Concentration and leaching of atrazine into drainage water in Gleyic Podzoluvisol

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ABSTRACT

The research objective was to investigate the influence of different pipe drainage systems on the concentration and quantity of atrazine leached in drainage water, as well as in surface (0–30 cm) and sub-surface (30–60 cm) soil horizons in the production of agricultural crops on drained Gleyic Podzoluvisol. Investigations involved four different variants of drainpipe spacing (15, 20, 25 and 30 m, i.e. four variants in four repetitions). In a particular trial year, the same crop was grown and the same agricultural practices applied in all trial variants. Maize was grown in the first two trial years and wheat in the last year. Soil and water samples were qualitatively and quantitatively analysed for the presence of atrazine by gas chromatography. Atrazine concentrations in drainage water ranged from those within the limits tolerated for drinking water (out of the growing season) up to 458 $ng.dm^{-3}$. Maximum quantity of leached atrazine was recorded in 1998 (0.500 $g.ha^{-1}$), namely, from atrazine application in May 1997 to March 1998 incl., 0.630 $g.ha^{-1}$ or 0.052% of the initially added quantity was leached. Maximum atrazine concentration in the surface soil layer was recorded in traces or it was not recorded at all (0.013 $mg.kg^{-1}$), whereas no atrazine was recorded in the subsurface layer during the trial period. Analysis of variance produced no statistically significant difference in the atrazine concentration and leaching in drainage water and in the surface soil layer between different variants of pipe drainpipe spacing.

Keywords: atrazine concentration; drainage water; drained Gleyic Podzoluvisol; leaching

Atrazine, developed in 1957 (Knusli 1970), has been used since the early sixties for weed control in different crops. It is still the leading herbicide for maize. Atrazine is one of the triazine herbicides most commonly applied to control broad-leaved weeds in maize (Hrlec 1990). Due to its good water solubility, atrazine is likely to be leached from the treated areas into surface and ground waters, as well as into the drinking-water bearing strata (Frank et al. 1989). Leaching and potential contamination of drinking water sources depend on atrazine sorption and its degradation in soil (Bacci et al. 1989). Owing to its chemical characteristics and persistence in soil, atrazine is one of the most ubiquitous pesticides in the environment (Capriel et al. 1985). The residues of experimentally administered atrazine can be found in various tissues, e.g. in the kidney, liver, lung, heart, brain, fat and muscles of rats (Bakke et al 1972, Santa Maria et al. 1986). Due to its relative persistence and frequent application, atrazine residues are often found in ground, surface, drain and drinking waters (Gojmerac et al. 1994, 1996, Vidaček et al. 1994, Šimunić et al. 1999). Though leaching through the plough-layer profile and runoff from the plough-layer surface are not the only ways in which atrazine gets into drinking water, this manner of water pollution is noteworthy.

For this reason, its use has been restricted or banned in some European countries.

The aim of the research carried out during 1996, 1997 and 1998 was:

– to determine the concentration and leaching of atrazine in drainage water, as well as in surface (0–30 cm) and sub-surface (30–60 cm) soil horizons for four different drainpipe spacings

– to determine, using the analysis of variance, whether there is a significant difference in the contents of atrazine in drainage water and surface (0–30 cm) and subsurface (30–60 cm) soil horizons between different drainpipe spacings

MATERIAL AND METHODS

The trial field Jelenšćak (Central Sava Valley) was set up on ameliorated Gleyic Podzoluvisol and for this purpose drainpipe spacing variants of 15 m (area 1425 m^2), 20 m (1900 m^2), 25 m (2375 m^2) and 30 m (2850 m^2) were studied (four variants in four replications). All variants were combined with contact hydraulic material – gravel. Drainpipes have the following characteristics: length 95 m, diameter 65 mm, average slope 3‰ and average depth 1 m, and they discharge directly into canals.

The same crop was grown and the same agricultural engineering practices were applied in all drainpipe variants and in each trial year (Table 1). Samples of drainage water and of soil were taken following seasonal dynamics. Drainage discharge was measured continually by means of automatic electronic gauges – limnimeters. Limnimeters were set up in each variant, at the drainpipe outlet into the open canal.

Atrazine was accumulated from soil samples in an ultrasonic bath by means of repeated extraction. Five grams of

Table 1. Agricultural practices and application dates for crops grown during the trial period

Year	Crop	Sowing	Protection		Harvest
			quantity (l.ha^{-1})	date	
1996	maize	May 22	Primextra (6 l.ha^{-1} = 1200 g.ha^{-1} atrazine)	May 23	November 28
1997	maize	May 6	Primextra (6 l.ha^{-1} = 1200 g.ha^{-1} atrazine) + Racer (0.9 l.ha^{-1})	May 9	October 27
1998	wheat	October 29			July 21

an air-dried and sieved (particle diameter < 1 mm) sample was extracted with 1 × 20 ml and 1 × 10 ml of the solvent mixture acetone: *n*-hexane 2:1. Bound extracts were evaporated under a nitrogen stream to 10 ml and centrifuged. Separated supernatant was evaporated to 1 ml under a nitrogen stream.

Qualitative and quantitative analyses of atrazine in water and soil extracts were done by capillary gas chromatography. Compounds were detected with detectors selective for nitrogen and phosphorus compounds.

The total annual quantity of atrazine leached was estimated based on the average concentration of atrazine (in and out of the growing season) and the average quantity of drainage discharge (in and out of the growing season). Data were statistically processed by means of the analysis of variance.

Soil characteristics

Drained Gleyic Podzoluvisol is located in the Sava river valley, on level relief, at an average altitude of 96.4 m a.s.l. Before the trial was set up, the area was utilised as a pasture, which was in association with swamp vegetation (Salix sp., Juncus sp. etc.).

Soil profile layering is: Ap-Bt,g-Gso-Gr. Depth to the impervious layer amounts to 1.3 m. Excess soil moistening prior to drainage was effected by ground and surface

Table 2. Major characteristics of ameliorated Gleyic Podzoluvisol

Horizon	Depth (cm)	Content of soil particles (%)		Porosity (%)	Capacity (%)		Permeability (m/day)	pH	
		silt	clay		water	air		H_2O	1M KCl
Ap	0–35	47	46	48	44	4	0.011	6.7	5.3
Bt, g	35–75	45	48	49	45	4	0.010	6.5	5.2
Gso	75–115	55	39	46	42	4	0.011	7.9	7.1
Gr	115–130	63	25	49	45	4	8.1	7.2	

Table 3. Major characteristics of ameliorated Gleyic Podzoluvisol per variants

Variant	Horizon	Depth (cm)	Content of soil particles (%)		Porosity (%)	Capacity (%)		Permeability (m/day)	pH	
			silt	clay		water	air		H_2O	1M KCl
15 m	Ap	0–35	48	45	47	43	4	0.011	6.8	5.3
	Bt, g	35–74	45	48	49	45	4	0.010	6.6	5.2
	Gso	74–115	55	38	45	41	4	0.011	7.9	7.1
	Gr	115–132	62	27	49	45	4	8.0	7.1	
20 m	Ap	0–35	47	47	49	45	4	0.011	6.7	5.3
	Bt, g	35–77	43	49	50	47	3	0.010	6.4	5.2
	Gso	77–112	54	41	47	43	4	0.011	7.8	7.0
	Gr	112–130	64	25	49	45	4	8.1	7.2	
25 m	Ap	0–35	46	47	49	45	4	0.011	6.7	5.2
	Bt, g	35–74	44	49	50	47	3	0.010	6.4	5.2
	Gso	74–116	55	40	47	44	3	0.011	7.8	7.0
	Gr	116–130	64	25	48	44	4	8.0	7.1	
30 m	Ap	0–35	48	45	47	42	5	0.011	6.7	5.3
	Bt, g	35–75	47	47	48	44	4	0.010	6.5	5.2
	Gso	75–117	56	37	45	40	5	0.011	7.9	7.1
	Gr	117–130	63	24	48	44	4	8.2	7.3	

Table 4. Total quantity and distribution of monthly precipitation (mm)

Year	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.	Total
1996	54.3	31.5	40.6	69.7	70.7	30.6	90.5	82.5	191.0	46.3	134.9	79.0	921.6
1997	43.8	54.5	25.5	45.4	73.3	81.3	102.8	62.8	29.8	76.0	126.5	85.6	807.3
1998	65.3	5.3	57.5	59.2	103.2	107.1	121.3	86.5	173.5	119.8	51.3	49.2	999.2

1.–12. months

waters. Major characteristics of ameliorated Gleyic Podzoluvisol are presented in Table 2 (average values), and major characteristics per variant in Table 3.

The soil is of silty clayey texture to the depth of 0.75 m. The clay content of this soil section is in the range of 46–48%, and the silt content is 45–47%. The soil depth of 0.75–1.15 m is of lighter texture. The silt component preponderates in soil texture (55%), while the clay content decreases (34%). Soil texture at depths over 1.15 m is silty loamy. The soil is porous, with the total pore volume of 48–49%. Soil water capacity is 42–45%. Air capacity is low (4%). Vertical hydraulic conductivity is very low (0.011 m/day).

Weather characteristics

Weather characteristics are presented in terms of the quantity and distribution of monthly precipitation (mm) and mean monthly and mean annual air temperatures (°C) – for the Meteorological Station in Kutina, which is 15 km from the field (Table 4).

During the three-year trial period, the total annual precipitation ranged between 807.3 mm (1997) and 999.2 mm (1998). Unevenly distributed annual precipitation was accompanied by uneven monthly precipitation, so maxima were recorded in summer and in autumn. The primary maximum occurred in autumn (September/November) and

Table 5. Monthly and total drainage discharge (mm) per variants

Variant	Year	1.	2.	3.	4.	5.	6.	7.	8.	9.	10.	11.	12.	Total
R 15	1996	32	17	23	26	8				**35**	**25**	**29**	25	220
R 20		32	20	22	25	7				**34**	**25**	**27**	30	222
R 25		32	19	22	24	9				**39**	**24**	**33**	22	224
R 30		38	19	21	25	5				**31**	**28**	**26**	32	225
R 15	1997	31	26	13	9	**11**	**12**					**24***	**34***	102
R 20		31	25	13	8	**10**	**12**					**24***	**36***	99
R 25		31	25	14	10	**13**	**16**					**23***	**32***	109
R 30		31	26	13	10	**12**	**17**					**22***	**33***	109
R 15	1998	**35**	**3**	**28**	**21**	**19**	**23**		7	36	55	17	28	330
R 20		**35**	**4**	**28**	**20**	**16**	**22**		6	36	59	19	24	329
R 25		**36**	**9**	**30**	**21**	**20**	**23**		6	37	57	20	24	338
R 30		**37**	**4**	**29**	**22**	**21**	**23**		7	38	58	21	24	341

Shaded fields refer to the time of crop growing
1.–12. months, * values of drainage discharge from November to December 1997, added to the growing season of 1998 (wheat grown)

Table 6. Quantities of drainage discharge in and out of the growing season, (mm) per variants

Variant	1996			1997			1998		
	out of growing season	in growing season	total	out of growing season	in growing season	total	in growing season	out of growing season*	total
15 m	131	89	220	79	23	102	187	143	330
20 m	136	86	222	77	22	99	185	144	329
25 m	128	96	224	80	29	109	194	144	338
30 m	140	85	225	80	29	109	193	148	341

* values of drainage discharge from November to December 1997, added to the growing season of 1998

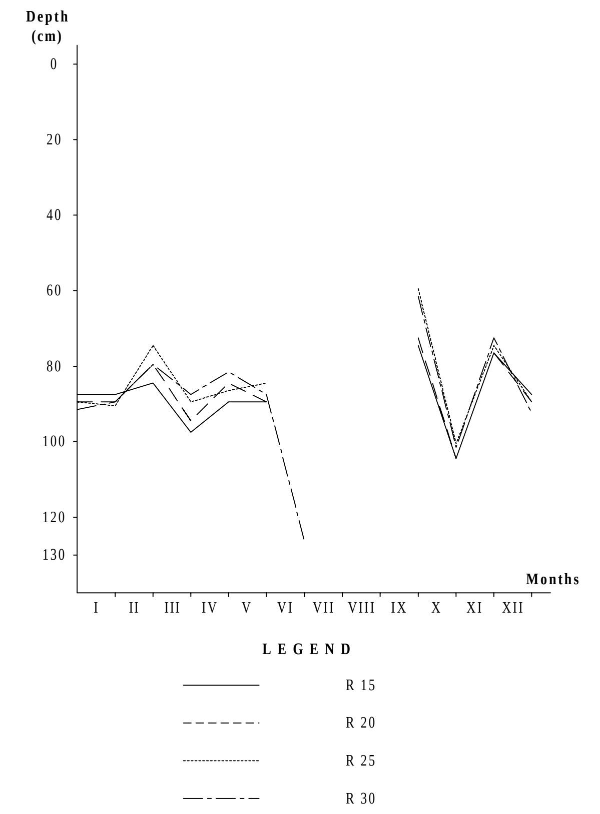

Figure 1. Fluctuations of groundwater per variants in 1996 year

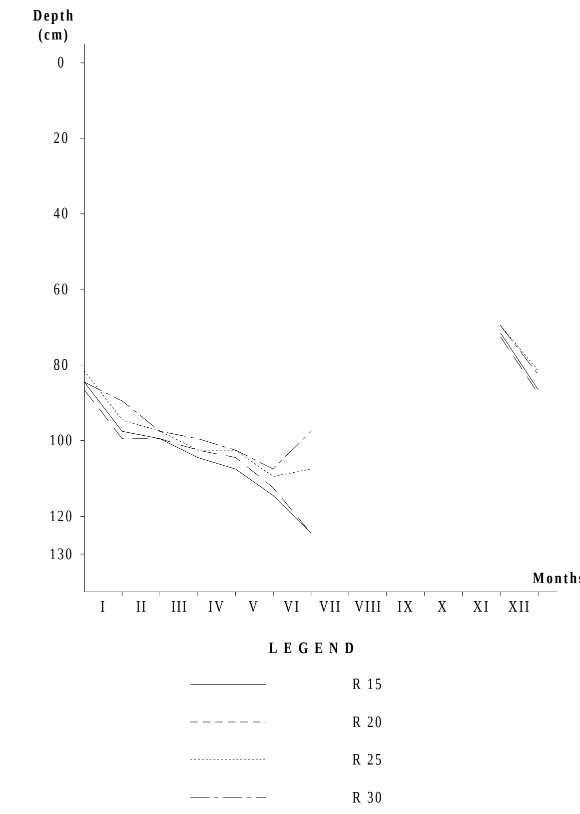

Figure 2. Fluctuations of groundwater per variants in 1997 year

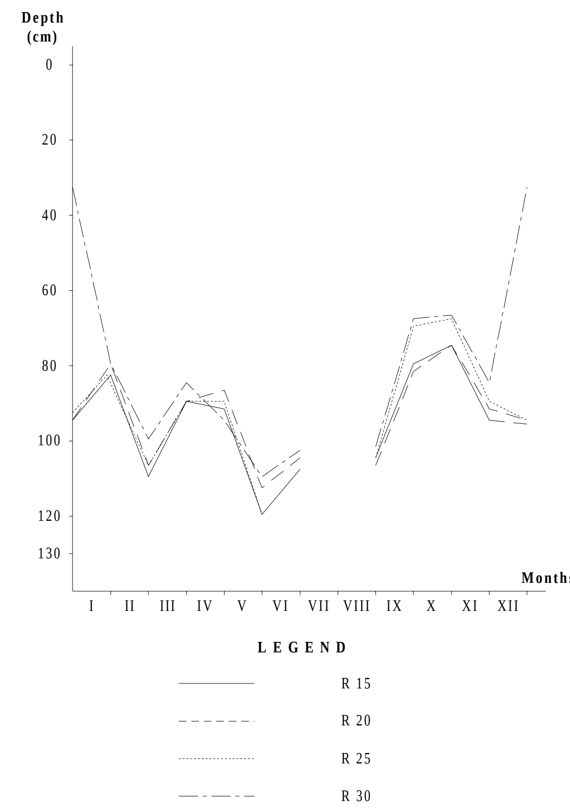

Figure 3. Fluctuations of groundwater per variants in 1998 year

the secondary maximum in summer (July). Air temperatures ranged from 10.3°C (1996) to 11.1°C (1998). On average, December, January and February were the coldest months while July, August and June were the warmest.

Hydrological relations

Drainage discharge, its duration and the groundwater level are important indicators of drainpipe efficiency for draining excess water from soil. It can be seen from Tables 5–7 and Figures 1–3 that there are certain differences in the total quantity of drainage discharge, its duration and the groundwater level both between the tested variants in each year, each growing and non-growing season, and between the trial years. The largest quantities of drainage discharge, its longest duration and the average most shallow groundwater level were recorded in all variants in 1998, when total precipitation was most abun-

Table 7. Total duration of drainage discharge (days) per variants

Variant	1996	1997	1998
R 15	185	149	197
R 20	190	153	198
R 25	202	159	206
R 30	206	161	207

Table 8. Average atrazine concentrations in drainage water in and out of the growing season (ng.dm^{-3}) per trial variants

Variant	1996		1997		1998
	out of growing season	in growing season	out of growing season	in growing season	in growing season*
15 m	35	212	47	423	255
20 m	22	210	49	446	262
25 m	49	219	29	458	256
30 m	47	218	9	415	259

F-test not significant between different variants

* values of drainage discharge from November to December 1997, added to the growing season of 1998 (wheat grown)

Table 9. Average total atrazine leaching into drainage water in and out of the growing season (g.ha^{-1}) per trial variants

Variant	1996			1997			1998
	out of growing season	in growing season	total	out of growing season	in growing season	total	in growing season*
15 m	0.046	0.189	0.235	0.037	0.097	0.134	0.477
20 m	0.030	0.181	0.211	0.038	0.098	0.136	0.485
25 m	0.063	0.210	0.273	0.023	0.133	0.156	0.497
30 m	0.066	0.185	0.251	0.007	0.120	0.127	0.500

F-test not significant between different variants

* includes the values of atrazine leached in November and December 1997

dant (Table 3), whereas the overall smallest quantities of drainage discharge, its shortest duration and the deepest groundwater level were recorded in all variants in 1997, the year with the least precipitation. The highest drainage discharge and the most shallow groundwater level were recorded in all variants in the autumn-winter and spring periods. The average lowest drainage discharge, its shortest duration and the average deepest groundwater level were recorded in all trial years at the drainpipe spacing of 15 m, while the least favourable state was recorded at the drainpipe spacing of 30 m. For these reasons, we consider the drainpipe spacing of 15 m to be the most efficient one and recommend its use in draining excess waters. Accordingly, the existing differences in the total quantity of drainage discharge, its duration and the groundwater level between particular years and between particular variants in the same year were caused by a number of factors, such as the different total annual quantity and distribution of precipitation, the crop grown and its different water requirements (evapotranspiration), as well as the different efficiency of each particular drainpipe system.

RESULTS AND DISCUSSION

Research results on atrazine concentration in drainage water and soil and its leaching with drainage water are presented in Tables 8–10, and in Figures 4 and 5.

Atrazine concentration and leaching in drainage water

It can be seen from Table 8 that different values were determined for atrazine concentrations in drainage water,

Table 10. Maximum atrazine concentrations in soil (mg.kg^{-1}) per variants

Variant	1996		1997		1998	
	0–30 cm	30–60 cm	0–30 cm	30–60 cm	0–30 cm	30–60 cm
15 m	–	–	0.010	–	0.007	–
20 m	0.003	–	0.013	–	0.008	–
25 m	–	–	0.011	–	0.009	–
30 m	0.003	–	0.010	–	0.008	–

– not detected, limit of detection is 0.002 mg.kg^{-1}

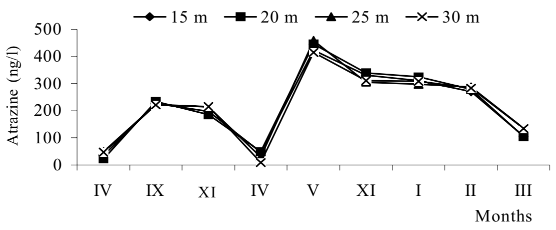

Figure 4. Concentration of atrazine in drainage water

both between the trial years and between different drainpipe spacings. This was certainly influenced by the date of atrazine application (Table 1), quantity and distribution of rainfall (Table 4), and the quantity of drainage discharge (Tables 5 and 6). Average values of atrazine exceeded the tolerated value of 100 ng.dm^{-3} in all variants in the growing season whereas lower values were recorded out of the growing season. Maximum atrazine concentrations in drainage water were recorded after its application and the first drainage discharge (September 1996 and May 1997, Figure 4). Gaynor et al. (1989) reported that in most cases atrazine was lost during the growing season than at other times. Differences in the maximum values of atrazine concentration recorded in 1996 and 1997 might be a consequence of atrazine degradation (1996). Frank et al. (1991) reported unequal duration of atrazine degradation in soil, from 149 to 684 days, depending on the temperature and microbiological activity of the soil. Atrazine concentration decreased with later drainage discharges (Figure 4). Namely, atrazine is very water-soluble and is readily transported with filtered soil waters (Albanis et al. 1988). Values of atrazine concentrations recorded in 1996 (out of the growing season) and in 1998 (growing season) result from atrazine applications in previous years, depending on the leaching speed and atrazine degradation. In the case of the mentioned atrazine rate (1200 g.ha^{-1}) applied to the studied soil type (Table 2) and the recorded quantities of precipitation and drainage discharge, atrazine concentration drops below the tolerated limit in 10–12 months. Albanis et al. (1988) reported that no atrazine residues were detected in water after 247 days.

Atrazine concentration in drainage water and the quantity of drainage discharge were used to calculate the quantity of atrazine leached (Table 9). The largest quantity of total atrazine leached occurred in the last trial year, the values ranging from 0.477 g.ha^{-1} to 0.500 g.ha^{-1}. However, if the atrazine values from 1997 are added, then the total quantity of atrazine leached amounts to 0.620 g.ha^{-1} to 0.630 g.ha^{-1} or 0.05% of its initially added quantity. Frank et al. (1991) recorded higher values of leached atrazine in drainage water and soil of predominantly sandy texture (0.2–1.9% of 3200 active ingredients). Albanis et al. (1988) reported different quantities of leached atrazine with respect to soil texture (0.54% in clay, 0.66% in loam and 0.47% in silt-loam).

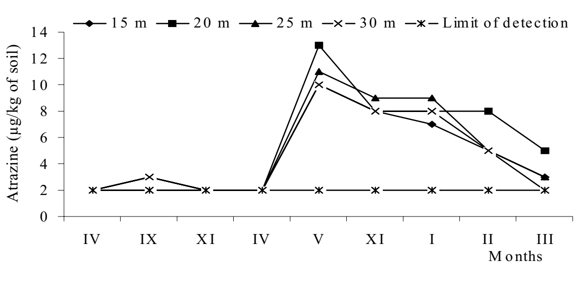

Figure 5. Concentration of atrazine in drainaged Gleyic Podzoluvisol

Atrazine concentration in soil

Atrazine concentration in soil is presented in Table 10, and the dynamics of its concentration in Figure 5. The soil content of atrazine is, in fact, the difference between atrazine input into and its output from the soil (losses of residues to water disappear by dispersion into air or by degradation) (Frank et al. 1991). It can be seen from Table 10 that, in 1996, atrazine was recorded in traces in the surface layer in some variants. It was not detected in the soil sub-surface layer of any trial variant. This may be due to the very low water-permeability of the sub-plough horizon (0.010 m/day, Table 2) so that on its way to the drainage ditch or contact material (gravel) water passes through the upper plough layer. Ritter et al. (1974) found that atrazine moved only slowly, up to 25 cm in one year. The same results are reported by Kozák and Vacek (1996), where soil atrazine concentration decreased with depth. Increased atrazine concentrations were recorded in all variants in 1997 (Figure 5) at the same atrazine rates as those applied in 1996. Atrazine content in soil decreased in the growing season of 1998. These different atrazine contents in soil may be a result of slow atrazine degradation and dissipation over winter months and probably their full arrest under frost, when the mean monthly temperatures were below 0°C (Frank et al. 1991). The tolerance limit for atrazine concentration in soil depends on the susceptibility of the next crop, as well as on the soil physical and chemical properties. For heavy clay soils, Šilješ (1980) gives the following tolerated atrazine concentrations in soil for the next crop in the crop rotation: oil rape 0.1 ppm (100 µg.kg^{-1}) and oats 0.25 ppm.

CONCLUSIONS

Atrazine concentrations and leaching in drainage water and in drained Gleyic Podzoluvisol of the trial field point to the following conclusions:

Atrazine concentrations in drainage water ranged from those within the limits tolerated for drinking water (out of the growing season) up to 458 ng.dm^{-3}.

Maximum quantities of leached atrazine were recorded in 1998 (0.500 g.ha^{-1}), when a total 0.630 g.ha^{-1} or 0.052% of the initially added quantity was leached from atrazine application in May 1997 to March 1998 incl.

Atrazine concentration in the surface soil layer was recorded in traces or not recorded (1996), up to the highest value recorded (0.013 $mg.kg^{-1}$), whereas no atrazine was recorded in the sub-surface layer during the trial period.

Analysis of variance showed no statistically significant difference in the quantity of atrazine, leached in drainage water and in the surface soil layer, between different variants of drainpipe spacing, that is, the existing differences in atrazine contents between variants are not significant.

From the aspect of the shortest duration of excess water drainage, the drainpipe spacing of 15 m is recommended.

Acknowledgments

These investigations were initiated and supported by the Ministry of Science and Technology of Croatia (project No. 178131, Systems of Management, Water – Soil – Plant) and the Water Management Agency Hrvatske vode, Croatia.

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Received on June 6, 2001

ABSTRAKT

Koncentrace a vyluhování atrazinu do drenážní vody v oglejené podzolové půdě

Byl sledován vliv různých trubkových odvodňovacích systémů na koncentraci a množství atrazinu vyluhovaného do drenážní vody a do povrchového (0–30 cm) a podpovrchového (30–60 cm) půdního horizontu při pěstování zemědělských plodin na hydromeliorované oglejené podzolové půdě. Byly použity čtyři varianty rozchodu drenážních trubek (15, 20, 25 a 30 m, tj. čtyři varianty ve čtyřech opakováních) a ve všech letech stejná plodina i agrotechnika. V prvních dvou pokusných letech jsme pěstovali kukuřici a v posledním roce pšenici. Pomocí plynové chromatografie jsme provedli kvalitativní a kvantitativní analýzu vzorků půdy a vody na přítomnost atrazinu. Koncentrace atrazinu v drenážní vodě se pohybovaly v mezích povolených pro pitnou vodu (mimo vegetační období) a dosahovaly 458 $ng.dm^{-3}$. Nejvyšší množství vyluhovaného atrazinu jsme zaznamenali v roce 1998 (0,500 $g.ha^{-1}$), a to v důsledku aplikace atrazinu v období od května 1997 do března 1998 včetně, kdy se z původně dodaného množství vyluhovalo 0,630 $g.ha^{-1}$, resp. 0,052 %. Maximální koncentrace atrazinu v povrchové vrstvě půdy se vyskytovala ve stopách nebo se vůbec neprokázala (0,013 $mg.kg^{-1}$), zatímco v podpovrchové půdní vrstvě jsme za celé pokusné období nezjistili žádné množství atrazinu. Analýza rozptylu nepřinesla mezi jednotlivými pokusnými variantami statisticky významné rozdíly v koncentraci ani ve vyluhování atrazinu do drenážní vody ani do povrchové vrstvy půdy.

Klíčová slova: koncentrace atrazinu; drenážní voda; odvodněná glejová podzoluvizem; vyluhování

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