:** Pesticides; Subsurface; Mixing; Loading; Spillage; Waste

### INTRODUCTION

The risk of point source pollution with pesticides at sites used for mixing and loading is obvious but has not previously been documented. The risk comes from the cleaning of sprayers and tractors, spillage of concentrated pesticides or spray solutions and foaming, together with leaking nozzles. Sites for filling and loading in Denmark are

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often covered with gravel and sand where the ability to adsorb and degrade pesticides is low. Further they are often located near open wells and well borings in the farm area or on farm yards. In Denmark, there is a potential for about 45,000 such sites plus 250 machine pools.

Spillage of concentrated pesticides may happen during the filling of sprayers. It is not possible to estimate the frequency of this but the risk should not be overlooked. The dumping of remnant of spray liquid in the sprayer may be another important source of pollution. In most sprayers between 5 and 40 L of spray solution will be left in the tank when the sprayer is "emptied" (Gummer Andersen, Personal comm., 2000). The pesticide concentration in the spray solution will often be between 1 and 5 g L<sup>-1</sup> [1], thus between 5 and 200 g active ingredient can be dumped. Wastewater without further treatment may be led directly to streams, municipal sewage systems or storage tanks, which may be old and in poor condition leading to leakage of pesticide polluted water.

According to Ganzelmeier [2] between 0.1 and 1.2 g pesticide was washed off a sprayer which could cover 18 m<sup>2</sup>. Cooper and Taylor [3] showed that pesticide washed off could cover 50 m<sup>2</sup>. Based on a rate of 1 kg ha<sup>-1</sup>, this amount is about 5 g. According to Ganzelmeier [2], internal cleaning of the sprayer may give pesticide remnants of between 1 and 9 g. In several European countries focus has been put on problems in connection with the filling and rinsing of sprayers [2,4–14].

At a depth of 6–10 m below a site for mixing and loading in an orchard, 80 and 390 µg L<sup>-1</sup> of mecoprop and dichlorprop, respectively were found 5 years after termination of use of the site [15]. In accordance with this, heavy pollution with phenoxyacids was found below a site on a machine pool even though it had not been used for 15–20 years [16,17]. This paper describes the findings of pesticides in ground water below mixing and loading sites in two Danish counties. Further, the potential for pollution from surface soil at some sites is determined by elution of surface soil with water.

## EXPERIMENTAL

### County of Bornholm

In the county of Bornholm, the determination of pollution of ground water at a depth of 1.5–6 m was performed below sites for filling and loading and farmyards at 10 machine pools and 5 nurseries/plantations. Every possible step was taken to sample around and not directly at the potential source of pollution. Sampling of water was performed in 20 mm PVC installed as driven wells (Geo Probe). Three to five samples taken from driven wells around one "hot spot" were mixed before analysis for 46 pesticides and metabolites by the commercial laboratory (Alfred Jørgensen).

Water samples were collected 2–3 weeks after installation of driven wells (November 1999), and most samples were taken between 2 and 4 m below soil surface. Field measurements of redox, conductivity and pH were performed.

### County of Storstrøm

The sampling was performed below 7 sites used for mixing and loading at three machine pools, two farms, one truck garden (outdoor vegetable production) and one general shop. Borings with filters 2–4 m below soil surface were established. Water samples were analysed for 23 pesticides and metabolites by the commercial laboratory

ROVESTA, Næstved. The 23 pesticides were selected in accordance with the Danish list of pesticides for control of water quality at water works [18]. Water samples were taken in autumn 1997, and were also analysed for oxygen and pH.

### Pollution of Soil Surfaces on Sites used for Mixing and Loading

Soil samples were taken in November 1999 on 6 farms at Zealand where the farmers volunteered for the survey. The sites were used either for mixing and loading, or cleaning sprayers or for both loading and cleaning.

Soil samples were taken from the top 0 to 10 cm. The soil was packed in glass columns (i.d. 5 cm, length 30 cm). To determine the potential for pollution of ground water, the columns were saturated with 0.01 M  $\text{CaCl}_2$  and left for 24 h before elution with 1060 ml 0.01 M  $\text{CaCl}_2$  in water, which was equal to 500 mm "precipitation" on the column surface.

Water samples were diluted 100 times prior to analysis to avoid the very low and insignificant concentrations and to limit the influence of matrix on the analyses. The samples were analysed for 43 pesticides and metabolites and for seven phenols by the commercial laboratory ROVESTA, Næstved.

## RESULTS

### County of Bornholm

In all ground water samples pesticide pollution above the drinking water level ( $0.1 \mu\text{g L}^{-1}$ ) was found. The highest concentrations and the highest numbers of different pesticides were found below sites at machine pools, where the highest concentrations were the phenoxyacid herbicides dichlorprop ( $750 \mu\text{g L}^{-1}$ ) and 2,4-D ( $800 \mu\text{g L}^{-1}$ ) (see Table I). The herbicides bentazone, mecoprop, isoproturon, dinoseb and MCPA were also found in relatively high concentrations. At 5 of the 10 sites between 15 and 30 different pesticides and metabolites were found of the 46 components analysed for, resulting in total concentrations as high as 1003 and  $1613 \mu\text{g L}^{-1}$  at two different locations. Pesticide concentrations at these levels and numbers were never detected below fields away from areas where sprayers are cleaned and loaded, showing the importance of point sources.

Table II shows how many times the different pesticides were detected on individual locations in Bornholm. The table shows that dichlorprop, mecoprop, 2,4-D, bentazone and isoproturon were found below 7–9 of the 10 machine pools, respectively.

TABLE I Pesticide concentrations (max. concentrations) in ground water 2–4 m below sites used for mixing and loading at 10 machine pools in County of Bornholm. Samples analysed for 43 pesticide compounds [19]

• $2.4 \mu\text{g L}^{-1}$ (Dichlorprop)	• $11 \mu\text{g L}^{-1}$ (Isoproturon)
• $18 \mu\text{g L}^{-1}$ (Dichlorprop)	• $800 \mu\text{g L}^{-1}$ (2,4-D)
• $750 \mu\text{g L}^{-1}$ (Dichlorprop)	• $4.0 \mu\text{g L}^{-1}$ (2,4-Dichlorophenol)
• $2.5 \mu\text{g L}^{-1}$ (Isoproturon)	• $14 \mu\text{g L}^{-1}$ (2,6-Dichlorobenzamide)
• $2.6 \mu\text{g L}^{-1}$ (Simazine)	• Glyphosate/AMPA: $2.8/1.6 \mu\text{g L}^{-1}$
• $0.83 \mu\text{g L}^{-1}$ (Cyanazine)	

TABLE II Pesticides and metabolites identified in ground water below sites used for mixing and loading at 10 machine pools in Bornholm. Numbers indicate number of findings at the sites [19]

• 2,4-Dichlorophenol (10)	• Malathion (4)	• Carbofuran (1)
• Simazine (10)	• Parathion-ethyl (4)	• Dichlobenil (1)
• Dichlorprop (9)	• Alachlor (3)	• DNOC (1)
• Mecoprop (8)	• Bromoxynil (3)	• Fluazifop- <i>p</i> -butyl (1)
• 2,4-D (7)	• Fenpropimorph (3)	
• 2,6-Dichlorobenzamide (7)	• Ioxynil (3)	• Metazachlor (1)
• Atrazine (7)	• Metamitron (3)	• Methabenzthiazuron (1)
• Bentazone (7)	• Propiconazole (3)	• Pendimethalin (1)
• Desisopropylatrazine (7)	• Triadimenol (3)	• Pirimicarb (1)
• Isoproturon (7)	• Trifluralin (3)	• Prochloraz (1)
• MCPA (7)	• Dicamba (2)	
• Desethylatrazine (6)	• Dimethoat (2)	
• Terbutylazine (6)	• Dinoseb (2)	
• 4-Chloro-2-methylphenol (5)	• Lenacil (2)	
• Cyanazine (4)	• Propyzamide (2)	
• Hexazinon (4)	• Bromacil (1)	

Below mixing and loading sites at nurseries and plantations, the pesticide concentration in ground water was much lower, and as could be expected fewer pesticides were detected compared to the results at machine pools. Table III shows the highest concentrations found.

TABLE III Pesticide concentrations (max. concentrations) in ground water below sites used for mixing and loading at nurseries and plantations [19]

• 0.56 $\mu\text{g L}^{-1}$ (Triadimenol)
• 0.18 $\mu\text{g L}^{-1}$ (Desisopropylatrazine)
• 3.0 $\mu\text{g L}^{-1}$ (Hexazinon)
• 0.15 $\mu\text{g L}^{-1}$ (Clopyralid)
• 7.30 $\mu\text{g L}^{-1}$ (2,6-Dichlorobenzoic acid)

Table IV shows how many times the individual pesticides were identified at the five nurseries and plantations. The metabolite from atrazine (DIA) and the insecticide dimethoate were identified at three of the five locations.

TABLE IV Pesticide concentrations in ground water below sites used for mixing and loading at nurseries and plantations in the County of Bornholm. Numbers indicate number of findings at the 5 sites [19]

• Desisopropylatrazine (3)	• Desethylatrazine (1)
• Dimethoate (3)	• Fenitrothion (1)
• 2,6-Dichlorobenzoic acid (2)	• Hexazinon (1)
• Clopyralid (2)	• Methabenzthiazuron (1)
• Simazine (2)	• Propyzamide (1)
• 2,6-Dichlorobenzamide (1)	• Terbutylazine (1)
• Atrazine (1)	• Triadimenol (1)
• Captan (1)	

### County of Storstrøm

Results from the County of Storstrøm also showed high concentrations (Table V), though the highest concentrations were below the findings from the machine pools in Bornholm. Single compounds constituted most of the total concentration on individual sites, e.g. below a site at a truck garden five pesticides at a total concentration of  $63.8 \mu\text{g L}^{-1}$  were determined and bentazone accounted for  $59 \mu\text{g L}^{-1}$ . The concentrations of individual pesticides differ very much from one site to the other indicating spillage or disposal of single compounds to be important for the pollution.

It appears, from Table V, that several pesticides were identified at each location. In total, 21 of the 23 pesticides analysed for were identified. Further, the table shows that the pesticides found in the highest concentrations varied from one location to the other. At three locations (one farm, one truck garden and one machine pool) concentrations above  $10 \mu\text{g L}^{-1}$  were found. The finding of 2,6-Dichlorobenzamide (BAM) as the highest at two locations is not unexpected since this compound is a metabolite of the herbicides (dichlobenil and chlorthiamid) which have been very widely used for total weed control in e.g. farmyards. Due to this risk, these herbicides are no longer registered in Denmark. The other herbicides found (mecoprop, dichlorprop, bentazone and isoproturon) are all among herbicides which were previously used extensively in Danish agriculture.

TABLE V Pesticide concentrations in ground water 2–4 meters below sites used for mixing and loading at farms, truck garden, machine pools and general shop in the County of Storstrøm. Samples were analysed for 23 pesticide compounds [20]

	<i>Number of compounds</i>	<i>Total concentration</i> $\mu\text{g L}^{-1}$	<i>Max. concentration</i> $\mu\text{g L}^{-1}$	<i>Compound (Max. conc.)</i>
Farm	7	0.4	0.2	Isoproturon
Farm	11	22.4	13	2,6-Dichlorobenzamide
Truck garden	5	63.8	59	Bentazone
Machine pool	5	30.2	27	Dichlorprop
Machine pool	7	6.0	5.1	Mecoprop
Machine pool	6	7.6	6.2	Dinoseb
General shop	7	1.1	0.3	2,6-Dichlorobenzamide

### Pollution of Soil Surfaces at Sites Used for Mixing and Loading

The potential for pollution of ground water from sites used for mixing and loading was determined at 6 farms. Soil sampled at the upper 0–10 cm from the sites was packed in glass columns and eluted with water. Several pesticides and relatively high concentrations could be identified in the leachate from the soils.

The water samples were analysed for 43 pesticides and metabolites and 24 different pesticides and metabolites were found. Though most concentrations were below  $10 \mu\text{g L}^{-1}$ , about 10% of the samples contained more than  $50 \mu\text{g L}^{-1}$  (Table VI). Propiconazole and isoproturon were found in most samples which is in good correlation with high use of these compounds. In 1996–98, the sale of isoproturon was 11, 12 in 17% of the total sale in Denmark and for propiconazole it was 8, 8 and 9%.

Large differences were seen between the concentrations in each sample of soil and between the different pesticides. This may indicate that in some cases spill of concentrated chemicals or of diluted spray solution is the cause of the pollution.

TABLE VI Pesticides leached out of topsoil from sites used for washing, mixing and loading at farms. Pesticide concentrations in water after elution of soil columns. Eight hundred gram soil was eluted with 1000 ml of water. Results are shown only if concentrations  $> 10 \mu\text{g L}^{-1}$  of the particular pesticide appeared in one or more samples [21]

<i>Pesticide or metabolite</i>	<i>Washing site</i> $\mu\text{g L}^{-1}$	<i>Loading site</i> $\mu\text{g L}^{-1}$	<i>Loading site</i> $\mu\text{g L}^{-1}$	<i>Loading–washing site</i> $\mu\text{g L}^{-1}$	<i>Washing site</i> $\mu\text{g L}^{-1}$	<i>Loading site</i> $\mu\text{g L}^{-1}$	<i>Washing site</i> $\mu\text{g L}^{-1}$	<i>Washing site<sup>a</sup></i> $\mu\text{g L}^{-1}$	<i>Loading site</i> $\mu\text{g L}^{-1}$
Atrazine				430				2.8	450
Bentazone	6.8							15	
Bromoxynil								129	
Desethylatrazine				15					7.2
Ethofumesate	9.2	6.7						3.2	14
Ioxynil								123	1
Isoproturon	7.6	8.6	1.7		3			1,060	4.6
Lenacil				13					
Pendimethalin	1.9	2.9	2.4						100
Pirimicarb	23							3.6	62
Propiconazole	4.3	4.6	3.1	3.8			1.5	7.1	12
Terbutylazine	31						1.8	34	4.1

<sup>a</sup>Soil was eluted by shaking in water since water could not penetrate through the column.

Ethofumesate, pendimethalin and terbuthylazine were found in four samples and atrazine and pirimicarb in three samples. Some of the sites had only very few pesticides and low concentrations, and one site used for filling was without registration of pollution.

Some sites are covered by concrete and polluted water is collected in reservoirs before distribution, or the water is led to tanks for manure. On two sites water was sampled from relatively small reservoirs. The one site had very low concentrations whereas the other, as shown in Table VII, contained high concentrations of isoproturon with  $11,710 \mu\text{g L}^{-1}$  ( $11.7 \text{ mg L}^{-1}$ ). This high concentration on the other hand is much lower than the concentration in the spray solution where  $1000 \text{ mg L}^{-1}$  is not unusual.

TABLE VII Pesticide concentrations in water from a reservoir collecting water from a site used for loading and washing of sprayers. The water collected is distributed on the soil surface [21]

<i>Pesticides</i>	$\mu\text{g L}^{-1}$
Bromoxynil	7.1
Dichlorprop	4.8
Fenpropimorph	2.4
Isoproturon	11,710
MCPA	5.4
Mecoprop	8.4
Pirimicarb	11
Propiconazole	27

On sites for mixing and loading, the two herbicides bentazone and isoproturon have been found in high concentrations in surface soil which is in accordance with the findings in ground water. Phenoxyacid herbicides are only rarely found in surface soil whereas they are important in the ground water findings in the counties. Possibly,

these herbicides are degraded in surface soil if not washed out to subsurface where the degradation rates are lower.

## DISCUSSION

The results from all three investigations clearly show that handling of pesticides and cleaning of sprayers can lead to spillage of pesticides. Both surface and subsurface are polluted with pesticides in concentrations which are much higher than those found in ground water monitoring programmes [22].

To elucidate the importance of the problem both the number of potential contaminated sites and the concentrations should be recognised. The general use of pesticides started in 1960 when 190,000 farms existed in Denmark. The number of farms is now reduced to about 45,000 and 250 machine pools. The investigations from County of Bornholm indicate that one should expect pollution from the major part of farms and machine pools, which may imply between 40- and 100,000 point sources in Denmark ( $1-2 \text{ km}^{-2}$  in Denmark). The number of point sources from only this source is thus a serious problem. The results from County of Storstrøm and other unpublished investigations further shows that other type of sites such as: Truck gardens, small waste disposal sites, storage and production sites are potential point sources for pollution with pesticides.

Concerning the mass of pesticides, pesticides spread on fields are significantly larger than spillage from point sources, but the concentration at the point sources are high. Results from County of Bornholm and County of Storstrøm show concentrations of pesticides in shallow ground water up to  $800 \mu\text{g L}^{-1}$ , which is 8000 times the European drinking water criteria. The concentrations however differs very much and at many sites only low concentrations of pesticides are observed. At this point we do not know the actual details about spreading of this type of pollution. However, there are several factors leading to the conclusion that many point sources pose a risk to the use of ground water as drinking water:

- Shallow ground water under point sources may be anaerobic (Indicated in unpublished investigations and field measurements at the investigations at County of Bornholm and County of Storstrøm). Therefore, many pesticides are only slowly degradable.
- Leakage of pesticides may introduce pesticides directly into the subsurface environment. In general, the degradation of pesticides is slow and the possibility for adsorption is low in this environment, compared to the surface soil.
- Lack of vegetation combined with coarse soil particles enhances infiltration to the ground water.

It is thus indicated that shallow drinking water wells close to potential point sources are at great risk. Further the point sources may even be a threat to drinking water wells installed in deeper aquifers. The number of potential point sources means that there often are several potential point sources close to drinking water wells.

It is shown [11] and [12] that point sources are important for pollution of surface water. To elucidate the threat from point sources to ground water in general, it is necessary to investigate: how widespread the high concentrations are, how fast they are degraded and distributed and to what extent will they pollute deeper ground



water and nearby wells and well borings. Further the findings imply the need for better farm practice.

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