DISPERSION OF GLYPHOSATE IN SOILS UNDERGOING EROSION

DISPERSION DE GLYPHOSATE DANS LES SOLS SOUMIS À L’EROSION

DISPERSIONE DI GLYPHOSATE NEI SUOLI INTERESSATI DA EROSIONE

Gorana Rampazzo Todorovic [(1)*], Axel Mentler [(1)], Nicola Rampazzo [(1)], Winfried E.H. Blum [(1)], Alexander Eder [(2)], Peter Strauss [(2)]

[(1)] University of Natural Resources and Applied Life Sciences, Wien
[(2)] Federal Agency for Water Management,
Institute for land and Water Management Research, Petzenkirchen
[*] Corresponding author: E-mail gorana.todorovic@boku.ac.at

Abstract

Different physical, chemical and biological processes influence the behaviour of organic contaminants in soils. A better understanding of the organic pollutant behaviour in soils would improve the environmental protection. One possible way for better attenuation of the risk of pollution in agriculture can be achieved through ta better-specified pesticide management based on the adaptation of the pesticide type and application rates to the specific environmental characteristics of the area of application. Nowadays, one of the actually most applied herbicide world wide is glyphosate. Glyphosate is highly water soluble and traces have been found in surface and groundwater systems. For a better understanding of the natural influence of erosion processes on glyphosate behaviour and dispersion under heavy rain conditions after application in the field, two erosion simulation experiments were conducted on two different locations in Austria with completely different soil types in September 2008. The results of the experiments showed that under normal practical conditions (e.g. no rainfall is expected immediatly after application), the potential adsorption capacity of the Kirchberg soil (Stagnic Cambisol, with about 16.000 ppm Fe-oxides) is confirmed compared to the low adsorption Chernosem soil (about 8.000 ppm pedogenic Fe-oxides). Considering the enormous difference in the run-off amounts between the two sites Pixendorf and Kirchberg soils it can be concluded how important the soil structural conditions and vegetation type and cover are for the risks of erosion and, as a consequence, pollution of neighbouring waters. In the rainfall experiments under comparable simulation conditions, the amount of run-off was about 10 times higher at Kirchberg, owing to its better infiltration rate, than at the Pixendorf site. Moreover, the total loss of glyphosate (NT+CT) through run-off at the Kirchberg site was more than double that at Pixendorf, which confirms the importance of the chemical and mineralogical nature of soils in the abatement and absorbency of glyphosate, and the poor results

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in case of erosive precipitations whwn soil structure and permeability are not favourable.
Key-words: *Glyphosate; erosion; soil tillage; environment*

Résumé

Le comportement de substances polluantes organiques dans les sols est influencé par différents procèssus physico-chimico-biologiques. Une compréhension plus profonde des comportements susdits permettrait d'améliorer les possibilités de protection du milieu. Une meilleure atténuation du risque de contamination en agriculture peut être atteinte à travers une gestion plusieurs spécifique des pesticides davantage basée sur une adaptation soignée aux caractères environnementaux du type de pesticide utilisé dans la zone d'application. Glyphosate est actuellement un des herbicides plusieurs utilisé à niveau mondial et il est hautement soluble dans l'eau de façon qu' traces en ont été retrouvées en systèmes aquifères superficiels et de flanc. Un des phénomènes naturels plus importants pour la dispersion de Glyphosate et l'érosion des sols. Au but de comprendre l'influence des procès naturels d'érosion mieux sur le comportement et la dispersion de Glyphosate dues aux pluies érosives fortes après l'application en champ, en 2008 deux essais de simulation érosive ont été menés en deux localités en Autriche avec des sols complètement différents. Les résultats des essais ont mis en relief comme en conditions d'application de champ normal, il vaut à dire sans danger de pluies immédiates après l'application, la capacité potentiel d'absorption du sol de Kirchberg (Stagnic Cambisol avec approximativement 16.000 mg d'oxydes de Fe pour kg de sol) il est confirmée et messe à la comparaison avec le sol Chernosem à la basse absorption, (8.000 mg d'oxydes de Fe pedogenetic pour kg de sol). Étant donné puis la différence énorme en quantité d'écoulement superficiel entre les deux sols Pixendorf et Kirchberg, l'importance des conditions structurales du sol, le type et le épaisseur de la végétation sur le risque d'érosion et risque conséquent de contamination des eaux adjacentes, et plus qu'évident. Les deux essais d'érosion, effectués sous conditions techniques de simulation identique, ils montrent comme la quantité d'écoulement superficiel au Kirchberg ait approximativement été 10 grandes fois de celle du sol de Pixendorf, qu'à le moment de l'essai il présentait un excellent infiltration par l'eau. En outre la perte totale de Glyphosate par l'écoulement superficiel au Kirchberg et plus restée du double qui au Pixendorf. En concluant, les résultats ont mis en évidence l'importance de la constitution chimique-mineralogique des sols en soin à l'atténuation et absorption de Glyphosate que poirier elle résulte vaine si les conditions du terrain, spécialement la structure et la perméabilité, ils ne sont pas optimaux en cas de précipitations érosives.
Mots-clés: *Glyphosate; érosion; travail du sol; environnement*

Riassunto

Il comportamento di sostanze contaminanti organiche nei suoli è influenzato da diversi processi fisico-chimico-biologici. Una più profonda comprensione di suddetti comportamenti porterebbe a migliorare le possibilità di protezione dell'ambiente. Una migliore attenuazione del rischio di contaminazione in agricoltura può essere raggiunta attraverso una gestione più specifica dei pesticidi basata su un più accurato adattamento del tipo di pesticida usato e delle quantità applicate alle caratteristiche ambientali dell'area di applicazione. Glyphosate è attualmente uno degli erbicidi più usati a livello mondiale ed è altamente solubile in acqua così che ne sono state rinvenute tracce in sistemi acquiferi superficiali e di falda. Uno dei fenomeni naturali più importanti per la dispersione di Glyphosate è l'erosione dei suoli. Al fine di comprendere meglio l'influenza dei processi naturali di erosione sul comportamento e la dispersione di Glyphosate dovuta a forti piogge erosive dopo l'applicazione in campo, nel 2008 sono stati condotti due esperimenti di simulazione erosiva in due località` in Austria con suoli completamente diversi. I risultati degli esperimenti hanno messo in rilievo come in condizioni di applicazione di campo normali (vale a dire senza pericolo di piogge immediate dopo l'applicazione), la capacità potenziale di assorbimento del suolo di Kirchberg (Stagnic Cambisol, con approssimativamente 16.000 mg di ossidi di Fe pedogenici per kg di suolo) e` stata confermata e messa a confronto con il suolo Chernosem a basso assorbimento (circa 8.000 mg di ossidi di Fe per kg di suolo). Considerando poi l'enorme differenza in quantità di scolo superficiale tra i due suoli Pixendorf e Kirchberg, l'importanza delle condizioni strutturali del suolo, il tipo e la densità di vegetazione sul rischio di erosione e conseguente rischio di contaminazione delle acque adiacenti, è più che evidente. I due esperimenti di erosione, effettuati sotto condizioni tecniche di simulazione identiche, mostrano come la quantità di scolo superficiale a Kirchberg sia stata approssimativamente 10 volte maggiore di quella del suolo di Pixendorf, che al momento dell'esperimento presentava un'ottima infiltrabilità per l'acqua. Inoltre la perdita totale di Glyphosate tramite lo scolo superficiale a Kirchberg è stata più del doppio che a Pixendorf. Concludendo, i risultati hanno messo in evidenza l'importanza della costituzione chimico-mineralogica dei suoli in riguardo all'attenuazione e assorbimento di Glyphosate, che però risulta vana se le condizioni del terreno, specialmente la struttura e la permeabilità, non sono ottimali in caso di precipitazioni erosive.

Parole chiave: *Glyphosate; erosione; lavorazione del terreno; difesa ambientale*

Introduction

Soils play an important role in the regulation of contaminants in ecosystems. Because the organic pollutants are often released in environment in enormous quantities there is a considerable risk for environmental pollution by these pollutants. A better understanding for the organic pollutant behavior in soils is important for the improvement of environmental protection. There is a need for more specified pesticide management based on the adaptation of the pesticide type and application rates to the specific characteristics of the area of application (Eriksson *et al.,* 2007; Crommentuijn *et al.,* 2000; Zablotowicz *et al.,* 2006;

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Peruzzo *et al.,* 2008; Warnemuende *et al.,* 2007; Albrechsten *et al.,* 2001; Papiernik SK, 2001).

The behaviour of organic contaminants in soils is generally governed by a variety of physical, chemical and biological processes, including sorption–desorption, volatilization, chemical and biological degradation, uptake by plants, run-off, and leaching (Mamy *et al.*, 2005).

Special focus of this presentation is given on glyphosate. Glyphosate is one of the actually most applied herbicide world wide, with about 60% of the global sales for non-selective herbicides (Candela *et al*., 2007). Moreover, glyphosate is highly water soluble and traces were found in surface- and groundwater systems (Eriksson *et al.,* 2007, Warren et al, 2003; Krueger *et al.*, 1999; Peruzzo *et al.,* 2008; Warnemuende *et al.,* 2007; Siimes *et al.*, 2006; Widenfalk *et al.,* 2007 ; Albrechsten *et al.,* 2001; Landry *et al.,* 2005; Papiernik, 2001; Stenrød *et al.,* 2007). The major degradation product of glyphosate is aminomethylphosphonic acid (AMPA) (Peruzzo *et al.,* 2008; Locke and Zablotowicz, 2004; Gimsing *et al.*, 2004). The glyphosate fate and behavior in soil are affected by different soil factors and processes, but depends also on interactions between herbicide and soil under the specific local conditions (Locke and Zablotowicz, 2004; Soulas and Lagacherie, 2001; Gimsing *et al.*, 2004).

Better understanding of glyphosate characteristics, environmental influence, specific soil parameters and soil microbes behavior can help estimation and better attenuation of time dependent adsorption/degradation rates and consequently risks for environmental pollution involved.

Fate of the organic pollutants in soils

Fate and behaviour of organic contaminants in soils is generally governed by different physical, chemical and biological processes (e.g. uptake by plants, volatilization, chemical and biological degradation, sorption–desorption, run-off, and leaching through soil profiles).

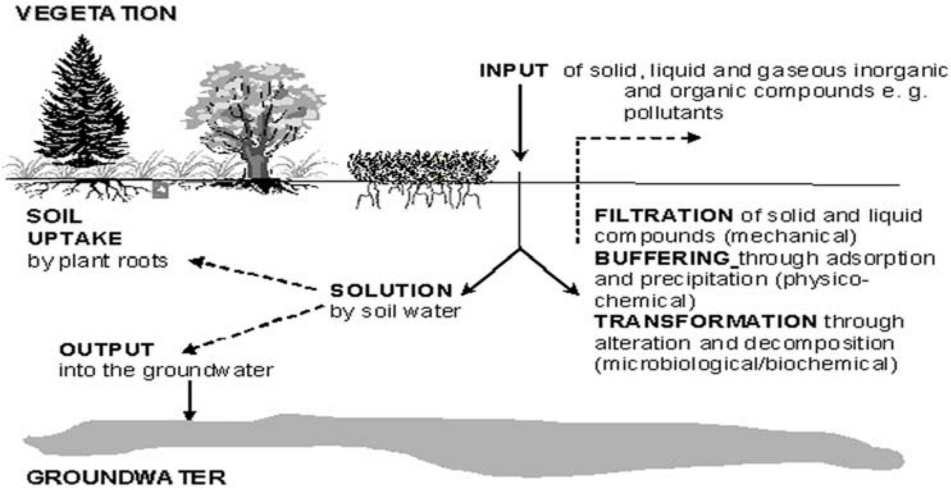
The attenuation of pollutant toxic concentration in soils is governed by processes of precipitation, sorption and immobilization (Fig.1). Due to these processes particular soil components can diminish the risk of water, air and food contamination with organic pollutants.

The distribution (horizontally and vertically) of organic pollutants in soils depends on their movement, degradation and sorption by soil solids which in turn depend on three general factors:

- Pollutant characteristics
- Chemical, biological and hydraulic properties of the soil
- Weather conditions

Moreover, distribution and behaviour of contaminants are also time dependent (e.g. different biodegradation phases; leaching and surface runoff-risk) (Nicholls, 1991, Schwarzbauer, 2005).

Figure 1 - *Attenuation of pollutants in soil environment.*



Glyphosate

Glyphosate is a post-emergency non-selective broad spectrum herbicide which blocks the shikimic acid pathway for biosynthesis of aromatic amino acids in plants, also in some microbes, but not in all soil microorganisms (Haghani K *et al.*, 2007). This herbicide is worldwide used in agriculture, railways, dam protection, surface water systems, urban areas (Nowack, 2003; Ternan *et al.*, 1998; Baylis, 2000). Usually the persistence is up to 170 with usual half life of 45-60 days (Peruzzo *et al.*, 2008), but there is also investigations which showed that half-life could be prolonged up to years (Carlisle and Trevors, 1978 in Zaranyika and Nyandoro, 1993; Eberbach, 1997). Its main metabolite is AMPA (Peruzzo *et al.*, 2008; Locke and Zablotowicz, 2004; Gimsing *et al.*, 2004).

Glyphosate sorption in soil

Strong sorption of glyphosate to soil particles may decrease degradation rate of glyphosate due to less bioavailability of bounded pesticide. Till now, the following ways of sorption were confirmed in numerous studies:

- Glyphosate adsorption on soil colloids;
- Fe/Al-oxides, clays, calcite, organic matter;
- Glyphosate complexation with metals;

Sorption of glyphosate in soil is mainly due to the inner sphere complex formation with metals of soil oxides, related to the soil phosphate adsorption capacity.
Possible binding mechanisms:

- Electrostatic bonds in extremely acid media

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- Hydrogen bonds with humic substances
- Especially, covalent bonds with Fe and Al oxides

Moreover, availability of glyphosate for decomposition is affected by the rate of desorption as well as by the soil processes and their influence on soil microbial population and activity (Eberbach, 1997; Gimsing *et al.,* 2004; Schnurer *et al.,* 2006; Sørensen et al., 2006; Rodrígez-Cruz et al., 2006; Kools *et al.,* 2005; Ghanem *et al.,* 2007).

Surface run-off

Since glyphosate is water soluble there is a potential risk of run-off in the case of erosive precipitations very soon after the herbicide application and in the regions with high risk of run-off. Pesticide residues in the bottom sediments of the surface water systems may be influenced by a number of factors including runoff potential and intrinsic properties of the pesticides (Krueger *et al.,* 1999; Eriksson *et al.,* 2007, Warren et al, 2003; Krueger *et al.,* 1999; Peruzzo *et al.,* 2008; Warnemuende *et al.,* 2007; Siimes et al., 2006; Widenfalk *et al.,* 2007).

Leaching-preferential flows

The sources and transport routes of pesticides to groundwater are multifaceted. They can include non-point sources such as agriculture, with pesticide leaching from top soil. In the study of Rueppel ML *et al.,* (1977), glyphosate was considered as pesticide I class and thereby posses 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 (Albrechsten *et al.,* 2001; Landry *et al.,* 2005; Papiernik SK, 2001; Stenrød M *et al*., 2007). Degradation potential of pesticide is lower in aquifers than in top soil.

Influence of glyphosate on shallow aquifers/aquatic/marine ecosystems

Influence of glyphosate on different water organisms in water systems was already in various investigations shown (Warren *et al.,* 2003; Feng *et al.,* 1990; Tsui and Chu, 2007; Peruzzo *et al.,* 2008; Amorós et al., 2007; Widenfalk *et al.,* 2007). There is still a need for more data for better understanding the influence and behavior of glyphosate and metabolites in water bodies. Interaction of these pollutants with different organisms in water ecosystems should be more investigated in different surface water systems.

Water erosion simulation field experiments (2008)

Site description. For a better understanding of the influence of erosion processes on glyphosate behaviour and dispersion under rainy conditions after application in the practice, two rain simulation experiments were conducted on two different locations in Austria with complete different soil types in September 2008. These locations are experimental fields where glyphosate is usually applied according to agricultural practices. The soil type in Pixendorf, Lower Austria, is classified as a

Degraded Chernosem, according to the WRB systematic (2006). The soil type in Kirchberg a.W., Styria, is classified as Stagnic Cambisol, according to WRB. Both soils have a rather high erosion risk due to their texture (silty in Pixendorf and fine sandy in Kirchberg) so erosion processes lead to loss of organic matter and clay downslope. Another main difference between the two soils consists in the amount of pedogenic iron-oxides (Kirchberg features 16.000 mg dithionite-extractable Fe/ kg soil and Pixendorf about 8.000 mg Fe/kg soil), pH (Kirchberg acid, Pixendorf slightly alkaline).

Experimental design. The rain simulation experiments took place in 3 field replications (1, 2, 3) of the Conventional Tillage (CT)- and the No Tillage (NT)-plots. The average slope in both sites was about 10%. Before starting the rain simulation, erosion plots were installed in each field repetition in a dimension of 2m x 2 m, see fig. 2. The culture type at time of the experiments was different in both sites (Pixendorf had a green cover after the wheat yield of July whereas Kirchberg stood immediately after the mais yield) but in both sites the vegetation cover degree was distinctly higher in the NT-plots.

Herbicide application. In the common agricultural practice the Round Up application consists in dissolving 4 liters Round Up Max (450 g glyphosate /L Round Up) in 200 liters of water and applied per ha (corresponding to a 2 % herbicide solution). This corresponds to an application of 1.800 g glyphosate ha^{-1} or 180 mg glyphosate m^{-2}. Consequently, for the rain simulation experiments a 2 % herbicide solution was prepared and sprayed homogeneously by a hand pump in the same concentration and amounts as in practice, e.g. 180 mg glyphosate / m^2 respectively 720 mg glyphosate / simulation plot. Immediately after application of Round Up the rain simulation started (worst case scenario) and lasted 60 minutes with an intensity of 30 mm. During the rain simulation, run-off-fractions at different time intervals were collected and cooled. Immediately after raining, soil samples were collected within the rain plot at 5 different depths (0 – 2 cm, 2-5 cm, 5-10 cm, 10-15 cm, 15-20 cm). The run-off-samples as well as the soil samples were immediately transported to the laboratory in refrigerator boxes. There the run-off samples were centrifuged in order to separate the liquid from the solid phase.
The rain simulator was designed as a portable out- and indoor equipment. The spray pattern got generated by full jet nozzles. The rainfall intensity was controlled with intermittent spraying. The opening cycles of the solenoid valves were fully programmable with computing equipment (Strauss *et al*., 2000).

Results

Figures 3 and 4 show the amount of run-off after each experiment. In both sites the CT plots produced the highest run-off amounts because of the lower vegetation cover. The amount of run-off in Kirchberg was 10 times higher than at Pixendorf because during the rainfall simulation the soil surface of Kirchberg was compacted

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and crusty, whereas Pixendorf had a very favorable crumby structure with a high infiltration rate.

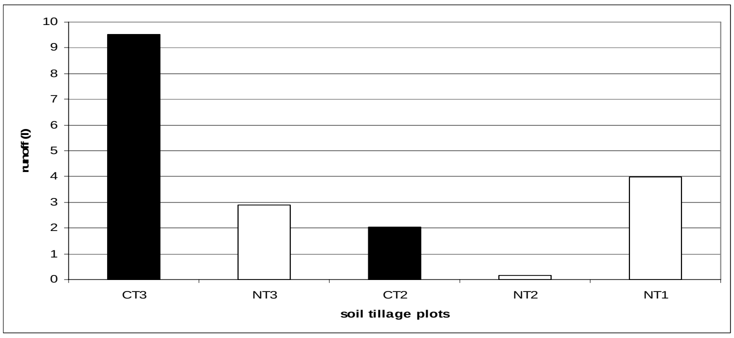

Figure 3

Total run-off of the CT- and NT plots in the 3 field repetitions at Pixendorf (Plot CT1 could not be exercised)

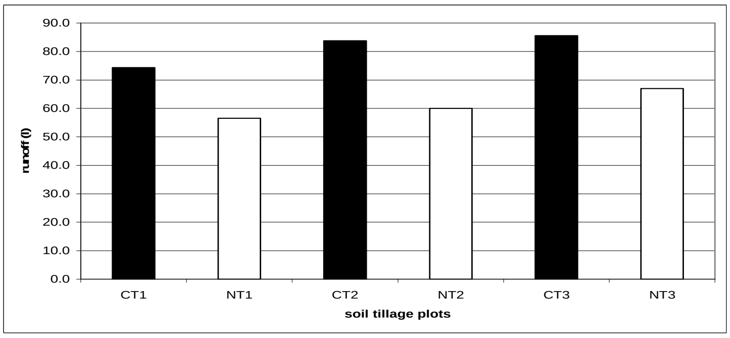

Figure 4

Total run-off of the CT- and NT plots in the 3 field repetitions at Kirchberg.

Figure 5 shows the time dependent glyphosate concentrations in run-off-fractions at Pixendorf. As it was expected the first fractions (respectively on the left) showed the highest concentrations in both variables CT and NT which got lower with time. The CT-plots showed distinguished higher concentrations than the NT-plots.

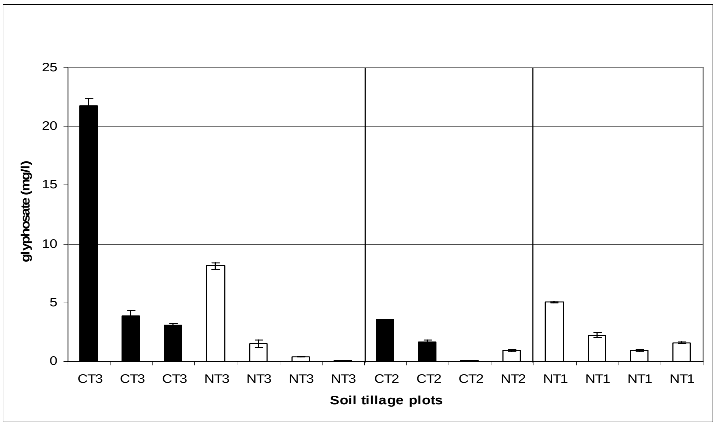

Figure 5

Glyphosate concentrations in run-off-fractions at different time intervals in the 3 repetitions (1, 2, 3) plots of Pixendorf (CT = Conventional Tillage, NT = No Tillage). (Plot CT1 could not be exercised.)

In concomitance with the concentration, the total amount of glyphosate washed out of the plots by run-off was much higher in the CT-plots, see fig.6, but still much lower than in Kirchberg, see fig.8.

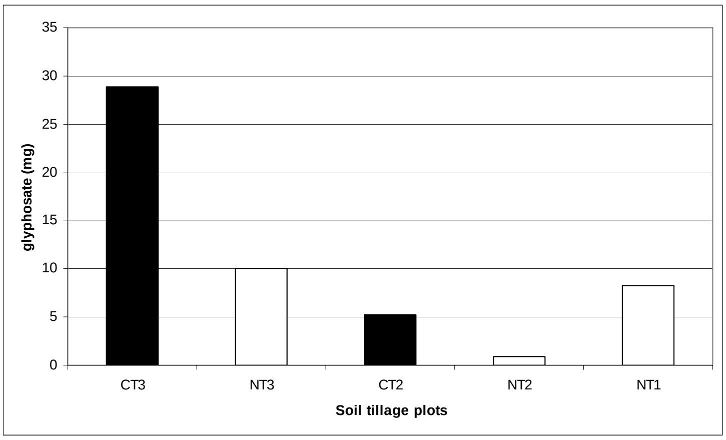

Figure 6

Total amounts of glyphosate in run-off at the 3 repetitions (1, 2, 3) plots of Pixendorf (CT=Conventional Tillage, NT = No Tillage). (Plot CT1 could not be exercised.)

The glyphosate concentration in run-off at Kirchberg is shown in fig. 7. In this case the run-offs of the NT-plots had a much higher concentration of glyphosate than the CT-plots. The reason for this is the nearly 100% plant cover of the NT-plots, where most of the applied glyphosate adheres to the photosynthetic active plant organs (still and leaves), get washed out through the rainfall und hardly infiltrates the soil surface. Moreover, the concentrations in the 3 field repetitions were very different because of the inhomogeneity of the field regarding soil surface structure, vegetation cover and micro relief.

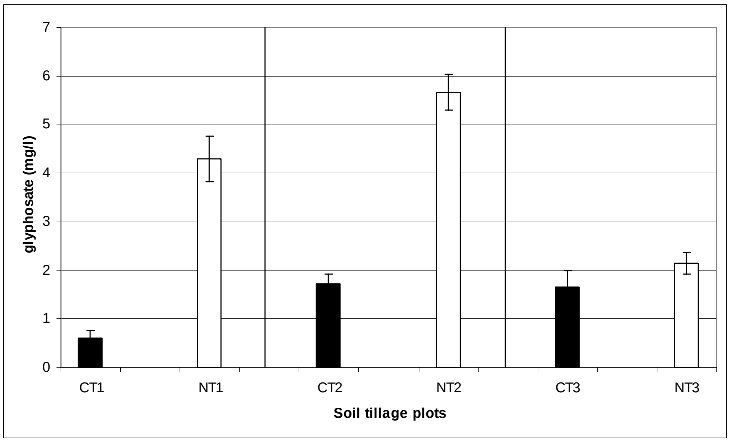

Figure 7

Average glyphosate concentrations in run-off at the 3 repetitions (1, 2, 3) plots of Kirchberg (CT=Conventional Tillage, NT= No Tillage).

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Figure 8 shows the total glyphosate amounts in the run-off of the erosion plots, where controversial to the site Pixendorf, the NT-plots had distinguished higher contents.

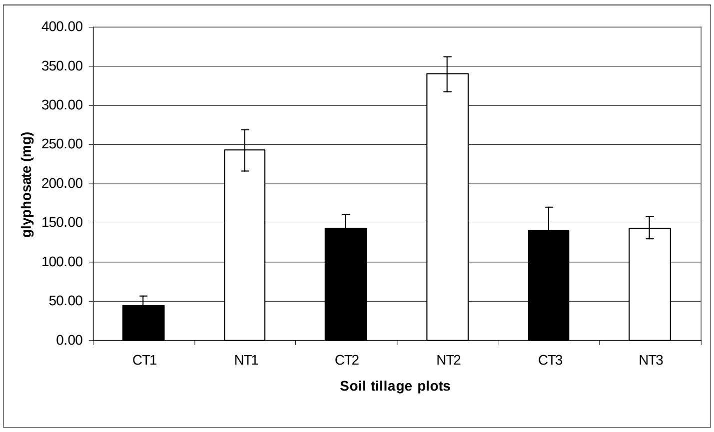

Figure 8

Total amounts of glyphosate in run-off at the 3 repetitions (1, 2, 3) plots of Kirchberg (CT= Conventional Tillage, NT= No Tillage).

A total balance of glyphosate for the rain simulation experiment in Pixendorf is shown in table 1.

Table 1 - *Distribution of glyphosate in % of the applied quantity into the 2m x 2m rain simulation plot (720 mg glyphosate = 100%) of Pixendorf. P.S. The plot repet.1-CT could not be exercised.*

Repetition 1	run-off-liquid %	run-off-solid %	soil %	rest %
NT (No Tillage)	8.22	0.009	74.1	17.7
CT (Conv. Tillage)	-	-	-	-
Repetition 2				
NT (No Tillage	0.93	0.00012	126.0	0.0
CT (Conv. Tillage)	5.29	0.0036	47.3	47.4
Repetition 3				
NT (No Tillage	10.00	0.006	50.0	39.5
CT (Conv. Tillage)	28.30	0.0081	34.4	37.2
Mean value NT	6.38	0.005	83.4	19.1
Mean value CT	16.80	0.0043	40.9	42.3

The 3 field repetitions were relatively inhomogenous. The CT-plots showed higher glyphosate % contents in aqueous run-off than the NT-plots. The glyphosate adsorption in soil was much higher in Pixendorf as compared to Kirchberg, because of the high infiltrability of the soil. The solid phase of run-off contented negligible amounts of glyphosate. The rate of detection in % of the applied glyphosate amounts at Puixendorf was relatively high, and much higher than at Kirchberg. In the soil of the NT-plot of repetition 2 up to 126 % of the application was detected. Evidently residual amounts from previous applications were detected. A total balance of glyphosate for the rain simulation experiment in Kirchberg is shown in table 2.

The 3 field repetitions were also in Kirchberg relatively ihnomogenous. The NT-plots showed higher glyphosate % contents in the aqueous run-off than the CT-plots. The glyphosate adsorption in the soil was relatively low.

Table 2 - *Distribution of glyphosate in % of the applied quantity into the 2m x 2m rain simulation plot (720 mg glyphosate = 100%) of Kirchberg.*

Repetition 1	run-off-liquid %	run-off-solid %	soil %	rest %
NT (No Tillage)	33.7	0.025	13.0	53
CT (Conv. Tillage)	6.3	0.07	0.1	93
Repetition 2				
NT (No Tillage	47.2	0.019	6.4	46.4
CT (Conv. Tillage)	19.9	0.06	17.8	62.2
Repetition 3				
NT (No Tillage	20.0	0.016	15.3	64.7
CT (Conv. Tillage)	19.6	0.052	33.1	47.2
Mean value NT	33.6	0.02	11.6	54.7
Mean value CT	15.3	0.06	17.0	67.5

The solid phase of run-off contented also in Kirchberg negligible amounts of glyphosate. The CT-plot of repetition 1 was extremely crusty because only about 7% (soil + run-off) of the applied glyphosate could be measured. Averagely (with exception of rep.1-CT) about 50 % of the applied glyphosate could be measured , respectively 50 % not found (rest).

Conclusions

By the performed investigations only hypothetical conclusions can be stated about the fate of the „rest" glyphosate. A part of glyphosate could remain adhering to the green plant organs through the adhesion substances added in Round Up. A part could have been leached to deeper soil horizons and than there adsorbed through the soil. A small loss out of the installed simulation plots through lateral fissures cannot be totally excluded.

A comparison of the glyphosate adsorption of the 2 investigated soils under is shown in fig. 9. Figure 9 shows clearly, that under normal practical conditions (e.g no rainfall is expected after application), the potential adsorption capacity of the Kirchberg soil (Stagnic Cambisol, with about 16.000 ppm pedogenic Fe-oxides) is confirmed compared to the low adsorption Chernosem soil (about 8.000 ppm pedogenic Fe-oxides). Considering the enormous differences in the run-off amounts between the two sites Pixendorf and Kirchberg it can be concluded how important the soil surface conditions and vegetation cover of the agricultural fields for erosion risk and pollution risk of surface water are. The Pixendorf-soil was less

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compacted and showed a higher pore volume and amount of macro pores than the Kirchberg-soil. These parameters affect infiltration, fast flow and preferential flow processes in the soil which then influence the glyphosate dispersion by run-off. In the rainfall simulation experiments under comparable simulation conditions, the amount of run-off at Kirchberg was app. 10 times higher than at the Pixendorf site, due to its better infiltration rate.

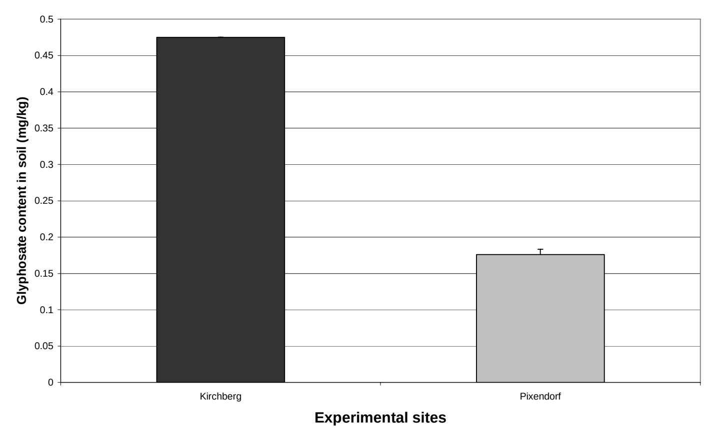

Figure 9

Amounts of glyphosate in different soils at 0-2 cm depth under normal agricultural field conditions. P.S. soil sampling immediately after Round Up application.

Moreover, the total loss of glyphosate (NT+CT) through run-off was more than double on the Kirchberg site, which confirms the higher risk of pesticide pollution for surface waters on the agricultural fields with higher erosion intensity. The results of the investigations have shown that a better understanding of glyphosate's behavior is needed (e.g. adsorption conditions, environmental influence, specific soil parameters, soil microbe's behavior). Based on this knowledge, time dependent adsorption, degradation rates and consequently risks for surface and groundwater involved can be estimated.

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