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Water quality variables and pollution sources shaping stream macroinvertebrate communities

Elisabeth Berger^{a,b,*}, Peter Haase^{a,c}, Mathias Kuemmerlen^a, Moritz Leps^a,
Ralf Bernhard Schäfer^d, Andrea Sundermann^{a,b}

^a Senckenberg Research Institute and Natural History Museum Frankfurt, Department of River Ecology and Conservation, Clamecystrasse 12, 63571 Gelnhausen, Germany

^b Goethe University Frankfurt am Main, Faculty of Biology, Department Aquatic Ecotoxicology, Frankfurt am Main, Germany

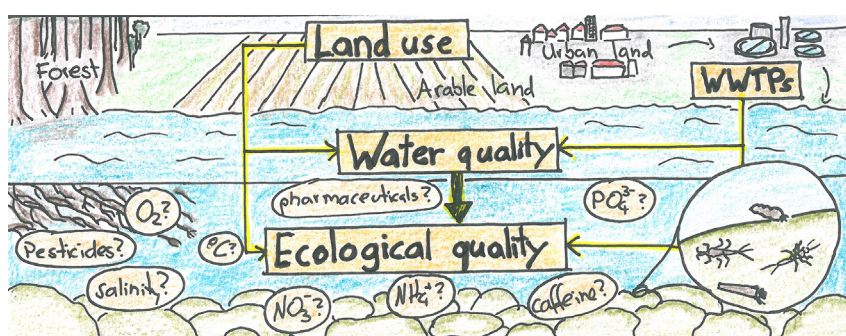
^c University of Duisburg-Essen, Faculty of Biology, Department of River and Floodplain Ecology, Essen, Germany

^d Quantitative Landscape Ecology, Institute for Environmental Sciences, University Koblenz-Landau, Landau, Germany

HIGHLIGHTS

- Stressor identification is important to prioritize management action in streams.
- Water quality and landuse variables were used to model ecological integrity.
- The use of indicator substances in multiple stressor analysis was assessed.
- Diffuse pollution and WWTPs clearly harmful; but not prioritized over each other.
- In-stream measures allow better prediction of ecological quality than land use.

GRAPHICAL ABSTRACT



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ABSTRACT

In 2015, over 90 percent of German rivers failed to reach a good ecological status as demanded by the EU Water Framework Directive (WFD). Deficits in water quality, mainly from diffuse pollution such as agricultural run-off, but also from wastewater treatment plants (WWTPs), have been suggested as important drivers of this decline in ecological quality.

We modelled six macroinvertebrate based metrics indicating ecological quality for 184 streams in response to a) PCA-derived water quality gradients, b) individual water quality variables and c) catchment land use and wastewater exposure indices as pollution drivers. The aim was to evaluate the relative importance of key water quality variables and their sources. Indicator substances (i.e. carbamazepine and caffeine indicating wastewater exposure; herbicides indicating agricultural run-off) represented micropollutants in the analyses and successfully related water quality variables to pollution sources. Arable and urban catchment land covers were strongly associated with reduced ecological quality. Electric conductivity, oxygen concentration, caffeine, silicate and toxic units with respect to pesticides were identified as the most significant in-stream predictors in this order. Our results underline the importance to manage diffuse pollution, if ecological quality is to be improved.

Abbreviations: GSI, German Saprobic Index; MMI, multi-metric index; EPT %, % Ephemeroptera, Plecoptera and Trichoptera; BMWP, Biological Monitoring Working Party; ASPT, Average Score Per Taxon; SPEAR %, % SPEcies At Risk; WWTPs, wastewater treatment plants.

* Corresponding author at: Senckenberg Research Institute and Natural History Museum Frankfurt, Department of River Ecology and Conservation, Clamecystrasse 12, 63571 Gelnhausen, Germany.

E-mail address: elisabeth.berger@senckenberg.de (E. Berger).

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However, we also found a clear impact of wastewater on ecological quality through caffeine. Thus, improvement of WWTPs, especially preventing the release of poorly treated wastewater, will benefit freshwater communities.

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1. Introduction

Freshwater ecosystems are hotspots for biodiversity and, at the same time, may well be the most impacted and endangered ecosystems in the world (Strayer and Dudgeon, 2010). They are thought to experience higher species extinction rates than marine or terrestrial systems (Collen et al., 2014; McLellan et al., 2014) since human activities and settlements have always been centred around freshwater: we need water to drink, clean, use it for irrigation, fishing and recreation, but also for navigation, hydroelectric power generation and waste disposal (Strayer, 2006). Therefore, several interests compete with the conservation of freshwater biodiversity. Nonetheless, EU member states agreed on the water framework directive (WFD) that had the ambitious aim to achieve a good ecological status for all European surface waters by 2015 (European Parliament and Council, 2000). However, approximately, 47% of all surface water bodies were estimated to fail that aim by 2015 (EEA, 2015) and a final deadline has now been moved to 2027 (European Parliament and Council, 2000). The pressures causing ecological deterioration in general are well known and include the channelization of rivers, damming, dredging for shipping, climate change, introduction of alien species, fishing, clearing of natural vegetation in the riparian zone, nutrient input leading to eutrophication, heavy metals, pesticides, industrial chemicals and emerging contaminants (Malmqvist and Rundle, 2002). An increasing number of projects attempt to rank multiple stressors regarding their relative contribution to ecological decline with the aim of supporting ecosystem management (i.e. Brack et al., 2015; Hering et al., 2015; Navarro-Ortega et al., 2015; Rico et al., 2016; Sundermann et al., 2013). The present study contributes to this aim, by investigating the relative importance of micropollutants compared to more traditional water quality parameters such as nutrients, temperature, pH, salinity and oxygen concentrations. The contamination of freshwater systems with thousands of industrial, agricultural and household chemicals is a major public concern, however long-term effects on aquatic flora and fauna are largely unknown (Boxall et al., 2012; Schwarzenbach et al., 2006). Micropollutants, typically occur at very low concentrations, hence their name, and enter surface waters through atmospheric deposition, polluted rainwater from roofs and sealed areas, untreated and treated wastewater as well as other diffuse sources such as field run-off carrying pesticides (Loos et al., 2013). In Europe, wastewater is typically only released after mechanical and biological treatment in conventional WWTPs (Jekel et al., 2015). However, conventional WWTPs were mainly designed to remove pathogens, carbon, nitrogen and phosphorus. Although they also biodegrade many micropollutants and remove non-polar compounds through sorption onto sludge, persistent polar organic compounds, including pharmaceuticals, personal-care products, hormones, and other industrial chemicals are often not completely removed and subsequently discharged to surface waters with the effluent (Loos et al., 2013; Margot et al., 2013). The release of micropollutants can be exacerbated when untreated wastewater is discharged after heavy rain leading to overflow of WWTPs with mixed sewage, i.e. collecting household wastewater and surface run-off (Launay et al., 2016). Thus, WWTPs are considered a major input path for micropollutants into streams (Margot et al., 2013). Laboratory and mesocosm studies have demonstrated biological impacts of several wastewater-associated compounds at environmentally relevant concentrations (Kidd et al., 2007; Lee et al., 2016; Niemuth et al., 2015). Consequently, some countries (e.g. Switzerland) started to upgrade their WWTPs with additional treatment steps such as ozonation and powdered activated carbon (Eggen et al., 2014) that successfully reduce the load of micropollutants in the effluent (Ashauer, 2016;

Hollender et al., 2009; Margot et al., 2013). In other countries extended treatment options are scrutinized (Völker et al., 2016) and potential benefits weighed against costs (Johnson and Sumpter, 2015; Joss et al., 2008). However, few studies have attempted to relate wastewater-associated micropollutants to ecological impacts in the field and to evaluate their relative importance in determining ecological quality compared to other water quality stressors (Berger et al., 2016; Burdon et al., 2016; Posthuma et al., 2016; Rico et al., 2016). Multiple stressor studies that aim to determine the influence of micropollutants and discriminate it from other variables encounter three major challenges. First large data sets with 'long' environmental stressor and ecological response gradients (large across site variability) are required to reach sufficient statistical power (Posthuma et al., 2016). A sensible solution that is increasingly applied is to exploit the wealth of governmental monitoring data. This option was followed in the present study. The second challenge is the 'curse of dimensionality', which refers to the inability to accommodate the very large numbers of micropollutants into one ecological model (Posthuma et al., 2016). Two approaches are most commonly used to overcome this problem; aggregation of the compounds in terms of toxic units (TU, Sprague, 1970) or in terms of the multisubstance Potentially Affected Fraction of species (msPAF, De Zwart and Posthuma, 2005). Both approaches require an estimation of the ecotoxicity of each measured micropollutant, which is commonly derived from species sensitivity distributions (SDD) for msPAF and single species toxicity data for TU, although variations are possible (Schäfer et al., 2013). Based on that information and the measured chemical concentrations, the number of variables (all separate micropollutants or groups of chemicals) is then reduced into composite variables characterizing the 'toxic pressure' of a site (i.e. TU_{organic} , $TU_{\text{pesticides}}$, $msPAF_{\text{organic}}$, $msPAF_{\text{metals}}$ etc.). Generally, the TU approach is more widely applicable, since it is less data-demanding and it was partly applied here. In addition, another approach based on indicator substances was assessed in the present study. Indicator substances are organic micropollutants that are representatives for a group of chemicals with similar characteristics regarding their application, input pathway into surface waters, physicochemical properties or reactivity (Jekel et al., 2015). Recently, a limited number of micropollutants has been suggested as indicator substances for a) contamination sources and b) the performance of removal processes during wastewater treatment (Jekel et al., 2015). For the first time we include several of these suggested source-specific indicator substances in a multiple stressor analysis. Since water quality variables such as nitrites, phosphates and major ions enter surface waters through agricultural run-off as well as WWTPs, indicator substances are also expected to help attributing them to pollution sources. The third challenge is multicollinearity. If two or more variables are highly correlated, even advanced statistical methods cannot disentangle their relative stressor influence and models can lead to unstable and erroneous results (Dormann et al., 2013). Given that a simulation study showed that even simple rules of thumb (omission of individual variables contributing to collinearity based on expert knowledge) perform as well as more sophisticated techniques (Dormann et al., 2013) we follow this strategy and report the correlation structure between water quality variables and consider it in the interpretation of results.

Thus, the primary objective of the present study was to identify the relative importance of key water quality stressors in determining the ecological integrity of streams as assessed through macroinvertebrate assemblages. Thereby, we also elucidated the relative importance of different pollution sources such as WWTPs and agricultural run-off. Although the nature of the present study is explorative, we hypothesized that micropollutants derived from WWTPs have a major influence on

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