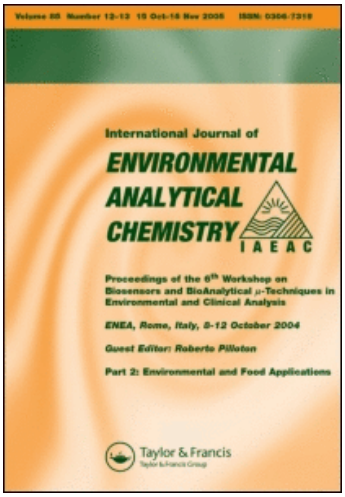

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Field Study of the Effect of Sewage Sludge Application on Atrazine Behaviour in Soil

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FIELD STUDY OF THE EFFECT OF SEWAGE SLUDGE APPLICATION ON ATRAZINE BEHAVIOUR IN SOIL

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Atrazine dissipation was followed in a hydromorphic soil and a loamy soil amended with different kinds of sewage sludge. The atrazine was applied 5 months after the organic amendments. The hydromorphic soil was drained and water samples were collected for atrazine analyses. The atrazine dissipation was slower in the plot amended with sewage sludge, but after 12 months, non significant differences were found between the control plot and the amended plot. The highest concentrations of atrazine and metabolites were found in the water drained from the control plot, and the biodegradation degree, estimated from the deethylatrazine to atrazine ratio, was higher than in the amended plot. In the loamy soil, the dissipation kinetics were similar for all the sewage sludges; however, significant differences were found in the distribution of the atrazine residues with depth.

KEY WORDS: Atrazine, soil dissipation, sewage sludge, water pollution.

INTRODUCTION

Organic matter influences the retention and, directly or indirectly, the degradation of most organic chemicals in soils. Some agricultural practices using organic amendments can modify the amount of organic matter and the nature and reactivity of the organic constituents. Exogenic organic matter (EOM) can modify the amount and activity of the microbial biomass, and the biodegradation of the organic chemicals, thus influencing the fate of chemicals present before or applied after the organic amendment application.

Addition of EOM to soil increases sorption of several pesticides[1-3], and reduces leaching of such chemicals[3-5]. The use of EOM could be a management practice to limit the pesticide pollution. However, EOM can contain soluble organic material which can form soluble complexes with freely dissolved pesticides; that can provoke competition between adsorbed phase and soluble organic matter and promote the desorption of pesticide previously adsorbed[6,7]. On the other hand, stable soluble complex between pesticide and soluble organic matter can increase the pesticide mobility in the soil[8]. The modification of the chemicals adsorption-desorption and of their apparent water solubility by soluble organic matter has been often reported[9,10].

In soils amended with EOM, chemical degradation can be increased or decreased, depending on EOM and pesticide properties. Usually, the degradation decrease is explained by an increased adsorption which makes the chemical less available for microbial degradation. However, contradictory results have been reported in the literature. Some works have shown a sewage sludge induced inhibition of the micro-

organisms responsible for pesticide degradation[11]; in other work, an increase of the degradation of the chemical after EOM addition was observed. This last effect was explained by the soil microbial activity increase due to easily degradable EOM[12], which enhances the chemical degradation by co-metabolism.

Sewage sludge application to soils is a common method of disposal. Such sludges are used in agriculture as organic amendments, but some environmental problems can occur because of their trace metal contamination[13,14].

The work presented in this paper is a study of how sewage sludge amendments can modify the behaviour of atrazine applied to two soils treated with different sewage sludges. Results describing agronomic consequences and trace element behaviour have been reported elsewhere[15,16].

MATERIALS AND METHODS

Experimental plots

The field experiments were conducted at two locations: at Rambouillet on a hydromorphic clay soil (*Aquic eutrochrept*) continuously cropped with corn (*Zea mays* L.) and at Grignon on a loamy soil (*Typic eutrochrept*) alternatively cropped with corn and wheat (*Triticum aestivum* cv.). The main soil characteristics are given in Table 1. The total soil microbial biomass was measured in the surface soil (0–30 cm) using the fumigation-extraction method[17]. The plot sizes at Rambouillet and Grignon were 1000 m^2 and 600 m^2 respectively. At Rambouillet, the soil was drained at the 80 cm depth and the drain location allowed the drainage water from each plot to be collected separately at the outlet. The soil water dynamic at Rambouillet had been previously studied using stable isotope tracer techniques[18].

Climatic conditions were similar for the two experimental sites. The monthly rain and temperature during the experiment are reported for Grignon in Figure 1. A frost period occurred at the beginning of December 1989, immediately after the sewage sludge application. The water balance (rain - potential evapotranspiration) was negative between February and October. The climatic conditions after atrazine applications were hot and dry, followed by a dry summer with a monthly rain average of 35 mm.

Table 1 Main soil characteristics of Grignon and Rambouillet sites.

Soil site	*Depths (cm)*	*Carbon (g kg^{-1})*	*pH (water)*	*Clay (g kg^{-1})*	*Silt (g kg^{-1})*	*Sand (g kg^{-1})*
Grignon	0–30	11.1	7.3	220	730	50
(*Typic eutrochrept*)	30–60	5.9	7.6	230	730	30
	60–90	3.5	7.9	270	705	25
	> 90	3.2	8.3	205	772	23
Rambouillet	0–25	8.9	6.7	215	351	434
(*Aquic eutrochrept*)	25–35	9.2	7.0	265	329	406
	> 35	2.4	7.2	551	177	272

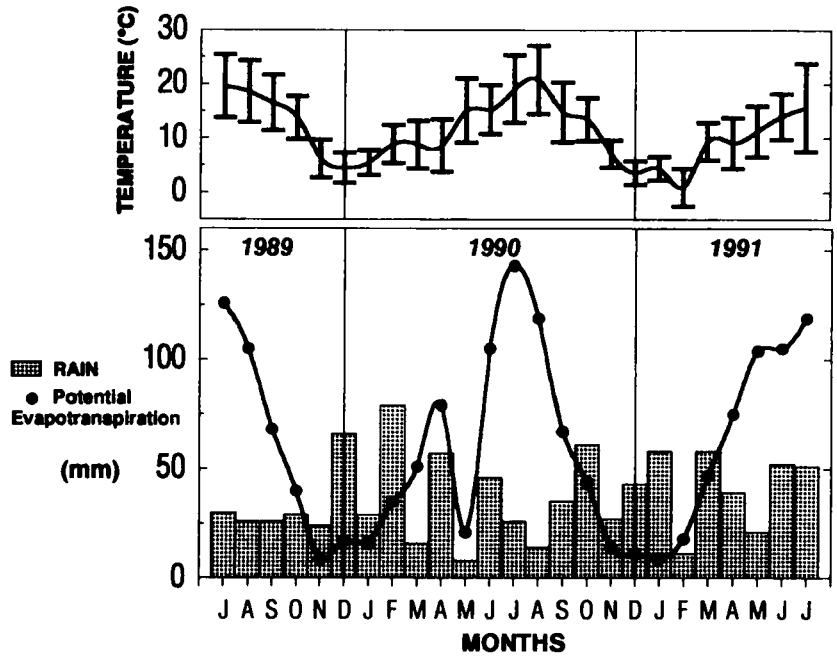

Figure 1 Climatic data (temperature, rain and potential evapotranspiration) from Grignon site during the experiment period.

Sewage sludges

Three types of sewage sludge were used. Two came from the sewage treatment plant of Achères (France). They were biological sewage sludges, with anaerobic methanical stabilisation, anf thermal treatment conditioning. We used the sewage sludge before thermal treatment (Achère Liquid sewage Sludge, ALS) and after thermal treatment (Achère Solid sewage Sludge, ASS). Only the first one was applied at Rambouillet. Another liquid sewage sludge was used at Grignon. It came from the municipal sewage plant of Plaisir (France) and was called Plaisir Liquid sewage Sludge (PLS). The main characteristics of the three sewage sludges are reported in Table 2. Sewage sludges were spread in November 1989. The applied amounts are given in Table 3. Control plots were untreated.

Table 2 Main sewage sludge characteristics of Achère solid (ASS) and liquid (ALS) sewage sludge and Plasir liquid sewage sludge (PLS).

	Dry matter	*C*	*N*		*N–NH$_4$*	*Cu*	*Pb*	*Cd*
Sewage sludge	*(%)*	*(g kg^{-1})**		*C/N*	*g kg^{-1}**	*(mg kg^{-1})**		
ASS	52.4	231	14	16	3	824	735	28
ALS	2.8	281	71	4	41	252	644	25
PLS	5.4	314	45	7	17	545	113	3

* Contents per kg dry weight

Table 3 Rate of sewage sludge application in Grignon and Rambouillet plots (t ha^{-1}).

Site		*Control*	*ASS*	*ALS*	*PLS*
Grignon	sewage	0	103	400	131
	dry matter	0	54.0	11.4	7.1
	carbon	0	12.5	3.2	2.2
Rambouillet	sewage	0	–	400	–
	dry matter	0	–	10.8	–
	carbon	0	–	3.0	–

Atrazine analysis

Atrazine (2-chloro-4-ethylamino-6-isopropylamino-1,3,5-triazine) was applied on April 30, 1990 at Rambouillet and May 9, 1990 at Grignon. The rate of application on each plot was calculated from amounts of atrazine collected on 24 glass sheets of 26 cm^2 each, placed on the soil surface during the spreading. The mean amount of applied atrazine was 1091 ± 110 g a.i. ha^{-1} at Rambouillet and 1021 ± 30 g a.i. ha^{-1} at Grignon.

The surface soils (0–10 cm) were sampled periodically during the 12 months, 7, 18, 36, 73, 163 and 350 days after the atrazine application at Rambouillet, and 2, 14, 30, 62, 150 and 352 days after the atrazine application at Grignon. At each date, 12 soil samples from each plot were collected and pooled. The atrazine was extracted in triplicate from the pooled soil sample. The soils were sampled at different depths (0–10, 10–20, 20–40 and 40–80 cm) 12 months after the atrazine application. All the soil samples were stored moist in glass containers and kept at -20°C until analysed. At Rambouillet, water samples were collected in the drains. No water could be sampled from April to December 1990 due to the dry climatic conditions.

Atrazine wa extracted from moist soil samples passed through a 2 mm sieve. Water content was measured at 110°C to express results on basis of dry soil. The extraction was performed with methanol at a soil: methanol ratio (weight : volume) of 1 : 2. Methanol extracts were obtained after 16 h shaking on a revolving agitator by filtration through Whatman papers N°5. The methanol extracts were evaporated under vacuum until dryness and the residues were dissolved in a known volume of toluene solution of metazachlor (2-chloro-N-(pyrazol-1-ylmethyl)acet-2,6-xylidide), used as an internal standard. Water samples were concentrated with C_{18}-BondElut cartridges. The cartridges were activated by prewashing with 5 ml of methanol and 5 ml of water. Water samples were passed through the activated C_{18}-cartridges under reduced pressure. The cartridges were dried with a flow of air for 15 min, and eluted with 5 ml of methanol. The extracts were evaporated to dryness and residues were dissolved in a toluene solution of internal standard.

The content of atrazine and dealkyl metabolites (deethylatrazine and deisopropylatrazine) in the toluene solutions were measured by gas chromatography with thermionic detector (Carlo Erba Ins. HRGC 5300). A fused silica capillary column (DB5, 30 m × 0.32 mm i.d., 0.25 μm film thickness, J. & W. Scientific) was used with He carrier gas at 1.7 ml min^{-1}. Samples were analysed with temperature programmation (80°C to 160°C at 20°C min^{-1}, then 160°C to 250°C at 5°C min^{-1} and 5 min at 250°C) using a splitless injector and a detector temperature of 250°C and 270°C respectively.

RESULTS AND DISCUSSION

Atrazine dissipation kinetics in the surface soil

Evolution of the atrazine contents in the surface soil at Grignon and Rambouillet are shown in Figure 2. In both soils, 20 to 40% of the applied atrazine were quickly dissipated during the 15 days after application, except in the Rambouillet ALS plot. The atrazine evolution in the different plots followed approximately first order kinetics until harvest in October. Atrazine dissipation graphs display some irregularities as frequently

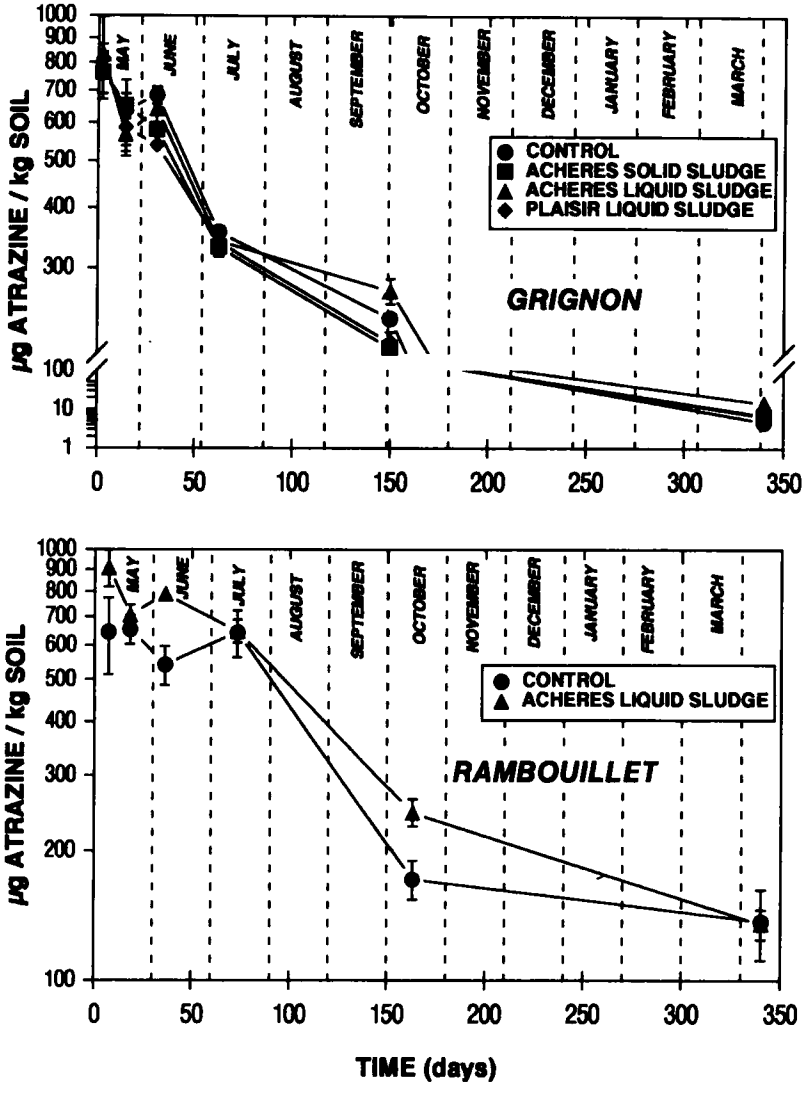

Figure 2 Dissipation kinetics of atrazine in the surface soils (0–10 cm) of Grignon and Rambouillet plots.

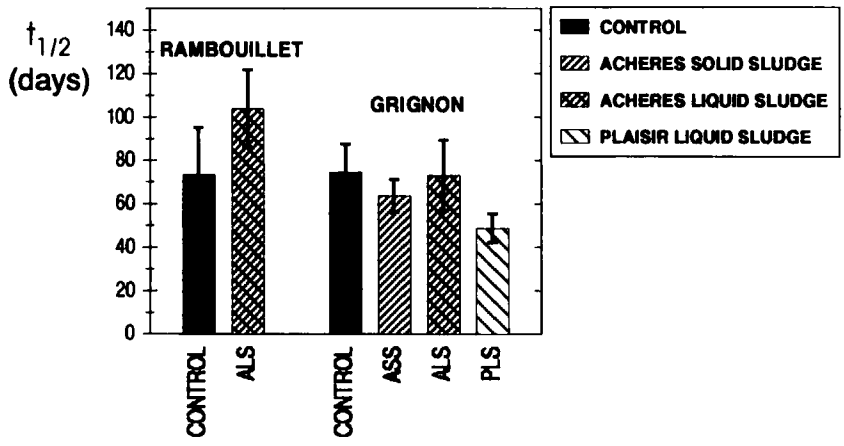

Figure 3 Atrazine half-life ($t_{1/2}$) estimation in surface soils (0–10 cm) of Rambouillet and Grignon plots. The dissipation kinetics were adjusted to first order kinetics.

reported in the literature. This could be due to time-variable atrazine extractability, or to but the heterogeneity of the sewage sludge and of the atrazine application.

At Grignon, the atrazine decreased quickly during the first 2 months: 20 to 30% of the applied atrazine was dissipated 14 days after the application. It disappeared more quickly in the PLS and ASS plots than in the ALS and control plots: after 5 months, at harvest date, more atrazine was found in the ALS plot. A comparison between dissipation rates can be done with half-life times calculated from the results obtained during the first months. Only in the PLS plot was the half-life time ($t_{1/2}$) significantly smaller than in the control plot : 48 ± 6 days in the PLS plot, 73 ± 16 days in the control plot (Figure 3).

Atrazine was more persistent at Rambouillet than at Grignon. In the control plot, 30 % of the applied atrazine was lost during the first week compared to only 5% in the ALS plot at Rambouillet (Figure 2). More atrazine was always found in the ALS plot than in the control plot showing that the liquid sludge treatment seemed to reduce the initial loss of atrazine and increase its persistence. The corresponding half-life times determined as indicated above, were significatively different at Rambouillet and Grignon, 73 ± 22 and 104 ± 18 days, respectively (Figure 3). At Grignon after 30 days, some differences appeared in the dissipation kinetics among the treatments; whereas at, Rambouillet, the ALS dissipation kinetics were already different from the control after only 7 days.

Atrazine volatilisation was not measured since it is likely to be of minor importance in the dissipation process of atrazine[19]. The longer atrazine persistence at Rambouillet than at Grignon could be related to a higher total microbial biomass at Grignon than at Rambouillet before the atrazine application. It represented 346 and 166 mg of C per kg of dry soil in the upper horizon respectively at Grignon and Rambouillet in October before sewage sludge spreading. It could be supposed that the amount of atrazine degrading micro-organisms was higher too, at least, those degrading by co-metabolism. Nevertheless, the atrazine degrading population could be similar in the two different soils, but its activity was mainly determined by the soil conditions[20]. The total soil biomass was not modified by sewage sludge treatment, and its mineralization was similar in the two soils during laboratory incubations[16].

Kinetics of atrazine dealkyl derivatives formation are shown in Figure 4. At Grignon, residues of these metabolites increased over approximately 14 days after the atrazine

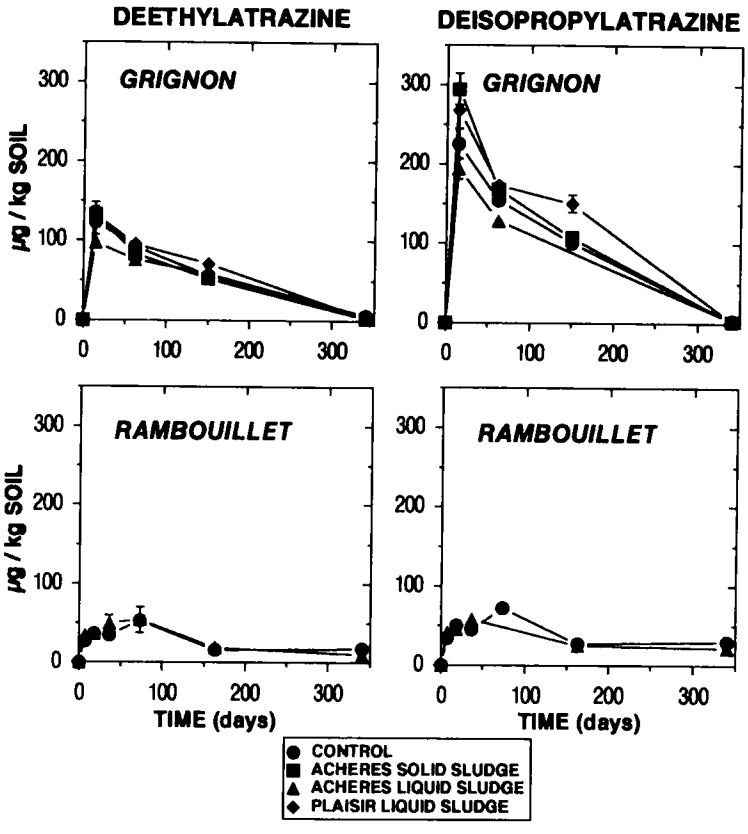

Figure 4 Evolution kinetics of deethylatrazine and deisopropylatrazine in surface soils (0–10 cm) of Grignon and Rambouillet plots.

application and then decreased continuously. At Rambouillet, on the contrary, dealkyl metabolites residues increased during the first 70 days. In both sites, the amounts of deisopropylatrazine was higher than those of deethylatrazine, mainly at Grignon. The presence of these metabolites indicates a biological degradation of the atrazine, whereas the presence of hydroxyatrazine would indicate a chemical degradation[25]. Usually more deethylatrazine than deisopropylatrazine is found because the ethyl chain is more quickly lost than the isopropyl, at least in laboratory experiments[21-23]. However, some micro-organisms produce atrazine dealkylation with preferential formation of deisopropylatrazine over deethylatrazine[24].

The dealkyl derivatives allow estimation of the importance of biological processes in the atrazine dissipation: an atrazine dissipation without dealkyl metabolites appearance could mean that the responsible processes were not biological. That is why the deethylatrazine and atrazine ratio (DAR), and the deisopropylatrazine and atrazine ratio (DiAR) in water or soils samples was used to describe the atrazine degradation process, and to decide if the water atrazine pollution was of a point or non-point nature[26,27].

At Grignon, the highest contents of the two dealkyl metabolites were always found in the PLS plot, the smallest in the ALS plot (Figure 4). The DAR was higher than 0.2 and increased slightly and progressively in the plots treated with the sewage sludges (Figure

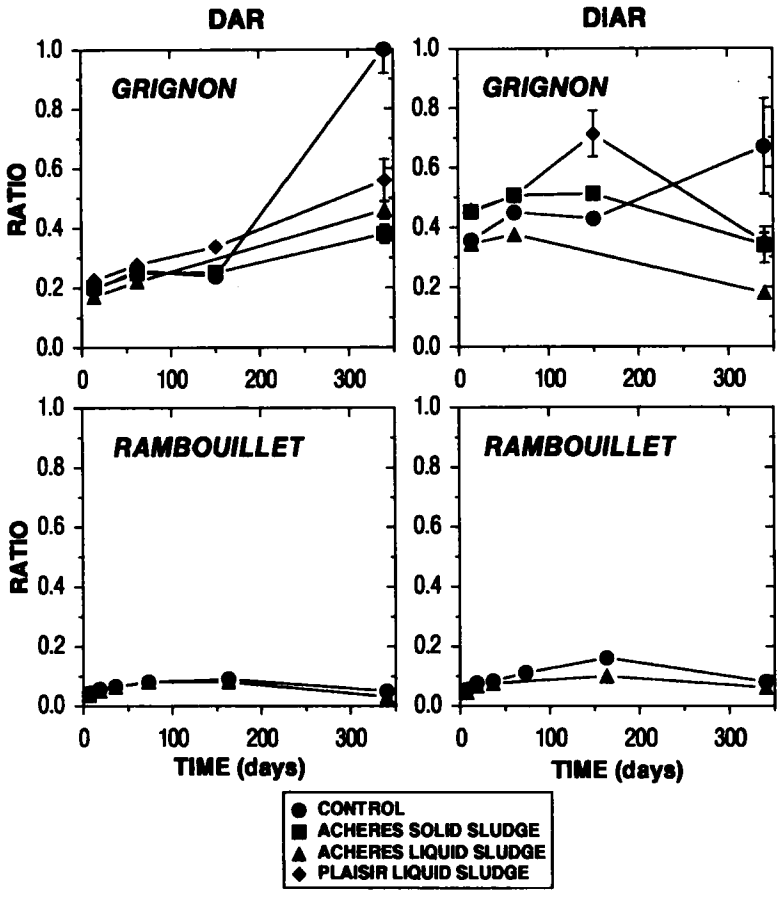

Figure 5 Evolution of the deethylatrazine - atrazine ratio (DAR) and the deisopropylatrazine - atrazine ratio (DiAR) with time in surface soils (0–10 cm) at Grignon and Rambouillet plots.

5). However, in the control plot, the DAR strongly increased to values larger than 1 at the end of the experiment, 12 months after the atrazine application. The DiAR also increased during 150 days and then decreased in the amended plots but still increased in the control plot. The highest DiAR were found in the PLS plot during the first month, and the smallest in the ALS one.

At Rambouillet, the dealkyl metabolites contents were not significantly different between the control and ALS plots (Figure 4). Similar DAR were found in the two plots; they remained smaller than 0.1, and slightly increased until 5 months (Figure 5). The DiAR and DAR evolution were similar. However, the DiAR were larger in the control plot than in the ALS plot.

At Rambouillet, the lower concentrations of dealkyl derivatives and the small DAR and DiAR values could indicate a less important biological degradation process compared to Grignon. Indeed, the mechanisms of atrazine degradation seemed different in the two sites. In each site, the sewage sludge treatment did not seem to modify them. The degradation pathways determine the fate of the residues in the environment. Chemical degradation of atrazine mainly produce the non phytotoxic hydroxyatrazine; it

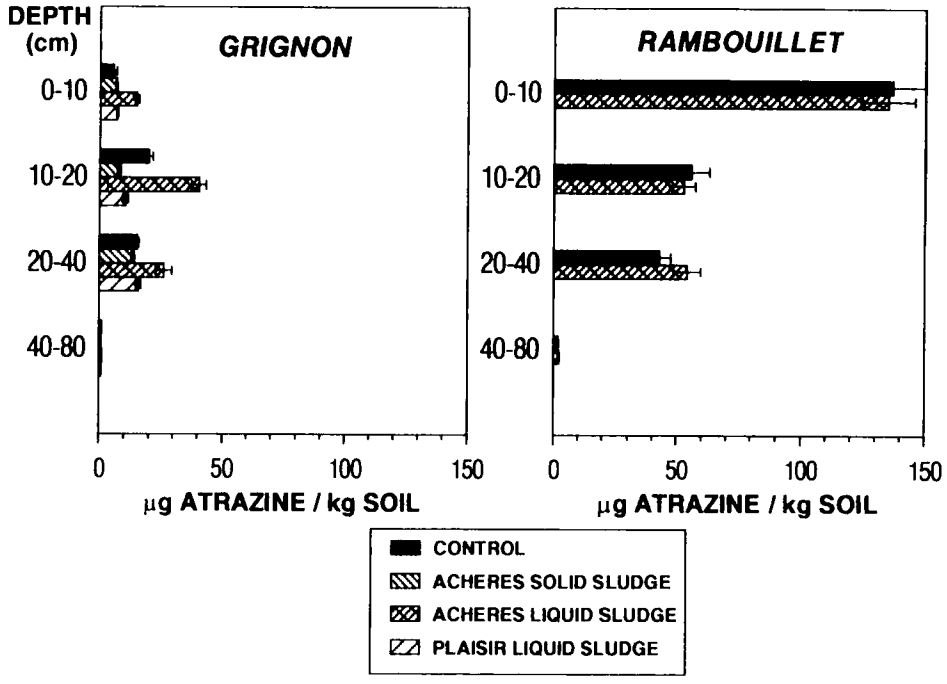

Figure 6 Distribution of atrazine content in the soil profiles of Grignon and Rambouillet plots, 12 months after the atrazine application.

is the most favourable intermediate for the mineralization of the triazinic ring to CO_2[28]. On the contrary, the dealkyl derivatives from the biological degradation of atrazine are also phytotoxic, although lesser than the atrazine[29]. On the other hand, removing the alkyl chains increases the intensity of the blocking processes occurring in the bound residues formation; the substitution of chlorine by a hydroxyl decreases it considerably[30].

Atrazine distribution in the soil profile 12 months after the application

Figure 6 shows the atrazine content distribution through the soil profile in the Grignon and Rambouillet plots. The atrazine dissipation in the upper horizon was more important at Grignon, and the atrazine was two to three times more persistent in the deep horizons of Rambouillet than in those of Grignon. The maximum amount of atrazine was found between 0 and 10 cm at Rambouillet, and between 10 and 20 cm at Grignon.

The atrazine persistence in the ALS plots at Grignon is greater at all depths than in the control plot. Similar results were not found with the other liquid sludge (PLS). The effect of sewage sludge varied with its nature and its origin. At Rambouillet, no effect of the liquid sludge was found in the upper horizons (0–10 and 10–20 cm); however, the atrazine amount was higher in the ALS plot below 40 cm. The ALS sludge seems to increase the atrazine transport at Grignon and at Rambouillet. This could be related to the migration of organic soluble materials from the sludge and is in agreement with laboratory experiments which showed that ALS sludge decreased atrazine adsorption on soil and favoured the desorption of atrazine previously adsorbed[7]. The longer persistence

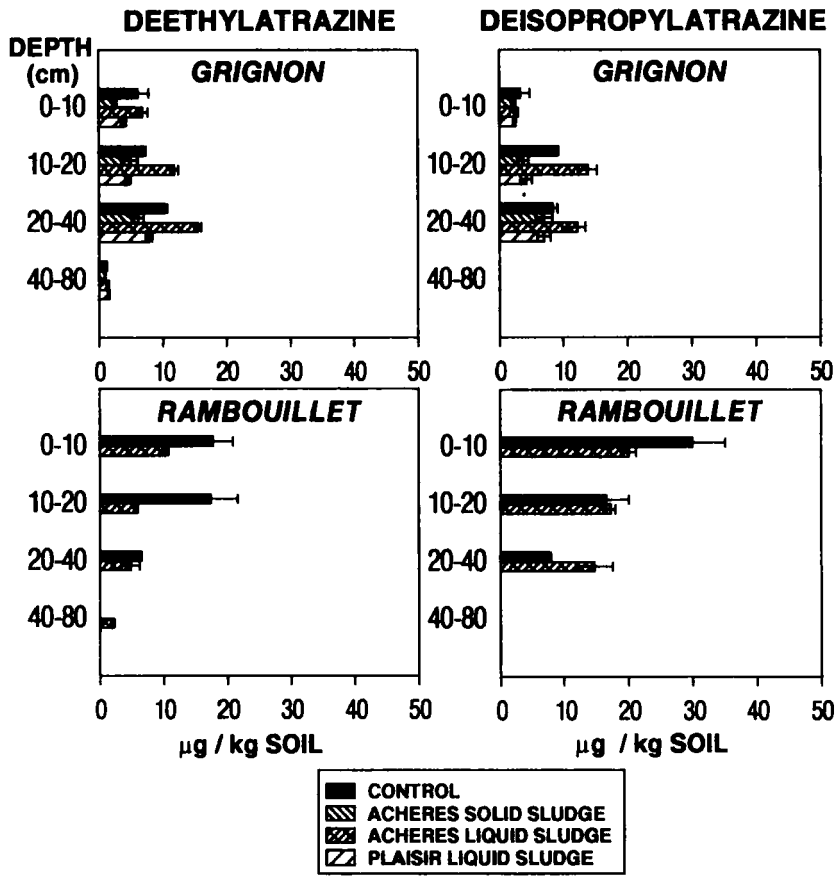

Figure 7 Distribution of deethylatrazine and deisopropylatrazine contents in the soil profiles of Grignon and Rambouillet plots, 12 months after the atrazine application.

of atrazine in the Rambouillet profiles, under hydromorphic conditions, could lead to important losses of atrazine to the water table when the water balance is positive. On the other hand, the ALS treatment favoured atrazine movements in soils, and could increase its soil persistence as the degradation decreased with depth[31–33]. In the hydromorphic Rambouillet soil, it could be more important because the atrazine degradation was slower under anaerobic conditions[23].

The distribution of the atrazine dealkyl metabolites in soil profiles was comparable to that of atrazine (Figure 7). Thus, as for the atrazine, the ALS treatment increased the persistence of these metabolites in the soil profile at Grignon. Amounts of deethyl- and deisopropylatrazine were comparable in all the soil samples; however, below 40 cm, only deethylatrazine was detected. At Rambouillet, the deethylatrazine content was higher in the control plot than in the ALS plot, until 40 cm, where it was only detected in the ALS plots. Deisopropylatrazine content was higher than deethylatrazine in Rambouillet plots, but it was not detected below 40 cm. In the Rambouillet control plot, deisopropylatrazine contents decreased more quickly than in the ALS plots: its content was higher in the control plot until 10 cm and then became higher in the ALS plots.

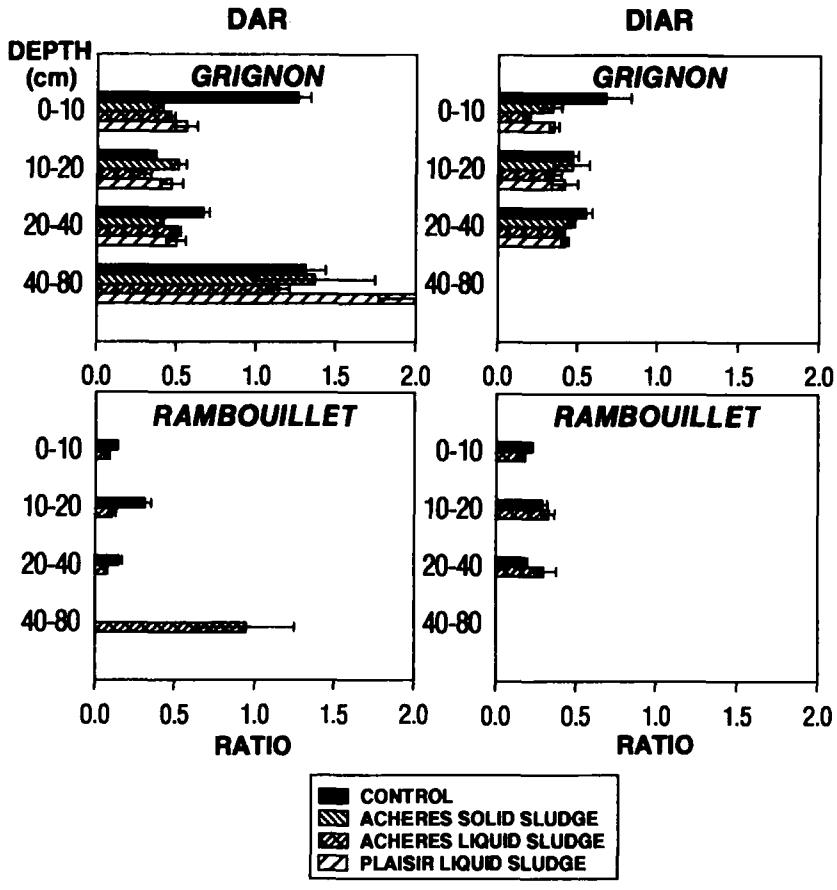

Figure 8 Variation of the deethylatrazine - atrazine ratio (DAR) and the deisopropylatrazine - atrazine ratio (DiAR) with depth in Grignon and Rambouillet plots, 12 months after the atrazine application.

Figure 8 shows the DAR evolution with depth in both sites. At Grignon, the DAR increased with the depth and reached values larger than 1 between 40 and 80 cm. These high DAR values showed an important biological degradation with a preferential migration of deethylatrazine. The higher migration of deethylatrazine as compared to that of atrazine may be related to its smaller adsorption in soils[34].

Atrazine and its dealkyl derivatives in drainage water sample

Climatic conditions during the sampling period did not allow collection of water in drains between April and December and monitoring was done from December to March, and then from January to May (Figure 9). The water sampling started 10 days after the sewage sludge application in November, 1989 and atrazine recovered during the first period corresponded to a treatment done by the farmer during the preceding corn cultivation. The atrazine found during the second period came from the treatment done in this experiment. The atrazine concentration was very variable but two general trends

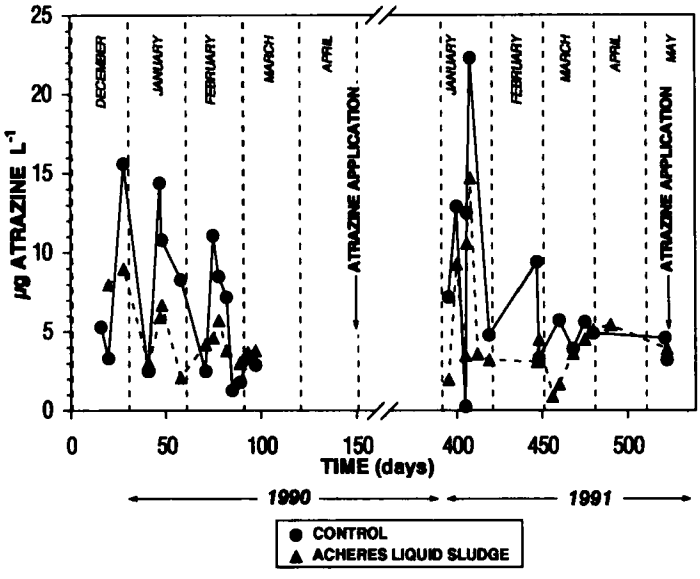

Figure 9 Atrazine concentration in drainage water of Rambouillet plots at different dates.

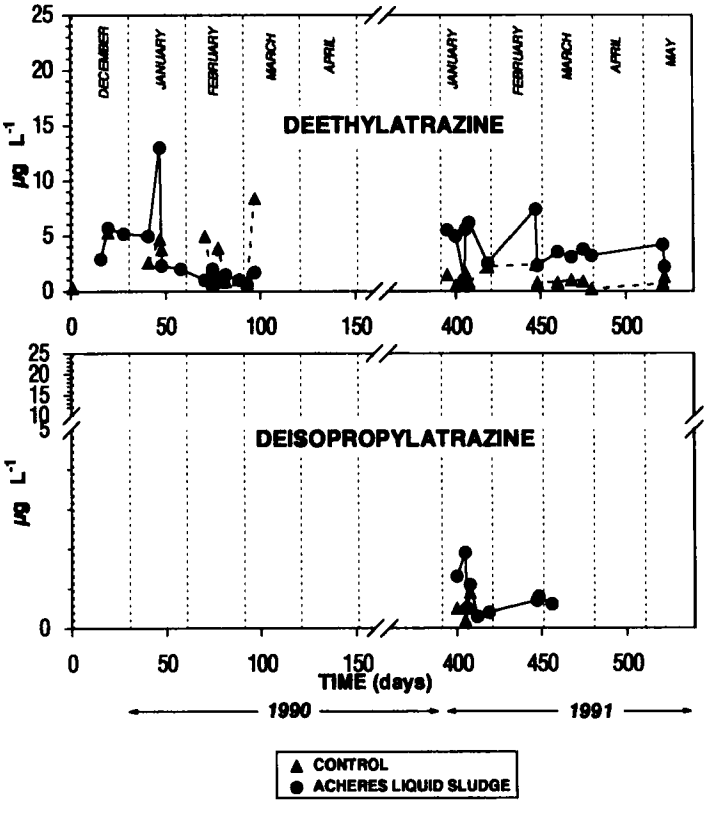

Figure 10 Contents of deethylatrazine and deisopropylatrazine in drainage water of Rambouillet plots at different dates.

could be observed. The concentration was always higher at the beginning of the drainage period, probably due to some piston-flow effect as suggested by the transfer characteristics of $^{18}O^{18}$. In the sludge treated plot, the concentration of atrazine was often lower than in the control plot while the amounts of atrazine remaining in the soil profiles were similar. Dealkyl atrazine metabolites were looked for in water samples (Figure 10).

Deethylatrazine was found in all the samples, in similar concentrations as atrazine. No difference was found between the two plots during the first drainage period. During the second one, the deethylatrazine amount was always higher in water from the control plot. The water content of deisopropylatrazine was smaller than that of deethylatrazine, which is classically reported[35,36]. It was only measurable during the second drainage period, and the amounts were higher in the control plot. These observations may be related to adsorption properties of atrazine dealkyl derivatives, which the adsorption coefficients follow the order: deethylatrazine<atrazine<deisopropylatrazine<hydroxyatrazine[33].

The DAR evolution in the drainage water samples is shown in the Figure 11. When the drainage started, the DAR in water samples from the control plot was higher than 1, which is usually found[27]. During the first drainage period, the DAR decreased with the time, and became smaller than 1 after February, which indicated a more important contribution of atrazine residues, probably from the surface soil. The smaller DAR in the ALS water samples could indicate the mobilisation of residues from the soil surface as the DAR was smaller in the upper horizons than in the deep ones. This mobilisation would be preferentially for deethylatrazine rather than for atrazine. No important biological degradation could occur because of the unfavourable climatic conditions. During the second drainage period, the DAR in water samples from the control plot were always higher than those of the ALS plot. Most often, they were higher than 1. It showed

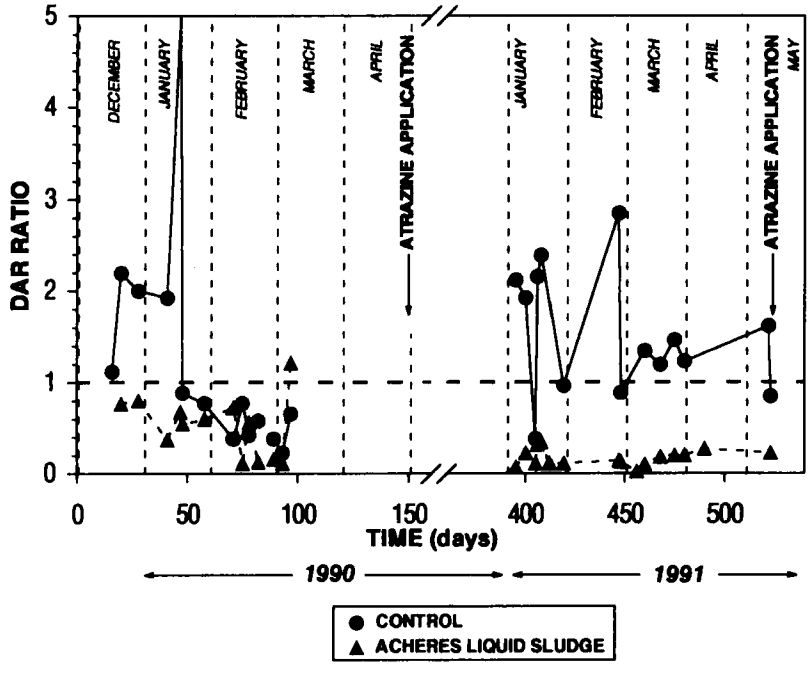

Figure 11 Evolution of deethylatrazine atrazine ratio (DAR) in water samples of Rambouillet plots.

that the atrazine was less degraded in the ALS plot than in the control plot which agreed with the dissipation kinetics of atrazine in soils from the two plots.

It was suggested[27] that the DAR could be used to assess the origin of the water pollution, a value greater than 1 corresponding to a non-point pollution and a value less than 1 to a point pollution. The greater the resident time in the soil, the more important is the biological degradation and thus the greater is the DAR. The results reported in Figure 11 showed that such a statement must be handled with care. Observed values of DAR in water samples from the sludge treated plot are less than 1 although there is non-point pollution.

CONCLUSION

The atrazine behaviour in soil may be modified by exogenic organic matters application. Both degradation and transport were affected. The effects of different kinds of sewage sludges applied before atrazine spreading depended on the soil and the sludge properties but could not be completely explained.

The long term persistence of atrazine was not changed while the dissipation kinetics of atrazine were modified, and in all the treatments, no modification of the herbicide efficiency was observed.

The evolution of the water drainage composition indicated that the transport of atrazine appeared to be reduced after a sewage sludge application. However, it was difficult to separate the sludge effect on the adsorption/desorption process and on the water flow characteristics.

Since non-point pollution of underground water is generally due to small amounts of chemicals, any practice modifing the behaviour of a soil applied chemical, should be considered carefully. Concerning sewage sludges, the impacts of added organic matter on the atrazine degrading soil microflora needs further research.

The atrazine dealkyl metabolites gave indications of the degree of biological degradation of the atrazine, particularly the ratio of their contents in relation to the atrazine content. Small differences were found among the treatments; only the soil samples of the control plots 12 months after the atrazine application showed a biodegradation degree higher than the soils from plots treated with sewage sludge. The biodegradation degree of the atrazine residues in waters from the Rambouillet ALS plot was smaller than these of the control plot; that could indicate modifications in the atrazine degradation, but selective transport modifications of the chemical could not be discarded.

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