



The concentration-discharge slope as a tool for water quality management

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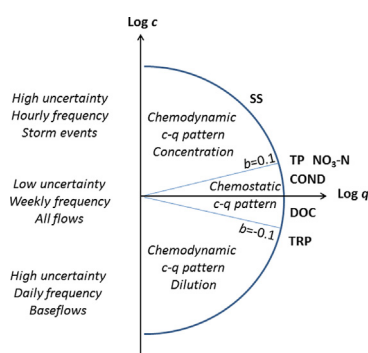
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HIGHLIGHTS

- High-frequency data help to understand the patterns in long-term data.
- Chemostatic responses lead to low errors in water quality parameters.
- Low-order agricultural catchments homogenize stream solute responses.
- Phosphorus and nitrogen chemostatic responses are driven by legacy stores.
- Concentration-discharge slope helps to prioritise monitoring and mitigation efforts.

GRAPHICAL ABSTRACT



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ABSTRACT

Recent technological breakthroughs of optical sensors and analysers have enabled matching the water quality measurement interval to the time scales of stream flow changes and led to an improved understanding of spatially and temporally heterogeneous sources and delivery pathways for many solutes and particulates. This new ability to match the chemograph with the hydrograph has promoted renewed interest in the concentration-discharge (c - q) relationship and its value in characterizing catchment storage, time lags and legacy effects for both weathering products and anthropogenic pollutants. In this paper we evaluated the stream c - q relationships for a number of water quality determinands (phosphorus, suspended sediments, nitrogen) in intensively managed agricultural catchments based on both high-frequency (sub-hourly) and long-term low-frequency (fortnightly-monthly) routine monitoring data. We used resampled high-frequency data to test the uncertainty in water quality parameters (e.g. mean, 95th percentile and load) derived from low-frequency sub-datasets. We showed that the uncertainty in water quality parameters increases with reduced sampling frequency as a function of the c - q slope. We also showed that different sources and delivery pathways control c - q relationship for different solutes and particulates. Secondly, we evaluated the variation in c - q slopes derived from the long-term low-frequency data for different determinands and catchments and showed strong chemostatic behaviour for phosphorus and nitrogen due to saturation and agricultural legacy effects. The c - q slope analysis can provide an effective tool to evaluate the current monitoring networks and the effectiveness of water management interventions. This research highlights how improved understanding of solute and particulate

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dynamics obtained with optical sensors and analysers can be used to understand patterns in long-term water quality time series, reduce the uncertainty in the monitoring data and to manage eutrophication in agricultural catchments.

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1. Introduction

Combating eutrophication is proving difficult and exposes gaps in our scientific understanding of hydrological and biogeochemical processes controlling stream concentrations of solutes and particulates. The relative importance of these processes and contribution of dominant sources and delivery pathways is captured by the concentration-discharge (c - q) relationship. The c - q relationship characterises solute/particulate change (dilution or concentration) with varying flow (Evans and Davies, 1998) and can be quantified as the slope b of the c - q regression relationship on logarithmic axes (Godsey et al., 2009). The c - q relationship is often complex due to hydrochemical variability but can generally be classified into two patterns: chemostatic ($|b| < 0.1$) and chemodynamic ($|b| > 0.1$) with either dilution ($b < -0.1$) or concentration ($b > 0.1$) pattern. The chemostatic c - q pattern, in which the concentrations are stable over a large range of flows has been observed for many solutes and particulates with an abundant source of the chemical in the catchment (Thompson et al., 2011) e.g. weathering bedrock (Ameli et al., 2017; Godsey et al., 2009; Hoagland et al., 2017) or agricultural soils and the unsaturated zone (Basu et al., 2011; Van Meter et al., 2017). These sources are also referred to as the legacy stores that can control the mobilisation and stream transport of the chemicals in the long-term and lead to transport-limitation (Basu et al., 2011). When the rate of the concentration change is larger than the flow change, a chemodynamic c - q pattern and source-limitation are observed with either concentrations increasing (concentration) or decreasing (dilution). Recent increased availability of high-frequency (sub-hourly) c and q data, due to deployment of optical sensors and wet-chemistry analysers, has led to improved understanding of the complex c - q patterns observed in water quality data. For example, recent work explains how both seasonal and storm-to-storm dynamics in source mobilisation and activation of different delivery pathways control the chemostatic and chemodynamic c - q responses (Bieroza and Heathwaite, 2015; Lloyd et al., 2016).

To date, the c - q relationship has been evaluated for a large range of chemicals both derived from weathering of bedrock and from agricultural land use. The studies of c - q dynamics in agricultural catchments focus on chemicals that are of major concern due to increasing eutrophication and hypoxia of inland and coastal waters: phosphorus (P) both as total P (TP) and total and soluble reactive P (TRP and SRP) (Basu et al., 2011; Bieroza and Heathwaite, 2015; Dupas et al., 2015), suspended sediments (SS) measured directly or with turbidity (TURB) as a proxy (Lawler et al., 2006), organic and inorganic nitrogen (N), particularly in the form of nitrate nitrogen ($\text{NO}_3\text{-N}$) (Bieroza et al., 2014; Dupas et al., 2016; Van Meter et al., 2017) and compounds that provide information on the general hydrochemical functioning of catchments: total and dissolved organic carbon (TOC and DOC) (Butturini et al., 2008; Hoagland et al., 2017) and specific conductivity (COND) (Bieroza and Heathwaite, 2015).

Hydrochemical data have been collected for over 150 years (Howden et al., 2010) and most of these long-term datasets are from rivers (>4th Strahler order) and collected at low frequency (typically monthly) (Tetzlaff et al., 2017). Therefore, this existing water quality sampling method is not suited to target the highly variable, in space and time, agricultural P and N pollution in low-order (<3rd Strahler order) catchments (Bieroza et al., 2014). Recent advances in situ water quality monitoring with optical sensors and wet-chemistry

analysers (Bieroza and Heathwaite, 2016; Flourey et al., 2017; Jordan et al., 2012; Rode et al., 2016) help to bridge this gap, but due to high-financial cost of the in situ technology per sampling site, new approaches that integrate the high- and low-frequency sampling are needed (Bieroza et al., 2014; Chappell et al., 2017; Jordan and Cassidy, 2011). To address this scientific and management need, we propose that the new knowledge of c - q dynamics obtained with high-frequency sampling can improve the understanding of hydrochemical patterns in readily available long-term datasets and can help to prioritise monitoring and mitigation efforts.

Specifically, we evaluated the uncertainty and variation in the c - q relationship for selected solutes and particulates, for a number of low-order small agricultural catchments in the UK, Norway and Sweden that are subjected to eutrophication pressures. We hypothesized that the c - q slope represents the catchment's tendency to store and transport chemicals and that it can be a useful tool in water management practice. Our objectives were to: 1) evaluate the variation in the c - q slopes for P, SS, $\text{NO}_3\text{-N}$, DOC and COND for both high- and low-frequency sampling, 2) evaluate the uncertainty in operational water quality parameters (mean, 95th percentile and load) derived from low-frequency datasets as a function of the c - q slope, 3) provide recommendations on how the c - q slopes can help to improve water quality management.

2. Methods

2.1. High-frequency datasets

Two high-frequency datasets were used in the analysis: HF1 (Leith catchment, UK, 2009–2014, hourly and sub-hourly, TP, TRP, TURB, $\text{NO}_3\text{-N}$ and COND) and HF2 (SE3 catchment, SE, 2017–2018, sub-hourly, TP, TRP, TURB, $\text{NO}_3\text{-N}$ and DOC) with continuous flow discharge measurements. Similar experimental setups were deployed in both cases, with stream water pumped to a small hut on the bank and the measurements conducted on unfiltered samples with wet-chemistry analysers (Systea's Micro Mac for HF1 and Hach Lange's Phosphax for HF2 giving TP and TRP) and in-line optical sensors for TURB and solutes (Systea's WaterWatch and Hach Lange's Nitratax for HF1 and s:can's Spectrolyser for HF2). The details of the HF1 experimental setup and principles of the in situ monitoring are given elsewhere (Bieroza and Heathwaite, 2015; Bieroza and Heathwaite, 2016; Bieroza et al., 2014). Both study catchments represent small, low-order catchments dominated by agricultural land use: grassland and livestock grazing in the Leith catchment (HF1 in Table 1) and arable land and crop production in the SE3 catchment (HF2 in Table 1). The catchments differ in terms of geology and soils, with sandstone and loam soils in HF1 and marine clay and heavy clay soils in HF2 with the effect on hydrology: intensive ground-surface water interactions (Krause et al., 2009) and subsurface flow pathways in HF1 (Bieroza et al., 2014) and overland, macropore and tile drainage flow pathways in HF2 (Ulén et al., 2011).

2.2. Low-frequency datasets

We collated long-term low-frequency water quality time series (TP, TRP and SRP, SS, $\text{NO}_3\text{-N}$, DOC and TOC and COND) for agricultural catchments subject to risk of eutrophication from three EU countries (UK, Norway and Sweden) spanning a range of climatic and soil conditions

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