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Tam V. Nguyen, Andreas Musolff, Pia Ebeling, Fanny J. Sarrazin, Jan H Fleckenstein, et al.. Long-term assessment of point source contributions to nitrate pollution in the Central European Rhine and Elbe river basins. *Environmental Research Letters*, 2026, 21 (4), pp.044018. <10.1088/1748-9326/ae44b0>. <hal-05529026>

**HAL Id: hal-05529026**

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Submitted on 26 Feb 2026

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To cite this article: Tam V Nguyen *et al* 2026 *Environ. Res. Lett.* **21** 044018

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## LETTER

## Long-term assessment of point source contributions to nitrate pollution in the Central European Rhine and Elbe river basins

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E-mail: [tam.nguyen@ufz.de](mailto:tam.nguyen@ufz.de) and [rohini.kumar@ufz.de](mailto:rohini.kumar@ufz.de)**Keywords:** point and diffuse nitrogen sources, stream water quality, nitrate pollution, wastewater effluent, European RiversSupplementary material for this article is available [online](#)

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1 July 2025REVISED  
3 February 2026ACCEPTED FOR PUBLICATION  
11 February 2026PUBLISHED  
24 February 2026Original content from  
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citation and DOI.**Abstract**

Understanding the contributions of diffuse and point sources to nitrate pollution is crucial for managing river water quality. We conducted a long-term modeling study for the Rhine and Elbe basins and their 146 subbasins from 1950 to 2021 to quantify the roles of diffuse and, in particular, point sources in driving stream  $\text{NO}_3\text{-N}$  concentrations. In both basins, simulated results show a decline in point source contributions from 1950 to 2000, followed by a relatively stable level at around 25% in the Rhine and fluctuations around 30% levels in the Elbe. The decline in the simulated stream  $\text{NO}_3\text{-N}$  concentrations in both basins after 1990 was largely driven by a decrease in point sources, and stream  $\text{NO}_3\text{-N}$  concentrations remained high ( $\sim 2 \text{ mg l}^{-1}$ ) during 2010–2021, even when point sources were excluded. At the subbasin level, changes in point source contributions and stream  $\text{NO}_3\text{-N}$  concentrations reflected the overall trends of their respective basins, although individual subbasins exhibited diverse patterns. In subbasins with high stream  $\text{NO}_3\text{-N}$  concentrations during 2010–2021, point source contribution accounted for around 30% (median values across subbasins), and the fractions of agricultural, urban, and industrial land cover were relatively high. These results highlight that point source management alone is not sufficient to reduce stream nitrate to a good ecological status ( $< 2 \text{ mg l}^{-1}$ ), and spatial targeted management is required to achieve good ecological status at both the regional and local levels.

**1. Introduction**

Eutrophication is a significant problem affecting the quality of surface and coastal waters around the world (Diaz and Rosenberg 2008, Boesch 2019, de Raús Maúre *et al* 2021). A key driver of eutrophication is the export of surplus nitrogen (N) from agricultural lands (diffuse sources) and effluent of N from treated wastewater treatment plants (WWTPs) and untreated wastewater (point sources). Understanding the relative contribution of N from diffuse and point sources in streams is essential for designing and prioritizing effective measures (O’Shea 2002, Pieterse *et al* 2003, Westphal *et al* 2020). This knowledge is

also crucial for assessing how well nutrient reduction efforts work, since managing point sources can hide the impact of diffuse sources, and vice versa (Lam *et al* 2010, Dupas *et al* 2018).

The role of diffuse and point sources in stream nitrogen (total N or nitrate) has been widely studied at global, regional, and local scales. For example, Van Drecht *et al* (2003) in their global scale analysis, found that point sources contributed 23% of total stream N in European rivers in the mid-1990s. Grizzetti *et al* (2008) reported that point sources accounted for 14%–50% of stream nitrate in major European basins (Danube, Rhine, Elbe, Weser/Ems, Meuse, Seine, and Rhône) between 1995 and 2002.

More recently, Wachholz *et al* (2022) and Ebeling *et al* (2021) documented long-term changes in stream nitrate concentrations and seasonality in German and French catchments, reflecting shifts in the dominance of point versus diffuse inputs. Sarrazin *et al* (2022) estimated that point sources contributed on average between 20 and 34% of stream nitrate across sub-basins in the Weser basin between 1960 and 2015. These studies, along with others (Pieterse *et al* 2003, Fuchs *et al* 2010, Lam *et al* 2010, Westphal *et al* 2020), demonstrate that the significance of diffuse and point sources can vary greatly between regions and over time. The aforementioned studies are limited to either short time scales (few years to less than a decade; Van Drecht *et al* 2003, Grizzetti *et al* 2008) or a few large basins (from 5000 km<sup>2</sup> to less than 50 000 km<sup>2</sup>; Sarrazin *et al* 2022) or do not quantify source contributions (Ebeling *et al* 2021, Wachholz *et al* 2022). A systematic study to understand the impact of diffuse and point source pollution across different spatial scales (from 100 km<sup>2</sup> to more than 100 000 km<sup>2</sup>) over extended periods is thus missing. Such an assessment allows for the identification of hotspot areas of nutrient pollution and the primary sources requiring targeted interventions.

The Central European region, especially Germany, has a long history of agricultural intensification and continuous improvement in wastewater treatment facilities (Batool *et al* 2022, Sarrazin *et al* 2024). The Rhine and Elbe Rivers are two major Central European rivers that drain into the European North Sea (Claussen *et al* 2009) and particularly the Wadden Sea, a UNESCO World Heritage site due to its unique ecological value. Recent reports indicate that nitrogen concentrations in most streams within these basins remain persistently high, exceeding the thresholds needed for good ecological status in rivers (2 mg l<sup>-1</sup> total N; Vigiak *et al* 2023). Eutrophication continues to inhibit natural aquatic vegetation in the Wadden Sea (van Katwijk *et al* 2024). Additionally, a recent review of the European Zero Pollution Action Plan (EC 2021) indicated that current EU policies should further reduce nutrient discharges into water from point sources (EEA 2024). These challenges highlight the importance of understanding the roles of both diffuse and point sources in influencing stream water quality in the Elbe and Rhine basins. To this end, we analyze the relative contribution of diffuse and point sources to stream nitrate (NO<sub>3</sub>-N) from 1950 to 2021 in the Rhine and Elbe basins and their subbasins. Specifically, we address the following questions:

1. How have point source contributions affected stream NO<sub>3</sub>-N in the Rhine and Elbe basins?
2. Which regions within these basins are hotspots of nitrogen inputs from point sources and would

benefit most from improved wastewater management?

3. Is improving point sources alone sufficient to reduce annual stream NO<sub>3</sub>-N concentrations to meet the good ecological status threshold of 2 mg l<sup>-1</sup> (Vigiak *et al* 2023)?

Through a long-term model-based assessment, our study provides valuable insights into the temporal and spatial dynamics of nutrient sources across different spatial scales in the Rhine and Elbe River basins. This is our main contribution, given that observation-based assessments are typically restricted to local scales, with only a few basins having continuous long-term stream NO<sub>3</sub>-N records. By quantifying the relative importance of diffuse and point sources, our study aims to identify key leverage points for reducing nitrate pollution and to better inform agricultural nutrient management. We also examine and discuss both the progress and remaining challenges of intervention activities for N pollution levels, and thereby our assessment can support policymakers to develop effective measures to improve water quality.

## 2. Method

### 2.1. Study area and data

The focus of our study is the Rhine and Elbe basins and their subbasins (figure 1). The Rhine and Elbe basins have an area of 272 950 km<sup>2</sup> (Rhine at Cologne: 144 415 km<sup>2</sup>; Elbe at Neu Dachau: 128 530 km<sup>2</sup>) and discharge into the Wadden Sea. About 60% of the Rhine and Elbe basins are located within German territory. The Rhine basin (within Germany) is located in the former West Germany, while the majority of the Elbe basin area (within Germany) is located within the former East Germany. There have been significant differences in the agricultural activities and wastewater treatment systems between West and East Germany due to their distinct political and economic systems (Sarrazin *et al* 2024). In total, our study consists of 80 subbasins in the Rhine and 66 subbasins in the Elbe (figure 1), including nested subbasins. The selection of the subbasins was done based on the availability of observational stream NO<sub>3</sub>-N and discharge data (see below). We note that the subbasins do not cover the entirety of the Rhine and Elbe main basins but represent a wide diversity of area, land use, topography, and hydro-meteorological characteristics (figures 1 and S9). The actual Rhine (163 660 km<sup>2</sup>) and Elbe (138 890 km<sup>2</sup>) basins are larger than the Rhine (144 415 km<sup>2</sup>) and Elbe (128 530 km<sup>2</sup>) basins considered in this study.

Data used in this study are at a yearly time step, including stream NO<sub>3</sub>-N concentration, N point and diffuse sources, and stream discharge. Yearly discharge-weighted stream NO<sub>3</sub>-N concentrations were derived from the QUADICA dataset (Ebeling

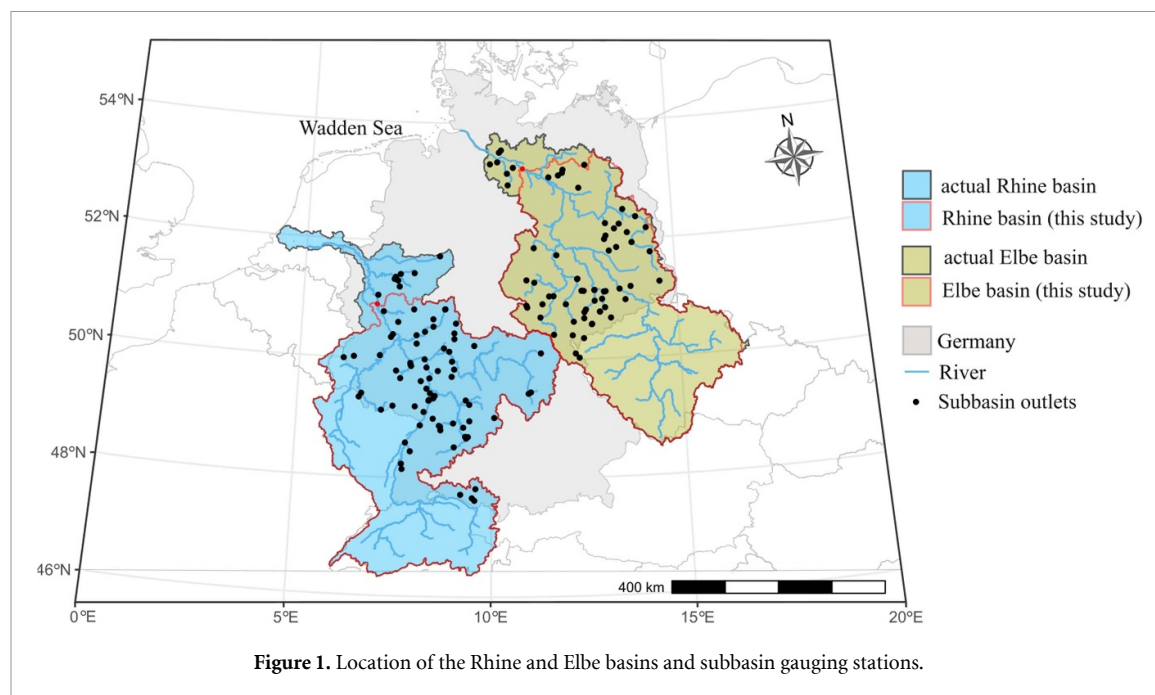


Figure 1. Location of the Rhine and Elbe basins and subbasin gauging stations.

*et al* 2022, 2026). The number of years with available stream  $\text{NO}_3\text{-N}$  data across study basins and subbasins ranges from 8 to 66, with a median of 20 years. Point source N data (1950–2019), including treated and untreated wastewater emissions, were taken from Sarrazin *et al* (2024) (ensemble mean of the point source data downscaled with population data) for areas within Germany and from Sarker (2024) for Switzerland and the Czech Republic. Within Germany, N diffuse source was provided by Batool *et al* (2022) for 1850–1949, and Häußermann *et al* (2020) for 1950–2021, as described in Ebeling *et al* (2026). For the catchment area outside Germany, the N diffuse source from Batool *et al* (2022) was adopted. N point and diffuse sources over 2020–2021 were assumed equal to 2019. Discharge data from 1950 to 2021 were taken from the QUADICA dataset (Ebeling *et al* 2026). For the two stations at the Elbe and Rhine outlets, we took discharge data from the Global Runoff Data Center. A more detailed data description is provided in the supporting information (text S1).

## 2.2. Nitrogen transport model and simulations

We developed the multiscale water quality model (mQM), in which we balance between process complexity, data availability, and the scale (yearly timestep at multi-decadal and catchment scale) of application. The mQM model simulates nitrogen processes using streamflow from observed or modeled data; and soil moisture from the meso-scale hydrologic model (Samaniego *et al* 2010, Kumar *et al* 2013). The model was used to reconstruct long-term continuous stream  $\text{NO}_3\text{-N}$  concentrations and separate the effects of point and diffuse sources, which could not be determined from incomplete or

discontinuous observations (figure S10). While the mQM model is primarily based on the ELEMENNT (Van Meter *et al* 2017) and StorAge Selection (SAS)-N (Nguyen *et al* 2022) models, it provides a more detailed representation of processes than either of these models individually. In addition, the mQM model also aims to improve process representation more than each of these models alone. For example, the mQM model provides a more detailed representation of the N transport process in the subsurface than the ELEMENNT model by using the SAS concept (Botter *et al* 2011, van der Velde *et al* 2012, Harman 2015, Nguyen *et al* 2022). It also provides a more detailed representation of the N process in the soil zone, accounting, among other factors, for the variability of soil moisture dynamics, compared with the SAS-N model, as described below (see also the model documentation in the supporting information). The model consists of three main compartments: the soil/root zone, the subsurface (including deeper soil and groundwater), and the stream. Effective precipitation (precipitation minus actual evapotranspiration) entering the soil zone is assumed to equal the sum of percolation to the subsurface and streamflow.

Similar to the ELEMENNT model, the mQM model considers different N pools within the soil zone, organic and inorganic N pools, while the SAS-N does not differentiate between soil N pools. Excess N surplus can enter the active organic, passive organic N pools as in the ELEMENNT model, or directly the inorganic N pool, as N surplus also contains inorganic N from inorganic fertilizer application. Active and passive organic N can mineralize and become inorganic mobile N. The passive organic N pool has a much slower mineralization rate than the active organic N pool representing a potential build-up of

soil biogeochemical N legacy (Six *et al* 2002, Van Meter *et al* 2017, Lutz *et al* 2022). This is in line with the representation of soil N pools in existing N transport models at smaller time steps (e.g. daily) such as HYPE (Lindström *et al* 2010), SWAT (Arnold *et al* 1998), INCA (Wade *et al* 2002), or LPJml (von Bloh *et al* 2018). A unique feature of mQM compared with ELEMNT, HYPE, SWAT, and INCA is the transport of inorganic N to deeper soil and groundwater based on the SAS concept (Botter *et al* 2011, van der Velde *et al* 2012, Harman 2015, Nguyen *et al* 2022). The SAS function describes how a catchment selects water of different ages for discharge, thereby influencing the transit time distribution and the nitrate concentration in discharge. The transit time distribution derived from the SAS function does not necessarily follow a specific distribution function, while the ELEMNT model assumes a predefined functional form of the transit time distribution (Van Meter *et al* 2017, Sarrazin *et al* 2022). Water with younger ages is typically linked to short and shallow subsurface flow (e.g. interflow from the deeper soil), whereas water with older ages is associated with longer and deeper flow paths, such as from the groundwater (Yang *et al* 2018) - which can be effectively represented in the SAS based transit time distribution approach. Similar to van der Velde *et al* (2010), we assume that the age of water is the age of nitrate; in other words, sorption-desorption processes is not accounted in this study. Inorganic N in both the soil and the subsurface can be denitrified following a first-order reaction (e.g. Beusen *et al* 2015, Van Meter *et al* 2017). Inorganic N from the subsurface mixes with N from point sources in the river and is transported to the basin outlets, subjected to instream N removal. A total of nine parameters need to be calibrated within mQM (table S1). More information about the model and underlying parameterizations can be found in the model documentation in the supporting information.

The model was established over the main Rhine and Elbe basins and 146 subbasins (figure 1) with the point and diffuse N source data described above. The initial conditions, warm-up (1850–1949), and calibration (1950–2011) are described in text S2. Each study basin (and subbasin) was parametrized (calibrated) and established independently. We identified suitable behavioral model parameters for each basin by first generating 100 000 parameter sets within the *a priori* range (table S1) using the Latin Hypercube Sampling approach. Next, the 100 behavioral simulations (parameter sets) with the lowest root mean square error values between the simulated and corresponding yearly stream NO<sub>3</sub>-N concentration observations were selected. In general, the model exhibited satisfactory performance in capturing observed variability of instream NO<sub>3</sub>-N concentrations (text S3), and the internal model results are also physically plausible (text S4). The performance of the mQM model is comparable to that of the existing SAS-N

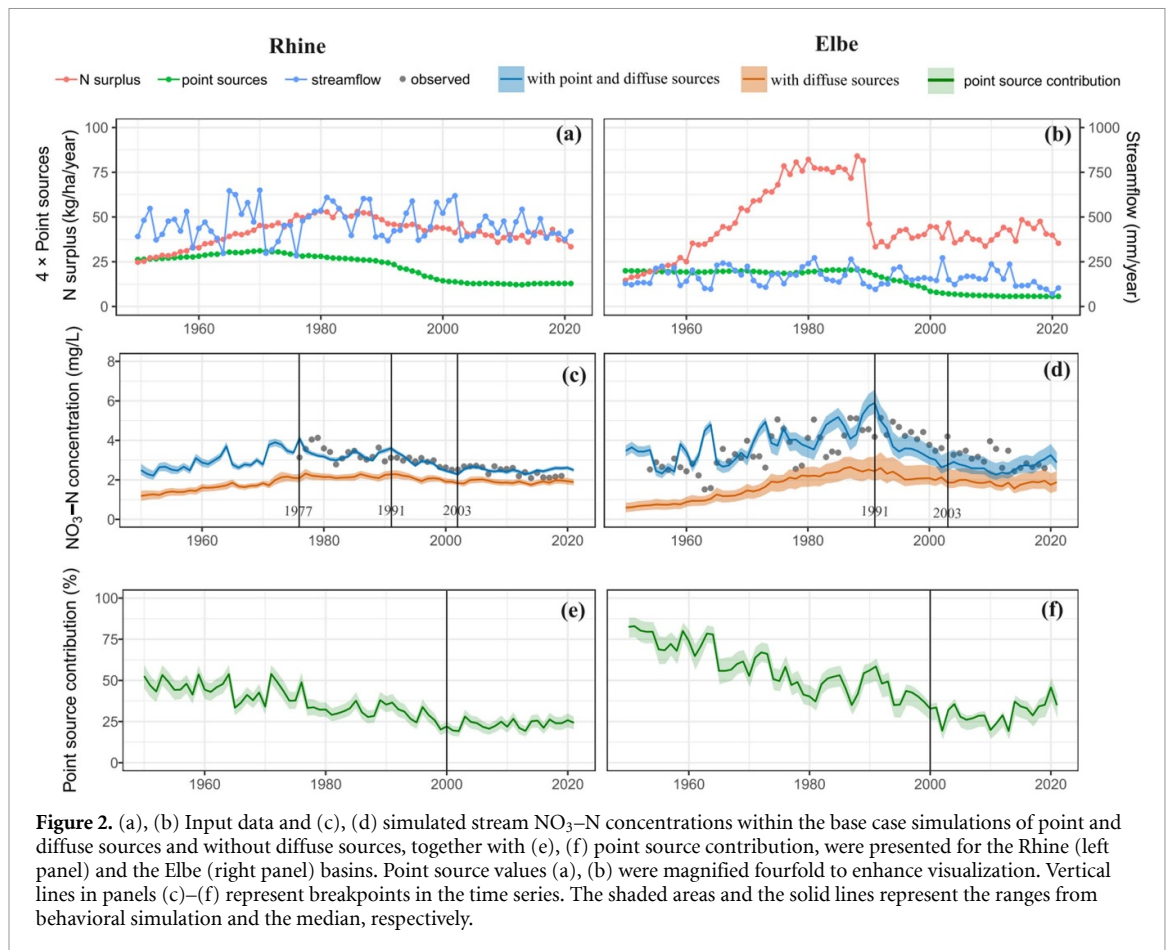
model (Nguyen *et al* 2022) for 28 common subbasins (text S3). Using these behavioral parameter sets, we performed a series of model simulations with diffuse source only to disentangle the role of point and diffuse sources on stream NO<sub>3</sub>-N. Then, we used the Mann-Kendall trend test (Mann 1945, Kendall 1975) to detect statistically significant trends ( $p$ -value < 0.05), and Sen's slope (Sen 1968) to estimate the magnitude of these trends. Furthermore, we analyzed the median NO<sub>3</sub>-N concentrations for each subbasin during two periods (1980–1989 and 2010–2021) to identify hotspots of point and diffuse sources before and after the EU Nitrate Directive (EEC 1991) and other regulations on wastewater (e.g. EEC 1991, AbwV 1997). Using median values over a multi-year period is more robust than relying on a single year (e.g. peak values), since single-year estimates are subject to greater uncertainty (e.g. from the data, model structure, and model parameters).

All simulations above (whether using diffuse sources only or both point and diffuse sources; hereinafter referred to as the base case) did not consider uncertainty in point and diffuse sources (Batool *et al* 2022, Sarrazin *et al* 2024). To account for such uncertainty, we reran the calibrated models at the subbasin levels with different point and diffuse source data to evaluate the impact of input data uncertainty on the main outcomes (text S5). In the remaining, we presented and discussed results from the base case without using the term 'base case' again unless otherwise stated to avoid repetitive use of this term. Comparative analysis from the model runs with different point and diffuse source data are presented in the supporting information (text S5), supporting the findings of the herein analyzed base case simulation results.

### 3. Results

#### 3.1. Stream NO<sub>3</sub>-N concentrations and point source contribution in the main basins

In the Rhine and Elbe basins, simulated stream NO<sub>3</sub>-N concentrations in the base case with point and diffuse sources were noticeably higher than those in the case with diffuse sources, especially before 1991 (figures 2(c) and (d)). Simulated concentrations in all cases showed significant increases ( $p$ -value < 0.05, Sen's slope  $s = 0.04$ – $0.06$  mg l<sup>-1</sup> yr<sup>-1</sup>) from 1950 to 1977 in the Rhine and to 1991 in the Elbe. After 1977 in the Rhine (figure 2(c)) and after 1991 in the Elbe (figure 2(d)), concentrations in both cases began to show divergent trends. For example, from 1977 to 2003, in the Rhine, simulated stream NO<sub>3</sub>-N concentrations with diffuse sources have very minor decreasing trends, while those with point and diffuse sources showed clear decreasing trends. The declines in stream NO<sub>3</sub>-N with point and diffuse sources were most pronounced from 1991 to 2003 (Rhine:  $p$ -value < 0.05,  $s = -0.10$  mg l<sup>-1</sup> yr<sup>-1</sup>; Elbe:



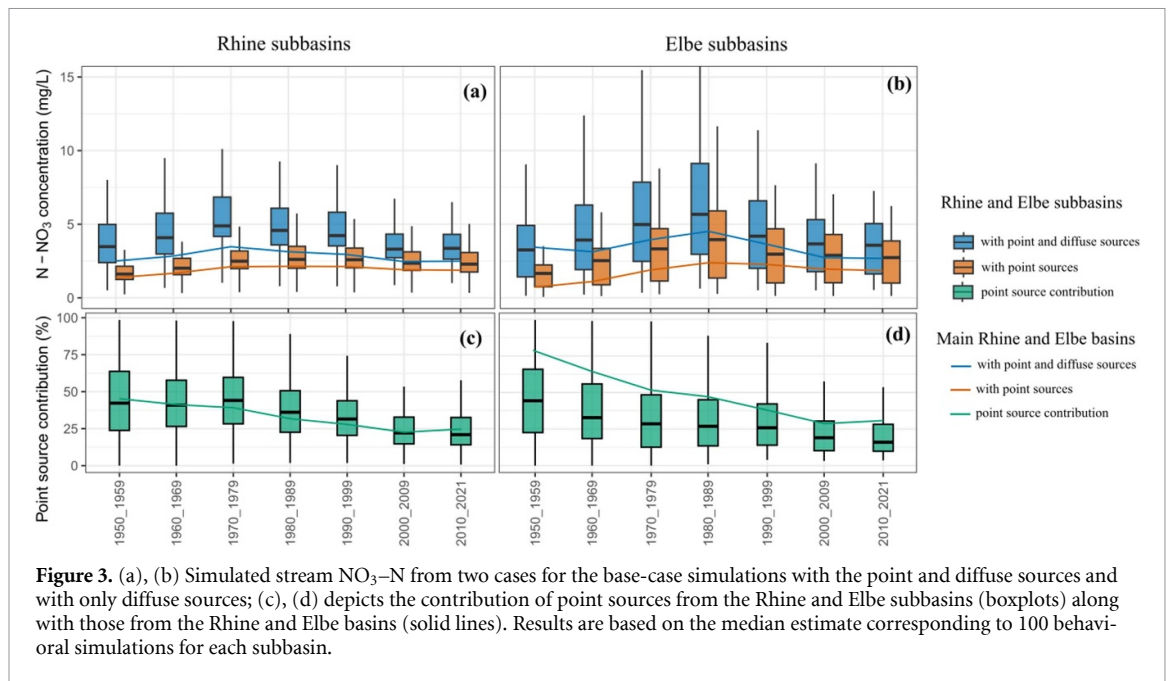
$p$ -value  $< 0.05$ ,  $s = -0.18 \text{ mg l}^{-1} \text{ yr}^{-1}$ ; figure 2(d)), corresponding with reductions in point source inputs (figures 2(a) and (b)). In both basins (figures 2(c) and (d)), simulated stream  $\text{NO}_3\text{-N}$  concentrations with diffuse sources averaged around  $2 \text{ mg l}^{-1}$  in the final two decades (1991–2021) of the simulation period, but concentrations exceeded this threshold with point and diffuse sources. The reduction in diffuse sources around 1990 (figures 2(a) and (b), red lines) only had a minor effect on the reduction of stream  $\text{NO}_3\text{-N}$  concentrations (figures 2(c) and (d), orange lines) in the Rhine and Elbe.

The point source contribution to stream  $\text{NO}_3\text{-N}$  loading in both basins (figures 2(e) and (f)) showed a decreasing trend from 1950 to 2000 (Rhine:  $s = -0.5\%$  per year; Elbe:  $-0.9\%$  per year; both with  $p$ -value  $< 0.05$ ). In the Rhine, point source contributions remained relatively stable in the recent decade, between 2010 and 2021 ( $p$ -value = 0.18,  $s = 0.1\%$  per year), while in the Elbe, they have increased slightly ( $p$ -value = 0.07,  $s = 0.3\%$  per year). During the 1950s, point source contributions in the Elbe were comparatively higher than in the Rhine by approximately 75% compared to 50%, which is likely due to lower diffuse source outputs in the Elbe basin during that time. Median stream  $\text{NO}_3\text{-N}$  concentrations from diffuse sources in the Elbe during the 1950s

were about  $0.8 \text{ mg l}^{-1}$ , compared to  $1.2 \text{ mg l}^{-1}$  in the Rhine. The recent increase in point source contributions in the Elbe (2010–2021) could also be attributed to changes in hydrologic conditions affecting stream  $\text{NO}_3\text{-N}$  concentrations (figures 2(b) and (f)). Notably, during the recent multi-year drought in Central Europe (Hari *et al* 2020, Moravec *et al* 2021), streamflow reductions were more pronounced in the Elbe River (figure 2(b)). Drought conditions can significantly compromise nitrogen retention and export behavior in catchments (Winter *et al* 2023).

### 3.2. Stream $\text{NO}_3\text{-N}$ concentrations and point source contribution in the subbasins

The overall temporal trends in simulated stream  $\text{NO}_3\text{-N}$  concentrations (represented by median values in the boxplots, figures 3(a) and (b) for the subbasins in both cases (with point and diffuse sources and with diffuse sources) mirrored those observed in the main Rhine and Elbe basins. For example, at the decadal scale, median stream  $\text{NO}_3\text{-N}$  concentrations in the Rhine increased from the 1950s to the 1970s in both cases (figure 3(a)). After the 1970s, concentrations with point and diffuse sources in the Rhine slightly declined until the 2000s and then remained stable into the 2010s, while concentrations with diffuse sources stayed relatively constant throughout. In



**Figure 3.** (a), (b) Simulated stream  $\text{NO}_3\text{-N}$  from two cases for the base-case simulations with the point and diffuse sources and with only diffuse sources; (c), (d) depicts the contribution of point sources from the Rhine and Elbe subbasins (boxplots) along with those from the Rhine and Elbe basins (solid lines). Results are based on the median estimate corresponding to 100 behavioral simulations for each subbasin.

both cases, the Elbe River displayed similar patterns, except that the increase extended from the 1950s until the 1980s rather than the 1970s (figure 3(b)). During 2010–2021, median  $\text{NO}_3\text{-N}$  concentrations in subbasins of both the Rhine and Elbe exceeded  $2 \text{ mg l}^{-1}$  in both cases (figures 3(a) and (b)).

The median trends of point source contributions to stream  $\text{NO}_3\text{-N}$  loading in the Rhine subbasins were similar to those in the overall Rhine basin (figure 3(c)), showing a decline from the 1950s to the 2000s, followed by relative stability thereafter. The Elbe subbasins showed a comparable decreasing trend in point source contributions from the 1950s to the 2000s (figure 3(d)). After the 2000s, median point source contributions in the Elbe subbasins showed only a minor reduction.

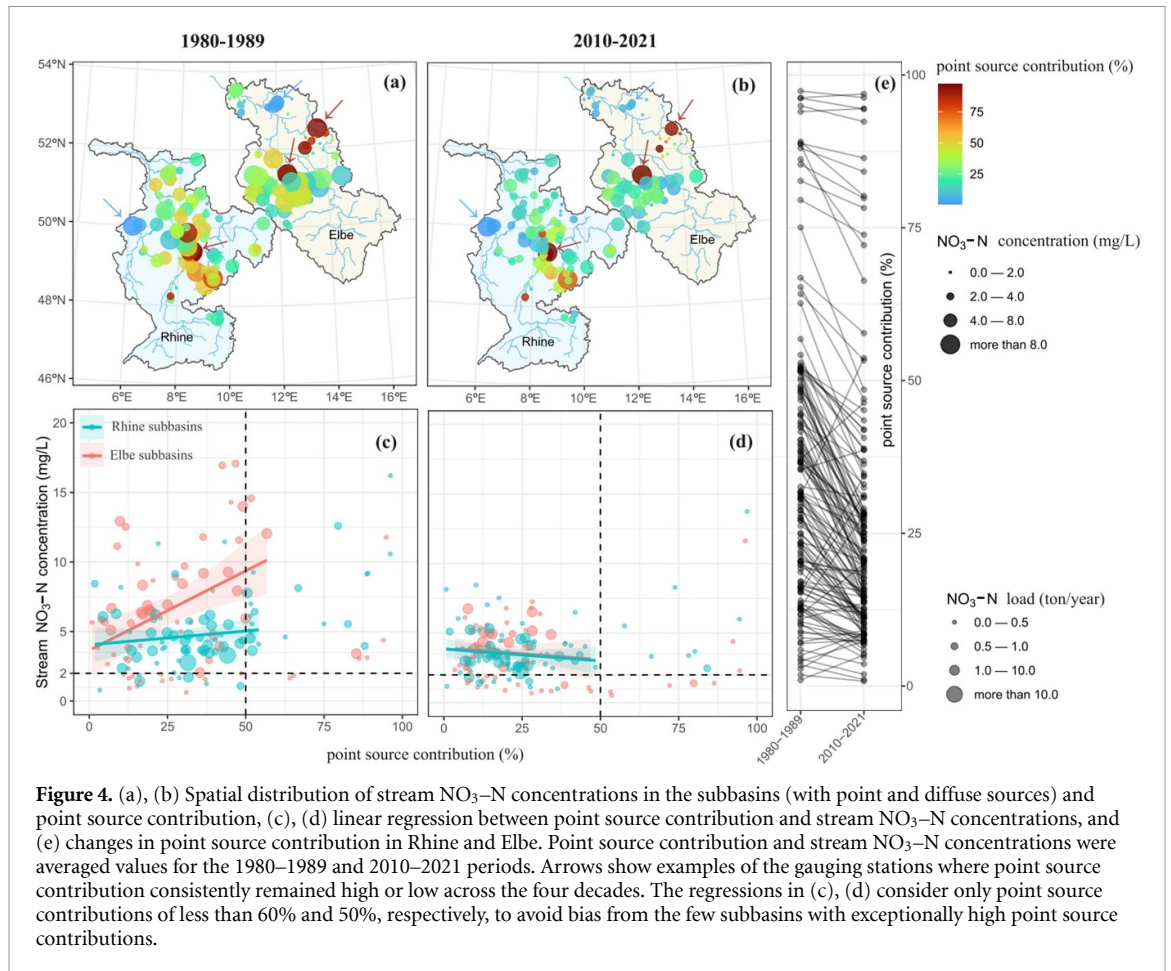
Figures 4(a) and (b) shows substantial variability in stream  $\text{NO}_3\text{-N}$  concentrations (with point and diffuse sources) and point source contributions across subbasins and periods. During both periods (1980–1989 and 2010–2021), 54 subbasins ( $\sim 37\%$  of all subbasins) consistently had point source contributions below 25%, while 10 subbasins ( $\sim 7\%$ ) consistently exceeded 75% (e.g. stations indicated by blue and dark red arrows in figures 4(a) and (b)). Basins where point source contributions consistently exceeded 75% included both small ( $37 \text{ km}^2$ ) and large ( $6256 \text{ km}^2$ ) basins. In general, most (130 or 89%) subbasins showed reductions in point source contributions over time (figures 4(b) and (c)). In contrast, 16 (11%) subbasins experienced substantial increases in point source contributions (figure 4(e)). Despite the increased point source contribution in the aforementioned 16 subbasins, their median stream  $\text{NO}_3\text{-N}$  concentrations decreased from  $4.9 \text{ mg l}^{-1}$

during 1980–1989 period to  $3.4 \text{ mg l}^{-1}$  during 2010–2021. In both periods, most Rhine and Elbe subbasins had point source contributions below 50% but stream  $\text{NO}_3\text{-N}$  concentrations exceeding  $2 \text{ mg l}^{-1}$  (figures 4(c) and (d)). During the 1980–1989 period, point source contributions were positively correlated with stream  $\text{NO}_3\text{-N}$  concentrations across the subbasins, especially in the Elbe (Pearson's correlation coefficient  $r = 0.36$ ,  $t$ -test with  $p$ -value  $< 0.05$ ; linear regression shown in figure 4(c)), indicating that higher point source contributions were associated with higher stream  $\text{NO}_3\text{-N}$  concentrations. This relationship weakened over time, with no statistically significant correlation found for 2010–2021 (figure 4(d)), partly due to the more dominant control of diffuse source exports (figures 3(c) and (d)). In both periods, subbasins with higher stream  $\text{NO}_3\text{-N}$  concentrations ( $\geq 2 \text{ mg l}^{-1}$ ; 125 and 116 subbasins in 1980–1989 and 2010–2021, respectively) had higher agricultural, urban, and industrial land cover fractions, as well as a higher number of urban WWTPs and population density (text S6). Both periods also show that subbasins with high stream  $\text{NO}_3\text{-N}$  concentrations ( $\geq 2 \text{ mg l}^{-1}$ ) and high point source contribution ( $\geq 50\%$ ) had clearly higher urban and industrial land cover fractions and population density (text S6).

## 4. Discussion

### 4.1. The role of point source in stream $\text{NO}_3\text{-N}$ concentrations

Results from our study show that point source contributions in the Rhine and Elbe basins, including most of their subbasins, declined from 1950 to 2000



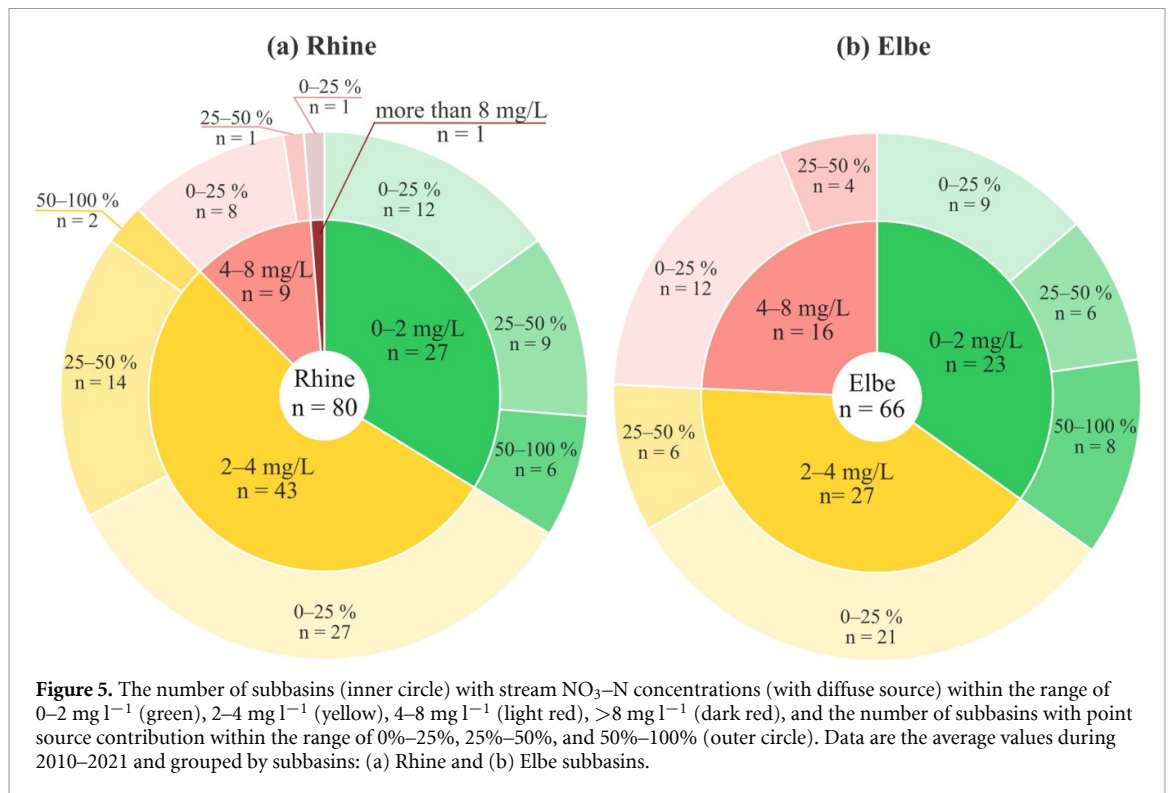
and then remained relatively stable at around 25%–30%. Point source contributions during the mid-1990s in the Elbe and Rhine basins align with the findings of Van Drecht *et al* (2003) for European rivers. Our results indicate that substantial improvements in stream  $\text{NO}_3\text{-N}$  concentrations from 1991 to 2003 were mainly driven by reductions in point sources, rather than by reductions in nitrogen surplus and associated diffuse inputs targeted under the EU Nitrate Directive.

The results also show that point sources were the dominant source of stream  $\text{NO}_3\text{-N}$  from 1950 to 1980 in the main Elbe basin, contributing more than 50% during this period, while point source contributions from the Rhine basin (and most of its subbasins) as well as most Elbe subbasins remained below 50%. The higher point source contribution in the main Elbe basin compared to its subbasins (figure 3(d)) may be explained by large WWTPs discharging directly into the higher-order river network. Large cities such as Berlin (3.4 million inhabitants), Leipzig (620 000), Dresden (574 000), Magdeburg (244 000), and Halle (227 000) together account for about 25% of the Elbe basin's total population but are not located in the subbasins. Therefore, the subbasins do not fully capture the substantial point source inputs from these highly urbanized areas, instead reflecting lower-populated regions.

#### 4.2. Is point source management alone sufficient?

In the Rhine and Elbe basins, point source management alone is unlikely to be sufficient to achieve good (aquatic) ecological status ( $<2 \text{ mg l}^{-1}$ ; Poikane *et al* 2019, Bouwman *et al* 2024), as 96 subbasins (about 66% of the subbasins) in the Rhine and Elbe exceeded  $2 \text{ mg l}^{-1}$  during 2010–2021, even in the absence of point sources (figure 5). In nearly all of these subbasins (except two in the Rhine; figure 5(a)), point source contributions were below 50%. This indicates that point source management alone is not sufficient to reduce  $\text{NO}_3\text{-N}$  levels to good ecological status in most Rhine and Elbe subbasins. However, 50 subbasins (about 34% of subbasins, figures 5(a) and (b)) could potentially reach good ecological status through point source management alone, assuming full N removal from point (WWTP) sources were achievable. Among these 50 subbasins, 11 subbasins need special attention for point source management, as both point source contribution ( $\geq 50\%$ ) and stream  $\text{NO}_3\text{-N}$  concentration ( $\geq 2 \text{ mg l}^{-1}$ ) were high. These subbasins were characterized by relatively high urban and industrial land cover fractions and high population density (text S6).

Overall, this underscores the need for integrated approaches that combine further point source reductions with effective measures to control diffuse (agricultural) nitrogen inputs if water quality targets in



the Rhine and Elbe basins and subbasins are to be met sustainably. Strengthening coordination between wastewater management and diffuse pollution will therefore be crucial for achieving long-term improvements in stream water quality. Together, these findings emphasize that focusing on only one source pathway will likely fall short of meeting ecological goals in complex, intensively used basins like the Rhine and Elbe.

#### 4.3. Implications for the existing and potential measures

The EU Nitrate Directive, which came into force in 1991, has likely contributed to the reduction in diffuse nitrogen surplus in the Rhine basin (western Germany), where it declined from  $49\text{ kg ha}^{-1}$  in 1991 to  $33\text{ kg ha}^{-1}$  in 2021. In contrast, the Elbe basin experienced a sharp decline in diffuse nitrogen inputs following the collapse of the eastern German and eastern European agricultural systems around 1990 (Gross 1996, Ehrhardt *et al* 2019). However, there has been no clear further reduction in nitrogen surplus in the Elbe since 1991 (figures 2(a) and (b)). Since 1992, only a minor decrease in  $\text{NO}_3\text{-N}$  export from diffuse sources has been observed in both basins, likely due to the accumulation of nitrogen in soils and the long travel times in groundwater that sustain high legacy exports (e.g. Nguyen *et al* 2022, Sarrazin *et al* 2022). In contrast, the reduction in point source inputs between 1991 and 2003 for both the Rhine and Elbe was driven by stricter regulations on wastewater treatment (EEC 1991, AbwV 1997) and improvements in treatment technologies. These reductions in

point source discharges led to an immediate decline in stream  $\text{NO}_3\text{-N}$  concentrations (figures 2(c) and (d)).

The revised urban waste water treatment directive (UWWTD; EU 2024) aims to further enhance nutrient removal efficiency, primarily targeting large WWTPs (with capacities over 100 000 population equivalents). This is expected to further reduce  $\text{NO}_3\text{-N}$  concentrations in the main Rhine and Elbe basins, which include many large WWTPs. However, a substantial number of subbasins (78 out of 146) that currently have high stream  $\text{NO}_3\text{-N}$  concentrations ( $>2\text{ mg l}^{-1}$ ; figure 4(d)) and notable point source contributions ( $>10\%$  during 2010–2021) do not have large WWTPs. Instead, these areas are served by smaller WWTPs that are subject to less stringent requirements and are not the primary focus of the revised UWWTD. This represents a significant challenge to the effectiveness of the new directive in improving water quality in these smaller rivers (subbasins). Our findings suggest that while stricter regulation of large point sources remains important, complementary measures addressing smaller treatment facilities and persistent diffuse pollution are needed to meet water quality goals basin-wide.

#### 4.4. Limitations and outlook

In this study, we focused exclusively on stream  $\text{NO}_3\text{-N}$ , while other nitrogen species were not considered due to limited observational data. We assumed that all nitrogen inputs (diffuse and point sources, representing total nitrogen) reach streams as  $\text{NO}_3\text{-N}$ . However, total nitrogen concentrations could have been higher than  $\text{NO}_3\text{-N}$  alone, especially before

2000. For example, observations from the Elbe River at Schnackenburg indicate that ammonium concentrations were comparable to  $\text{NO}_3\text{-N}$  concentrations before 2000 but became negligible thereafter (UBA 2025). Future studies could expand this work by incorporating multiple nitrogen species (e.g.  $\text{NH}_4\text{-N}$ , organic N) where data are available, to better allow long-term assessment of full nitrogen dynamics. Furthermore, future efforts could consider using a parameter regionalization approach, which may be particularly useful in basins with no stream  $\text{NO}_3\text{-N}$  observations or having only a few years of data that are insufficient for a detailed model calibration.

Additionally, while the model adequately captured long-term trends in stream  $\text{NO}_3\text{-N}$  concentrations in both the Rhine and Elbe basins (figures 2(c) and (d)), the year-to-year variability in  $\text{NO}_3\text{-N}$  concentrations is not adequately represented, especially in the Elbe basin. For example, observed data showed an enrichment pattern (high streamflow with high  $\text{NO}_3\text{-N}$  concentration), which could be because years with higher discharges had more outflows from the top soil layer with high nitrate concentrations. In the model, we could represent this by a time-variant SAS function with a stronger preference for young water in years with higher discharges. The simulated peak concentrations in both basins in years with lower discharges, in contrast to observed data, especially before 1991 when point source inputs were high (figures 3(a)–(d)), are mainly due to lower dilution of point sources. This suggests that instream N removal in years with lower discharge could be higher, but it was not accounted for in the model (e.g. Schulz *et al* 2023). The modeling approach employed here assumes a uniform stream nutrient removal fraction represented by one calibrated parameter. Although this assumption is consistent with previous studies (e.g. Van Meter *et al* 2017, Nguyen *et al* 2022), it does not account for the temporal variability in removal processes. Wachholz *et al* (2024) showed that uptake of dissolved inorganic nitrogen in the Elbe River was higher before 1990 and remained lower thereafter, likely due to shifts in nitrogen speciation (higher  $\text{NH}_4\text{-N}$  before 1990) and changing seasonality, as also described in Ebeling *et al* (2021). Moreover, the assumption of a fixed instream processing does not capture the positive dependency of instream denitrification and  $\text{NO}_3\text{-N}$  concentrations as described by Mulholland *et al* (2008) and Böhlke *et al* (2009). This dependency may have an effect on the simulation that excludes the impact of point sources.

Future modeling efforts could consider incorporating more process-based representations of stream nitrogen transformations and sub-annual variations to better capture nitrogen export and retention dynamics over time. Furthermore, the current setup of mQM does not explicitly consider direct surface runoff, which is a minimal part (below 2%) of the total runoff at the annual time scale (Wolters *et al*

2023). Future model developments at finer temporal and spatial resolutions (e.g. daily or sub-daily; gridded levels) should account for direct surface runoff, as it could be a dominant component of localized total runoff at certain extreme events (Tarasova *et al* 2020). Further analysis can also consider using instream stable isotopes along with  $\text{NO}_3\text{-N}$  data to better constrain the internal model parameterization of transit time distributions and (denitrification) reaction rates (Borriero *et al* 2024).

## 5. Conclusion

Understanding the role of diffuse and point source contributions to stream  $\text{NO}_3\text{-N}$  pollution is critical for effective management strategies to tackle water quality challenges. The Rhine and Elbe are two of the main Central European river basins discharging  $\text{NO}_3\text{-N}$  into the Wadden Sea, where eutrophication has been a major environmental issue for several decades. Here, we conducted the long-term assessment of point and diffuse source contributions to stream  $\text{NO}_3\text{-N}$  concentration of the Rhine and Elbe basins and their subbasins from 1950 to 2021.

In both the Rhine and Elbe basins, our results show a decline (more pronounced in the Elbe) in point source contributions from 1950 to 2000. Since 2000, point source contributions in the Rhine remained relatively stable at around 25% and fluctuated around 30% in the Elbe. The decline in stream  $\text{NO}_3\text{-N}$  concentration from 1991 to 2003 in the Rhine and Elbe basins was mainly attributed to the decline in point sources, as revealed through separating the effects of diffuse and point sources on stream  $\text{NO}_3\text{-N}$  concentrations. In contrast, the significant reduction of diffuse sources in both basins, especially in the Elbe, around 1991, only caused a minor reduction in stream  $\text{NO}_3\text{-N}$  concentrations due to legacy effects. Consequently, stream  $\text{NO}_3\text{-N}$  concentrations remained relatively stable at ( $\sim 2 \text{ mg l}^{-1}$ ) during the last two decades (2003–2021) of the simulation.

In the Rhine and Elbe subbasins, the median temporal changes in stream  $\text{NO}_3\text{-N}$  concentrations and point source contributions reflected the overall trends of their respective basins, although individual subbasins exhibited diverse patterns. In subbasins with relatively high stream  $\text{NO}_3\text{-N}$  concentrations during 2010–2021, point source contribution was generally low (median value of 30%) and diffuse source contribution was high. These are subbasins within the study domain with higher levels of human impact in terms of higher agricultural, urban, and industrial land cover fractions and the number of urban WWTPs.

Our findings suggest that point source management alone will not be sufficient to reduce  $\text{NO}_3\text{-N}$  concentrations to a good ecological status ( $< 2 \text{ mg l}^{-1}$ ; Vigiak *et al* 2023) in most subbasins. However, in about 27% of subbasins (figures 5(a) and

(b)), point source management alone could achieve this target, assuming complete elimination of point source loads. This underscores the need for both diffuse and point source management, in a spatially targeted manner, to achieve a good ecological status not only at the regional level but also at the local level.

## Acknowledgment

This work was partially funded by the EU project NAPSEA (HORIZON-CSA, proposal 101060418).

## Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI: <https://doi.org/10.4211/hs.aca9f19613894b6e8cbd53d99424b1bf> (Nguyen *et al* 2025). The model input data are provided in the QUADICA v2 database by Ebeling *et al* (2026). Further information can be available from the corresponding authors.

## Conflict of interest

The authors declare that there are no conflicts of interest in the submission of this work.

## Ethics statement

The authors declare that there are no ethical issues arising from this work.

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