Monitoring of pesticide leaching from cultivated fields in Denmark

Walter Brüsch, Annette E. Rosenbom, Nora Badawi and Preben Olsen

The Danish Pesticide Leaching Assessment Programme (PLAP) was initiated in 1998 by the Danish Parliament in order to evaluate whether the use of approved pesticides will result in an unacceptable contamination of the groundwater, if applied under field conditions in accordance with current Danish regulation. In this programme, water samples from variably saturated soil and groundwater collected at five cultivated fields are analysed for selected pesticides and their degradation products. The PLAP results are summarised and evaluated in yearly reports and used by the Danish Environmental Protection Agency in the regulation of pesticides in Denmark (Brüsch *et al.* 2015). In order to represent typical farming scenarios in Denmark, the test fields are situated on meltwater and marine sands, and on tile-drained clayey soils in till areas.

Methods

The five cultivated PLAP fields (1.2–2.4 ha), representing different soils and hydrogeological settings, spread across Denmark (Fig. 1) are located at Silstrup, Estrup and Faardrup with tile-drained clayey soils, and at Tylstrup and Jyndevad with sandy soils (Lindhardt *et al*. 2001). The groundwater table is shallow at all fields, which enables a rapid detection of any pesticide leaching to the groundwater (Table 1). The PLAP fields are farmed according to conventional agricultural practice, and pesticides are applied in the maximum permissible doses and as specified in the regulations.

Water samples are collected weekly from drainage at the clayey till fields, and monthly from standard teflon suction cups in the unsaturated zone at the sandy fields, and from horizontal and vertical groundwater monitoring wells at all fields. The wells are installed in buffer zones surrounding the fields in order to avoid artificial transport pathways for pesticides and their degradation products from the surface to the groundwater. The vertical wells are located downstream from the field (Fig. 2), except for one upstream vertical well, which is used to determine the upstream influx to the groundwater beneath the field. The horizontal wells are installed at the clayey till fields at depths of 2–3.5 m under the pesticide-treated areas, and at the sandy fields just beneath the fluctuating groundwater table. Detection of pesticides or their degradation products can be directly related to the specific pesticide application to the PLAP fields by monitoring both the variably and fully saturated soil and accounting for potential upstream influx.

In the drainage from the clayey till fields, the weighted average concentration of pesticides is based on flow-proportional sampling. In the two sandy soils, the weighted average pesticide concentration leached to the suction cups at 1 m depth is estimated from the detected concentrations and estimated percolation on a monthly basis (Brüsch *et al.* 2015).

The analytical programme includes relevant pesticides and their degradation products as well as inorganic compounds such as chloride, nitrate, phosphate and bromide,

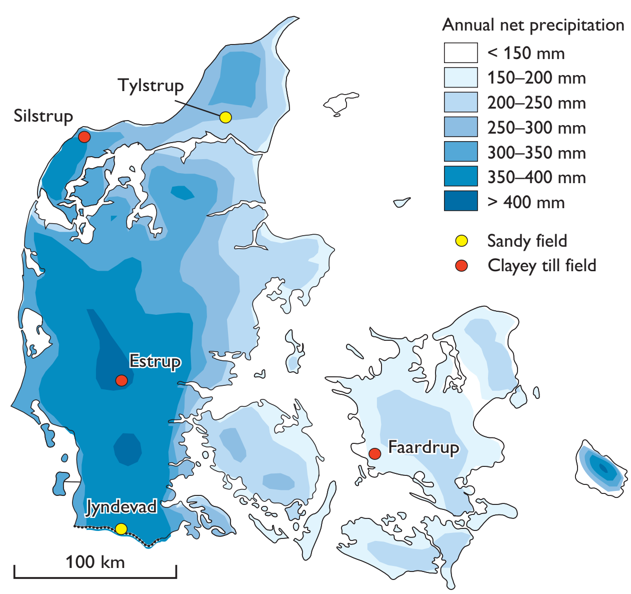

Fig. 1. Annual net-precipitation in Denmark and the location of the five PLAP fields (*http://www2.mst.dk/Udgiv/publikationer/1992/87-503-9581-5/pdf/87-503-9581-5.pdf*; Rosenbom *et al*. 2015). Tylstrup and Jyndevad are located in sandy areas with marine sand and glaciofluvial sand, respectively. Silstrup, Estrup and Faardrup are situated in areas dominated by clayey till, and the three fields are drained. The sediments were deposited during and after the last glaciation.

Table 1. Characteristics of the five pesticide leaching assessment fields

	Tylstrup	Jyndevad	Silstrup	Estrup	Faardrup
Precipitation (mm/y)*	668	858	866	862	558
Potential evapotranspiration (mm/y)*	552	555	564	543	585
Area (ha)	1.1	2.4	1.7	1.3	2.3
Tile drain	No	No	Yes	Yes	Yes
Depth to tile drain (m)			1.1	1.1	1.2
Deposited by	Saltwater	Meltwater	Glacier	Glacier	Glacier
Sediment type	Fine sand	Coarse sand	Clay till	Clay till	Clay till
Topsoil classification	Loamy sand	Sand	Sandy clay loam	Sandy loam	Sandy loam

* Based on the period 1961–1990, modified from Lindhardt *et al.* (2001).

which is used as a tracer. The pesticides are generally analysed for two years following application, but the monitoring continues if significant leaching occurs. To evaluate the pesticide leaching, the water balance, including the percolation through the variably saturated soil, is assessed for all five PLAP fields using the numerical model MACRO (Larsbo *et al.* 2005) based on long-term detailed monitoring of climate, crop-growth, soil water content, groundwater table, and if present, drainage flow (Rosenbom *et al.* 2015).

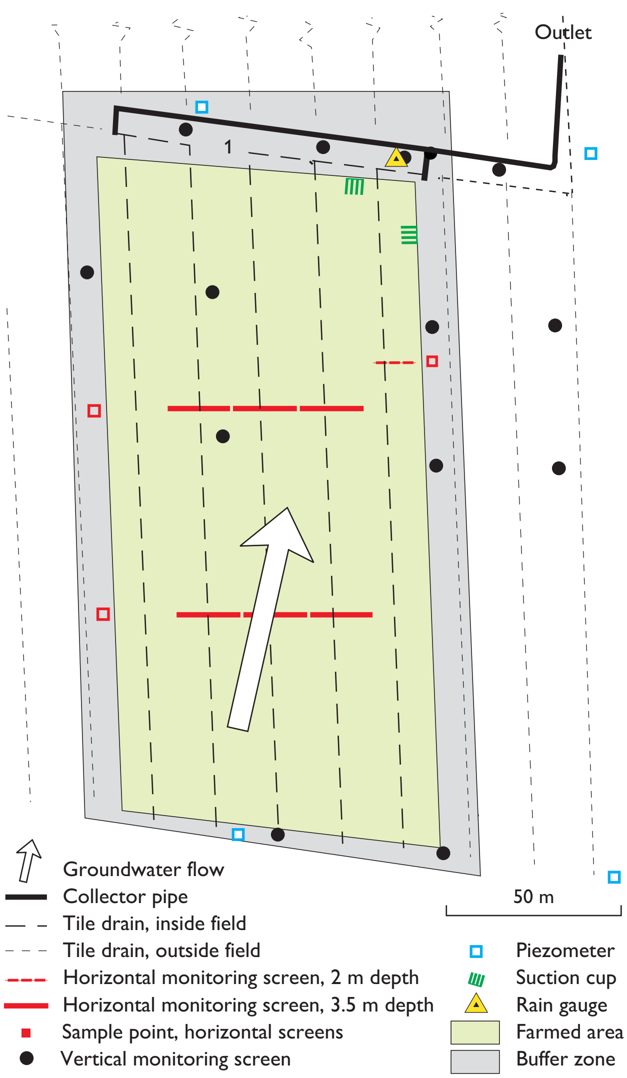

Fig. 2. Overview of the Silstrup field and its technical installations.

Monitoring results

According to the legislation of the European Union, the maximum permissible concentration of any pesticide in groundwater is 0.1 µg/l (Council of the European Union 1994). This limit is not based on health investigations but was the analytical detection limit when the legislation was made in the 1980s, and was chosen to ensure that drinking water did not contain measurable amounts of pesticides. During the latest monitoring period from July 2012 to June 2014, a total of 7378 single analyses of different pesticides or their degradation products were carried out on water samples collected at the five sites. The leaching risk of 22 pesticides and 17 degradation products was evaluated after applying the specific pesticide on specific crops. Of these 39 pesticides and their degradation products, 21 were not detected in any of the water samples.

During the entire monitoring period from May 1999 to June 2014, 51 pesticides and 52 degradation products were analysed. These are listed in the Appendix. The monitoring data showed leaching of 17 of the applied pesticides and their degradation products through the soil to tile drains or suction cups in average concentrations exceeding 0.1 µg/l. These are marked with asterisks in the Appendix.

The results of the monitoring also showed leaching of an additional 17 pesticides, but in low concentrations, marked by † in the Appendix. Although the concentrations exceeded 0.1 µg/l in several water samples collected from suction cups and tile drains at 1 m depth, the average leaching concentrations did not exceed 0.1 µg/l on an annual basis.

In groundwater samples, twenty-one pesticides or their degradation products were only detected at concentrations

Table 2. Total number of pesticides analysed, detected, and detected below 0.1 μg/l in all sample types

		Fine-grained sand	Coarse-grained sand		Clayey till	
		Tylstrup	Jyndevad	Silstrup	Estrup	Faardrup
Pesticides	Detections	16	19	39	45	38
and	Detections >0.1 μg/l	6	9	22	31	21
metabolites	Detections in %	28.1	32.8	59.1	77.6	66.7
	>0.1 μg/l in %	10.5	15.5	33.3	53.4	36.8
Groundwater avg	Nitrate-N	15.5	11.9	3.0	0.4	8.5
	Chloride	49.9	15.6	29.5	11.7	27.1
Drainage avg	Nitrate-N	ns	ns	2.1	3.5	11.2
	Chloride	ns	ns	30.3	26.6	27.5

Samples collected from suction cups, drainage and groundwater in the five PLAP fields between 01 January 2000 and July 2012.
Average nitrate and chloride concentrations from groundwater and drainage in the period January 2011 – July 2012.
Avg: average concentration in mg/l.
ns: no samples.

below 0.1μg/l or not at all. These are marked by § in the Appendix.

At the three clayey till fields, several pesticides were detected in the drainage, whereas the frequency of detection in the groundwater monitoring screens beneath the tile drain system was lower and varied considerably between the three fields. In the two sandy fields, fewer pesticides and degradation products were generally detected, both in the variably saturated soil and in groundwater (Table 2). The different leaching patterns in the sandy and clayey till fields can be attributed to specific hydrological, geological and geochemical conditions. The subsoil C horizon beneath the tile drains at the Estrup field shows low permeability with few macropores (Kjær *et al.* 2005; Rosenbom *et al.* 2015) in contrast to the Faardrup and Silstrup fields, where the clayey till is characterised by fractures and heterogeneity. Hence the fewer records of pesticides and degradation products in the groundwater at Estrup than at Faardrup and Silstrup can be related to the low permeability at the former site.

A comparison between the clayey till fields shows that the number of water samples containing pesticides and degradation products was higher at Silstrup and Estrup (35 and 40%, respectively) than at Faardrup (15%). This can be attributed to different hydro-geochemical conditions and the low net precipitation at Faardrup. The leaching pattern for non-pesticides shows that the average concentration of nitrate-N was much higher in both groundwater and drainage at Faardrup than at the other two fields (Table 2; Ernstsen *et al.* 2015). However, the average chloride content in both drainage and groundwater at Faardrup was higher than at Silstrup (Table 2), due to an up-concentration in the infiltration water caused by the low precipitation at Faardrup. The occurrence of precipitation and subsequent percolation within the first month after application were generally higher at Silstrup and Estrup than at Faardrup (Table 1).

At the clayey till fields, 59–78% of the different applied pesticides and their degradation products were detected in drainage water or groundwater (Table 2), while only 28–33% of them were detected at the sandy fields. High pesticide concentrations dominated at the three clayey till fields, with 33–53% of the detections exceeding 0.1 μg/l, while only 11–16% of the detections at the two sandy fields exceeded the threshold limit. However, the limit of 0.1 μg/l is only relevant for groundwater and not for drainage water.

The average nitrate concentrations were high in the groundwater of the sandy fields and lower at the clayey till fields (Ernstsen *et al.* 2015). However, a high average nitrate concentration was recorded in both the drainage and groundwater from the Faardrup field where the precipitation is low. This is probably because the uppermost part of the till is characterised by high permeability. It is therefore apparent that the pesticide and nitrate concentrations both reflect the geochemical conditions of groundwater and drainage water.

Further details regarding PLAP can be found in Kjær *et al.* (2002, 2003, 2004, 2005, 2007, 2008, 2009, 2011), Rosenbom *et al.* (2010), Brüsch *et al.* (2013a, 2013b, 2015), Ernstsen *et al.* (2015) and Rosenbom *et al.* (2015). For further information please visit: *http://pesticidvarsling.dk/monitor_uk/index.html*.

Conclusions

The results presented here provide an overall picture of the detections of pesticides and their degradation products in soil and groundwater in five monitored cultivated fields representing typical Danish farming activities on clayey and sandy soils in the period from 1999 to 2014. The overall pesticide leaching detected in the monitoring programme is an outcome of the pesticide selection, hydraulic conditions, type of agriculture and the geochemical conditions such as the redox potential, aerobic conditions and hence the leaching of nitrate-N and potential persistence of individual pesticides. For instance, the leaching of pesticides is more pronounced in fractured clayey soils than in sandy soils due to fast transport in anaerobic fractures in the former soils, in contrast to slower matrix transport in the more aerated sandy soils. This is illustrated by the high number of recorded pesticides in drainage water and groundwater from clayey till soils due to bypassing of the topsoil by rapid leaching through well-connected macropores such as wormholes and fractures (Rosenbom *et al.* 2015). The occurrence of pesticides in samples from the two sandy soils is probably specifically linked to the application of persistent pesticides such as metalaxyl-M applied to potatoes.

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Appendix. PLAP analyses from May 1999 to June 2014. Part A

Pesticide	Analyte		Tile drain and suction cup					Groundwater			
			Samples	Det.	≥0.1	Max.		Samples	Det.	≥0.1	Max.
Aclonifen	Aclonifen	†	111				§	298			
Amidosulfuron	Amidosulfuron		144	3	1	0.11	§	332			
	Desmethyl-amidosulfuron		24					88			
Aminopyralid	Aminopyralid	†	133				§	261			
Azoxystrobin	Azoxystrobin	*	717	139	16	1.4		1798	3		0.04
	CyPM	*	740	390	144	2.1		1910	69	5	0.19
Bentazone	Bentazone	*	1051	350	43	43		2603	81	8	0.6
	2-amino-N-isopropyl-benzamide		561	4		0.06		1295	1		0.02
Bifenox	Bifenox		303	17	3	0.38		751	7	1	0.1
	Bifenox acid	*	278	55	47	8.6		702	29	23	3.1
	Nitrofen		303	11	4	0.34		751			
Boscalid	Boscalid	†	56				§	111			
Bromoxynil	Bromoxynil		528	5	3	0.6	§	1122			
Chlormequat	Chlormequat	†	95	2		0.017	§	190			
Clomazon	Clomazone		224	1	1	0.28	§	598			
	FMC 65317		216	1	1	0.3		577			
Clopyralid	Clopyralid		219	7	4	4.094	§	520	1		0.026
Cyazofamid	Cyazofamid	†	100				§	262			
Desmedipham	Desmedipham	†	287					580	1		0.033
	EHPC		199					383			
Diflufenican	Diflufenican		109	32	14	0.49		324	1	1	0.47
	AE-B107137	*	121	19	1	0.13		333	1		0.016
	AE-05422291		109					324			
Dimethoate	Dimethoate	†	515	1	1	1.417		1253	1		0.085
Epoxiconazole	Epoxiconazole		330	14	2	0.39		999	1		0.011
Ethofumesate	Ethofumesate	*	519	70	17	12		1095	36	7	1.4
Fenpropimorph	Fenpropimorph	†	657	2		0.038		1531	2		0.029
	Fenpropimorph acid		636	2	1	0.25		1435			
Flamprop-M	Flamprop-M-isopropyl		520	38	1	0.109		1204	1		0.024
	Flamprop		525	23	1	0.35		1212			
Florasulam	Florasulam	†	146					351			
	Florasulam-desmethyl		109					130			
Fluazifop-P-buthyl	Fluazifop-P-butyl		128					232			
	TFMP	*	184	53	24	0.64		555	87	16	0.29
	Fluazifop-P		451	11	4	3.8		1109	7	1	0.17
Fludioxonil	CGA 192155	†	11				§	48			
	CGA 339833		11					48			
Fluroxypyr	Fluroxypyr	*	521	4	3	1.4		1273	2		0.072
Glyphosate	Glyphosate	*	1091	429	136	31		2216	77	5	0.67
	AMPA	*	1092	632	142	5.4		2217	37		0.08
Iodosulfuron-methyl-natrium	Metsulfuron-methyl		332	1		0.054		842			
	Iodosulfuron-methyl	†	60				§	250			
Ioxynil	Ioxynil		527	24	7	0.25		1128	1		0.01
Linuron	Linuron	†	67				§	271			
Mancozeb	ETU		44	7		0.038		200	2		0.024
	EBIS		7					25			
MCPA	MCPA		354	14	3	3.894		916	1		0.019
	2-methyl-4-chlorophenol		354	2	1	0.24		912			
Mesosulfuron-isopropyl	Mesosulfuron-methyl		153	13		0.059	§	411			
	Mesosulfuron		119					119			
Mesotrione	Mesotrione	†	50				§	156			
	AMBA		50					156			
	MNBA		50					156			

Fifty-one pesticides and 52 degradation products analysed in the PLAP programme in the period May 1999 – June 2014. The columns show the number of water samples analysed, number of detections, and detections in concentrations ≥ 0.1µg/l in water samples from the variably-saturated zone (drainage and suction cups), and in groundwater (vertical and horizontal groundwater wells).

Det: number of detections. ≥0.1: number of detections ≥0.1µg/l. Max: maximum concentration in µg/l.

*: Pesticides and their degradation products leached through soil to tile drains or suction cups in average concentrations above 0.1 µg/l.

†: Pesticides not detected or detected only in a few samples above their threshold concentrations at 1 m depth.

§: Pesticides and their degradation products not detected or only detected in a few samples in groundwater.

Appendix. PLAP analyses from May 1999 to June 2014. Part B

Pesticide	Analyte		Tile drain and suction cup					Groundwater			
			Samples	Det.	≥0.1	Max.		Samples	Det.	≥0.1	Max.
Metalaxyl-M	metalaxyl-M		207	15		0.037		592	79	23	1.3
	CGA 108906	*	215	175	69	4.8		593	468	128	2.7
	CGA 62826	*	216	100	25	1.2		593	147	8	0.68
Metamitron	Metamitron	*	515	103	31	26.369		1095	53	7	0.63
	Desamino-metamitron	*	518	129	23	5.549		1094	78	16	1.3
Metrafenone	Metrafenone		136	20		0.072		273	1		0.04
Metribuzin	Metribuzin		97	2		0.024		414	1		0.014
	Diketo-metribuzin		340	256	63	0.69		552	479	336	1.372
	Desamino-diketo-metribuzin	*	255	81	51	2.1		551	256	18	1.831
	Desamino-metribuzin	*	91					392			
Pendimethalin	Pendimethalin		694	89	30	32		1811	1		0.052
Phenmedipham	Phenmedipham		288					580	2		0.025
	MHPC		288	2	1	0.19		580	1		0.053
	3-aminophenol		109					245			
Picolinafen	Picolinafen		117	18		0.07		193			
	CL153815	*	117	31	11	0.5		193			
Pirimicarb	Pirimicarb		887	62		0.077		2120	6		0.035
	Pirimicarb-desmethyl-formamido	*	707	29	13	0.379		1638	2		0.076
	Pirimicarb-desmethyl		780	8		0.053		1911	3		0.042
Propiconazol	Propiconazole		899	32	3	0.862		2084	3		0.035
Propyzamid	Propyzamide	*	257	27	8	1.6		754	10	2	0.14
	RH-24644		257	19		0.051		754	2		0.032
	RH-24580		257	2		0.016		754			
	RH-24655		233	1		0.017		690			
Prosulfocarb	Prosulfocarb		199	6	1	0.18		516	5		0.032
Pyridat	Pyridate		39					116			
	PHCP		125	4	4	2.69		373	14	4	0.309
Rimsulfuron	Rimsulfuron		117					367			
	PPU	*	502	388	74	0.29		1519	432	13	0.23
	PPU-desamino		502	186	6	0.18		1519	107		0.089
Tebuconazole	Tebuconazole	*	289	47	17	2		784	8	2	0.12
	1.2.4-triazol	*						16	7	1	0.17
Terbuthylazine	Terbuthylazine	*	513	213	56	11		1324	88	23	1.9
	Desethyl-terbuthylazine	*	612	365	88	8.3		1664	261	33	0.94
	Desisopropylatrazine		414	156	2	0.44		996	92		0.047
	Hydroxy-terbuthylazine	*	384	136	18	0.99		940	34		0.069
	2-hydroxy-desethyl-terbuthylazine	*	342	128	28	6.3		850	9		0.092
Thiacloprid	Thiacloprid	†	47				§	100			
	Thiacloprid-amide		47	1		0.012		100			
	M34		55					100			
	Thiacloprid sulfonic acid		56					100			
Thiamethoxam	Thiamethoxam	†	132				§	359			
	CGA 322704		132					359			
Triasulfuron	Triasulfuron	†	82				§	301			
	Triazinamin		393					1103	1		0.042
Tribenuron-methyl	Triazinamin-methyl	†	569	2		0.042	§	1523			
Triflusulfuron-methyl	Triflusulfuron-methyl	†	95					288			
	IN-E7710		95	5		0.014		288			
	IN-M7222		95					288	1		0.052
	IN-D8526		95					288			

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