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Vegetative buffer zones as pesticide filters for simulated surface runoff

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Abstract

Losses of pesticides from agricultural land to surface waters can cause environmental harm to fish and other aquatic organisms. Vegetated buffer zones (BZ) between agricultural land and surface waters have proved to be effective filters for sediments and sediment-bound nutrients. It is therefore, likely that BZ also can be effective filters for pesticides, especially sediment-bound pesticides. The retention of glyphosate, fenpropimorph, propiconazole and soil particles was studied in short-term BZ experiments with simulated surface runoff. Runoff water containing pesticides and soil particles was added directly to the BZ. The BZ was 5 m wide and consisted of natural grass/herbaceous vegetation. To calculate retention efficiency of pesticides and particles through the BZ, surface runoff was collected before entering and after passing the BZ. The average removal efficiency was 39, 71, 63 and 62% for glyphosate, fenpropimorph, propiconazole and soil particles, respectively. Aminomethylphosphonic acid (AMPA), which is a degradation product of glyphosate, constituted only a small part of glyphosate (about 10%) in this short-term experiment. Based on this study BZ can serve as contributors to reduce pesticide input to surface waters.

Keywords: Vegetative buffer zone; Pesticides; Retention efficiency; Surface runoff; Soil particles

1. Introduction

The intensification of agriculture in Norway after the Second World War has led to an increased use of pesticides, and hence increased risk of pesticide pollution to surface waters. Norwegian authorities aim to develop methods to minimize the risk of pesticide residues in food and the environment (Ministry of Environment, 1997). According to the Plan of Action for pesticide reduction 1998–2002 "the amount of pesticides in small streams and surface waters should

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be reduced as far as possible and not exceed levels which can be harmful to the environment" (Ministry of Agriculture, 1998).

Knowledge about how adsorption, mobility and degradation of pesticides is influenced by Nordic climate and soil conditions is vital. Low temperature and high water content in soil may for instance reduce the degradation rates for pesticides (Vink and vanderZee, 1996). Adsorption of some pesticides to soil may be correlated to the clay size fraction (Riise et al., 2001; Wu et al., 2003). Adsorption and mobility will greatly affect the transport pathway of the pesticide and the possibility to leach (Jones et al., 1995; Marshall et al., 1996). The distribution coefficient (K_d value) expresses the ratio between the content of pesticide

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adsorbed to the soil and the mass concentration of pesticide in the aqueous solution (Tomlin, 1994). The $K_{\rm d}$ value for a given pesticide determines the degree of particle-bound transport. The major transport pathway for soil particles and particle-bound pesticides is surface runoff. Particle-bound pesticides are generally believed to have a lower potential for leakage to watercourses than pesticides with lower affinity to soil (Wauchope, 1978). Investigations in 13 different Nordic soils show that glyphosate has a high K_d value (Greve et al., 1998). Fungicides like fenpropimorph and propiconazole also have low mobility in soil (Roy et al., 2000; Roseth and Haarstad, 2002). However, particle-bound pesticides (e.g. glyphosate, propiconazole and fenpropimorph) have been detected in watercourses in Norway (Ludvigsen and Lode, 1999).

Vegetated buffer zones (BZ) located along stream banks have proved to be effective filters for surface runoff from agricultural land (e.g. Dillaha et al., 1989; Magette et al., 1989; Correll, 1997; Syversen, 2002a). The dominating retention process in surface runoff buffer zones is trapping of sediments and sediment-bound nutrients (Haan et al., 1994; Syversen, 2002a). It is likely that pesticides adsorbed to soil particles will also be trapped through surface runoff buffer zones. However, other processes like sorption of pesticides to soil and organic matter during the runoff process may also contribute to decreased concentration of pesticides in runoff water after passing a BZ. Additionally, uptake by plants may remove pesticides from the BZ. Internationally there is a lack of knowledge regarding BZ and their effect on pesticides (Correll, 1997).

The objective of the present study was to estimate the reduction in concentration of particle-bound pesticides (glyphosate, fenpropimorph and propiconazole) in the case of high surface runoff (simulated) through the buffer zones. A similar study was carried out looking at natural runoff events (Syversen, 2003).

2. Materials and methods

2.1. Field site descriptions

The experimental site was established in the south-eastern part of Norway (60° N), about 70 km north-east of Oslo (Fig. 1). The slope of the agricultural field and the BZ were 14% and the soil type



Fig. 1. Location of the experimental site, Norway.

was levelled silty clay loam with 45% clay, 52% silt, 3% sand, and 1.5% organic matter. The BZ soil profile except the top layer (0–30 cm depth) had a body structure remaining from the levelling process. The top layer has started a development of a platy structure. A considerable activity of earthworms is observed, especially in the top layer. Root development was observed down to 70 cm depth. The vegetation consisted of various grasses (*Circium arvense* (L.) Scop.), *Elytrigia repens repens* (L.) Desv. Ex Nevski, *Phleum pratense pratense* (L.), *Deschàpsia cespitòsa cespitòsa* (L.) Beauv, *Festuca pratensis* Huds.). The buffer zone width was 5 m.

2.2. Experimental design

The simulation experiment consisted of four short-term experiments (about 5 h runoff per experiment). The runoff water containing selected pesticides and particles was added directly to the upper boarder of the sloping 5 m wide BZ through a perforated supply gutter (Fig. 2, Table 1). Water from a nearby stream was pumped into a 10001 tank located upside the BZ, where soil particles and pesticides were added to the water. The concentration of

Table 1 Design and pr	rocedures used	in the simulation exp	periments	
Experiment	Season	Experimental area (m)	Soil texture in added particles	Amount of surface runoff water (l)

Design and procedures used in the simulation experiments					
Experiment	Season	Experimental	Soil texture in	A	

		alea (III)	added particles	runon water (1)	
1A–E ^a	Autumn	5×5	Silty clay loamb	5000	Grab samples
2A-C ^a	Autumn	5×5	Silt loam ^c	3000	Grab samples
3A–E ^a	Autumn	5×7.5	Silty clay loam ^b	5000	Volume proportional composite samples
4A–E ^a	Early summer	5×7.5	Silty clay loam ^b	5000	Volume proportional composite samples

^a Each of A-E represents one 10001 runoff.

^b Originates from the experimental site.

c 24% clay, 58% silt and 18% sand.

soil particles added, represented a "normal to high" runoff event in the area, and varied between 1250 and 5000 mg suspended solids/l. The pesticide concentration was related to the concentration of soil particles and represented a normal pesticide application in the area, which was 4000 ml Round UpEco/ha (active pesticide: glyphosate-360 g/l) and 1000 ml Tilt-Top/ha (active pesticides: fenpropimorph-375 g/l and propiconazole-125 g/l). The added concentration of glyphosate was about $12-23 \,\mu g/g$ soil, about $3 \,\mu g/g$ soil of fenpropimorph and about $1 \mu g/g$ soil of propiconazole. The exact concentration of soil particles and pesticides was analysed both in inlet and outlet water.

The BZ was saturated with water prior to the experiment to create surface runoff. The simulated runoff discharge into the BZ was about $0.41 \,\text{s}^{-1}$ and was held constant during all experiments. Discharge from the BZ occurred as surface runoff and preferential runoff. Only results from surface runoff are presented in this



Fig. 2. Design of the simulation experiment.

paper. The outlet water was collected in a gutter. The average retention time through the BZ was 4 min.

Water sampling technique

To simulate natural variation in weather conditions and application time for pesticides, four experiments were carried out. One experiment during early summer and three in autumn. Some adjustments were made to the experimental design throughout the experimental period (Table 1).

Soil samples were dried and crushed before being added to the pesticide solution in all experiments. The soil/pesticide mixture was left for 24 h to obtain adsorption-desorption equilibrium of pesticides to soil particles (OECD Guideline, 1984). In the 10001 tank a circulation pump kept the soil particles and pesticides in solution during the experiment. Inlet samples were collected from the supply gutter simultaneously with outlet samples from the collecting gutter. Grab samples were taken manually with 1 inlet- and 2-3 flowproportional outletsamples per 10001 water added. Volume proportional composite samples at the inlet were sampled from a hole in the perforated supply gutter. At the outlet volume proportional composite samples consisted of a 1/21 sample taken manually after stirring of each 101 bucket collected at the outlet. The 1/21 samples were mixed thoroughly and sampled. For volume proportional composite samples one sample from inlet and one from outlet were sampled from each 10001 of water used in the simulation.

2.3. Analytical laboratory methods

The water samples were analysed for glyphosate, fenpropimorph, propiconazole, suspended solids (SS), loss on ignition and soil texture. Glyphosate and aminomethylphosphonic acid (AMPA) analysis were carried out in acidified (pH 2) water samples (11), and stored at 4 °C. Only the supernatant from the samples was prepared for analysis. The samples were concentrated in two ion exchangers, derivatised twice and analysed by gas chromatography with mass-spectrometric detection (GC/MS-SIM). The detection limit was 0.01 μ g/l. For fenpropimorph and propiconazole 500 ml of the water samples was extracted twice with dicloromethane after addition of NaCl. The samples were analysed by gas chromatography (GC-MULTI M03). A detailed description of the method is given by Holen et al. (2002).

The water samples were analysed for suspended solids and loss on ignition according to Norwegian standards (Norwegian Standard, 1983). Loss on ignition was measured to get an estimate of the organic matter content.

For texture analysis an exact volume (about 11) of the water sample was filtered through a $0.45 \,\mu\text{m}$ preweighed Millipore filter. Due to filter clogging, 1–8 filters were usually needed per sample. The filters were dried at 40 °C for 24 h and weighed after drying. Some of the suspended matter was removed from the filter surface and oxidised by H₂O₂ to remove organic matter. The soil samples including aggregates were dispersed in an ultrasonic bath, before being injected to a Coulter LS 230 laser. This instrument analysed particle sizes in the range of 0.04–2000 μ m. The laser diffraction texture analysis is a technique that measures the light scatter from an ensemble of particles. Compared to traditional methods, the laser diffraction technique gives high resolution curves.

Textural classes are displayed in Table 2. Due to very low content of sand in the samples, coarse silt and sand are treated as one class in the discussion. Only samples from experiments 1, 3 and 4 were analysed for texture.

Table 2

Limits	for	particle	sizes-texture	analysis

Fine	Medium	Coarse	
Clay (µm) 0.06–0.2	0.2–0.6	0.6–2	
Silt (µm) 2–6	6–20	20–60	
Sand (mm) 0.06–0.2	0.2–0.6	0.6–2	

Soil particles and particle-bound pesticides were removed from some samples (experiments 3 and 4) by centrifugal power (7500 rpm in 10 min). Only the soluble phase was analysed for glyphosate, fenpropimorph and propiconazole in these samples.

2.4. Effects of soil particles on recovery of glyphosate

Because only the supernatant was analysed for glyphosate, a separate experiment was designed in order to investigate the recovery of glyphosate as a function of level of suspended solids, particle size of SS in the water samples, and application rate of glyphosate. The factorial design included three levels of glyphosate (8, 15 and 30 ug/l), five levels of SS (100, 500, 1500, 3000 and 5000 mg/l) and two levels of clay content in SS (46 and 60%; which corresponded to the clay content in the experimental field and in the runoff water from the experimental field). Results were analysed by multivariate regression with recovery as an output variable. Recovery of the analyses itself was 87%. Total recovery of glyphosate (R) was calculated as follows:

$$R = \left(\text{glyphosate-analysed} + \text{AMPA-analysed} \right) \times \left(\frac{169}{111} \right) \left(\frac{100}{\text{glyphosate-added}} \right) \times \left(\frac{100}{87} \right)$$
(1)

where 169/111 is the relative molecular weight of glyphosate and AMPA.

The data-analyses showed no statistically significant effect (P > 0.05) of different levels of glyphosate and clay content on the recovery of glyphosate. The effect of SS was significant (P < 0.0001). The logarithmic transformation of SS versus recovery of glyphosate fitted better ($R^2 = 0.92$) than the linear relationship.

$$R = -9.64\ln(SS) + 113 \tag{2}$$

Glyphosate recoveries presented later in this paper were adjusted according to this relationship.

2.5. Recovery of fenpropimorph and propiconazole

Standards with known concentration of fenpropimorph and propiconazole were analysed simultaneously with the water samples. The recovery of fenpropimorph varied from 97 to 103%, with an average of 98% (n = 4), while the recovery of propiconazole varied from 111 to 113%, with an average of 119% (n = 7). Corrections for recovery of fenpropimorph and propiconazole were made.

2.6. Calculations of retention through the buffer zone

To calculate retention of pesticides and particles through the BZ, information on input and output from the zone is needed. The removal efficiency in % (Rem) for pesticides and particles in the buffer zone was calculated according to:

$$\operatorname{Rem} = \left(\frac{C_{\rm in} - C_{\rm out}}{C_{\rm in}}\right) \times 100 \tag{3}$$

where C_{in} and C_{out} are concentrations of soil particles and pesticides in mg/l and μ g/l, respectively.

Data were analysed by one-way ANOVA, bivariate linear and multivariate regression models. Statistical differences were tested at 5% level (P < 0.05).

3. Results and discussion

3.1. Removal efficiency of soil particles and particle-bound pesticides

Fig. 3 shows the concentrations of glyphosate, fenpropimorph, propiconazole and soil particles into and

after passing the BZ. The figure represents results from all experiments (average, median and standard deviation). The decrease in concentration from inlet to outlet of the BZ were statistically significant for glyphosate, fenpropimorph and soil particles, but not significant for propiconazole. The average removal efficiency for glyphosate, fenpropimorph, propiconazole and soil particles was 39, 71, 32 and 62%, respectively. AMPA which is a degradation product of glyphosate, constituted only a small part of glyphosate (about 10%) in this experiment and is therefore not presented. Low removal efficiency for propiconazole was caused by one sample in experiment 3D (Fig. 4), where the outlet value from the BZ was 5-25 times higher than any other outlet value for propiconazole. The inlet concentration in this case, was similar to the other inlet values for propiconazole. The removal efficiency for propiconazole increased from 32 to 63% and became statistically significant if this outlier was removed from the dataset.

Experiment 1 had significantly lower removal efficiency for particles than experiments 2–4 (Fig. 4). However, there was no statistically significant differences between the experiments for any of the pesticides. The low or negative particle removal efficiency in experiments 1D and 1E were part of a decreasing removal efficiency from start to end of experiment 1. Moreover, the added amount of particles to the BZ was also decreasing throughout experiment 1. Fig. 5 shows the regression between added amount



Fig. 3. Concentration of glyphosate (Glyphos., $\mu g/l$), soil particles (SS, g/l), fenpropimorph (Fenpr., $\mu g/l$) and propiconazole (Prop., $\mu g/l$) into (In) and after passing a 5 m wide buffer zone (Out). The figure shows mean (bar), standard deviation and median concentrations for all experiments. n = 10-18 for input values and 10–33 for output values.



Fig. 4. Development of removal efficiency (%) for suspended solids, glyphosate, propiconazole and fenpropimorph through buffer zone experiments 1–4. A represents the first 10001 runoff of each experiment, B the second, C the third, D the fourth and E the fifth.

of particles to the BZ and the removal efficiency of particles for experiments 1–4. There is a significant increase (P = 0.005) in removal efficiency with increasing amount of particles added.



Fig. 5. A linear regression model between added amount of soil particles (g/l) into the buffer zone and the removal efficiency of particles (%) in the buffer zone. The linear fit illustrates the increase in removal efficiency with increasing amount of particles added (P = 0.005).

These results support previous results from BZ with natural runoff and constructed wetlands, where increased input of particles to the BZ/wetland gave increased retention of particles (Braskerud et al., 2000; Syversen, 2002b). This phenomenon is explained by the high input of particles that often occurs when surface runoff is high. High runoff can cause detachment of coarse particles, which easily trap in the BZ. High particle concentration can also cause aggregation of small particles into larger ones, which are trapped in the BZ. The particle size distribution in the inlet water was not change as a function of total amount of particles. However, there was a significant increase in relative clay content in the outlet water with increasing amount of particles added to the buffer (P = 0.0054), indicating that relatively higher amount of the coarser fractions were trapped in the BZ.

The organic matter content of suspended solids did not change throughout the BZ. As the organic matter content of the experimental soil was low, it was not

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expected to influence the removal efficiency of pesticides in this experiment.

The selection of pesticides in this investigation was based on their adsorption to soil mineral particles and organic matter. The distribution coefficients (K_d) of the experimental soils varied from 461 to 1539, 11 to 22 and 12 to 17 ml/g soil for glyphosate, propiconazole and fenpropimorph, respectively (Syversen et al., 2000; Thorstensen and Linjordet, unpublished). The $K_{\rm d}$ values show high adsorption of glyphosate and moderate adsorption of fenpropimorph and propiconazole to the experimental soils. Retention of glyphosate is therefore, expected to be mainly related to trapping of sediments. Surprisingly, the removal efficiency of fenpropimorph and propiconazole (excluding the outlier) were more similar to the removal efficiency of particles than the removal efficiency of glyphosate. Based on the K_d values the opposite was expected. Other processes of glyphosate retention may have effect in the BZ, like e.g. sorption to the soil surface. According to Greve et al. (1998) glyphosate is adsorbed to clay particles. There was a significant increase in removal efficiency for glyphosate with increasing relative amount of clay in outlet water from

the BZ (P = 0.02) indicating that this increase in glyphosate removal was not adsorbed to clay particles in runoff. Fig. 6 shows the opposite situation for fine silt (P = 0.04), medium silt (P = 0.02) and sand and coarse silt (P = 0.05), where the removal efficiency for glyphosate was decreasing with increasing relative amount of these fractions in outlet water. These results may indicate that glyphosate was largely adsorbed to fine and medium silt (not significant for sand and coarse silt). There was a significant decrease in relative content of medium silt through the buffer zone, while there was no significant differences between inlet and outlet values for fine silt (Fig. 7), indicating that fine silt particles did not trap in the BZ. It is therefore, likely that glyphosate is adsorbed by fine and medium silt, but only trapped by sedimentation of medium silt in the BZ. This may explain the relatively low retention efficiency for glyphosate. Retention of glyphosate in solution are discussed later.

There is no significant correlation between texture in outlet water and removal efficiency for fenpropimorph and propiconazole. Earlier studies by Riise et al. (2001) have suggested that organic carbon, mainly associated with the coarse size fraction,



Fig. 6. A linear regression model for removal efficiency of glyphosate (%) with texture (in vol.%) in outlet water from the buffer zone. All trends were significant (P < 0.05), except for sand and coarse silt, which are slightly significant (P = 0.05).



Fig. 7. A linear regression model for difference in texture (in vol.%) between in (1) and out (2) of the buffer zone (BZ). The linear fit illustrates the increase/decrease in percentage of each fraction throughout the buffer zone. The trends were significant for clay (P = 0.0015), medium silt (P < 0.0001) and sand and coarse silt (P < 0.0001). The trend was not significant for fine silt (P = 0.74). Fine, medium and coarse clay show the same trend as the whole clay fraction.

seems to be the most important factor for the adsorption of propiconazole. In addition, Wu et al. (2003) found that the clay fraction are an important carrier for propiconazole. It is not possible to draw any conclusions regarding binding of propiconazole and fenpropimorph to particles of different sizes from our experiments. Adsorption of pesticides to soil and vegetation in the BZ may be important retention processes, though they were not investigated in this study.



Fig. 8. Average removal efficiency (n = 6) in soluble phase of glyphosate, propiconazole, and fenpropimorph through the buffer zone (based on centrifugated samples).

3.2. Removal efficiency of pesticides in solution

Removal efficiency of pesticides in solution was evaluated based on analysis of the supernatant of centrifugated samples (Fig. 8). Removal efficiency varied from 24 to 70% for glyphosate, 32 to 78% for propiconazole and 61 to 73% for fenpropimorph. Retention of soluble pesticides in the BZ are most probably explained by the adsorption of pesticides to soil surface or plant residues in the BZ. This process is not fully documented and need further investigation.

4. Conclusions

This study demonstrated the effect of BZ on minimising simulated surface runoff of particles, particle-bound and soluble pesticides. K_d values of the pesticides included in the four experiments showed moderate to high adsorption to the experimental soil. However, a great part of the pesticides were soluble during the experiment. Average total removal efficiency of glyphosate, propiconazole, fenpropimorph and soil particles in simulated runoff water passing a 5 m wide BZ was 39, 63, 71 and 62%, respectively. The removal of glyphosate was less than that of fenpropimorph and propiconazole. Low removal efficiency of glyphosate is probably due to adsorption of glyphosate to the smallest particle size fractions, which have lower trapping efficiency in BZ. Trapped pesticides were in both soluble and particle-bound form. Removal efficiency of soluble fractions through the BZ was 24-70% for glyphosate, 32-78% for propiconazole and 61-73% for fenpropimorph. There were no significant differences in removal efficiency for any of the pesticides between the experiments, which were carried out both during spring and autumn. The study showed that BZ can be effective filters for particles and pesticides.

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