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# The legacy of pesticide pollution: An overlooked factor in current risk assessments of freshwater systems

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ABSTRACT

We revealed a history of legacy pesticides in water and sediment samples from 19 small streams across an agricultural landscape. Dominant legacy compounds included organochlorine pesticides, such as DDT and lindane, the organophosphate chlorpyrifos and triazine herbicides such as terbutylazine and simazine which have long been banned in the EU. The highest concentrations of legacy pesticides were found in streams draining catchments with a large proportion of arable farmland suggesting that they originated from past agricultural applications. The sum of toxic units ($SumTU_{D.magna}$) based on storm water samples from agriculturally impacted streams was significantly higher when legacy pesticides were included compared to when they were omitted. Legacy pesticides did not significantly change the predicted toxicity of water samples to algae or fish. However, pesticide concentrations in bed sediment and suspended sediment samples exceeded safety thresholds in 50% of the samples and the average contribution of legacy pesticides to the $SumTU_{C.riparius}$ was >90%. Our results suggest that legacy pesticides can be highly significant contributors to the current toxic exposure of stream biota, especially macroinvertebrate communities, and that those communities were primarily exposed to legacy pesticides via the sediment. Additionally, our results suggest that neglecting legacy pesticides in the risk assessment of pesticides in streams may severely underestimate the risk of ecological effects.
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## 1. Introduction

Publication frequency of articles characterising the contamination dynamics of freshwater systems in space and time has increased over the past decade in recognition of the need to increase realism of current exposure and risk assessments to support an informed management of these systems. Pesticides in particular have received increasing attention given their suggested important role in the global loss of freshwater biodiversity and ecosystem functioning (Beketov et al., 2013; Malaj et al., 2014; Rasmussen et al., 2012; Schäfer et al., 2012). In this article, we subdivide pesticides into those still registered for agricultural use in the European Union and in Denmark (referred to as contemporary pesticides) and those that have been discontinued or banned for usage in conventional agriculture (referred to as legacy pesticides).

Pesticides applied to agricultural fields may reach surface water through a series of different pathways with surface runoff and tile drains being widely accepted as the most important routes for contemporary pesticides (Schulz, 2004). These transport routes are primarily initiated during heavy precipitation events and lead to transient peak concentrations often exceeding current ecological quality criteria (Bundschuh et al., 2014; Liess and von der Ohe, 2005; Schulz, 2004). In contrast, legacy pesticides may enter surface water continuously via groundwater inflow (Barth et al., 2007; Gilliom, 2007; McKnight et al., 2015), atmospheric deposition

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(Konstantinou et al., 2006; Weber et al., 2010) or through continuous leaching from agricultural soils and landfills (Aliyeva et al., 2013). Consequently, legacy pesticides may generate a relatively constant exposure regime in surface waters. The yearly flux of legacy pesticides to freshwater ecosystems may comprise up to several percent of the historical yearly applied amounts in a catchment (Barth et al., 2007). Importantly, pesticides and their residues may persist and even accumulate in sediments of freshwater ecosystems (Dai et al., 2014; Kuivila et al., 2012; Nowell et al., 2013).

Factors controlling the fate of a pesticide in agricultural landscapes include a variety of chemical and environmental properties of the pesticide (e.g. degradation rate, adsorption to organic carbon and water solubility), climatic factors (e.g. temperature and precipitation), soil characteristics, topography and agricultural practices (Leonard, 1990; Wauchope, 1978). More than 20,000 pesticide products have entered the market since registration became legislatively required in 1947, and it is therefore not surprising that the combined effect of multiple factors influencing the environmental transport and fate of each pesticide generates highly complex exposure profiles of pesticide mixtures in time and space (Konstantinou et al., 2006; Wauchope, 1978). However, pesticides that are currently applied in the highest quantities are also those that occur most often in surface waters with the more water soluble and persistent compounds reaching the highest concentrations (Bundschuh et al., 2014; Kreuger and Tornqvist, 1998; Li et al., 2013; Moschet et al., 2014). Therefore, current pesticide usage is often used to guide the prioritisation of active ingredients included in monitoring programmes and research activities. Moschet et al. (2014) showed that a stringent focus on EU priority pollutants or a subset of the active ingredients applied in the highest quantities on the national level may seriously underestimate predicted toxic pressures in streams. Whereas Moschet et al. (2014) aimed to document that an extensive pesticide screening (249 active ingredients) translates into significantly higher predicted mixture toxicities compared to screenings restricted to fewer pesticides ($\leq$36), the authors did not distinguish between the toxic contribution of contemporary and legacy pesticides. Based on water samples mainly analysed for herbicides and four sediment samples mainly analysed for insecticides, McKnight et al. (2015) suggested that legacy pesticides could still be prominent players driving observed impairments of freshwater invertebrates, and the authors urged for more extensive studies that allow for quantifying the predicted toxicological potency of legacy pesticides in comparison to current use pesticides. To our knowledge, such an extensive study of the potential toxicity of legacy pesticides to aquatic biota relative to that of contemporary pesticides is still lacking despite a substantial body of literature addressing the occurrence, concentrations and predicted toxicities of selected legacy pesticides (Aliyeva et al., 2013; Gilliom, 2007; McKnight et al., 2015; Weber et al., 2010). The novelty element is therefore to quantify the possible toxicity of legacy pesticides as an integral part of current risk assessment. Such an integration has a number of potentially vital implications for the usability of risk assessment, including that i) contemporary regulatory actions are only targeting substances that are still in use; ii) it gives an increased explanatory power in river quality assessment by quantifying the impact of current unknowns, which will additionally reduce the potential underestimation of the role of pesticides as stressors in stream ecosystems, which is currently most likely the case (Beketov et al., 2013; Malaj et al., 2014), and iii) it provides highly needed insight into pesticide exposure profiles in time and space that may be used as improved benchmarks for the interpretations of ecological response parameters.

This article aims to compare the toxicity of legacy pesticides and their metabolites to those of contemporary pesticides in 19 Danish 1st and 2nd order streams situated in agricultural landscape covering a range of agricultural intensity, local climate and soil types. Water samples were collected during base flow and peak flow for pesticide analyses, and bulk sediment and suspended sediment were sampled to optimize detections of pesticides with low water solubility. In more detail, our objectives were to: i) characterize the occurrence of legacy pesticides in Danish headwater streams, ii) estimate the predicted toxicity of legacy pesticides and their residues using the Toxic Units (TU) approach, iii) evaluate the relative contribution of legacy pesticides and their residues to the summed TU of contemporary pesticides, and iv) evaluate which legacy pesticides are of highest ecotoxicological concern.

## 2. Methods

### 2.1. Study streams

Nineteen Danish 1st and 2nd order streams (Fig. S1) were sampled for pesticide occurrences. Nine streams with <50% agricultural land-use in a two-sided buffer extending 2000 m upstream of the sampling site were selected in addition to 10 streams with expected high impact of pesticides (conventional agriculture >60% in the two-sided 100 m buffer). Furthermore, all study sites complied with the following selection criteria: i) forest should occupy <50% of a two-sided 50 m buffer extending from the study site and 2000 m upstream, ii) proportional coverage of silt and mud in stream substrates (indicative of drainage ditches) should be <50%, and iii) no influence from waste water treatment plants, but scattered settlements may influence the chemical water quality. Detailed information on the study streams and catchments is provided in Table S1). In this article, we refer to the nine streams with expected low agricultural impact as controls and the ten streams with expected high agricultural impact as agricultural streams. All catchments are characterised by loam or sandy loam, low elevation and precipitation ranges from ca. 800–850 mm year$^{-1}$ for central Jutland and on Funen and 700–750 mm year$^{-1}$ on Zealand.

Base flow discharge was calculated as the product of the mean stream width, mean depth and mean water velocity, based on measurements at ten transects along a 100 m stream reach extending upstream from the sampling point (depth and velocity measured at 0, 25, 50 and 75% of the width of each transect). Moreover, yearly mean discharge was estimated as the product of yearly mean discharge coefficients (L s$^{-1}$ km$^{-2}$), calculated for national hydrological monitoring stream sites geographically/geologically selected as representative for the study streams, and catchment area for the study streams (km$^2$). In a few cases national monitoring sites could not be regarded as truly representative, and yearly mean discharge was designated as > base flow (Table S1). The proportion of conventional agriculture was quantified for the catchments of each study stream and for a two-sided 100 m buffer extending 2000 m upstream of the sampling site were quantified in ArcGis 10.1 for windows.

### 2.2. Pesticide sampling

Sampling was conducted during May–August in 2012 coinciding with the main pesticide application season in this part of Europe. Dissolved phase pesticides were sampled with: i) manual grab samples in August during low flow conditions to optimize detections of pesticides originating from groundwater inflow (one sample per stream) and ii) event-triggered water samplers

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