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A review of rapid transport of pesticides from sloping farmland to surface waters: Processes and mitigation strategies

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Abstract

Pesticides applied to sloping farmland may lead to surface water contamination through rapid transport processes as influenced by the complex topography and high spatial variability of soil properties and land use in hilly or mountainous regions. However, the fate of pesticides applied to sloping farmland has not been sufficiently elucidated. This article reviews the current understanding of pesticide transport from sloping farmland to surface water. It examines overland flow and subsurface lateral flow in areas where surface soil is underlain by impervious subsoil or rocks and tile drains. It stresses the importance of quantifying and modeling the contributions of various pathways to rapid pesticide loss at catchment and regional scales. Such models could be used in scenario studies for evaluating the effectiveness of possible mitigation strategies such as constructing vegetated strips, depressions, wetlands and drainage ditches, and implementing good agricultural practices. Field monitoring studies should also be conducted to calibrate and validate the transport models as well as biophysical-economic models, to optimize mitigation measures in areas dominated by sloping farmland.

Key words: pesticide; rapid transport; sloping farmland; overland flow; subsurface flow DOI: 10.1016/S1001-0742(11)60753-5

Introduction

The fate of pesticides applied to sloping farmland and surface water contamination by the discharged pesticides is of considerable environmental concern in hilly or mountainous regions. There have been a number of studies in which overland flow induced by a single rainfall/storm event led to detection of pesticides (e.g., azinphos-methyl, chlorpyrifos, endosulfan, atrazine, and alachlor) in rivers at concentrations exceeding target water quality limits (e.g., Richards and Baker, 1993; Dabrowski et al., 2002). However, pesticide transport processes from sloping farmland to surface waters in hilly or mountainous regions have been poorly documented. Elucidating these processes is a prerequisite for developing mitigation strategies against possible surface water pollution in such regions.

Figure 1 is a schematic representation of various pesticide transport routes to surface waters from sloping farmland with an impermeable layer, and tile drains beneath the plough layer. The surface waters include streams and ditches directly adjacent to agricultural fields, large rivers, lakes and reservoirs. Sloping farmland typically has an impermeable layer (e.g., fragipan, bedrock) below its topsoil. In both sloping farmland types, rapid transport of pesticides to surface waters can occur during storm events, or in response to irrigation. Pathways of the rapid transport include (1) overland flow in response to heavy rainfall or irrigation, (2) rapid leaching through preferential flow pathways such as cracks and fissures in soil, (3) subsurface lateral flow through preferential paths above the impermeable layer or tile drains, and (4) direct point losses through spray drift, spillage and the equipment-cleanup (Flury et al., 1995; Cessna et al., 2001; Dabrowski and Schulz, 2003). Pesticide leaching in soil is an integral part of the transport processes. Pesticides are transported through the soil profile in both dissolved and colloid-associated forms, and the transportation is affected by such processes as preferential vs. matrix water flow, uptake by plants, sorption to the solid phases, and biodegradation (Schmidt and Pestemer, 1980; Torstensson, 1980).

In this article, we review existing literature relevant to the transport of pesticides from sloping farmland to surface waters. We emphasize the significance of rapid transport pathways in pesticide loss. We also discuss possible measures to minimize such losses. This is of great importance given the increasing frequency of heavy rainfall events over the past few decades in many areas around the world (Easterling et al., 2000).

1 Sources of pesticide pollution: point vs. non-Pesticides and their metabolites enter surface waters from

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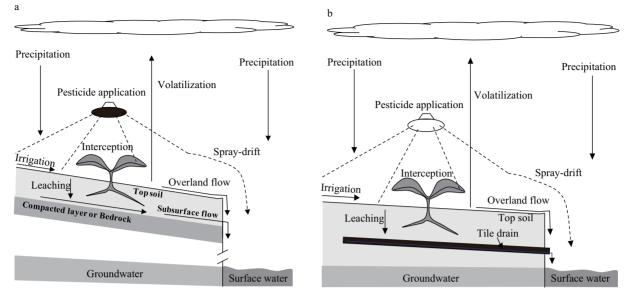


Fig. 1 Schematic representation of pesticide transport routes from sloping farmland with an impermeable layer (a) and tile drains (b) beneath the plough layer into water bodies.

sources which may be categorized as either diffuse or point. In accordance with Carter (2000), diffuse sources are defined as those deriving from agricultural application of pesticides onto farmland. They may result from rainfall- or irrigation-generated surface runoff, throughflow/interflow/subsurface lateral flow, tile drain flow, leaching or spray drift. Point sources of pesticides include farmyard runoff resulting from accidental spills or improper disposal of pesticides, and direct entry into surface waters, such as inappropriate waste disposal or surface water overspray.

The relative importance of various diffuse-source contributions to the total pesticide load has been characterized (Ng and Clegg, 1997; Rawn et al., 1999). At the catchment scale, however, studies estimating the contributions of point and diffuse sources are relatively scarce. Point sources reportedly account for 20%–80% of the pesticide load in rivers in different catchments throughout Europe (Neumann et al., 2003; Leu et al., 2004a).

2 Sorption of pesticides in soil

Soil sorption is one of the most important processes affecting the fate of pesticides in the environment (McCarthy and Zachara, 1989). Sorption retards the transport of dissolved pesticides, but it can enhance the transport of particulate or colloid-associated forms if rainfall or irrigation triggers the discharge of suspended matter. A typical example of the latter is soil erosion, in which the overland flow transports soil particles carrying sorbed pesticides directly to surface water (e.g., Dabrowski et al., 2002). Recent recognition of colloid-associated transport of sorbing contaminants in the subsurface environment (Kretzschmar et al., 1999) further emphasizes the importance of elucidating sorptiondesorption behavior of pesticides under the conditions prevailing in the surface as well as subsurface environment.

Pesticide physicochemical properties (e.g., solubility,

polarity, polarizability, charge characteristics) together with soil chemical properties govern pesticide sorption in soil (Gevao et al., 2000). Clay mineralogy mainly determines the sorption of cationic pesticides such as glyphosate and paraquat, while the sorption of non-ionic pesticides (e.g., chlorpyrifos and diuron) usually depends on the soil organic matter content (Kookana et al., 1998; Wauchope et al., 2002). Biotic or abiotic degradation, volatilization, hydrolysis and photolysis of pesticides are known to depend upon sorption affinity of the pesticides to the principal sorbents (e.g., organic matter and clay minerals) in soil (Kookana et al., 1998).

To predict pesticide transport, either in dissolved or particulate form, the tendency of pesticides toward sorption needs to be evaluated. The tendency is expressed in terms of the sorption coefficient (also called distribution coefficient), K_d , defined as the ratio of the pesticide concentration in the sorbed phase to that in the aqueous solution phase. Since pesticide sorption is often positively correlated with soil organic matter content, the sorption coefficient normalized to soil organic carbon content, K_{OC} , is also commonly used.

Determination of K_d is commonly practiced by batch sorption experiments, in which sorption by soil is calculated from the difference in the initial and final pesticide concentrations in an aqueous solution (e.g., Wauchope et al., 2002). For the weakly sorbing hydrophilic pesticides that pose a greater risk of groundwater pollution, however, it has been argued that the K_d determined by conventional batch sorption is prone to considerable experimental error (Ahmad et al., 2005). An alternative method, utilizing the piston-like displacement of an equilibrated solution in unsaturated soil, has been proposed, and successfully applied to the determination of K_d for weakly sorbing pesticides (Ahmad et al., 2005; Ochsner et al., 2006). For further details about pesticide sorption in soil, readers are referred to Kookana et al. (1998) and Gevao et al. (2000) No. 3

3 Pesticide phases – dissolved vs. particulate

Pesticide is lost from farmland toward water bodies in various forms including dissolved, sorbed on dissolved organic carbon, and sorbed on suspended or colloidal particles (Flury, 1996). Pesticides in each phase have different modes of transport and mobility. In particular, sorption on dissolved organic carbon can effectively increase the mobility of some pesticides (e.g., atrazine, 2,4-dichlorophenoxyacetic acid) and lead to significant transport of these compounds through soil profile (Gao et al., 1998; Li et al., 2005). However, Suba and Essington (1999) reported on the contrary that the sorption of fluometuron and norflurazon on dissolved organic carbon under conservation tillage did not cause higher leaching of these chemicals.

Sorption of pesticides on dispersed particles potentially enhances their transport under circumstances where those particles are transported by water flow (e.g., Vinten et al., 1983; Worrall et al., 1999). This form of transport is known as colloid-facilitated (or particle-facilitated) transport (Kretzschmar et al., 1999). For pesticides showing a high sorption on organo-mineral soil particles, such as glyphosate, trifluralin, paraquat or organochlorine pesticides, pesticide transport by surface runoff is principally associated with the suspended soil particles generated by water erosion (Wauchope, 1978). This is consistent with the concentration of parathion being considerably higher in suspended particles than in the aqueous phase in agricultural streams during rainfall-induced surface runoff events (e.g., House et al., 1992).

Pesticide distribution among different soil particle size fractions is also important in predicting the fate of pesticides in environment, because different-sized particles differ in their pesticide sorption capacity, settling velocity and resultant transport distance and deposition pattern. For instance, Wu et al. (2003) found that, among various size fractions, a colloidal fraction of $< 0.16 \,\mu\text{m}$ exhibited the highest K_d value of 113 L/kg for propiconazole whereas the bulk sample showed a considerably lower K_d of 27 L/kg. They noted that highly mobile particles in the < 2µm fraction can be important carriers of pesticides in the sediment load to rivers.

It has been recognized that soil organic matter plays a key role in the formation of non-extractable pesticide residues (Gevao et al., 2000). Questions remain, however, about how representative the matrix of a real soil is after undergoing the exhaustive and aggressive extraction procedure with organic solvents (Mordaunt et al., 2005). A realistic estimate of the solubility/availability of pesticides, together with the sorbent properties and mobility of the eroded soil particles in finer fractions, is of fundamental importance for predicting the transport of pesticides in the environment.

4 Soil pore system: macropores vs. soil matrix pores

Vertical transport of pesticides in the soil profile is critical-

ly dependent on the way water flows in soil. Water may mainly flow uniformly through soil matrix, or it may concentrate in preferential pathways, leaving the water in the soil matrix effectively immobile. The latter type of water flow, referred to as preferential flow, is of particular interest in relation to the rapid transport of pesticides from farmland. The soil pore system in conjunction with the intensity of water input into the soil together dictate the mode of water flow.

Pores in soil may be divided into macropores and soil matrix pores. Macropores can be defined as pores having an equivalent cylindrical diameter of > 1 mm (Luxmoore, 1981), and consist of inter-aggregate pores, cracks/fractures formed by soil tillage or soil shrinking due to dry-wet cycles, various cylindrical bio-conduits created by soil fauna and plant roots. In soils where the soil matrix has a relatively low unsaturated hydraulic conductivity, water flow is predominated by gravity rather than capillarity, and the flow is preferentially through macropores. Within a wide variety of hydrogeologic settings, water can move quickly to subsurface soil layers through macropores with little interaction between its suspended particles, dissolved and particle-associated contaminants and the soil matrix (Flury, 1996).

5 Flow/transport routes

5.1 Overland flow

Overland flow is a water flow process by which pesticides are transported in dissolved and suspended forms along the surface of sloping land. Overland flow can be categorized into either infiltration-excess overland flow or saturationexcess overland flow, based on mechanisms by which it occurs. The infiltration-excess type of overland flow occurs when the rainfall rate exceeds the rate of water infiltration into soil. This type of overland flow is observed during storm events, typically in silty soils which are prone to soil structure degradation by mechanical compaction or raindrop impact. The saturation-excess type of overland flow, on the other hand, occurs as a result of water saturation in soils having impermeable subsurface horizons and in areas with a shallow groundwater table. This type of overland flow is initiated at the foot slope upon saturation of the soil receiving downslope subsurface flow. For this reason, the foot slope area is more vulnerable to pesticide loss via overland flow than the rest of the catchment (Müller et al., 2006).

The significance of overland flow in the pesticide loss is affected by the following factors: (1) the length of time between a pesticide application and the first subsequent rainfall or irrigation event; (2) the intensity and duration of the rainfall or irrigation; (3) the soil moisture conditions prior to the rainfall or irrigation; (4) the amount and the method of pesticide application; (5) the topography (e.g., slope, length) of the land; (6) the chemical properties (e.g., $K_{\rm d}, K_{\rm OC}$) and degradation rate of the pesticide; (7) soil chemical properties, and (8) vegetation type and density (Shipitalo and Owens, 2003).

The amount and intensity of rainfall strongly affect the contribution of overland flow to total pesticide losses to surface waters. For example, Southwick et al. (2009) reported that metolachlor loss via overland flow during normal rainfall periods was 4.5%-6.1% of the amount applied, whereas leaching accounted for 0.10%-0.18%. A reduction of 35% in rainfall led to a 97% reduction in surface runoff loss of metolachlor and a 71% increase in leachate loss. The first overland flow event usually causes the highest pesticide loss, especially after a long dry period during which numerous pesticide applications have been made (e.g., Riise et al., 2004). A number of studies consistently observed the highest pesticide (e.g., endosulfan) concentrations in receiving water when irrigation occurs soon after a pesticide application (e.g., Kennedy et al., 2001). Based on modeling, Bach et al. (2001) postulated that overland flow is a major path of rapid pesticide losses in Germany.

Pesticide losses by overland flow can take place in dissolved or particle-associated forms. Pesticides with a high affinity to soil sorbents are likely to migrate in the particle-associated form, owing to the erosion of pesticideenriched finer soil particles by overland flow (Ghadiri and Rose, 1991). For strongly-sorbing pesticides with a K_{OC} greater than 1000 L/kg, however, the erosioninduced particle delivery in overland flow acts as the main loss pathway (e.g., Wu et al., 2004). For water-soluble pesticides, losses in the dissolved form are considered far more important than those in the particle-associated forms (Leonard, 1990). Müller et al. (2002) found that the loss of moderately water-soluble triazines pesticides from fallow Hamilton clay loam with a 10% slope was predominantly via overland flow, with 99% of the loss in the dissolved forms. For the more strongly sorbing pendimethalin, 35% of the loss was associated with eroded sediment.

5.2 Preferential flow to tile drains

Preferential water flow through macropores to tile drains plays an important role in the rapid transport of pesticides to surface waters. The existence of macropores may lead to bypassing of the topsoil matrix, which is the most active part of soil in pesticide sorption (e.g., Larsson and Jarvis, 2000) and degradation (e.g., Charnay et al., 2005). Preferential water flow is strongly influenced by the complex interplay between rainfall patterns, soil moisture content at the time of application, and soil water repellency (Jarvis et al., 2008; Lewan et al., 2009). In structured loamy and clayey soils, the rainfall intensity and pattern are among the most important factors influencing pesticide transport by macropore flow (Novak et al., 2001; Lewan et al., 2009). Heavy rainfall soon after a pesticide application may lead to significant and rapid losses to surface waters via macropores and tile drains. Stone and Wilson (2006) reported that preferential flow contributed 11% and 51% of the total tile drain flow for two storm hydrographs, with positive correlations between the glyphosate concentrations in the tile drainage and the contribution of preferential flow.

Another factor that may influence the response of tile drain flow, and hence rapid pesticide loss, is the proximity of the capillary fringe from the groundwater table (i.e., the zone of tension saturation) to the land surface. If the capillary fringe rises to the land surface, small amounts of rainfall may cause large increases in tile drain flow due to the saturation and the hydraulic connectivity of tile-drainsoil system (Stone and Wilson, 2006).

Pesticide transport by preferential flow to drains, coupled with transport of mobile particles such as clay particles and organic colloids through soil macropores, can lead to high transient pesticide concentrations in agricultural ditches and small rivers (Brown et al., 2004; Leu et al., 2004b). The transport is storm-driven, and the initial storm after a pesticide application produces higher pesticide concentrations in the tile drain flow than subsequent storms (Kladivko et al., 1991; Stone and Wilson, 2006).

Flury (1996) noted that seasonal or annual loss of pesticides through tile drains usually accounts for < 0.1% to 1% of the applied mass, but occasionally the loss can reach up to 4%. Kladivko et al. (2001) found that while the drain flow volumes in the humid Midwestern United States accounted for 0–40% of the annual rainfall, pesticide losses via tile drains were usually less than 0.5% of application, and that overland flow transported a greater amount of pesticides to surface waters than tile drain flow. In areas where overland flow is not significant because of their soil's high infiltration capacity, tile drain flow may be of greater significance in transporting pesticides to surface waters (Stone and Wilson, 2006).

5.3 Subsurface lateral flow

In regions where soil is underlain by impervious subsoil or rocks, water that has percolated downward through the soil changes its flow direction, producing subsurface lateral flow. Such flows often make a considerable contribution to rapid pesticide discharge from farmland, particularly when the soil immediately above the impervious layer contains macropores. For example, structured loamy and heavy clay soils are often underlain by relatively impermeable geological media (e.g., consolidated soil horizons and rocks), resulting in the occurrence of rapid subsurface lateral flow above the impermeable layer (McDaniel et al., 2008; Zhu et al., 2009). The subsurface lateral flow may be very rapid, with a mean pore velocity as high as 18.7 m/day on the top of a fragipan, and may account for up to 90% of the rainfall received on site during late winter/early spring (McDaniel et al., 2008). Pesticides with a smaller tendency toward sorption usually have a greater potential to enter surface waters through subsurface lateral transport (Kookana et al., 1998).

5.4 Leaching

In sandy soils with low organic matter content as well as in the macropore flow-dominated loamy or clayey soils, weakly sorbing and/or persistent pesticides are most at risk of leaching under high precipitation conditions (Reichenberger et al., 2007). Preferential pathways in structured soils may provide a short circuit from the soil surface not only to the surface waters via tile drains but also to the groundwater. Pesticides may be concentrated

· 65

along the walls of the preferential paths. Leaching through preferential pathways explains the detection of strongly sorbing pesticides such as chlorpyriphos in groundwater (e.g., Kookana et al., 1998).

Flood irrigation also enhances pesticide leaching (Domagalski and Dubrovsky, 1992). It has been reported that flood irrigation in a farm led to leaching of pesticides into the shallow Gulf Coast aquifer (Chang et al., 2008).

Among the worst cases of pesticide leaching. Hall et al. (1989) reported that 8.4% of the applied simazine and 9.6% of atrazine leached more than 1.22 m into an untilled silty clay loam after 1000 mm of rainfall, and Flury et al. (1995) detected 4.3% of the applied atrazine leached more than 0.5 m into a tilled loamy soil after 90 mm of cumulative infiltration. The rapid transport of a fraction of pesticides in both cases was ascribed to preferential water flow through cracks or root channels in soils.

5.5 Direct point losses

Direct point losses of pesticides occur through spray drift, spillage, the cleanup of pesticide application equipment, and other operations. Spray-drift during pesticide application is affected by such factors as equipment design, pressure and droplet size, spray type, and meteorological conditions (Gil and Sinfort, 2005). Spray drift is an important route for pesticides into surface waters and should be taken seriously in view of the directness of the input and the high pesticide bioavailability (Schulz et al., 2001). Its contribution to surface water pollution in European countries is, however, thought to be rather small (e.g., Neumann et al., 2002; Ropke et al., 2004) although contributions from spray-drift were observed in ditches (Meli et al., 2007). The importance of rapid direct point losses, including spray-drift, tank filling, spillages, faulty equipment, washing, waste disposal and overspray of surface waters, has been confirmed by monitoring campaigns (Carter, 2000; Holvoet et al., 2007).

6 Mitigation strategies to reduce pesticide input to surface waters

6.1 Grassed/vegetated buffers/barriers, depressions, waterways and wetlands

A number of studies have verified that vegetated buffer/filter strips (VBSs/VFSs) are effective in reducing overland flow and soil erosion (e.g., Patty et al., 1997). VBSs/VFSs reduce pesticide loss by (1) facilitating the deposition of particles which sorb pesticides, (2) enhancing pesticide retention/sorption by increasing the time available for infiltration, (3) sorbing dissolved-phase herbicides to the grass, grass thatch and soil surface, and (4) reducing the volume of overland flow containing dissolved and particle-associated pesticides (Schmitt et al., 1999; Krutz et al., 2004).

Vegetated buffer strips are shown to have high removal efficiencies for pesticides and sediments (e.g., Syversen and Bechmann, 2004; Vianello et al., 2005). Popov et al. (2006) reported reductions in the total load of atrazine

by 40%-85% by adopting vegetated biofilters. A review by Sabbagh et al. (2009) documented load reductions for pesticides including metribuzin, isoproturon, metolachlor, atrazine, cyanazine, terbuthylazine, lindane, didlufenican, and pendimethalin of 11%-100% by VFSs ranging in width from 0.5 to 20.1 m. Syversen and Bechmann (2004) found average removal efficiencies by VBSs of 39%, 71%, 63% and 62% for glyphosate, fenpropimorph, propiconazole and soil particles, respectively.

Performance of VFSs for pesticide trapping depends on the hydrologic conditions (i.e., precipitation, infiltration and overland flow), the VFSs design (i.e., width, slope, and density and height of vegetation cover), and characteristics of the particles and pesticides (e.g., Sabbagh et al., 2009). It should be noted that the environmental fate of the pesticides and their metabolites retained in VBSs/VFSs has rarely been evaluated. Concerns remain regarding the subsequent release of the trapped pesticides (e.g., Delphin and Chapot, 2001).

During the occurrence of overland flow caused by heavy rainfall events, large water volumes may be produced in a short time, which in some cases will not be retained by any of the widely used buffer strip/zones (Schulz, 2004). More effective measures to reduce pesticide loss associated with overland flow/soil erosion include grassed depressions, waterways, ditches and wetland, which can effectively reduce the volume and velocity of overland flow, prevent gullying, and retain sediments and harmful substances from adjacent fields (Rodgers and Dunn, 1992; Briggs et al., 1999). Moore et al. (2001) suggested wetland buffer travel distances of 100-400 m (for fields 4, 40, and 400 ha in size) would be necessary to effectively mitigate metolachlor runoff from potential contaminating of receiving surface water. More detailed information on the efficacy of vegetated buffers in trapping sediment and reducing pesticide losses to surface waters were discussed by Krutz et al. (2005) and Liu et al. (2008).

Assessing the cost/impact of VBSs/VFSs would be a prerequisite to any widespread promotion of them as a tool for reducing pesticide losses to surface waters. Sieber et al. (2010) assessed the cost/impact ratios of various riparian buffer strip widths by integrating a biophysical model and an economic model. Gutsche and Roßberg (1997) used the Synoptic Assessment Model for Pesticides (SYNOPS) to estimate potential eco-toxicological risk and impact caused by spray drift and runoff of pesticides into surface waters. Costs in converting agricultural land to riparian buffer strips were estimated by Kreins and Cypris (1999) using the agricultural sector (cost) model Regionalized Agricultural and Environmental Information System for the Federal Republic of Germany (RAUMIS). The cost/impact ratios depended on the size of the VBSs/VFSs. Sieber et al. (2010) found that a 3-m wide buffer strip reduced the pesticide risk by 60% whereas extending 30-m wide strips to 50 m reduced the risk by only 2%.

Good agricultural practices (GAPs) here refer to agricul-tural management practices aiming at minimizing of the

movement of pesticides to surface waters during rain or irrigation, and are among the mitigation measures to reduce rapid pesticide loss. Examples of GAPs with respect to pesticide application are the use of drift-reducing nozzles (Campbell et al., 2004), band spraying on row crops (Baker et al., 1995), application restrictions for vulnerable soils and/or wet climates, and simply keeping a sufficient distance, e.g., between 2 and 200 m, from the adjacent water bodies when spraying (Holvoet et al., 2007).

Restricting pesticide applications, particularly in the risk areas, could be an effective approach to reducing surface water contamination (Leu et al., 2004b). This is because pesticide losses are much more sensitive to the variability of field-specific characteristics than the differences in compound-specific properties of the pesticides (Leu et al., 2004b). Reduction in pesticide application may be achieved by suggesting to farmers when to use pesticides (Campbell et al., 2004), and employing biological control (Corrales and Campos, 2004) or an integrated approach to pest management (Mansingh et al., 2007).

de Snoo and de Wit (1998) found that a 3-m wide no-spray cropped buffer decreased drift deposition in a ditch by at least 95% in the Netherlands. Under typical agricultural conditions in Italy, spray drift in vineyards occurs at a distance more than 24 m, which may be taken as the minimum width of no-spray zone required for avoiding direct pesticide contamination of surface water (Vischetti et al., 2008). Models have also been used to estimate spray drift and the required minimum no-spray zone from the edge of the surface waters (e.g., Holterman and van de Zande, 2003). Field evaluation experiments under realistic and representative conditions need to be conducted to validate the models. Social-economic factors should also be taken into account in determining the minimum width of no-spray buffer zones along the edge of surface waters.

Another mitigating GAP concerns tillage practice. Tillage alters soil hydraulic properties, and hence the pathways of water flow and pesticide transport through the soil. In soils where preferential flow is significant, conventional tillage can reduce pesticide leaching by disrupting continuous macropore flow paths (Isensee et al., 1990). In contrast, in soils where water flow is mainly through the soil matrix, conventional tillage may enhance pesticide leaching as compared with no-tillage and reduced tillage (Gish et al., 1995; Sadeghi et al., 1998). This has been attributed to weaker pesticide sorption associated with the lower soil organic carbon content in the conventionally tilled soils (Sadeghi et al., 1998). Conservation tillage (i.e., reduced or zero tillage) increases retention/sorption of pesticides in topsoil, particularly by its organic components, while decreasing the availability of pesticides for biological degradation, leading to enhanced persistence in soils. This persistence of pesticides is partially compensated for by more intensive microbial activity under conservation tillage. The effects of stronger sorption of pesticide in reduced tillage systems may be counter-balanced by the increased preferential transport in the soils having improved macropore connectivity (Larsbo et al., 2009).

Tillage practice also influences pesticide transport to

surface waters via overland flow. The pesticides transported by overland flow may be in dissolved form or particle-associated form. Adoption of conservation tillage systems can reduce the transport of strongly sorbing pesticides. In zero tillage, for example, crop residues left on the soil surface after harvest protect soils from erosion (Ritter, 2001), thereby reducing losses of the pesticides sorbed on the soil particles. Moderately sorbing, watersoluble herbicides are, however, transported primarily in dissolved form so that the losses may only be reduced with management practices that reduce runoff volume (Wauchope, 1978).

Tile drainage, a common agricultural water management practice, can also be a mitigation practice in areas with a shallow groundwater table or seasonally perched water tables. Tile drainage was found to reduce the overland flow volume by 38%, resulting in a reduction of metolachlor and atrazine losses by 56% compared with no tile drainage (Southwick et al., 1990). As described before, tile drains can act also as rapid pathways for pesticide transport if water flow to the tiles is predominated by preferential flow through macropores. Gaynor et al. (1995) noted that other factors, such as the timing of overland flow or tile drain discharge relative to the time of pesticide application, and the antecedent soil moisture conditions prior to an overland flow event, are more important in controlling pesticide loss than tillage practice. The effectiveness of tile drainage to mitigate pesticide loss is thus dependent on management practice, soil type, soil hydraulic properties, pesticide solubility and other environmental factors (Jury, 1986).

7 Research needs in sloping farmlanddominated catchments and regions

On sloping farmland prone to soil erosion and underlain by an impermeable layer, overland flow, preferential flow to tile drains and subsurface lateral flow can transport significant amounts of pesticide in dissolved and particleassociated forms to surface waters. Pesticide transport processes under such conditions, however, have not been fully elucidated, particularly in hilly or mountainous regions. For example, in the upper reaches of Yangtze River in China, more than 65% of the total farmland is on slopes where a loamy soil is underlain by purplish shale and forms a typical binary structure of soil-bedrock. The soil is particularly vulnerable to water erosion, owing to intensive cultivation and the area's wet climate (Zhu et al., 2009). According to Zhu et al. (2009), the average discharge of the subsurface lateral flow in the rainy season accounted for 63% of total runoff (i.e., overland flow plus subsurface flow discharge). Thus, there has been increasing concern about possible rapid transport of pesticides via overland and subsurface flows toward the branches and confluences of the Yangtze River. To the best of our knowledge, however, there have been no reports on the pesticide loss to surface waters in these areas at catchment and regional scales. Elucidating pesticide transport processes from sloping farmland is a prerequisite for evaluating the

risk of surface water contamination by pesticides, and developing appropriate mitigation strategies in such areas. We identify below the research needs related to pesticide loss in catchments dominated by sloping farmland.

7.1 Elucidating and modeling rapid pesticide transport at different scales

Existing knowledge about pesticide transport processes obtained in the laboratory and fields need to be upscaled for predicting and assessing the risk of surface water contamination by pesticides. Flow and transport processes in soil are heavily dependent on the geometry and hydraulic properties of soil pores, particularly those of macropores which show large spatial and temporal variability (Beven, 1991; Jury and Flüler, 1992). Large variations in transport behavior were observed between undisturbed soil columns in the laboratory (e.g., Jarvis et al., 1994; Lennartz, 1999). Even in a one-dimensional network, this has caused difficulties in upscaling from the column to the field scale, as parameters for the macropore flow are often poorly identifiable (e.g., Akhtar et al., 2003). Upscaling the existing knowledge is a challenging task in elucidating pesticide losses from sloping farmland.

The contributions of overland flow and subsurface lateral flow to the total pesticide loss from sloping farmland need to be evaluated. Capel et al. (2001) found in the large catchments (> 10^7 ha) that less than 2% of the applied pesticide was ultimately transported from agricultural fields to surface waters, with pesticide loss occurring primarily during and immediately after the application period. In the regions dominated by sloping farmland, a larger fraction of pesticides is expected to be transported to surface waters, owing to a larger contribution from overland flow and/or subsurface lateral flow. Overall pesticide loss in a catchment should be evaluated by taking into account weather conditions, soil type, land use, properties of the pesticide and point sources.

Relative contributions of the dissolved and particleassociated form to the total pesticide transport depend on rainfall patterns, land topography, soil hydraulic and chemical properties and the chemical properties of pesticides (McCarthy and Zachara, 1989; Worrall et al., 1999; Wu et al., 2004). Pesticide phases are likely to greatly influence the mobility in different circumstances. Their contributions to the pesticide losses from sloping farmland remain to be determined.

Spatial differences in pesticide losses in a catchment should also be elucidated. Pesticide losses are mainly driven by a few discharge events during and shortly after application. The highly dynamic pattern is probably due to the fact that pesticide enters surface waters via various rapid flow pathways. Therefore, spatial differences in the loss behavior are likely to be strongly affected by spatial variations in soil susceptibility to surface saturation and subsequent rapid transport from fields via overland flow, subsurface lateral flow, and tile drain flow (Blanchard and Lerch, 2000; Leu et al., 2004a, 2004b). Despite the complexity of the flow processes governing pesticide transport from fields to water bodies, the temporal dynamics of pesticide concentrations in surface waters often exhibits a fairly simple pattern (Siber et al., 2009). Monitoring studies on pesticide losses to surface waters are necessary to see if this also holds at the sloping farmland-dominated catchments of hilly or mountainous regions.

When a pesticide migrates with a limited water-soil matrix contact time, the influences of hydrology may dominate over the differences in sorption and biodegradation behaviors (Larsson and Jarvis, 2000). It remains to be determined and modelled to what extent sorption and biodegradation affect pesticide losses from sloping farmland where rapid water flow is expected.

Modeling pesticide transport via preferential flow through structured agricultural soils in sloping farmland should be pursued. The past decade has seen a considerable progress in modeling pesticide transport in structured soils using two- or three-domain models such as RZWQM (Root Zone Water Quality Model) (Ahuja et al., 2000), MACRO (Jarvis et al., 1994), and HYDRUS-1D (Šimůnek et al., 2005). Preferential pesticide transport models were also coupled with Geographical Information Systems and groundwater flow models for application at the catchment and regional scales. However, appropriate parameterization of preferential flow and pesticide processes remains an obstacle to model applications (Köhne et al., 2009), leaving a challenging subject in agricultural landscapes characterized by complex topography, high spatial variability of land use and strong surface-subsurface flow interaction such as those in hilly or mountainous southwestern China.

7.2 Release of pesticides retained in mitigating buffer zones and potential counter measures

Pesticides trapped by VBSs/VFSs, vegetated depressions, waterways and wetland may be released or biodegraded to more mobile compounds under certain conditions, and this topic therefore needs further attention. Krutz et al. (2004) found that the mobility of metolachlor metabolites was greater than metolachlor in VFSs and cultivated soil. Higher organic carbon content in VFSs relative to cultivated soil, however, may limit subsequent transport of the compounds from the vegetated filter strip. There is a clear need for research to investigate potential release of trapped pesticides through dissolution, desorption, and resuspension of soil particles in buffer zones and for developing counter measures against such pesticide release.

7.3 Integrated approaches for reducing pesticide loss and evaluation by field monitoring

Research needs also exist for integrated approaches for reducing pesticide loss and evaluation by field monitoring. Most of the foregoing studies have focused mainly on isolated mitigation approaches, and very few direct comparisons have been made of the effectiveness in reducing rapid pesticide losses toward water bodies. Evaluating the combined effects of several mitigation strategies at the regional/catchment scales has been scarcely attempted. Extensive monitoring data and intensive observation of integrated good agricultural practices are essential for assessing the effects of GAPs in a catchment. To optimize the field monitoring schemes, combined use of historical monitoring data and appropriate models is recommended. As an important element of a successful field campaign for implementing integrated mitigation measures, a participatory approach needs to be adopted to motivate farmers not only to minimize pesticide use and optimize application method, but to evaluate the environmental, health and economic benefits from taking the measures.

8 Conclusions

This article provides an overview of issues relevant to rapid pesticide transport from sloping farmland to surface water bodies and of possible mitigation strategies. The main conclusions are:

Rapid pesticide transport via overland flow, subsurface lateral flow, and leaching through preferential pathways in soil as well as direct losses through spray drift and spillage is very likely to make a major contribution to the pesticide losses to surface waters from sloping farmland and in farmland underlain by an impermeable layer.

Rapid transport of pesticide toward surface waters may be reduced by implementing simple practices, such as constructing vegetated buffers and barriers at relatively low cost. The effectiveness of the practices should be assessed using a calibrated, validated biophysical-economic model with a novel cost/impact ratio indicator.

At agricultural catchments of hilly or mountainous regions where sloping farmland is dominant and the soil is prone to overland flow as well as fast subsurface lateral flow, there is a clear need to identify possible rapid pesticide transport routes, and evaluate their contributions in pesticide losses toward surface waters.

Field experiments including mitigation practices and monitoring should be performed at different scales, from plot and slope scales to the catchment scale. The field experiments should be combined with scenario-based modeling exercises to gain insight into which measures or integrated approaches are the most effective, and to help accurately build catchment/regional management plans.

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