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Int. Agrophys., 2014, 28, 93-100
doi: 10.2478/intag-2013-0031

Influence of soil tillage and erosion on the dispersion of glyphosate and aminomethylphosphonic acid in agricultural soils

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Received July 2, 2013; accepted November 27, 2013

Abstract. Erosion processes can strongly influence the dissipation of glyphosate and aminomethylphosphonic acid applied with Roundup Max® in agricultural soils; in addition, the soil structure state shortly before erosive precipitations fall can be a key parameter for the distribution of glyphosate and its metabolite. Field rain simulation experiments showed that severe erosion processes immediately after application of Roundup Max® can lead to serious unexpected glyphosate loss even in soils with a high presumed adsorption like the Cambisols, if their structure is unfavourable. In one of the no-tillage-plot of the Cambisol, up to 47% of the applied glyphosate amount was dissipated with surface run-off. Moreover, at the Chernozem site with high erosion risk and lower adsorption potential, glyphosate could be found in collected percolation water transported far outside the 2x2 m experimental plots. Traces of glyphosate were found also outside the treated agricultural fields.

Keywords: glyphosate, aminomethylphosphonic acid, erosion, adsorption, soils

INTRODUCTION

There is growing concern about identifying and understanding the mechanisms that control the fate of chemicals as a source of environmental contamination, especially in soils and water. Therefore, there is a need for more specific pesticide management based on the adaptation of the pesticide type and application rates to the characteristics of the area of application (Peruzzo *et al.,* 2008).

Glyphosate, among the non-selective herbicides, is one of the most applied organophosphonates worldwide with 60% of the global sales (Candela *et al.*, 2007). The major sustainable agricultural systems in South America and USA are fundamentally based on a technological package that combines no-till and glyphosate in the cultivation of maize (Aparicio *et al.,* 2013; Cerdeira and Duke, 2006; Christoffoleti *et al.,* 2008.) The no-tillage system (direct drill) is becoming more and more important also in the Austrian agricultural practice, tending to substitute the plough. This leads to a considerable growth of weeds within the NT-plots (about 80-100% of weed cover), which would seriously impede the germination of maize plants; therefore, Roundp Max® is used.

The active ingredient in Roundup Max®, a post-emergency non-selective broad spectrum herbicide, is glyphosate (N-(phosphonomethyl)glycine). As a polar, highly water-soluble substance, glyphosate binds in different ways to soil components (Eberbach, 1997; Gimsing *et al.*, 2004; Ghanem *et al.*, 2007) but can be degraded, desorbed, or leached out of the soil. Traces of glyphosate and its major degradation product aminomethylphosphonic acid (AMPA) were found in surface-water and groundwater environments (Degenhardt *et al*., 2012; Gimsing *et al*., 2004; Landry *et al*., 2005; Peruzzo *et al*., 2008).

Like for many other organic contaminants, the distribution of glyphosate in soils and the environment is strongly governed by different soil constituents and processes as well as by specific local site conditions (Eberbach, 1997; Gimsing *et al.,* 2004; Mamy *et al.,* 2005; Soulas and Lagacherie, 2001). The investigation of the pathways of glyphosate and AMPA in the environment and their interaction with different soils is therefore of major interest for assessing their

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environmental impact, but sometimes controversial. It seems to be that particularly pedogenical Fe-(and Al-) oxides have a strong capacity of glyphosate adsorption through covalent bonds (Gimsing and Borggaard, 2002; Pessagno *et al.*, 2008; Rampazzo *et al*., 2013); therefore, the transport of glyphosate and AMPA from land to water environments seems very limited.

Regarding the pathway of leaching-preferential flows, the sources and transport routes of pesticides to groundwater are complex. In the study of Rueppel *et al.* (1977) glyphosate was considered as a pesticide with no propensity for leaching. On the other hand, in certain soils and under different conditions, there is a risk for groundwater contamination as it is shown in some other investigations (Landry *et al.,* 2005).

Since glyphosate is highly water soluble, an additional risk of dissipation of glyphosate and AMPA in the soil-water environment is the herbicide application at sites and soils susceptible to serious erosion processes.

Nevertheless, the knowledge about loss of glyphosate and AMPA through surface run-off, especially following extreme erosive precipitation events very soon after herbicide application before the plants can absorb it is insufficient and the importance of this transport as related to ecological risk assessment has previously been scarcely investigated, (Borggaard and Gimsing, 2008; De Jonge *et al.,* 2000; Donald *et al.,* 2005; Giesy *et al.,* 2000; Peruzzo *et al.,* 2008; Zaranyika and Nyandro, 1993).

Therefore, the aim of this work was to investigate the influence of heavy precipitations falling very shortly after application of Roundup Max® (worst case scenario) in field conditions on the dissipation of glyphosate and AMPA. In addition, the effect of different tillage systems was investigated.

MATERIAL AND METHODS

The experiments were carried out at two agricultural fields (Pixendorf, Lower Austria and Kirchberg, Styria) where the following soil tillage systems were compared in 3 field replications:

- conventional tillage (CT) with plough with and without cover crop during winter period;
- no-tillage (NT) with cover crop during winter period.

The investigated soils were a Chernozem from loess at Pixendorf and a sandy stagnic Cambisol from tertiary carbonate free sediments at Kirchberg (WRB, 2006).

In order to investigate the influence of erosion and tillage on glyphosate and AMPA, two rain simulation experiments were conducted in 3 field replications (1, 2, 3) within the conventional tillage (CT)- and the no-tillage (NT)-plots. For this, Roundup Max® was applied onto rain simulation soil plots (2x2 m, delimited by metallic fences inserted into the soil and with a downslope run-off collection outlet) according to the common agricultural practice (2% herbicide solution, corresponding to 4 l Roundup Max® diluted in 200 l deionized water corresponding to 180 mg glyphosate m^{-2}). In both sites, the vegetation cover degree was typically higher in the NT-plots (80-100% of weed cover) than in the CT-plots (only few yield residues of maize) and the application was carried out in sunny and not windy weather shortly before starting the rain simulation experiment (worst case scenario). The average slope in both sites was 12-15% at the Cambisol and 10% at the Chernozem. Both sites are known as rather erodible. The soil surface of the Chernozem immediately before the rain simulation was crumby; in turn, the cambisol had a crusted, dry, and cracky surface.

The rain simulator was designed as a potable out- and indoor equipment, the spray pattern was generated by full jet nozzles, the rain fall intensity was controlled with intermittent spraying. The opening cycles of the solenoid valves were fully programmable with computing equipment (Strauss *et al.*, 2000). The crucial elements for the construction of the simulator were:

- nozzles (½ HH30WSQ, ½ HH40WSQ, Spraying System, USA);
- solenoid valves: 220-V, 3/2 way (131, Bürkert, Germany);
- pressure regulators: P16A, Wilkerson Corp., USA;
- water suction pumps (0830 Semadeni, Ostermundingen, Switzerland);
- steering interface and steering programme, self-made equipment;
- drop size distribution: median volumetric size 2.1 mm, kinetic energy: 17 J mm^{-1}.

During 60 min of rain simulation with 30 mm, run-off-fractions were collected at different time intervals at the Chernozem and averagely at the Cambisol and cooled in boxes. In the laboratory, the run-off samples were immediately centrifuged in order to separate the liquid from the solid phase. Immediately after the rain simulation, soil samples were collected within the simulation soil plots of 2x2 m at different depths (0-2, 2-5, 5-10, 10-15, and 15-20 cm at the Chernozem and at 0-2 and 2-5 cm at the Cambisol). Glyphosate and AMPA were analyzed according to Rampazzo *et al.*, 2013. All physical and chemical analyses on soil samples were carried out according to the standard methods.

RESULTS AND DISCUSSION

The Chernozem shows the development from loess with typical silty texture (topsoil 0-20 cm, 12% clay, 65% silt, 23% sand, pH 7.3, 15% $CaCO_3$ and 3% OM), whereas the Cambisol is a loamy sandy soil (topsoil 0-20 cm, 14% clay, 33% silt, 53% sand, pH 5.7, no $CaCO_3$ and 3% OM).

Previous investigations (Rampazzo *et al*., 2013) showed that pedogenic Fe-oxides (dithionite and oxalate-extractable Fe) seem to be important absorbers for glyphosate in soils. The Chernozem exhibited a low content and the Cambisol a high content of Fe oxides and therefore the expected sorption capacity for glyphosate and AMPA was theoretically higher at the Cambisol (Table 1).

Figure 1 shows the amount of total (liquid and solid) run-off after the rain simulation experiments on the Chernozem. Before glyphosate and AMPA were analyzed, a separation of the solid and liquid run-off phase in the laboratory was carried out.

The CT-plots produced the highest run-off amounts because of their lower protecting weed cover, causing a splash of the surface by the erosive precipitation with consequent loss of infiltrability. On the other hand, the amount of run-off at the Chernozem was 10 times lower than the Cambisol because of its crumby structure with a better infiltration rate during the rainfall simulation, whereas the soil surface of the Cambisol was compacted and crusty. The different amounts of run-off between the 3 field replications of the Chernozem (Fig. 1) were due to the inhomogeneity of the field conditions. Consequently, the total amount of glyphosate washed out of the plots by liquid run-off at the Chernozem was much higher in the CT-plots than in the NT-plots (Fig. 2).

A fractionation of the time-dependent glyphosate contents in run-off-fractions of the Chernozem at time intervals of 15 min is shown in Fig. 3a. As it was expected, the first fraction showed the highest contents in both variables CT and NT and then decreasing with time. The CT-plots showed again higher glyphosate contents than the NT-plots, which instead showed higher glyphosate concentration (less dilution) at the same time (Fig. 3b).

According to Gjettermann *et al.,* 2011, desorption kinetics are important for evaluating the significance of dissolved and particle-facilitated transport of glyphosate. Consequently, the separation from water and solid phases should be done within a short time of minutes. We managed to do this within 30 min from field sampling. The contents of glyphosate and AMPA in the solid phase of run-off in the Chernozem are shown in Figs 3c,d.

The glyphosate contents retained by the run-off sediment is an analogue to that in the total and fractionated run-off (Figs 2, 3a), where the first collected fraction of run-off sediment contains the highest amounts of glyphosate which then generally decreases in the following fractions and the CT-plots shows higher amounts than the NT-plots. Analogous is the distribution of AMPA in the sediment (Fig. 3d). Since the loss of glyphosate by run-off was higher in the CT-plots (Fig. 3a), the amount of glyphosate and AMPA adsorbed by the Chernozem immediately after the rain simulation experiments was consequently higher in the NT-plots (Fig. 4).

Moreover, there is a clear depth function of the adsorption of glyphosate and AMPA through the soil immediately after Roundup Max® application and rainfall simulation at the Chernozem. The glyphosate and AMPA contents clearly

T a b l e 1. Fe-oxide distribution in the investigated soils

Site	Soil type (WRB)	Depth (cm)	Fe_o (mg kg^{-1})	Fe_d (mg kg^{-1})	Fe_o/Fe_d
Pixendorf	Chernozem	0-5 5-20	983 1 040	7 970 8 378	0.12 0.12
Kirchberg	Cambisol	0-5 5-20	3 422 3 726	14 843 15 032	0.23 0.25

Fe_o – amorphous (weakly crystallized) Fe-oxides, oxalate-soluble; Fe_d – well crystallized Fe-oxides, dithionite-soluble.

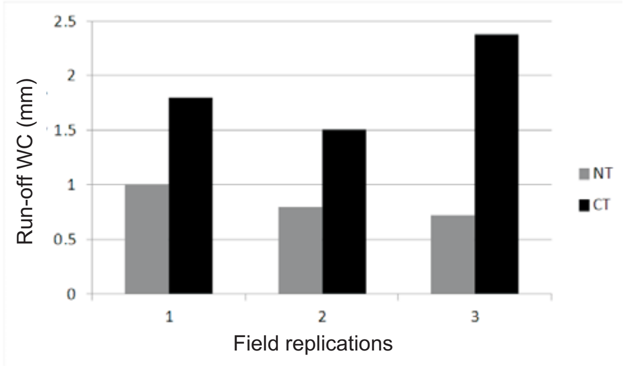

Fig. 1. Chernozem: total run-off of the conventional tillage (CT) and no-tillage (NT) plots in the 3 field replications, WC – water column.

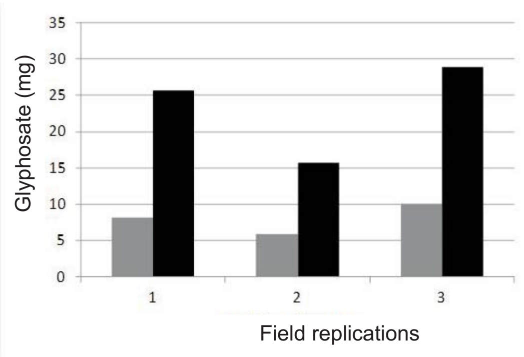

Fig. 2. Chernozem: total amounts of glyphosate in liquid run-off at the 3 field replication plots. Legend as in Fig. 1.

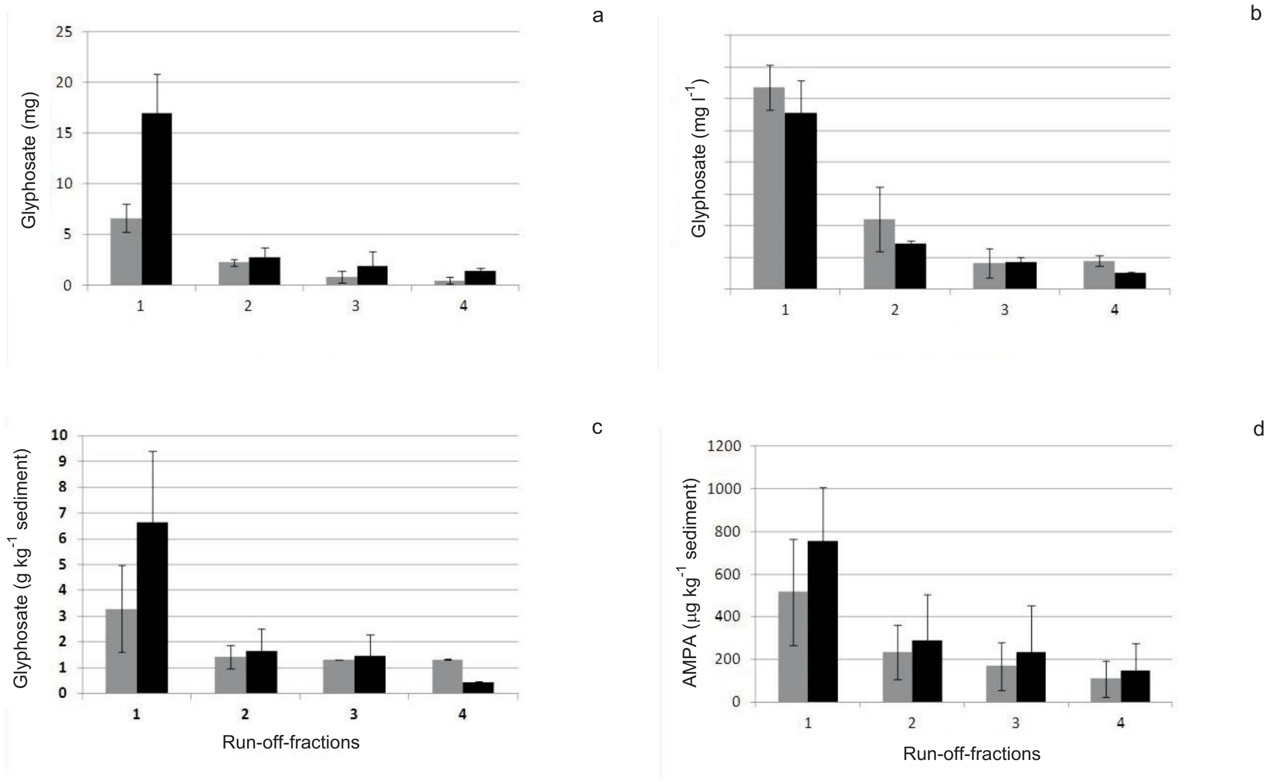

Fig. 3. Chernozem: a – glyphosate amount, b – glyphosate concentrations in liquid, and c – glyphosate contents, d – AMPA contents in the solid phase of run-off-fractions at 15-min intervals (average of 3 field replications). Legend as in Fig. 1.

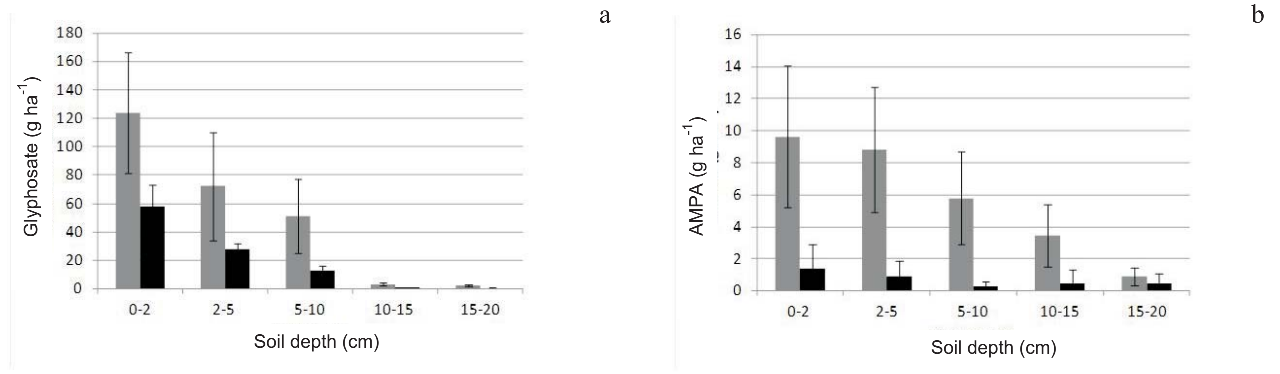

Fig. 4. Chernozem: a – glyphosate contents, and b – AMPA contents in the soil within the rain simulation plots (average value from the 3 field replications). Legend as in Fig. 1.

decreased with soil depth. As shown in a previous study by Mamy *et al.* (2005), this could be explained because the two compounds are very quickly adsorbed by the soil compounds, probably also depending on the physical soil conditions and water flow during rainfall. In fact, the Chernozem had a favourable crumby structure in the NT-plots, with no cracks, no preferential flow, and optimal conditions for water retention in the upper soil layers at the moment of the rainfall simulation experiment, so that more than 50% of the adsorbed glyphosate was retained in the first 5 cm of the soil. The fact that AMPA could already be detected 1 h after the Roundup Max® application underlines the quick glyphosate degradation in soil, as reported by Mamy *et al.* (2005) as well.

The total (liquid and solid) amount of surface run-off in the Cambisol is shown in Fig. 5. The Cambisol had a dry, crusty, and very deeply cracky soil surface of the CT-plots before starting the rainfall simulation and therefore the first

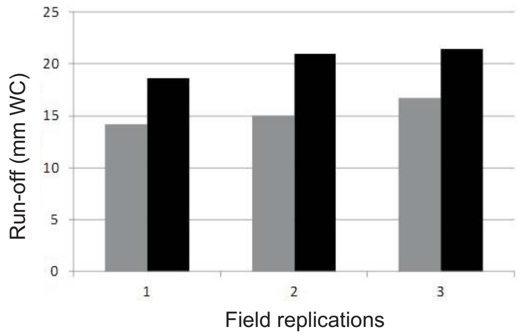

Fig. 5. Cambisol: total run-off of the CT- and NT-plots in the 3 field replications, mm WC – mm water column. Legend as in Fig. 1.

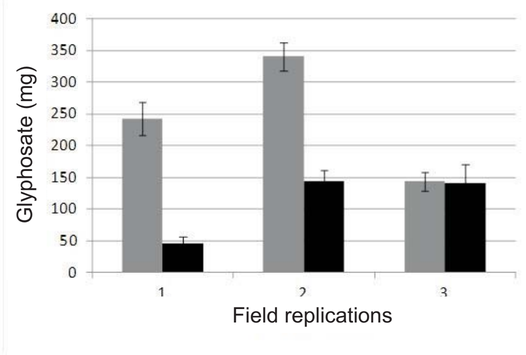

Fig. 6. Cambisol: total amounts of glyphosate in liquid run-off at the 3 field replication plots. Legend as in Fig. 1.

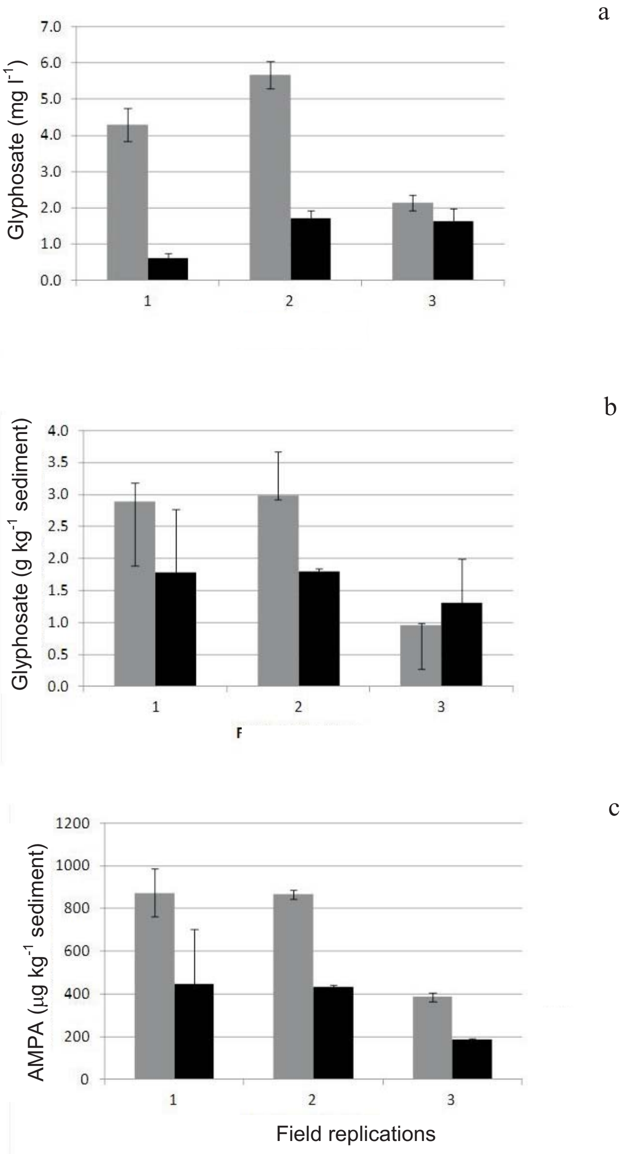

Fig. 7. Cambisol: a – glyphosate concentrations in liquid, b – glyphosate, and c – AMPA contents in the solid phase of run-off at the 3 field replications (average of the 60 min rain simulation). Legend as in Fig. 1.

amount of the precipitation quickly infiltrated in the cracks, but very soon a splash process and loss of infiltration took place due to the fine sandy texture and low surface protection by weeds. This led to a higher surface run-off of the CT-plots than the NT-plots.

Consequently, Figs 6 and 7a show that the contents and concentrations of glyphosate in the liquid run-off of the NT-plots of the Cambisol were much higher than in the CT-plots. In the dry and cracky soil surface of the CT-plots, it took some time before run-off started and glyphosate could easily enter deeper into the soil; on the other hand, the NT-plots had a nearly 100% weed cover, as reported also by Locke and Bryson (1997); consequently, this might buffer potential effects of glyphosate in the soil (Locke *et al.,* 2008). In this study, most of the applied glyphosate adhered to the photosynthetically active plant organs (stem and leaves) immediately after application; consequently, glyphosate was literally washed out of the 2x2 m simulation plots with run-off and had less time to infiltrate the soil surface (Fig. 8a).

Based on the high content of pedogenical Fe-oxides (15 000 mg Fe$_d$ kg^{-1}, Table 1), high soil adsorption of glyphosate was expected for the Cambisol. The surprisingly high loss of glyphosate by surface run-off (in one of the 3 field replications about 47% of the applied glyphosate) measured in this study confirmed the crucial effect of soil structure and preferential flow on the dissipation of glyphosate after heavy erosive precipitations, which were also be observed by other scientists (Borggaard and Gimsing, 2008; De Jonge *et al.,* 2000; Gjettermann *et al.,* 2009; Siimes *et al.,* 2006, Vereecken, 2005).

The contents of glyphosate and AMPA in the solid phase of run-off at the Cambisol are shown in Figs 7b, c, respectively.

The concentrations of glyphosate and AMPA in the solid phase of run-off at the Cambisol are similarly distributed to the corresponding aqueous fractions of run-off; they are mostly higher in the NT-plots than in the CT-plots.

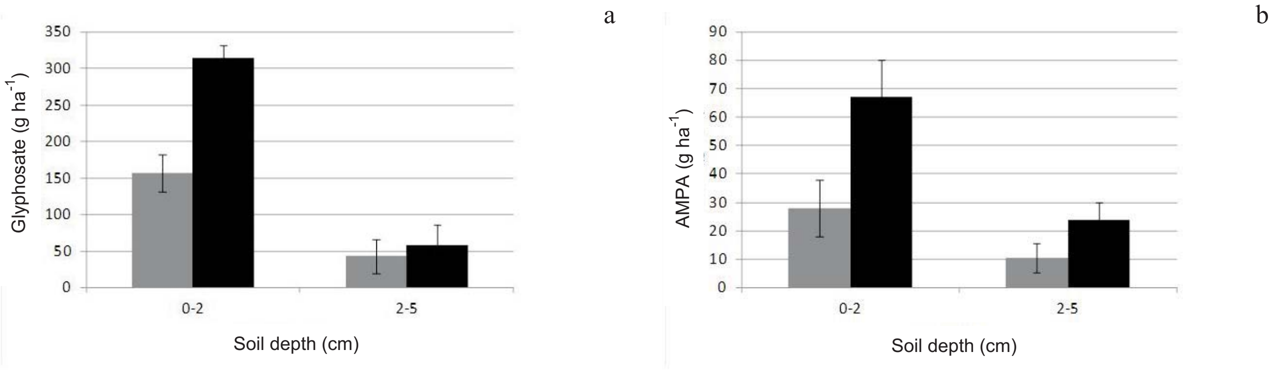

Fig. 8. Cambisol: a – glyphosate and b – AMPA contents in the soil within the rain simulation plots (average value from the 3 field replications). Legend as in Fig. 1.

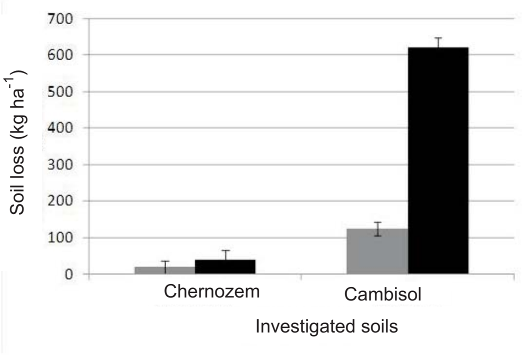

Fig. 9. Total soil loss of the investigated soils after the rain simulation experiments (averages of 3 field replications). Legend as in Fig. 1.

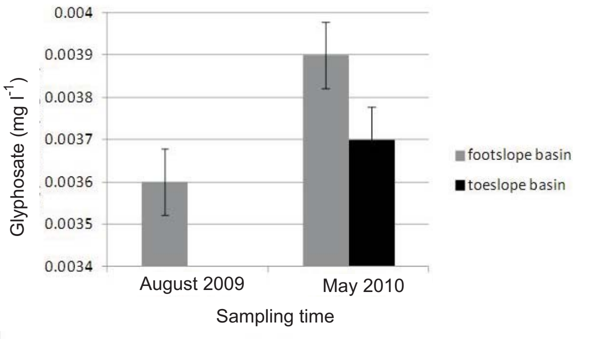

Fig. 10. Glyphosate concentrations in natural run-off retention basins outside the experimental fields.

Figure 8 shows the content of glyphosate and AMPA adsorbed by the soil immediately after the rain simulation experiments at the Cambisol.

Immediately after the rain simulation experiment, a very clear distribution in the soil appears: glyphosate and AMPA are first adsorbed in the upper 0-2 cm of the soil and only a small amount reaches the next soil depth of 2-5 cm. In general, the NT-plots show a clearly lower content of glyphosate and AMPA as compared to the CT-plots. This is explained by the respectively higher glyphosate contents in run-off of NT-plots (Fig. 6).

The soil losses of the Chernozem and Cambisol through erosion processes are shown in Fig. 9.

At both sites, the soil loss from the CT-plots, measured as sediment in the surface run-off, was higher than from the NT-plots because of the much lower vegetation cover before the simulation experiment, splash, and reduction of infiltration. The loss of the Cambisol soil was 10 times higher than that of the Chernozem. The reason for this is that the two experimented soils had a completely different soil structure and surface conditions before starting the rain simulation. The Chernozem had a very friable, crumby, permeable structure after the wheat yield. The Cambisol stood right after the corn yield, the soil surface was crusty and less permeable, except for shrinking cracks which swelled during the experiment.

The Chernozem at Pixendorf and surroundings is generally known as a location with high erosion risk because of the high silt amount (> 60 mass %) and especially with corn crop, where deep gully erosion forms. The erosion rills discharge downslope to an artificial run-off retention basin at the footslope of the experimental field. This basin can run over and flow downwards on different paths and is collected through further toeslope retention basins. Water samples from both retention basins were analyzed and traces of glyphosate, which were surely not connected with rain simulation experiments, were found (Fig. 10).

Moreover, soil percolation water samples at the footslope of the experimental field were collected at two depths from previously installed stations and analyzed for glyphosate and AMPA. Since the rainfall simulation experiment was conducted at the topslope whereas the percolation water samples were collected at the footslope (100 m distance) at the same time, it seems unlikely that the measured amounts of glyphosate and AMPA were influenced by the rain

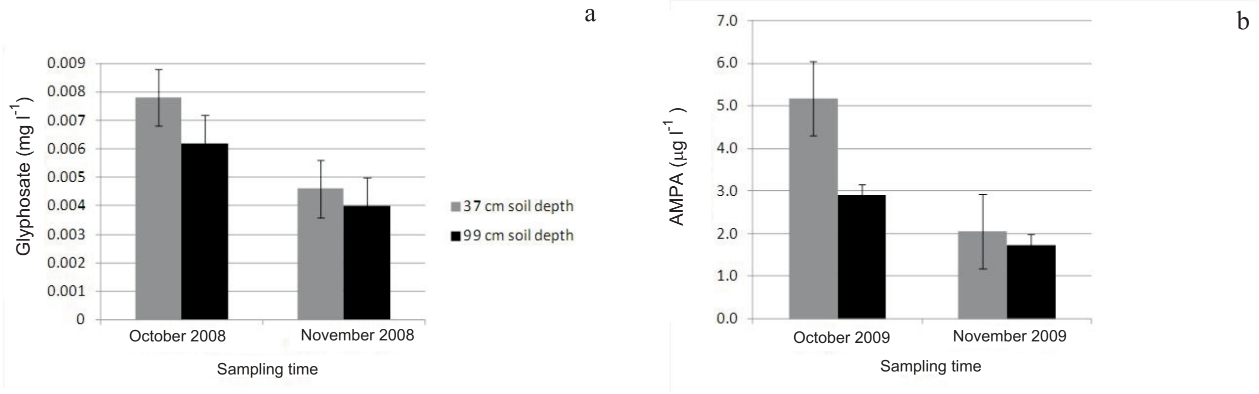

Fig. 11. Concentrations of: a – glyphosate, and b – AMPA in percolation water at 2 different times and soil depths.

simulation, but are probably residual amounts from previous field application, confirming the possibility of dissipation through natural processes.

The results show that small amounts of glyphosate and its metabolite can dissipate through soil percolation, mainly depending on the physico-chemical adsorption and structural properties of soils, and were found in collected soil solutions at two different depths far out of the 2x2 m precipitation plots (Fig. 11).

CONCLUSIONS

1. The rain simulation experiments clearly showed that even in a potentially high glyphosate adsorbing soil like the Cambisol, erosion and surface run-off can lead to severe glyphosate loss if the soil structure state *eg* compaction degree, crusting, infiltrability, pore size distribution, in the case of erosive precipitations shortly after Roundup Max® application, is unfavourable. In this study, in one of the NT-plot repetitions, up to 47% of the applied glyphosate amount were dispersed with run-off.

2. Traces of glyphosate in collected percolation soil water at Pixendorf, probably from previous conventional field application of Roundup Max®, confirmed the general low glyphosate adsorption capacity of Chernozems from Loess and the risk of transport towards groundwater.

3. Analysis of water from run-off retention basins in the landscape in the surroundings of the investigated Chernozem confirmed that through high erosion processes, especially in maize crop, glyphosate is partly transported outside the treated agricultural fields.

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