A quantitative typology for distinguishing active and legacy source contributions to stream water quality

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INTRODUCTION

Hydrochemical constituents in streams may originate from currently active sources at the land surface and/or legacy sources retained in soil, slow-flowing groundwater, and sediments from earlier active source inputs, waste deposits, and/or land contamination.

These different types of sources and their contributions to total hydrochemical concentrations and loads in streams need to be distinguished for effective pollution mitigation and water quality improvement. The work synthesized and presented here has developed and tested a new general methodology for such distinction that quantifies the concentration and load contributions of each source type (Figure 1, Chen et al., 2021; Destouni et al., 2021).

METHODOLOGY

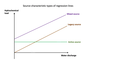

Figure 1. Schematic illustration of regression line types expected for load contributions versus water discharge of hydrochemical constituents originating from different types of (active, legacy, or mixed) sources. Source: Destouni et al. (2021).

The source-related differences in load vs discharge regression lines illustrated in Figure 1 emerge from relatively simple, first-order mechanistic consideration of active and legacy sources of a hydrochemical constituent that may prevail over a catchment (Destouni et al., 2021). If the active sources on average emit a constituent at relatively stable average input rate, while constituent retention may also occur at a relatively stable average rate in the subsequent hydrological transport through the catchment, then the average active-source load contribution at catchment outlet would also be relatively stable as the outlet discharge varies over time (green type of regression line in Figure 1). The output load stability follows from catchment-scale mass balance, implying that temporally stable average input and retention rates must on average lead to a balanced temporally stable average constituent load at catchment outlet (i.e., a balanced average constituent amount leaving the outlet per unit time). The outlet concentration must then vary with discharge to maintain this stable average outlet load (product of concentration and discharge). For diffuse legacy sources, in contrast, if their constituent release into the average mobile water flow through the catchment follows relatively stable zeroth-order release kinetics, then their average concentration contribution would be independent from discharge rate and remain on average temporally stable. This concentration stability implies that the legacy source contribution to average constituent load must vary more or less linearly with discharge (brown type of regression line in Figure 1). Considerable mixed contributions from both types of sources would be additive with load vs discharge behaviour represented by the purple regression line type in Figure 1.

DATA & RESULTS

The proposed methodology is based on characteristic differences that can be mechanistically hypothesized to emerge between possible well-fitting regression lines to data points of load (product of concentration and discharge) versus water discharge for hydrochemicals originating from active, legacy, or mixed sources (Figure 1). To test this methodology and its underlying mechanistic hypothesis, we have used stream monitoring data series of discharge and nutrient (total nitrogen, TN, and total phosphorous, TP) loads from 33 catchments draining into the Baltic Sea along the coast of Sweden (Figure 2). To further test the methodology also for other hydrochemical constituents and catchment scales, we have also used data for loads of chloride and metals (copper, lead, and zinc) from smaller sub-catchments within the most heavily populated Swedish water management district Northern Baltic Proper (NBP, outlined in red in the map shown left in Figure 3).

For TN and TP, only 3-4 (of 37) catchments emerge as having mixed sources with 32-59% active source contributions to total loads, and only 1 catchment, just for TN, has dominant active sources. For metals, 1-3 (of total 11) catchments have mixed sources with average 1-3% load contributions from active sources. In contrast, corresponding results for chloride show mostly mixed-source catchments (18 of 19), however still with only 19% active-source contributions to total load on average. The quantified source types can also be spatially mapped, for example for TN along the Swedish coast (Figure 4, with similar results also obtained for TP), and for chloride and metal legacies over the Swedish NBP district (Figure 5). The source mapping comparison with known mining and mining waste areas (for the metals), urban areas (for chloride), and other indicators of human activity (population density, agricultural land share, for the nutrients) in the study catchments support the applicability of the tested methodology and underlying load vs discharge behaviour hypothesis (Chen et al., 2021; Destouni et al., 2021). The methodology applications illustrate an overall dominance of legacy sources for the studied hydrochemical constituents and catchment settings of different scales in the Swedish regional conditions.

NUTRIENTS

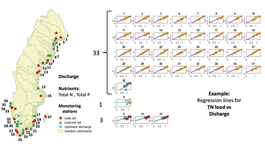

Figure 2. (Left) Nutrient and discharge monitoring stations along the coast Sweden. (Right) Regression lines for total nitrogen (TN) load versus discharge data over the study period 2003–2013, and associated source attribution based on Figure 1 implying 33 catchments dominated by legacy sources, 1 catchment by active sources, and 3 catchments by mixed sources. All data are normalized with their respective average values; the value 1 on either axis thus represents the average value of that variable. Source: Chen et al. (2021).

CHLORIDE & METALS

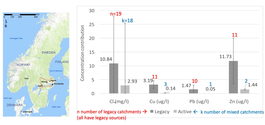

Figure 3. (Left) The Swedish water management district Norther Baltic Proper (NBP) outlined in red. (Right) Resulting multi-catchment statistics for that district of average legacy and active concentration contributions at catchment outlet. Legacy sources are dominant in all catchments, while only a few mixed catchments have considerable active source contributions. Error bars show ±1 standard deviation among the catchments in each category. Source: Destouni et al. (2021).

CONCLUDING REMARKS AND SOURCE MAPS

For all studied hydrochemical constituents and Swedish regional catchment settings of different scales, legacy sources emerge as dominant in terms of concentration values, contribution fractions to total loads, and number of catchments with considerable legacies, compared with corresponding results for active sources (Figure 3, and source maps in Figures 4-5 below). The legacy contributions also correlate well with human-activity indicators in the catchments: urban areas for chloride, agricultural land share for TN, population density for TP, historic mining and mine waste areas for the metals. This indicates the legacy contributions as being largely anthropogenic rather than contributed from natural background concentrations.

In general, for any studied constituent, it is essential to note that legacy source dominance in currently measured stream concentrations does not imply that the currently active sources are unimportant, but only that their main contributions are likely to be seen first in future stream monitoring. Today's active sources may be the legacy sources of the future, so all sources are important and require mitigation, but with effects notable over different time scales.

This is why it is essential to distinguish and quantify the contributions and geographic locations of the different source types, in order to select and apply the different methods needed to mitigate each source type, and to appropriately anticipate, monitor, and follow up effect evolution over time. The developed and tested methodology provides a relatively simple novel way for doing this. It can be used to screen commonly available stream monitoring data for distinction, quantification, and mapping of active and legacy contributions of any hydrochemical constituent in and across various hydrological catchment settings.

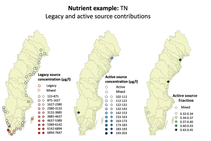

Figure 4. Calculated and mapped contributions to total nitrogen (TN) concentrations at various catchment outlets along the whole coastline of Sweden from (left) legacy sources and (centre) currently active sources, and (right) contribution fractions from the active sources in the few catchments found to have mixed source contributions rather than dominanant legacy source contributions. Source: Chen et al. (2021).

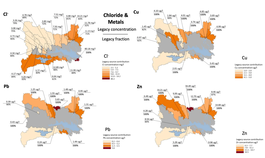

Figure 5. Spatial distribution of temporal average legacy concentration contributions (top number) and associated average load fractions from legacy sources (bottom number) for each (colored) subcatchment within the studied Swedish water management district of Northern Baltic Proper for chloride (top left) and the metals copper (top right), lead (bottom left), and zinc (bottom right). Source: Destouni et al. (2021)

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ABSTRACT

Hydrochemical constituents in streams may originate from currently active sources at the surface and/or legacy sources retained in soil, slow-flowing groundwater and sediments from earlier surface inputs, waste deposits, and land contamination. These source contributions need to be distinguished for effective pollution mitigation and water quality improvement. This study outlines a methodology for such distinction based on some general mechanistic differences in stream concentration and load behavior versus discharge between the contributions from these different types of sources. The methodology is applied to and tested on stream concentration data for chloride, nitrogen, phosphorous, copper, lead, and zinc, and corresponding data for water discharge, measured over recent decades (time series of 1-3 decades) in multiple Swedish hydrological catchments of different scales (10-37 catchments depending on substance, chloride/nutrient/metal, and its monitoring). Mixed sources are indicated in most (18 of 19) catchments for chloride (with average 19% active-source contribution to total load), but only in 3-4 (of 37) catchments for total nitrogen and total phosphorous (32-59% active), and 1-3 (of 11) catchments for copper, lead, and zinc (1-3% active). Only 1 catchment (of 37) is indicated to have dominant active sources for total nitrogen, and most catchments thus have dominant legacy source contributions for all substances. The legacy contributions correlate well with human activity indicators in the catchments (urban areas for chloride, agricultural land share for nitrogen, population density for phosphorus, historic mining and mine waste areas for the metals), indicating that they are largely anthropogenic. The developed and tested methodology is relatively simple and can be used to screen commonly available stream monitoring data for distinction of active and legacy contributions of any hydrochemical constituent in and across various hydrological catchment settings.

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