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OPINION

Zones of untreatable water pollution call for better appreciation of mitigation limits and opportunities

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This opinion piece addresses subsurface legacy sources and their role in mitigation of large-scale water pollution and eutrophication. We provide a mechanistic theoretical basis and concrete data-based exemplification of dominant contributions from such sources to total recipient loads. We specifically develop a diagnostic test to detect such contributions, recognizing that they are inaccessible and associated with long transport times that tend to evade detection when homogeneous catchment models are calibrated to typically heterogeneous catchments. Dominant legacy-source contributions are also in practice untreatable within the commonly short time frames given for compliance with environmental regulation. We therefore argue that, for considerable water quality improvements to be achieved within such short time frames, mitigation measures need to be spatially differentiated and directed to (sub)catchments without major legacy sources. The presented diagnostic test identifies dominant prevalence of such sources where there is linear temporal correlation between the nutrient/pollutant loads and the water discharges from a (sub)catchment. Confidence in this identification may be strengthened by independent quantification of long transport times and records of temporally extended presence of nutrient/pollutant sources in the same (sub)catchment.

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eutrophication, legacy sources, long time lags, mitigation, pollution, slow transport pathways

1 | INTRODUCTION

Over the last century, the world population has increased more than threefold (Roser & Ortiz-Ospina, 2017). Associated human activities, for example, for food and energy production, have also expanded and intensified and, with them, so have the leaching and waterborne spreading of nutrients and pollutants to various water environments, including ground, surface and coastal water. For example, annual loads of organic industrial water pollutants in different countries are spatially–geographically well correlated with country population; this has been found in particular for the industrially well-developed countries in Europe, North America, and Oceania ($R^2 = 0.98$) with high average load level, and to somewhat lower degree in Asia, Central and South America, Middle East and Northern Africa ($R^2 = 0.76$) with lower average load level (Destouni, Hannerz, Prieto, Jarsjö, & Shibuo, 2008). Furthermore, annual average concentrations of nitrogen and phosphorous have also been found to be spatially well correlated with population among countries and sub-catchments in large European transboundary drainage basins of the Baltic Sea, Northern Europe, and the Sava River, south-eastern Europe ($R^2 = 0.82$ for total/dissolved inorganic nitrogen and $R^2 = 0.90$ for total phosphorous; Levi, Cvetkovic, & Destouni, 2018).

Such findings indicate close links between the waterborne loads or concentrations of pollutants and nutrients from hydrological catchments and the human population and associated activities prevailing within the catchments. To mitigate human-driven water pollution and eutrophication problems caused by these links, far-reaching policy, regulation and international agreements have been implemented in various parts of the world, such as the Water Framework Directive (WFD) across the EU (European Commission, 2000), the Helsinki Convention (HELCOM) (http://www.helcom.fi) and Baltic Sea Action Plan (BSAP) in the Baltic region (Helsinki Commission, 2007), and the Great Lakes Water Quality Agreement of Canada and the United States (DePinto, Young, & McIlroy, 1986; Scavia et al., 2014). However, actual mitigation results of these efforts have been largely disappointing, with water quality targets not met even after many years of mitigation efforts and after regulation deadlines have passed for meeting the targets (Arheimer & Brandt, 2000; Destouni, Fischer, & Prieto, 2017; Meals, Dressing, & Davenport, 2010).

Possible reasons for not achieving mitigation targets may include (some combination of):

1. Mitigation measures required by regulation/agreement are not implemented. Authorities responsible for following up the implementation should then know and act on this.
2. Implemented mitigation measures are only effective after a long period of time. Such delays may be expected and accounted for in water quality policy and regulation—or not, which implies unrealistic expectations of the time frame needed to achieve targeted effects.
3. Implemented mitigation measures are not effective at any time. Responsible authorities may then have unrealistic expectations of the impacts of their policy decisions.

In this study, we focus on the last two (2, 3) situations of society trying its best to mitigate but not managing to do so, at least not in the given/expected time frame for mitigation compliance. For example, the wide-ranging water quality policies WFD and BSAP, applying across the EU and the Baltic region, respectively, expect major water quality improvements to be achieved from policy application, starting in 2009 for the WFD and in 2007 for the BSAP, until their common compliance deadline in 2021, that is, over a period of 12–14 years. However, available data show only small changes so far, and then mostly deterioration of water quality in classified moderate-to-bad status waters (Destouni et al., 2017), for which both the WFD and the BSAP require major improvements. Only a few years are now left to achieve most of the water quality improvements required by these key policies. To credibly claim that these requirements will be reached in these remaining few years until 2021, demonstrable improvement should be observable by now over the regulated areas, which is not the case based on available data (Destouni et al., 2017).

As one possible explanation for such situations, we propose and discuss here the possibility that some essential aspects of large-scale waterborne nutrient and pollutant release and transport are not yet sufficiently well understood. Previous studies (Darracq & Destouni, 2005; Destouni, Persson, Prieto, & Jarsjö, 2010) have suggested misleading interpretations due to insufficient account of the full spatiotemporal spectrum of source releases and the subsequent transport through whole catchments. In particular, major subsurface accumulation and storage legacies may control nutrient and pollutant concentrations and loads in and from catchments, for example, due to slow transport (Darracq, Lindgren, & Destouni, 2008; Destouni et al., 2010) and/or retention (Törnqvist et al., 2015) with long time lags (Wang & Burke, 2017) in the soil and groundwater system. Such subsurface controls are out of sight and may also tend to be out of mind, for example, in pollution mitigation practices (Destouni et al., 2010) and applications of homogeneous transport models to heterogeneous catchments (Kirchner, 2016; Persson, Jarsjö, & Destouni, 2011). In this study, we outline and discuss how subsurface legacies and their implications can be quantified and identified based on commonly available discharge and concentration data. We also concretely exemplify dominant contributions of legacy sources to total recipient loads, and argue for the need of such explicit quantification and identification in efforts to mitigate large-scale nutrient and pollutant loads.

2 | REPRESENTING DIFFERENT SOURCE CONDITIONS

The focus of this study is illustration of the main discussion issues for some particular hydrological settings, rather than a review of all possible settings. To simply and briefly exemplify how different source releases and associated transport patterns can be distinguished by different model representations, we consider a concrete case of nonreactive (chloride) transport in a relatively data-rich Swedish boreal catchment. This is the Kringlan catchment (with area of 295 km^2 and longest line-distance to outlet of 30 km), located in east-middle Sweden at the western boundary of the major Norrström drainage basin that comprises the capital city of Stockholm (see fig. 2 of Soltani & Cvetkovic (2017), for Kringlan catchment map and location within the Norrström basin and Sweden). The whole Norrström basin has been investigated in previous studies of large-scale nutrient transport through its stream network (Darracq & Destouni, 2005) and entire subsurface–surface water continuum (Darracq et al., 2008; Destouni et al., 2010). The Kringlan catchment within the Norrström basin is particularly well suited for the present study because long-term data series are available over the period 1985–2007 (23 years) for daily precipitation and water discharge (Swedish Meteorological and Hydrological Institute, 2011a, 2011b) and average monthly chloride concentration in both precipitation and discharge (Swedish University of Agricultural Sciences, 2011). Kringlan data have been used in previous studies of tracer (chloride) load distribution and associated water/tracer travel time and age distributions under different scenarios (cases) of pathway length distribution in the catchment (Soltani & Cvetkovic, 2017). In this study, we use the data for another type of scenario analysis and evaluation.

The evaluation is based on how modeled scenario results can represent actual observations, and the analysis focuses on clarifying the relative importance of different possible source input conditions, including:

1. areal leaching with precipitation from continuously renewed sources at the land surface of the catchment;
2. legacy source releases throughout the whole transport-active domain of the catchment; or
3. some combination of the above.

The modeling used here to quantify these conditions is not aimed at providing a new model tool, but at demonstrating and sustaining the arguments being made. For (100% contribution of) source Scenario 1, we use the following common and relatively simple, yet mechanistic model representation of (nonreactive, ideal) tracer loading $L_{\text{out-1}}$ [MT^{-1}] and associated flux concentration $C_{\text{out-1}}$ [ML^{-3}] in the water discharge Q_{out} [L^3T^{-1}] into the recipient at the catchment outlet at time t [T] (cf. eq. 7 in. e.g., Soltani & Cvetkovic (2017)):

$$L_{\text{out}-1}(t)=\int_{t_0}^{t} C_{\text{in}}(t-t_0-\tau)Q_{\text{in}}(t-t_0-\tau)g(\tau)d\tau, \tag{1a}$$

$$C_{\text{out}-1}(t)=\frac{L_{\text{out}-1}(t)}{Q_{\text{out}}(t)}=\frac{\int_{t_0}^{t} C_{\text{in}}(t-t_0-\tau)Q_{\text{in}}(t-t_0-\tau)g(\tau)d\tau}{Q_{\text{out}}(t)}, \tag{1b}$$

where C_{in} [ML^{-3}] is the input tracer concentration in the water input flow Q_{in} [L^3T^{-1}] at time $(t - t_0 - \tau)$ [T], which is the rate of precipitation [LT^{-1}] spatially integrated over the catchment area A_c [L^2]. The time scales t_0 [T] and τ [T] are the investigation (and for simplicity also assumed tracer input) start time and the travel time of waterborne tracer through each transport pathway to the catchment outlet, respectively. Furthermore, $g(\tau)$ is the distribution of all tracer travel times, from all input points at the land surface through the ensemble of all transport-active pathways to the outlet of the catchment.

For (100% contribution of) source Scenario 2, a legacy source is assumed to be widely spread and prevail more or else everywhere in the transport-active catchment domain, accumulated by biogeochemical and physical mass transfer/immobilization processes and slow transport acting on some fraction of the total tracer amount that has been entering the catchment for some time. Tracer is further continuously remobilized and released from this diffuse source into the mobile aqueous phase at some rate k [T^{-1}] along the total pathway lengths through the transport domain. This transport situation has different initial and boundary conditions and must be represented by a different type of release-transport model than Equation 1 (based on tracer input only at land surface points and starting first at time t_0).

A relevant simple yet still mechanistic model representation of spatially ubiquitous release and transport of solute from diffuse internal sources has been derived and used for describing release and transport of dissolved metals from mining waste deposits (Eriksson & Destouni, 1997) and (organic and inorganic) carbon from the subsurface to the stream network of a catchment (Jantze, Lyon, & Destouni, 2013). In analogy, we can also for the ubiquitous legacy source of Scenario 2 express the tracer loading $L_{\text{out-2}}$ and flux concentration $C_{\text{out-2}}$ at the catchment outlet as (cf. e.g., eqs 8a and 10 in Eriksson & Destouni (1997), and 3b and 5 in Jantze, Lyon, & Destouni (2013)):

$$L_{\text{out}-2}(t)=\frac{A_c\,C_0^*}{n}k\int_{0}^{t-t_0} q_{\text{out}}(t;\tau)g(\tau)d\tau\approx\frac{Q_{\text{out}}(t)\,C_0^*}{n}kE[\tau], \tag{2a}$$

$$C_{\text{out}-2}(t)=\frac{L_{\text{out}-2}}{Q_{\text{out}}}=\frac{C_0^*}{n}kE[\tau], \tag{2b}$$

where n is the porosity [−] and C_0^* [ML^{-3}] (per unit bulk soil/aquifer volume) is the bulk concentration of tracer in the source by the investigation start time t_0, with $1/k$ quantifying a characteristic time scale for total source depletion under assumed zeroth-order kinetics $C^*=C_0^*(1-kt)$. For simplicity, we assume n, C_0^* and k as essentially constant and independent of τ. Furthermore, $q_{out}(t;\tau)$ [LT^{-1}] is the specific water discharge contribution of each transport pathway to total discharge $Q_{out}(t)$ at the catchment outlet at time t. Assuming that the temporal variation of $Q_{out}(t)$ (rather than the spatial, length-dependent variability of τ among pathways) largely determines the main variation of q_{out}, we express $q_{out}(t;\tau) \approx Q_{out}(t)/A_c$ and move this out from the integral over τ. With time characteristics of interest being $\tau < t - t_0 < 1/k$ through most of the transport domain (such that, $\int_0^{t-t_0} g(\tau)d\tau \approx \int_0^{\infty} g(\tau)d\tau = 1$), the remaining integral expression quantifies the expected travel time $E[\tau] \equiv \int_0^{\infty} \tau g(\tau)d\tau \approx \int_0^{t-t_0} \tau g(\tau)d\tau$.

Based on the model Equations 1 and 2, we can also represent the combined source Scenario 3, with contributions from both land-surface and legacy source inputs. The total tracer loading L_{out} and flux concentration C_{out} at the catchment outlet can then be expressed as:

$$L_{out}(t) = \gamma L_{out-1}(t) + (1-\gamma)L_{out-2}(t), \tag{3a}$$

$$C_{out}(t) = \frac{L_{out}(t)}{Q_{out}(t)}, \tag{3b}$$

where the dimensionless factor $0 \leq \gamma \leq 1$ quantifies the relative contribution of each source type to L_{out}.

3 | RECOGNIZING DOMINANT LEGACY SOURCES

Based on available data, we can directly see that more source inputs of chloride than just the precipitation deposition are active in the Kringlan catchment, because the cumulative output load is increasingly greater than the cumulative chloride input by

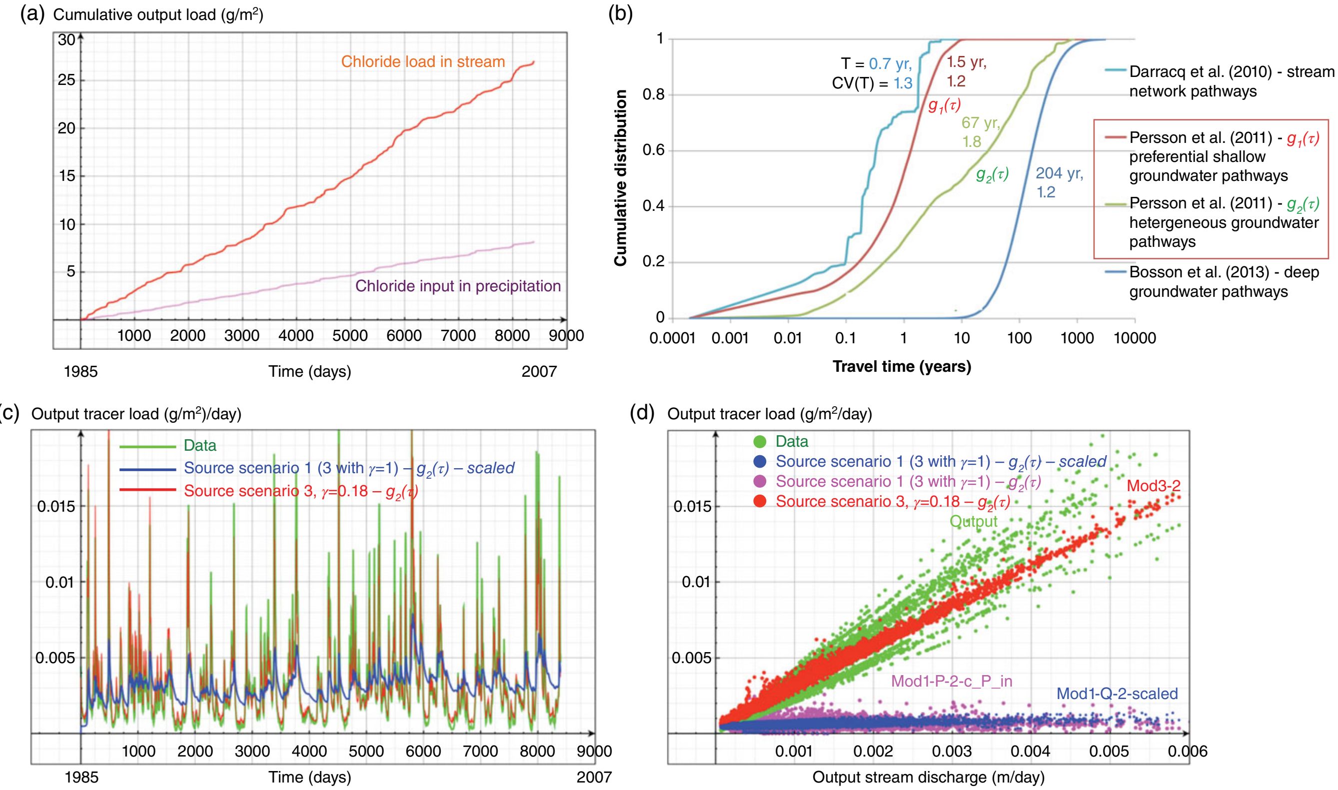

FIGURE 1 Data and model components and results for the Kringlan catchment. (a) Cumulative area-normalized chloride input with precipitation and loading out from the catchment as function of time. (b) Travel time distributions $g(\tau)$ used in the modeling of chloride transport through the catchment, specifically $g_1(\tau)$ and $g_2(\tau)$ in this panel, which are obtained from Persson et al. (2011) for the Swedish catchment Forsmark (see τ maps corresponding to $g_1(\tau)$ and $g_2(\tau)$ in Figure 2a,b, respectively); these are compared with other travel time distributions calculated for other pathway conditions in the same catchment in Darracq, Destouni, Persson, Prieto, & Jarsjö (2010) and Bosson, Selroos, Stigsson, Gustafsson, & Destouni (2013). (c) Data- and model-based area-normalized chloride loading out from the catchment as function of time. (d) Data- and model-based area-normalized chloride loading from the catchment as function of area-normalized water discharge (runoff); see further corresponding flux concentration results and additional result discussion in Figure S1 and Appendix S1

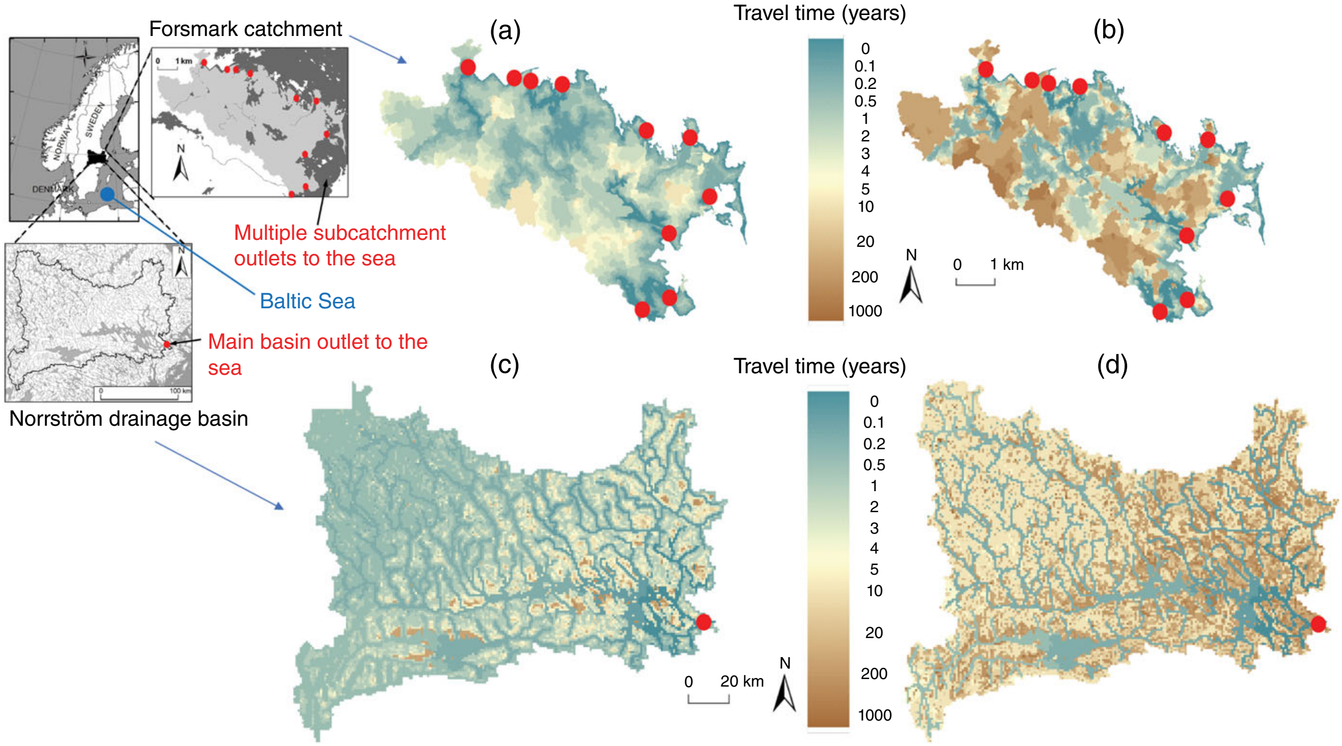

FIGURE 2 Maps of simulated travel time τ under different pathway assumptions for catchments of different scales. The upper-left map inserts show the locations within Sweden and in relation to the recipient Baltic Sea of the small Forsmark catchment (τ maps in panels a,b) and the major Norrström drainage basin (τ maps in panels c,d); red filled circles indicate catchment outlets to the sea. Travel time maps for the Forsmark catchment correspond to the distributions $g_1(\tau)$ and $g_2(\tau)$ in Figure 1b based on results in Persson, Jarsjö, and Destouni (2011) and Darracq, Destouni, Persson, Prieto, and Jarsjö (2010) (a,b), and maps for the Norrström basin are based on results in Darracq et al. (2010). The travel time simulations assume no contribution (a,c) or considerable contribution (b,d) of slow transport pathways through relatively deep and/or low-gradient/conductivity subsurface zones in each catchment

precipitation (Figure 1a). Additional chloride inputs occur in this region from common use of de-icing salt, fertilizers and manure (Thunqvist, 2004). These types of sources are currently active and continuously renewed at the land surface, but both these and direct precipitation have also contributed chloride for some time, possibly leading to accumulation of inaccessible legacy sources in the subsurface.

However, the simple mass balance assessment in Figure 1a does not aid in distinguishing between contributions to total output load from current sources at the land surface and legacy sources from older inputs in the subsurface. For such distinction, we use Equation 3 with different values of the source contribution factor γ, combined with different previously estimated travel time distributions $g(\tau)$, typical for different transport conditions in this boreal region (Figure 1b). From the illustrated distributions in Figure 1b, we use the two subsurface ones $g_1(\tau)$ (red with $E[\tau] = 1.5$ years, Figure 1b) and $g_2(\tau)$ (green with $E[\tau] = 67$ years, Figure 1b) to test model ability to reproduce the output load data in Figure 1c. The observed load variations (green curve, Figure 1c) are poorly reproduced by source Scenario 1 (blue curve), implying that they cannot be explained solely by input from current sources at the land surface. However, the observed variations are relatively well reproduced by the combined source Scenario 3, represented by Equation 3 with $\gamma = 0.18$ and travel time distribution $g_2(\tau)$ (red curve, Figure 1c). This modeled Scenario 3 implies load contributions of 18% from current sources at the land surface (Scenario 1) and 82% from diffuse subsurface legacy sources (Scenario 2). The fit of source Scenario 3 is also nearly as good (not shown) for Equation 3 with the alternative distribution $g_1(\tau)$ and $\gamma = 0.26$, implying similar results of 26% loading from current land surface sources and 74% loading from subsurface legacy sources.

A contribution of around 70%–80% of total load from diffuse legacy sources is thus a robust result, regardless of the exact form of travel time distribution $g(\tau)$. Greater load contribution than around 20%–30% from current land surface sources (Equation 1 or 3 with $\gamma = 1$) has an adverse effect on reproducing observed load variations. This is true also if $L_{\text{out-1}}$ from Equation 1 with tracer input flux given from precipitation flux and concentration data is scaled up (thus representing additional land surface sources) to reach the same average output load level as the data (blue curve, Figure 1c).

Moreover, Scenario 3 with a dominant contribution of around 80% loading from legacy sources (red symbols, Figure 1d) reproduces well the strong correlation exhibited between observation-based data on daily load L_{out} and daily discharge Q_{out} (green symbols, Figure 1d). In contrast, Scenario 1 model of only current land surface sources cannot reproduce this correlation, regardless of used travel time distribution $g(\tau)$ or whether the model is scaled (blue symbols, Figure 1d) or unscaled (pink

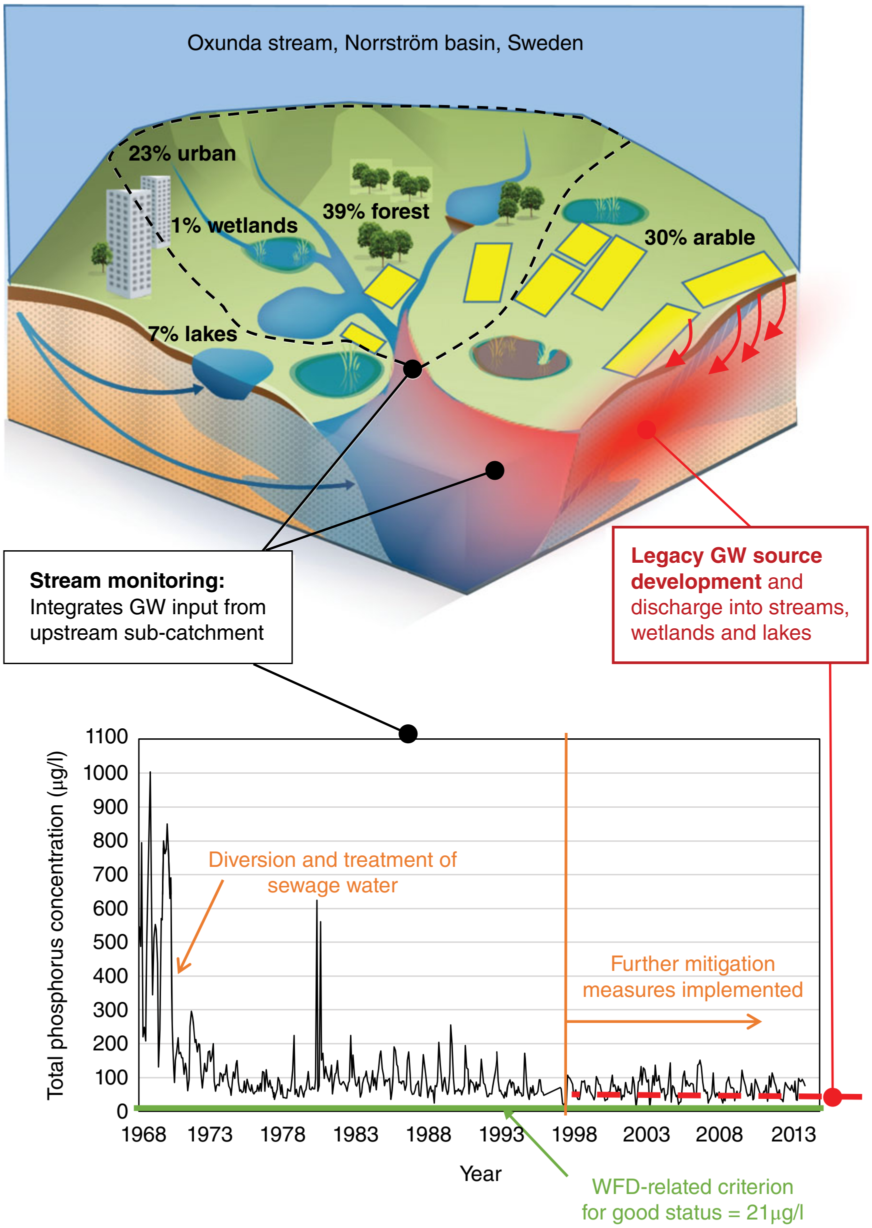

FIGURE 3 (a) Schematic catchment illustration with quantification of land-use shares for the Oxunda catchment, within the Norrström basin, Sweden. (b) Measured monthly concentrations of total phosphorous in the Oxunda stream of Oxunda catchment. The concentration time series in (b) is based on data and results reported in Andersson, Petersson, and Jarsjö (2012). Panel (b) also shows the stable average concentration since 1998 (dashed red line), the timing of implemented mitigation measures (orange line and text), and the WFD- related phosphorous concentration target for good water status (green line)

symbols, Figure 1d); see also corresponding concentration C_{out} results in Figure S1 (Supporting information). The often-found linear correlation between solute load and water discharge, for example, for nutrients (Destouni et al., 2017; Levi et al., 2018) and carbon (Jantze et al., 2013), thus emerges here mechanistically from the robust best-model result of Equation 3 with small γ. This result suggests major load contributions from diffuse legacy sources as a main underlying cause for this correlation and the associated relatively stable $C_{out} = L_{out}/Q_{out}$ level across different Q_{out} conditions (Figure S1).

4 | MANAGEMENT IMPLICATIONS

The dominance of legacy source contributions for chloride as a nonreactive tracer should be due to physical processes (since abiotic or biotic geochemical processes do not affect nonreactive tracer), including slow transport and/or mass transfer and immobilization–remobilization processes (e.g., diffusive mass transfer between mobile and immobile subsurface water zones). These physical processes can affect various waterborne substances, including nutrients and pollutants, with at least slow advective transport acting on and affecting all types of solute.

The travel time distributions $g_1(\tau)$ and $g_2(\tau)$ considered above (Figure 1b) represent different pathway possibilities with regard to slow advective transport. Whereas $g_1(\tau)$ assumes that most subsurface flow and associated waterborne transport in a catchment follows relatively fast pathways (through shallow, high-gradient/conductivity zones), the alternative $g_2(\tau)$ accounts for the possibility that a considerable flow-transport fraction follows much slower pathways (through deeper and/or lower-gradient/conductivity zones; Persson, Jarsjö, & Destouni, 2011). Both of these pathway possibilities may be supported by available data for catchments of different scales (Darracq et al., 2010; Persson et al., 2011). Associated travel time τ scenarios can then be calculated from all possible input points to nearest stream in a catchment, based on available catchment data for topography, soil/aquifer characteristics and flow conditions. Such τ calculations provide statistical distributions $g(\tau)$ (Figure 1b) and corresponding maps of τ distribution over a catchment (Figure 2). The catchment examples in Figure 2 show that the τ mapping can help to identify catchment zones with particularly slow subsurface transport. Dominant legacy sources may have accumulated in such zones, if also considerable historic-to-current land surface sources have prevailed there, feeding pollutants and nutrients into these zones (e.g., indicated by high subcatchment population density; Destouni, Hannerz, Prieto, Jarsjö, & Shibuo, 2008; Levi, Cvetkovic, & Destouni, 2018).

One concrete example of much delayed or even nonexistent response to mitigation measures, likely due to legacy source accumulation, can be illustrated by the case of another relatively well-investigated Swedish catchment, Oxunda (Andersson, Petersson, & Jarsjö, 2012; Figure 3). As with the Kringlan catchment above, the Oxunda catchment (270 km^2) is also located within the major Norrström drainage basin, in its eastern part, where many areas are quantified and mapped to have particularly slow groundwater transport pathways (Figure 2c,d). The Oxunda catchment and overall eastern Norrström basin part are under high population pressures from the expanding city of Stockholm, in addition to agricultural water-quality pressures (see quantified land-use shares in Figure 3a). Phosphorous concentration is monitored at multiple locations (17 lakes and 6 stream monitoring points, including the catchment outlet point) along the Oxunda stream and lake system, as part of inter-municipal water-management cooperation for this catchment. The stream and lake measurements provide integrated measures of the quality of groundwater feeding into the stream–lake network from different catchment zones, reflecting then also the possible contributions from legacy sources in the groundwater system (schematic illustration, Figure 3a). In 2017, none of the lakes or main stream branches in the Oxunda catchment reach the good ecological status required by the WFD; current classifications range between poor and moderate status.

Figure 3b shows that stream concentrations of total phosphorous at the catchment outlet decreased rapidly in the early 1970s, from previously high levels (~300 μg/L and above), as a consequence of removing discharges of untreated sewage water directly into the stream (Andersson, Petersson, & Jarsjö, 2012). Thereafter, extensive additional measures have been taken since 1998 to mitigate the impacts of remaining water-quality pressures, including phosphorous loads. These measures include improved handling of storm water, reduction of considerable source inputs from animal farms, creation of buffer zones between agricultural land and surface waters, and mitigation of lake sediment leakage (Andersson et al., 2012). However, as mentioned above, not a single recipient water body has so far reached good water good status in response to these measures. There are also no notable effects on total phosphorous concentration at the catchment outlet (Figure 3b); the average concentration level remains stable around 50 μg/L since 1990s, which is well above the limit of 21 μg/L for achieving good ecological water status in compliance with the WFD. The near-complete lack of phosphorous concentration response (Figure 3b; dashed red line) to the land-surface and surface-water oriented measures is consistent with dominant contributions from subsurface legacy sources, which are not affected by these measures. Such legacy source contributions can also more generally explain failures to meet WFD-related goals in the regional Norrström basin (Andersson, Jarsjö, & Petersson, 2014; Darracq et al., 2008) and elsewhere in Sweden (Destouni et al., 2017).

The framework and analysis results presented here suggest ways forward to identifying catchment zones with dominant legacy sources. Available water discharge data, combined with concentration and load data for nutrients/pollutants of interest can be used for such identification. Data-given temporal correlation between loads L_{out} and discharges Q_{out} from a (sub)catchment is then a key indicator of dominant legacy-source zones within that (sub)catchment. In other words, a linear temporal L_{out}–Q_{out} correlation (and associated relatively stable average level of concentration C_{out}) suggests water pollution legacies that may be untreatable in practice within the relatively short time frames given for regulation compliance (e.g., 12–14 years in the WFD and BSAP) and with limited resources available for achieving the mitigation targets.

Confidence in legacy-source identification by such L_{out}–Q_{out} correlation may also be strengthened by independent supporting indicators. For example, physically based flow and transport modeling (Bosson et al., 2013; Darracq et al., 2010; Persson et al., 2011; Soltani & Cvetkovic, 2017) and/or isotope-based (e.g., δ^{18}O, δ^2H) water-age dating (Bethke & Johnson, 2008) may quantify zones of slow flow/transport/mass-transfer in the heterogeneous subsurface part of a (sub)catchment. Associated long travel (and total residence) times of water/tracer (and reactive solute) (Cvetkovic, Carstens, Selroos, & Destouni, 2012) imply possible accumulation of waterborne nutrients and pollutants in such zones over time (Darracq et al., 2008; Destouni et al., 2010; Törnqvist et al., 2015; Wang & Burke, 2017). Furthermore, temporally extended presence of sources at the land surface (Darracq et al., 2008) (or proxy indications of such presence, e.g., by population data (Destouni et al., 2008; Levi et al., 2018)) implies sustained long-term feeding of nutrients and pollutants from the surface into the subsurface accumulation zones, which can thereby build up to constitute today's legacy sources.

Other studies have shown that calibration of frequently used homogeneous flow-transport models to heterogeneous catchments (with "real-world" spread in their travel time distribution) may lead to large and systematic estimation errors in basic transport quantities, such as average travel time (Kirchner, 2016). In particular, aggregation errors imply that such models tend to considerably underestimate mean travel times (Kirchner, 2016), with important implications for estimates of pollutant and nutrient loads to downstream surface and coastal waters in heterogeneous catchments (Persson et al., 2011). Zones of slow subsurface transport and their contributions to nutrient and pollutant loads and concentrations may then evade detection and attention; this may apply even if homogeneous catchment models are seemingly successfully calibrated to available tracer data (e.g., Cl^-, $\delta^{18}O$, $\delta^{2}H$), emphasizing the need for independent data checks, such as the diagnostic test and other indicators outlined above. More generally, not recognizing such prohibitively difficult mitigation conditions associated with ubiquitous and dominant legacy sources may be a main cause for common failure to reach legally required and/or international agreed targets of good water quality.

5 | CONCLUSIONS

We have outlined a process-based diagnostic test to detect dominant contributions of subsurface legacy sources to total nutrient/pollutant load from any (sub)catchment. This involves checking available (sub)catchment data for possible linear temporal correlation between L_{out} and Q_{out} (Figure 1d), and associated relatively stable average level of C_{out} (Figure S1). Confidence in legacy-source identification with this test may also be strengthened by independent quantification of slow transport pathways with associated long solute travel times (Figure 2b,d) and records of temporally extended nutrient/pollutant sources (or proxies for these) in the same (sub)catchment. Such identification opens opportunities to meet regulation requirements of short-term water quality improvements by spatially differentiated mitigation measures, directed to (sub)catchments without dominant legacy sources. To be able to implement such differentiated mitigation strategies, relevant data must be available for quantifying and assessing the spatial distribution of legacy-source indicators.

Furthermore, if new sources do not continue to feed into and keep up legacy pollution levels, depletion of legacy sources should lead to water quality recovery also in areas of currently untreatable water pollution. However, this will happen on longer time scales (of several decades or more) than those commonly required for regulation compliance. It is a major challenge to decide how available mitigation resources should be shared between efforts for short-term mitigation compliance in areas without major legacies, and long-term source reduction efforts in areas with such legacies. This challenge needs to be recognized and addressed in further research.

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CONFLICT OF INTEREST

The authors have declared no conflicts of interest for this article.

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section at the end of the article.

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