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Spatial and temporal patterns of pesticide concentrations in streamflow, drainage and runoff in a small Swedish agricultural catchment

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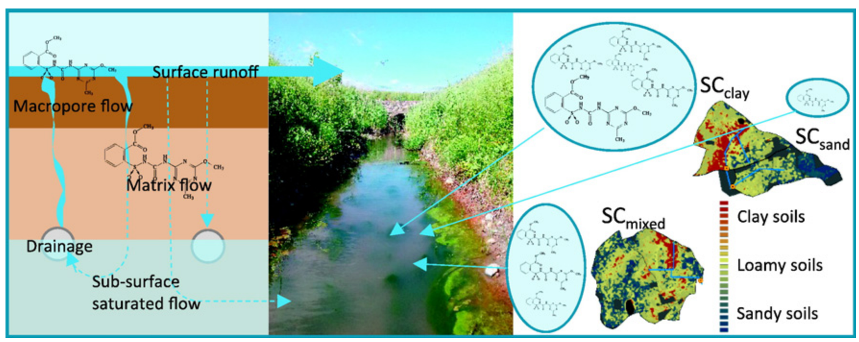
HIGHLIGHTS

- Pesticides were sampled in streams, drainage and runoff in three small sub-catchments.
- Detections were most frequent in the catchment with a large proportion of clay soils.
- Only two compounds found at small concentrations in an area dominated by coarse soils.
- The spatial pattern of detections in streamflow was consistent in three sampling years.
- Losses characterized by fast macropore flow and long-term subsoil storage of residues.

GRAPHICAL ABSTRACT

This paper investigated whether spatial variation in pesticide occurrence in the stream draining a small Swedish agricultural catchment could be related to spatial variation in soil properties, and also assessed the relative importance of surface and subsurface transport pathways.



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ABSTRACT

A better understanding of the dominant source areas and transport pathways of pesticide losses to surface water is needed for targeting mitigation efforts in a more cost-effective way. To this end, we monitored pesticides in surface water in an agricultural catchment typical of one of the main crop production regions in Sweden. Three small sub-catchments (88–242 ha) were selected for water sampling based on a high-resolution digital soil map developed from proximal sensing methods and soil sampling; one sub-catchment had a high proportion of clay soils, another was dominated by coarse sandy soils while the third comprised a mix of soil types. Samples were collected from the stream, from field drains discharging into the stream and from within-field surface runoff during spring and early summer in three consecutive years. These samples were analyzed by LC-MS/MS for 99 compounds, including most of the polar and semi-polar pesticides frequently used in Swedish agriculture. Information on pesticide applications (products, doses and timing) was obtained from annual interviews with the farmers. There were clear and consistent differences in pesticide occurrence in the stream between the three sub-catchments, with both the numbers of detected compounds and concentrations being the largest in the area with a high proportion of clay soils and with very few detections in the sandy sub-catchment. Macropore flow to drains was most likely the dominant loss pathway in the studied area. Many of the compounds that were detected in drainage and stream water samples had not been applied for several years. This suggests that

Abbreviations: CSA, critical source area; LC-MS/MS, liquid chromatography and tandem mass spectrometry; SC_{clay}, one of three investigated sub-catchments in the present study with the largest proportion of clayey soils; SC_{mixed}, one of three investigated sub-catchments in the present study with a mixture of clayey and sandy soils; SC_{sand}, one of three investigated sub-catchments in the present study with a large proportion of sandy soils; a.i., active ingredient; LOD, limit of detection; LOQ, limit of quantification; K_{foc}, Freundlich adsorption coefficient; DT50, dissipation half-life; PPDB, pesticide properties database; GUS, groundwater ubiquity score.

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despite the predominant role of fast flow pathways in determining losses to the stream, long-term storage along the transport pathways also occurs, presumably in subsoil horizons where degradation is slow.

1. Introduction

Reducing losses of pesticides to surface waters from agricultural land still remains a major challenge worldwide. For example, in Sweden, 46% of all samples from national monitoring campaigns carried out in the period 2002–2013 contained one or more compounds at concentrations larger than eco-toxicological guideline values (Lindström and Kreuger, 2015). Losses of pesticides to surface water occur through spray drift, surface runoff and erosion, subsurface saturated flow (base-flow) and discharge from drainage systems (Brown and van Beinum, 2009; Burgoa and Wauchope, 1995; Holvoet et al., 2007). A few studies at the catchment scale have shown that losses of pesticides to surface water may originate from a relatively small fraction of the agricultural landscape (Doppler et al., 2012; Freitas et al., 2008; Frey et al., 2009; Leu et al., 2004). These 'critical source areas' (CSA) are thought to represent areas of land that are highly susceptible to the fast transport processes that are triggered when the soil surface is at or close to saturation, primarily infiltration excess (Hortonian) surface runoff or rapid subsurface flows through soil macropores to subsurface drainage systems (e.g. Blanchard and Lerch, 2000; Holvoet et al., 2007; Leu et al., 2010). Clearly, cost-effective improvements in water quality would result if mitigation efforts were focused on those areas of land that are the most vulnerable to pesticide losses. At the regional scale, Blanchard and Lerch (2000) showed that pesticide detections in surface water were correlated with broad runoff classes derived from soil maps. Therefore, it seems possible that high-resolution digital soil maps could serve as a simple 'proxy' method to enable farmers and their advisors to identify CSA's and effectively target mitigation practices. However, it is not yet clear whether the location of CSA's would also depend on compound properties related to sorption strength and persistence. Gassmann et al. (2015) investigated this question using a catchment-scale simulation model. They concluded that the locations of CSA's were relatively stable regardless of pesticide properties, but supporting experimental evidence from field studies is still lacking. Temporal variations in weather conditions may also introduce additional complexity, as the relative importance of different flow paths to surface water as well as the size of any given CSA may depend on antecedent soil moisture contents and rainfall characteristics (e.g. Doppler et al., 2014).

Although the diffuse pathways by which pesticides are transported to surface waters are known, only a few studies have quantified their relative importance at field or landscape scales. Buttle and Harris (1991) found surface runoff to be the dominating loss pathway of metolachlor from a loamy field in Ontario, Canada, with an average slope of 6%. Measured concentrations and loads transported in surface runoff from two downslope plots were higher than those from two uphill plots in the first few runoff events when the water table in the two lower-lying plots was still high. They also measured higher concentrations in runoff from one of the uphill plots than from the other, owing to the steeper slope of the former plot and wheel tracks running parallel to the direction of the slope (Buttle and Harris, 1991). Riise et al. (2004) found similar or larger concentrations and greater total pesticide losses in drainage than from surface runoff in an artificially-levelled field with a silty clay loam soil. In a controlled-application experiment, Leu et al. (2004) concluded that surface runoff was the main contributing loss pathway of corn herbicides in a small agricultural catchment in Switzerland. In sub-catchments of the studied area where surface runoff was prevented from reaching the brook, or where topography was flat enough for ponded water to infiltrate, rapid transport in macropores to drainage pipes was the dominant loss pathway (Leu et al., 2004). Very few studies have compared losses through spray drift with those from surface runoff and drainage. However, in a small catchment in France dominated by vineyards, Lefrancq et al. (2014) found that the loads of fungicides transported in runoff increased at the catchment scale compared with the plot scale due to spray drift deposition on roads where runoff coefficients are generally much larger. Little is known about the relative importance of different flow pathways for losses of pesticides to surface waters in Sweden. Most of the arable land in Sweden is relatively low-lying and flat or only gently undulating and approximately 50% of the agricultural area is systematically subsurface drained. As a result, it is presumed that drainage is the dominant loss pathway of pesticides to surface waters, while surface runoff mainly occurs during snowmelt on frozen soil in winter. However, data that might confirm this assumption are limited. Measurements of pesticide concentrations and loads in surface runoff and drainage have been reported for only a few plot- and field-scale studies (Larsbo et al., 2016; Ulen et al., 2012; Ulen et al., 2014), while targeted monitoring data at the catchment scale is lacking. Here, we present the results of a study conducted in a small agricultural catchment in the south of Sweden, where pesticide concentrations in drainage, surface runoff and stream discharge were monitored at multiple locations during three consecutive growing seasons. Our main aims with this study were to i.) assess whether spatial differences in pesticide losses to surface water at the small catchment scale could be related to differences in soil properties based on high-resolution digital soil maps and ii.) determine the relative importance of surface and subsurface transport pathways for these losses.

2. Material and methods

2.1. Study site, selection of sub-catchments and soil mapping

The E21 catchment is located in southern Sweden close to Lake Vättern (Fig. 1a) and has an approximate area of 16 km^2, of which 92% is agricultural land mainly used for crop production. Mean annual temperature and precipitation recorded at a nearby meteorological station (Swedish Meteorological and Hydrological Institute, SMHI, stations 8522 and 8427, located approximately 10.5 and 8 km from the studied area, respectively) for the years 1961–1990 are 6.0 °C and 477 mm, respectively. Mean annual discharge at the catchment outlet (SMHI station 2406) for the years 2002–2015 is 155 mm. The soils in the area have developed from glacial and postglacial fluvial deposits and glacial till and textures range from loamy sand to clay. The topography is gently undulating with elevations ranging from 100 to 131 m.a.s. and most of the agricultural fields are systematically subsurface drained. Surface drain inlets are also common. The stream network comprises open ditches with the catchment outlet located to the south-east (see Fig. 1). E21 is one of four catchments representative of the main agricultural regions in Sweden included in the Swedish environmental monitoring program for pesticides. In this monitoring program, data is also collected through annual interviews with the farmers on crops grown and management practices such as pesticide use. The interviews are carried out by local extension officers and are guided by a questionnaire. This data is available for the E21 catchment since 2002.

Three smaller sub-catchments within E21 were selected for this study. These sub-catchments were delineated manually in ArcMap v. 10.0 (Esri Inc. Redlands, CA, USA) guided by a digital elevation model (5 m resolution) as well as the location of structural features (e.g. roads, field edges etc.) that are known to constitute topographical barriers to flow. These datasets were obtained from the Land Survey of Sweden and the Swedish Board of Agriculture. In those cases where maps of subsurface drains were available, this information was used