



## Critical source areas for herbicides can change location depending on rain events



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### ABSTRACT

During rain events, herbicides can be transported from their point of application to surface waters, where they may harm aquatic organisms. Since the spatial variability of herbicide losses to streams can be large, the identification of critical source areas could help to target mitigation measures efficiently to those locations where they reduce herbicide pollution the most. We performed a controlled herbicide application on wheat (isoproturon) and corn fields (atrazine, S-metolachlor and sulcotrione) in an agricultural head-water catchment (about 1 km<sup>2</sup>) in the Swiss Plateau to investigate the spatial variability of herbicide losses. We performed spatially distributed discharge measurements and high-resolution water sampling during rain events after herbicide application to determine herbicide loads and loss rates for individual fields. The dry weather conditions after the wheat herbicide application resulted in a very low isoproturon loss rate (0.005% of the applied amount). In contrast, the corn herbicide application was followed by several rain events with varying intensities and magnitudes causing loss rates of 0.26, 0.16 and 0.26% for atrazine, S-metolachlor and sulcotrione, respectively. The spatial differences in loss rates between fields were about a factor of three in most events. However, the spatial loss pattern varied between events implying that being a critical source area is not a temporally stable field property. No correlations were observed with several field characteristics (connectivity of the fields to the stream, the tendency for topsoil saturation, field-specific herbicide dissipation rates and sorption affinities). However, the data suggest that critical source areas may depend on the type of rain event because infiltration and saturation-excess runoff affects different parts of the catchment.

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### 1. Introduction

In modern agriculture, a wide variety of pesticides is used to increase crop productivity. They encompass a broad range of chemicals and are used to control weeds, to fight plant diseases, insects, arachnids and other pests. Pesticides can enter the water system, where they can harm aquatic organisms even in low concentrations. Small streams in catchments with intensive crop production are especially at risk (Liess and Schulz, 1999), as diffuse pollution

from agricultural fields causes major inputs to the stream in these areas (Leu et al., 2010; Stehle et al., 2011). Pesticides mainly enter surface waters during rain events, when they are mobilized and transported with fast runoff (surface runoff and preferential flow to subsurface drainage systems), with the pathway through groundwater and baseflow being of little importance for most pesticides (Thurman et al., 1991). Several cases showed that herbicide loss rates (relative to the applied amount) from different fields within a given catchment can differ by over an order of magnitude (Louchart et al., 2001; Leu et al., 2004b; Gomides Freitas et al., 2008). This implies that a relatively small proportion of a catchment can cause the major part of surface water pollution with herbicides. The same has been observed for diffuse pollution of surface waters with phosphorus (Pionke et al., 1996, 2000). The areas that contribute a large fraction of the pollution load are called critical source areas (CSAs) or contributing areas (Pionke et al., 1996). The insight that not all parts of a catchment have the same relevance for diffuse pollution offers efficient mitigation options, because actions on a small

*Abbreviations:* CSA, critical source area; LOD, limit of detection; LOQ, limit of quantification; PLE, pressurized liquid extraction; DT<sub>50</sub>, half-life time; DT<sub>90</sub>, time needed to degrade 90% of the initial amount.

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proportion of the area can strongly reduce the substance input to the stream. An area has to fulfill three conditions to become a critical source area for pesticides: (1) Pesticides have to be applied on the area (or reach the area by drift in relevant amounts). (2) The area has to be hydrologically active, i.e. the relevant mobilization and fast transport processes do occur. (3) The area has to be directly connected to the stream such that fast flow reaches the stream without relevant retention processes (Frey et al., 2009). The spatial extent of the CSAs ( $A_{CSA}$ ) can be interpreted as the spatial intersection of the areas of a catchment where each condition is fulfilled:

$$A_{CSA} = A_{source} \cap A_{active} \cap A_{connect} \quad (1)$$

with  $A_{source}$  representing the source area of a given compound,  $A_{active}$  the hydrologically active area, and  $A_{connect}$  the part of the catchment in direct connection to the stream network. For pesticides, the source areas  $A_{source}$  correspond primarily to the areas of pesticide applications but may encompass also additional surfaces where pesticide are deposited in relevant amounts due to drift deposition. The compound's chemical properties can modify  $A_{source}$  in space and time. Degradation and sorption both determine the amount of substance that is available for transport at the time of rainfall (Louchart et al., 2001). There may be substantial spatial variability in sorption and degradation rates of pesticides in soils (Ghafoor et al., 2011a,b) within fields and small catchments possibly affecting the spatial CSA distribution. Earlier studies in the Swiss Plateau (Leu et al., 2004b; Stamm et al., 2004), however, indicate that degradation rates and sorption coefficients did not vary strongly between fields in the corresponding study catchment and could not account for observed spatial differences in herbicide loss rates (Leu et al., 2004b, 2005; Gomides Freitas et al., 2008). In those studies the spatial variability of herbicide loss rates was attributed to the susceptibility of the fields to generate fast flow and the connectivity of the fields with the stream.

Even though the chemical properties of the pesticides may not necessarily determine the spatial pattern of losses, they are important in determining the pesticide mobilization and transport behavior. While the pesticide half life in soil determines the amount of pesticide that is present in soil at the time of rainfall (e.g., Louchart et al. (2001)), the sorption behavior can affect both mobilization and transport. For many pesticides it has been shown that sorption equilibrium is only reached after weeks or months and therefore kinetic sorption has to be considered (see Vereecken et al., 2011 for a recent review of pesticide sorption studies). Several field studies have shown that sorption strength influences pesticide losses to streams and tile drains, leading to lower loss rates and lower peak concentrations for substances with stronger sorption (Louchart et al., 2001; Leu et al., 2004a; Gomides Freitas et al., 2008; Brown and van Beinum, 2009).

A reliable spatial prediction of CSAs is necessary if site-specific mitigation measures like reduced application rates or changes in crop rotations should be implemented in practice. The relevant scale for a site-specific management of CSA is the sub-field to small subcatchment scale (i.e. fractions of a hectare to few hectares under typical Swiss conditions). However, there are only few comprehensive field data sets available that allow a quantification of spatial differences in herbicide losses at this scale (Leu et al., 2004b; Gomides Freitas et al., 2008). The quantification of spatial differences is not possible with conventional monitoring data because the different fields are usually not sprayed at the same day with the same substances. Therefore different weather conditions after application and different substance properties would strongly influence the results and prevent a meaningful spatial interpretation. To quantify spatial differences, a controlled application (same substances applied at the same day on different fields) and spatially distributed sampling are required.

In this paper we present the results of a controlled herbicide application in the catchment of a first order stream where we quantified the spatial differences of herbicide loss rates at the scale of fields and small subcatchments. We selected a catchment with a high variability of soil types ranging from well-drained Cambisols to poorly drained Gleysols (high tendency for topsoil saturation) to test the hypothesis that the soil moisture regime determines the generation of fast flow (surface runoff and preferential flow to tile drains) and therefore the spatial variability of herbicide losses. Although soil texture and organic matter content did not vary to large degree across the catchment, the treated fields vary strongly in the percentage of areas with a high tendency for topsoil saturation and hence also in their drainage density. We therefore expected a high spatial variability of herbicide loss rates because previous studies in the region had demonstrated saturation-excess runoff is a major process for herbicide transport under the prevailing soil and climatic conditions.

In a previous paper (Doppler et al., 2012), we have demonstrated that the main process of herbicide mobilization and the initial transport mechanism is surface runoff on the fields. However, overland flow hardly reaches the stream directly but it is redirected into the wide-spread subsurface drainage systems. Surface connectivity to the open stream, which means a topographic situation such that surface runoff generated within the catchment can flow on the soil surface into the stream without being retained in a topographic depression, is very low in our catchment (4.4% of the area). However, hydraulic shortcuts like manholes in topographic depressions or storm drains on roads and farmyards establish the connectivity for surface runoff by routing it to subsurface drains or storm sewers connected to the stream. This direct connectivity is complemented by indirect connectivity which means that runoff may be transmitted through macropores to underlying drains (Doppler et al., 2012) from local depressions. Such conditions of direct and indirect connectivity are widespread in the Swiss Plateau.

This paper focuses on quantifying the spatial heterogeneity of herbicide losses within the catchment and investigates whether the spatial patterns observed can be explained by the site-specific factors and herbicide properties like dissipation rates.

## 2. Materials and methods

### 2.1. Site description

The Eschibach catchment is located in the northeast of Switzerland (Fig. 1). The catchment area is 1.2 km<sup>2</sup>, topography is moderate with altitudes ranging from 423 to 477 m above sea level. The twenty-year mean annual precipitation at the closest permanent measurement station (Schaffhausen, 11 km north of the catchment, 1989–2008) is 900 ± 165 mm (Meteoschweiz, 2009). The soils developed on moraine material with a thickness of around 10 m which is underlain by Freshwater molasse (Süßwassermolasse; (Einsele, 2000; swisstopo, 2007). Soils in the center of the catchment are poorly drained Gleysols. Well drained Cambisols, and eroded Regosols are located in the higher parts of the catchment. Topsoil texture is rather homogeneous in the catchment with clay contents between 20 and 30% and silt contents of around 30% (FAL, 1997). The dominant land use is crop production (75% of the area), around 13% of the catchment is covered by forest, and a small settlement area is located in the southeast of the catchment. Forty seven percent of the agricultural land is drained by tile drains. The stream system consists of two branches, an open ditch that was partly built as recipient for the drainage water, and the main branch of the stream that runs in a culvert (Fig. 1). Further details on the catchment can be found in (Doppler et al., 2012). Table 1 shows some soil characteristics of the experimental corn fields.

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