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Significance of urban and agricultural land use for biocide and pesticide dynamics in surface waters

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ABSTRACT

Biocides and pesticides are designed to control the occurrence of unwanted organisms. From their point of application, these substances can be mobilized and transported to surface waters posing a threat to the aquatic environment. Historically, agricultural pesticides have received substantially more attention than biocidal compounds from urban use, despite being used in similar quantities.

This study aims at improving our understanding of the influence of mixed urban and agricultural land use on the overall concentration dynamics of biocides and pesticides during rain events throughout the year. A comprehensive field study was conducted in a catchment within the Swiss plateau (25 km^2). Four surface water sampling sites represented varying combinations of urban and agricultural sources. Additionally, the urban drainage system was studied by sampling the only wastewater treatment plant (WWTP) in the catchment, a combined sewer overflow (CSO), and a storm sewer (SS). High temporal resolution sampling was carried out during rain events from March to November 2007.

The results, based on more than 600 samples analyzed for 23 substances, revealed distinct and complex concentration patterns for different compounds and sources. Five types of concentration patterns can be distinguished: a) compounds that showed elevated background concentrations throughout the year (e.g. diazinon >50 ng L^{-1}), indicating a constant household source; b) compounds that showed elevated concentrations driven by rain events throughout the year (e.g. diuron 100–300 ng L^{-1}), indicating a constant urban outdoor source such as facades; c) compounds with seasonal peak concentrations driven by rain events from urban and agricultural areas (e.g. mecoprop 1600 ng L^{-1} and atrazine 2500 ng L^{-1} respectively); d) compounds that showed unpredictably sharp peaks (e.g. atrazine 10,000 ng L^{-1}, diazinon 2500 ng L^{-1}), which were most probably due to improper handling or even disposal of products; and finally, e) compounds that were used in high amounts but were not detected in surface waters (e.g. isothiazolinones).

It can be safely concluded that in catchments of mixed land use, the contributions of biocide and pesticide inputs into surface waters from urban areas are at least as important as those from agricultural areas.

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1. Introduction

Surface waters are exposed to a wide variety of anthropogenic organic compounds. Biocides and pesticides belong to the most harmful class of these compounds since they are designed to control the occurrence of pests, weeds, and other unwanted organisms in both urban and agricultural areas (e.g. Liess and Schulz, 1999; Chèvre et al., 2006). The term pesticide refers to compounds used for plant protection (mainly in agriculture). While the term biocides, at least in Europe, is used for all compounds that are used for non-plant protection purposes (EU-98/8/EG, 1998; SR-813.12, 2005). Nevertheless, pesticides and biocides often belong to the same chemical classes or may even be identical molecules. Approximately 270 biocides and 400 pesticides are currently registered in Switzerland (BLW, 2007; FriedliPartner et al., 2007). Recent studies in Switzerland and Denmark showed that biocides and pesticides are used in similar quantities (Lassen et al., 2001; BLW, 2007; FriedliPartner et al., 2007). While agricultural applications of each compound are limited to specific crops, their use in urban environments is more diverse. For example, biocides are used as in-can preservatives in cosmetics, but also as material protection agents on facades and roofs (Schoknecht et al., 2003; Burkhardt, 2006). Pesticides may be applied in gardens, on lawns, and even on sealed areas, although this is prohibited in Switzerland.

Biocides and pesticides may be lost from the site of application to surface waters and occur in measurable concentrations throughout the year (e.g. Hoffmann et al., 2000; Gerecke et al., 2002; Neumann et al., 2002; Blanchoud et al., 2007; Singer et al., submitted for publication). There are two main mechanisms for agricultural pesticide losses: diffuse losses from agricultural soils after pesticide applications to crops, or spills on roads and farmyards washed off to the urban drainage system (point-source losses). In this study, the term "loss" refers to the transport of a compound from the application site to surface waters. These two types of losses have been studied intensively (e.g. Kreuger, 1998; Müller et al., 2002; Leu et al., 2005; Freitas et al., 2008). In general, it was found that for diffuse losses pesticide concentrations increase with increasing discharge during rain events in the application season. Spills, on the other hand, may cause extremely high but short-lived concentration peaks independently of the discharge dynamics.

Urban losses are usually point losses; they enter surface water systems either via wastewater treatment plants (WWTPs), storm sewers (SS), or combined sewer overflows (CSOs). Losses from urban areas have not been studied as intensively as losses from agriculture. There are a few studies on losses from buildings (Jungnickel et al., 2008; Burkhardt et al., 2009), some on pesticide losses from lawns and gardens (Templeton et al., 1998; Blanchoud et al., 2004), and some on losses from CSOs (Gasperi et al., 2008). There are also several studies on losses via WWTPs (Gerecke et al., 2002; Müller et al., 2002; Singer et al., submitted for publication). However, in Switzerland and other European countries, some farms are connected to the urban drainage system, so losses via WWTPs may also represent agricultural sources (Gerecke et al., 2002; Müller et al., 2002).

The compounds found in surface waters and their mixtures pose a potential threat to the aquatic environment because they are designed to control the occurrence of target organisms (Liess and Schulz, 1999; Fleeger et al., 2003; Chèvre et al., 2006). To take appropriate measures to reduce these losses, it is crucial to understand the contribution of the different urban and agricultural sources of these compounds, their entry paths, and the dynamic contributions of urban and agricultural losses. Few studies have comprehensively compared the overall contribution of urban and agricultural pesticide sources (Hoffmann et al., 2000; Jongbloed et al., 2004; Skark et al., 2004; Blanchoud et al., 2007; Woudneh et al., 2009). They concluded that both urban and agricultural sources play an important role as sources of biocides and pesticides, but pointed out that understanding of the transport from urban sources to surface waters is still limited. However, these studies had some limitations which made a simultaneous assessment of all sources difficult: either the losses had only been estimated on the basis of user data, or they had been quantified with weekly composite samples that make it difficult to differentiate between the dynamics of different sources, and none of the studies included biocides used as material protection agents in buildings.

The aim of this study is thus to identify urban and agricultural sources of 14 important biocides and pesticides and to understand their transport to surface water in a catchment of mixed land use as typically found across the Swiss plateau. Specifically, we address: a) the short-term dynamics of compounds of different usage during single discharge events; b) the seasonal dynamics of compounds of different usage; and c) how the cumulative effects of all sources and pathways control the overall contamination patterns as a function of land use. A comprehensive field study concept was designed in order to achieve these aims. To disentangle the relevance of different sources for the overall biocide and pesticide contamination, various data needed to be collected and combined. Key aspects of the data collection effort included a sufficiently high temporal resolution of sampling, spatially distributed sampling sites, a selection of representative biocides and pesticides, and the selection of relevant tracer compounds for wastewater. The findings of the study are transferable to other similar sites based on specific information of land use, crop statistics, or information on the urban drainage system.

2. Materials and methods

2.1. Field study concept

The field study concept includes the following aspects to allow a comprehensive analysis of the contributions of different urban and agricultural sources:

- The urban drainage system lies completely within the hydrological catchment.
- All important sources are present and are spatially distinguishable. For urban areas, different components of