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# Long-term data reflect nitrogen pollution in Estonian rivers

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## ABSTRACT

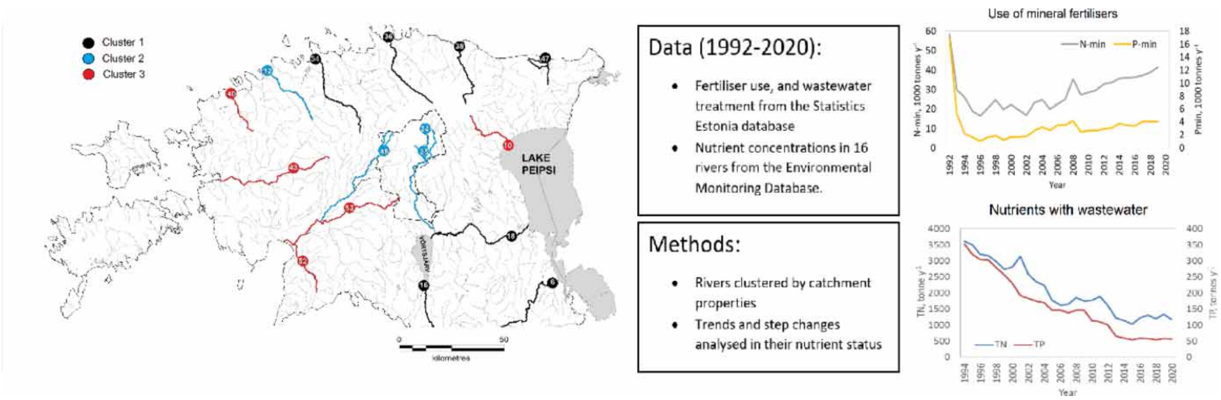
We analysed long-term (1992–2020) changes in fertiliser use, wastewater treatment, and river water nutrient status in Estonia (N-E Europe) in the context of changing socio-economic situations and legislation. We hypothesised that improved regulation of fertiliser usage and wastewater treatment are reflected as declining riverine nutrient concentrations, with the largest relative improvements occurring in catchments with initially high proportions of point source loading. We analysed nutrient dynamics in 16 rivers differing by catchment land use, population and livestock densities. Data on fertiliser use and wastewater treatment originated from the Statistics Estonia database, and riverine nutrient concentrations from the State Environmental Monitoring Database. We clustered the rivers by their catchment properties and analysed trends in their nutrient status. Point source nutrient loading reductions explained most of the decline in riverine nutrient concentrations, whereas application of mineral fertilisers has increased, hindering efforts to reach water quality and nutrient load targets set by the EU Water Framework Directive and the Baltic Sea Action Plan. Highest nitrogen concentrations and strongest increasing trends were found in rivers within the Nitrate Vulnerable Zone, indicating violation of the EU Nitrates Directive. To comply with these directives, resource managers must address non-point source nutrient loading from river watersheds.

**Key words:** manure, mineral fertilisers, organic fertilisers, point and non-point sources, wastewater treatment

## HIGHLIGHTS

- Drop in point source loading explained the decline in riverine nitrogen (N) and phosphorus (P) since 1994.
- Fertiliser and wastewater management measures failed short to meet the water quality and nutrient load targets set by the EU Water Framework Directive.
- Highest N concentrations and strongest increasing trends were found in rivers within the nitrate vulnerable zone violating the EU Nitrates Directive.

## GRAPHICAL ABSTRACT



## INTRODUCTION

Anthropogenic nutrient loading in aquatic systems is a combination of discharges from point and non-point sources. Point sources, such as industries and wastewater treatment plants, are easier to identify and implement management approaches to reduce nutrient losses. Runoff from agricultural areas is the major cause of non-point source nutrient pollution worldwide and has long been considered as a major driver of eutrophication in downstream rivers and lakes (Barcala *et al.* 2020; Xia *et al.* 2020). In Europe, reduced loading from point sources has shifted agriculture into the key role in nutrient losses from rural areas to surface waters over recent decades (European Environment Agency 2005).

By 2050, the global N pollution level is projected to rise by 50% of that in 2010, with agriculture accounting for 60% of this increase; nearly half of N fertiliser applied to fields is lost via gaseous emissions or discharges to water bodies (Martínez-Dalmau *et al.* 2021). To alleviate eutrophication, it is often necessary to reduce the inputs of both N and P (Paerl *et al.* 2016).

Different dynamics in N and P concentrations have been observed in Europe. For example, P concentrations increased until 2004 in the Vltava River, Slapy reservoir (Czech Republic), then decreased, while N concentration increased throughout the period (1960–2019; Kopáček *et al.* 2021). N concentrations and losses from agricultural catchments decreased in Denmark and Sweden from 1988 to 2011, likely due to implementation of agro-environmental measures (Stålnacke *et al.* 2014). However, increasing N concentrations and catchment losses were detected in the Baltic countries, coinciding with intensification of agriculture in the Baltic States (Stålnacke *et al.* 2014). These results indicated that targeted strategies towards reducing N losses from agricultural land would improve surface water quality.

In the Baltic Sea catchment, a major part of the nutrient load originates from diffuse sources, mainly agriculture. Point sources now comprise 4–5% of nutrient loads to the Baltic Sea and has decreased substantially in recent decades (HELCOM 2018). Riverine discharges contribute 70.3% of total N (TN) and 94.8% of total P (TP) inputs to the Baltic Sea (HELCOM 2018).

Major socio-economic changes took place in Estonia in the early 1990s. The re-establishment of Estonia's independence closed a large part of the Russian markets to Estonian production, and agricultural production deceased. At the same time, lands expropriated in the 1940s were returned to their former owners and their descendants. As many of them lacked the skills and opportunities to cultivate the land, large areas of former agricultural land lied fallow, and few farmers could afford to buy fertilisers. These changes led to smaller meadows and pastures along riverbanks and lake shores being overgrown with reeds and brushwood. Estonia implemented the Water Act in 1994 and adopted European directives to replace the soviet-era laws (Supplementary material, Table 1). The spreading of fertilisers on frozen fields and snow was banned in 2001. After joining the EU, water consumption taxes increased, which reduced water extraction and the amount of wastewater (Supplementary material, Figure 3). With the support of EU funds, new water treatment plants were constructed, and existing plants were reconstructed. All of these measures were expected to improve water quality in the rivers.

After the economic downturn in the 1990 and 2000s, agriculture recovered in Estonia due to EU subsidies (although smallest within the EU). These investments encouraged agricultural redevelopment, and fertilisers were again affordable for farmers. Concentration of agriculture by large landowners enabled new agronomic techniques (e.g., catch crops, granular fertilisers) and smart dosing to avoid over-fertilisation.

Here, we analyse trends in long-term changes in wastewater treatment, N and P fertiliser use, and river water nutrient status in the context of changing legislation and socio-economic status. We hypothesise that (1) more precise regulation of fertiliser use and improved wastewater treatment have led to declining TN and TP concentrations, and (2) the largest water quality improvements occurred in catchments with a high proportion of point source pollution.

## STUDY AREA

Estonia (45,339 km$^2$) has approximately 2,000 km$^2$ of surface waters (Figure 1). Slightly more than half of the land is coniferous and mixed forests, with agricultural land (23%) and wetlands (22%) comprising much of the rest. In central Estonia, large limestone areas are characterised by karst phenomena, where the thickness of glacial drifts can be insignificant, and the groundwater is unprotected. The karst area largely overlaps with the nitrate vulnerable zone (NVZ), which includes areas designated as being at risk from agricultural nitrate pollution (Figure 1). The NVZ in Estonia was determined considering soil and ground conditions, ground and surface water vulnerability, and intensity of agriculture (Water Act 2019) and covers 3,250 km$^2$, or 7.5% of the land area.

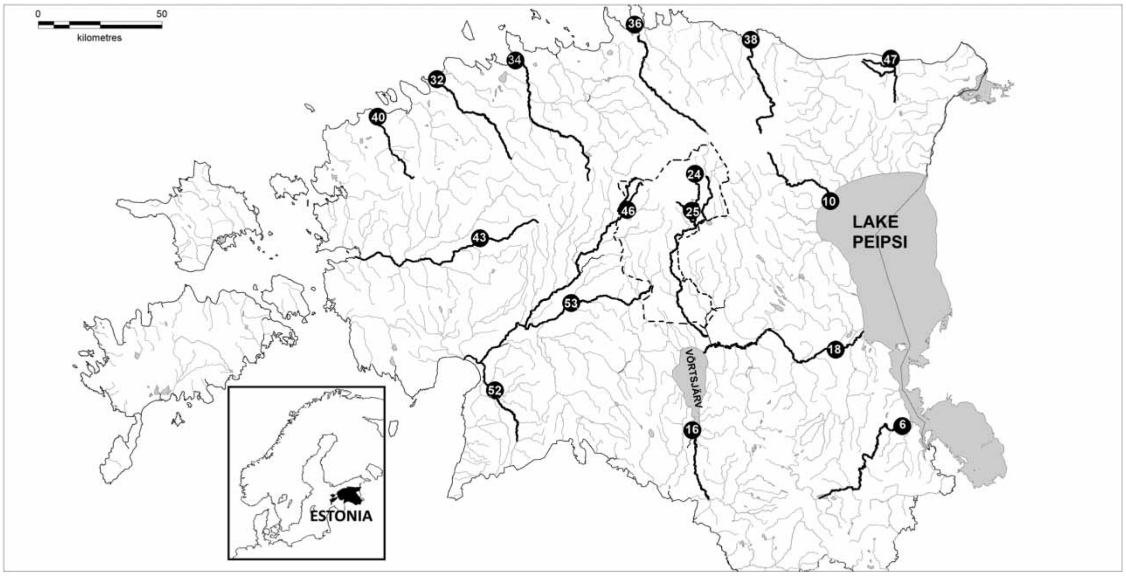



**Figure 1** | Location map of the study area and water quality monitoring stations in the rivers. Site numbers according to the Environmental Monitoring Information System (KESE 2021) ($n = 16$). Nitrate vulnerable zone surrounded with dashed line.

Long-term (1981–2010) annual average air temperature is between 4.9 and 7.1 °C (Estonian Weather Service 2021). Snow cover lasts from 61 to 130 days, with high spatial variation (Tarand *et al.* 2013; Viru & Jaagus 2020). Estonia is sparsely populated, with 30.6 inhabitants per km$^2$ (2021).

There are approximately 31,000 km of natural running waters, and their density is 0.72 km km$^{-2}$. The total annual runoff is about 12 km$^3$, accounting for about 40% of annual precipitation (Timm *et al.* 2019).

## Rivers

TN and TP concentrations were analysed in 16 rivers, which were selected based on a sampling frequency of at least four times per year (Table 1; Figure 1). Six rivers flow directly into the Baltic Sea, three into Lake Peipsi, one into Lake Võrtsjärv, and the other six are tributaries of different rivers. Three sampling sites (Nos 24, 25, and 46) were situated in the NVZ. Details on catchment land use of these sites are provided in Iital *et al.* (2010). According to the EU Water Framework Directive (Council Directive 2000/60/EC 2000, WFD) typology in Estonia, based on catchment size and water colour (humic matter content measured as permanganate oxygen demand), the selected sites belonged to four different WFD types (Table 1).

TN and TP concentrations in river water were obtained from the Environmental Monitoring Information System (KESE 2021). Historical data on daily flow rates of rivers for the period 1992–2020 were obtained from the Estonian Weather Service (2021).

## Data collection

Data on agricultural activities were obtained from the Statistics Estonia database (2021). Data on N and P in livestock manure (PM0646: *Nitrogen and phosphorus in livestock manure*) were available only for 6 years (2014–2019) and are not directly comparable with previous data from 1992 to 2011 (KK19: *Nitrogen and phosphorus brought into the soil with organic fertilisers*) because the methodology for measurement changed. No data were available for 2012 and 2013 (Supplementary material, Figure 1). Data for the numbers of livestock and poultry were available from 1992 to 2020 (PM09: *Livestock and poultry*). In accordance with the Ministry of Rural Affairs Regulation No. 71 (2014; *Estimated nutrient values for different types of manure, methodology for calculating manure storage capacity and conversion factors for livestock units*; Riigiteataja I: 16.07.2014, 8; Appendix 9, *Coefficients for converting animals into livestock units based on animal faeces*), the amount of manure produced by livestock and poultry was calculated. The number of livestock units (LSUs) for each year back to 1992 was also calculated (Supplementary material, Figure 2). Consistent with the available 6 years of data, one LSU produced an average of 120 kg N and 23 kg P per year. These numbers were multiplied by the LSU numbers for the years 1992–2013 to generate a homogenous time series for N and P applications via manure. Data on application of N and P mineral fertilisers,

**Table 1** | Sampling sites in rivers sorted by catchment size

| River/sampling site | Site number | WFD type | Catchment type cluster number | Catchment area, km² | Number of water samples | Population density, in h/km² | Forest, % | Wetland, % | Arable, % | Pasture and grassland, % |
|---|---|---|---|---|---|---|---|---|---|---|
| Emajõgi Kavastu | 18 | 3C | 1 | 8,539 | 348 | 21.9 | 42 | 11 | 19 | 23 |
| Väike-Emajõgi mouth | 16 | 3C | 1 | 1,270 | 256 | 24.1 | 46 | 6 | 13 | 32 |
| Võhandu mouth | 6 | 3C | 1 | 1,144 | 347 | 29.8 | 42 | 7 | 10 | 37 |
| Pirita mouth | 34 | 2C | 1 | 794 | 287 | 83.4 | 44 | 15 | 16 | 21 |
| Keila mouth | 32 | 2C | 3 | 682 | 345 | 34.1 | 38 | 12 | 24 | 22 |
| Reiu Lähkma | 52 | 2B | 2 | 548 | 177 | 12.2 | 62 | 17 | 9 | 10 |
| Kunda mouth | 38 | 2C | 1 | 528 | 344 | 15.5 | 49 | 13 | 17 | 20 |
| Vihterpalu mouth | 40 | 2B | 2 | 474 | 233 | 3 | 57 | 26 | 8 | 9 |
| Valgejõgi mouth | 36 | 2C | 1 | 453 | 345 | 27.3 | 47 | 21 | 13 | 16 |
| Avijõgi mouth | 10 | 2C | 2 | 366 | 344 | 8.2 | 58 | 14 | 13 | 14 |
| Pühajõgi mouth | 47 | 2C | 1 | 196 | 348 | 183.1 | 48 | 10 | 19 | 13 |
| Saarjõgi Kaansoo | 53 | 2B | 2 | 191 | 145 | 2.9 | 61 | 17 | 13 | 9 |
| Velise Valgu | 43 | 2B | 2 | 135 | 160 | 2.1 | 66 | 13 | 11 | 10 |
| Vodja Vodja[a] | 46 | 1C | 3 | 52 | 151 | 10.4 | 22 | 19 | 35 | 24 |
| Preedi Varangu[a] | 24 | 1C | 3 | 34.8 | 166 | 8.4 | 35 | 8 | 34 | 22 |
| Oostriku Oostriku[a] | 25 | 1C | 3 | 29.7 | 143 | 12.1 | 23 | 4 | 53 | 17 |

*Note:* Site numbers according to the Environmental Monitoring Information System (KESE 2021). Coding of Water Framework Directive (WFD) type names: 1 – catchment area $<$ 100 km²; 2 – catchment area 100–1,000 km²; 3 – catchment area $>$ 1,000 km²; B – brown waters, chemical oxygen demand $>$ 25 mg O/l; C – clear waters, chemical oxygen demand $<$ 25 mg O/l.
[a]Nitrate vulnerable zone (NVZ).

use of water, and N and P discharged with waste waters for 1992–2020 were obtained from the Statistics Estonia database (PM065: *Use of mineral fertilisers for the production;* KK47: *Water use by field of water use;* KK23: *Waste water treatment*; KK25: *Pollution load to surface water bodies*).

### Data analysis

River sites differed by catchment characteristics (area, population density, percentages of land use categories; see Table 1), so clustering was used to typify the catchments. In contrast to the WFD typology, which is based on natural features (catchment size, water colour), our clustering approach considered long-term human impacts reflected in population density and land use categories in the catchment. For cluster analysis, catchment characteristics for all 16 river sites were standardised by subtracting the mean of each variable and dividing the residuals by the standard deviation. Using the k-means clustering procedure in STATISTICA 64 (Version 13, TIBCO Software Inc. 2017), the sites were grouped into three clusters. The number of clusters was optimised using the Elbow method (Kodinariya & Makwana 2013). K-mean clustering was selected because it shows the role of different factors in defining the clusters. Differences between clusters by individual factors were determined using the Fischer LSD test at 95% confidence.

Trends in river time series were analysed with the Mann–Kendall non-parametric test, and step changes in means were evaluated using the Distribution-Free CUSUM non-parametric test. A step change identifies an inflection point in a time series characterised by the largest difference between the means in two parts of the record. Both tests were run using trend/change detection software (TREND) provided in the e-Water Toolkit (https://ewater.org.au/products/ewater-toolkit/, accessed 20 December 2021).

## RESULTS AND DISCUSSION

### Changes in agriculture and fertiliser application

Both the extension and intensity of Estonian agriculture declined after re-establishing its sovereignty in 1992 (Supplementary material, Table 2; Figure 2). The land area fertilised with mineral and organic fertilisers, as well as the use of fertilisers per

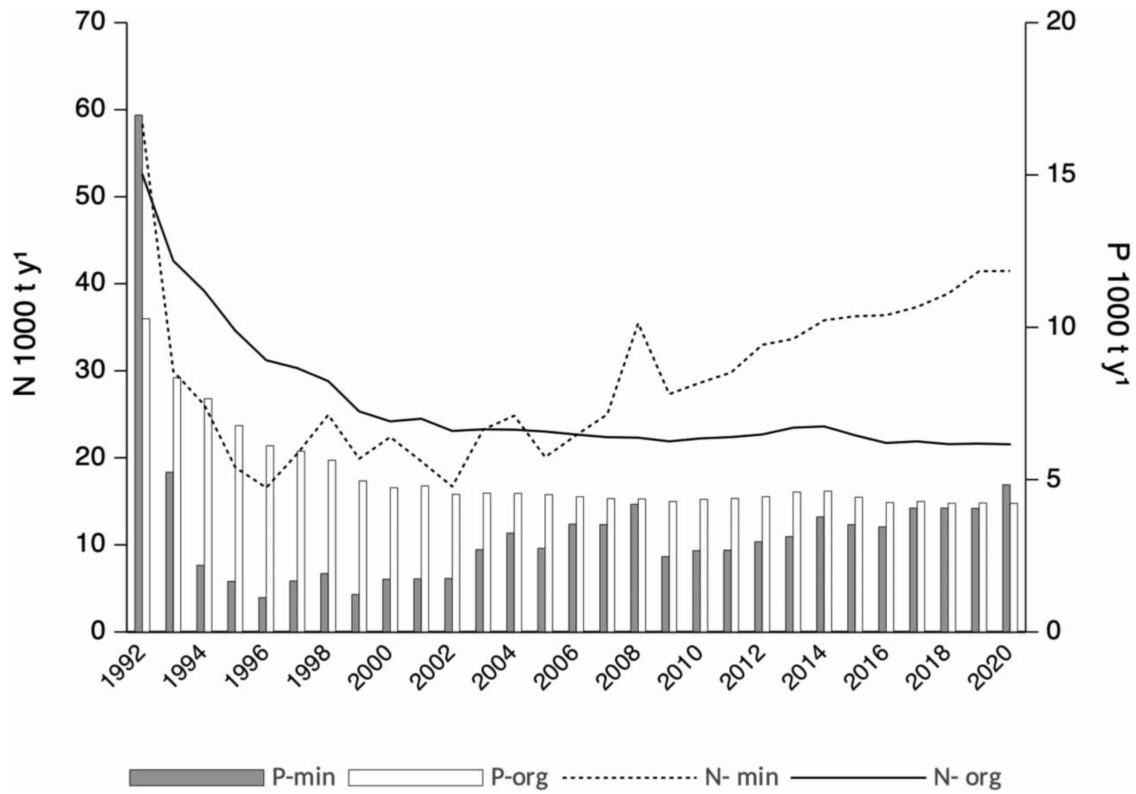



**Figure 2** | Use of N and P in mineral (N-min and P-min) and organic fertilisers (N-org and P-org) in Estonia (1992–2020).

hectare, decreased more than two-fold. The application of mineral fertilisers decreased initially, reaching a minimum in 1996, because farmers could not afford agrochemicals (Lehtsaar & Jullinen 1998). This situation promoted the development of organic farming (Zobena 1998), which is still popular in the Baltic countries and exemplifies how societal practices can be facilitated by economic situations. Altogether, the use of fertilisers decreased more than 3.5-fold. Numbers of livestock also decreased, and the amount of manure declined accordingly (Supplementary material, Figure 1), resulting in less N and P being applied to fields. This trend reversed in the early 2000s, and more than 20,000 t of N and 4,000 t of P are applied with organic fertilisers annually (Figure 2; Supplementary material, Table 2).

After Estonia joined the EU in 2004, the amount of arable land and utilised agricultural area, as well as the use of mineral fertilisers, increased with the aid of agricultural subsidies. There was a brief decrease in fertiliser usage after the global economic recession in 2008. By 2020, mineral N and P fertiliser applications had increased by 2.5 and 3.7 times, respectively, over 1996 values. In recent years, mineral N applications (around 40,000 t $year^{-1}$) exceeded that of organic N. Mineral and organic P were applied almost equally at around 4,000 t $year^{-1}$ in recent years, but mineral P fertiliser applications have doubled since 2009 (Figure 2). In 2019, the amount of LSU in Estonia was 180,000 (Supplementary material, Figure 2), the area fertilised with mineral and/or organic fertilisers was 6,198 $km^2$, and 63,119 t of N and 8,269 t of P were applied on agricultural land (Supplementary material, Table 2).

## Nutrient discharges with wastewater

From 1994 to 2020, TN and TP discharges decreased three- and six-fold, respectively, with a step change ($p < 0.01$) in 2004 (Figure 3) due to a rapid increase in wastewater treatment plant construction in the early 2000s (Supplementary material, Figure 4). Since 2013, N and P discharges have stabilised at 1,200–1,300 and 60 t $year^{-1}$, respectively. A larger decrease in P compared to N in wastewater has led to a distinct increase in the N/P ratio (Figure 3). Although the total water usage, as well as industrial and domestic water use, decreased by half during the 1990s (Supplementary material, Figure 3), the volume of wastewater did not decrease substantially during the study period (Figure 3). Water usage in Estonia is primarily cooling water in thermal power plants, followed by industrial and domestic uses (Supplementary material, Figure 3).

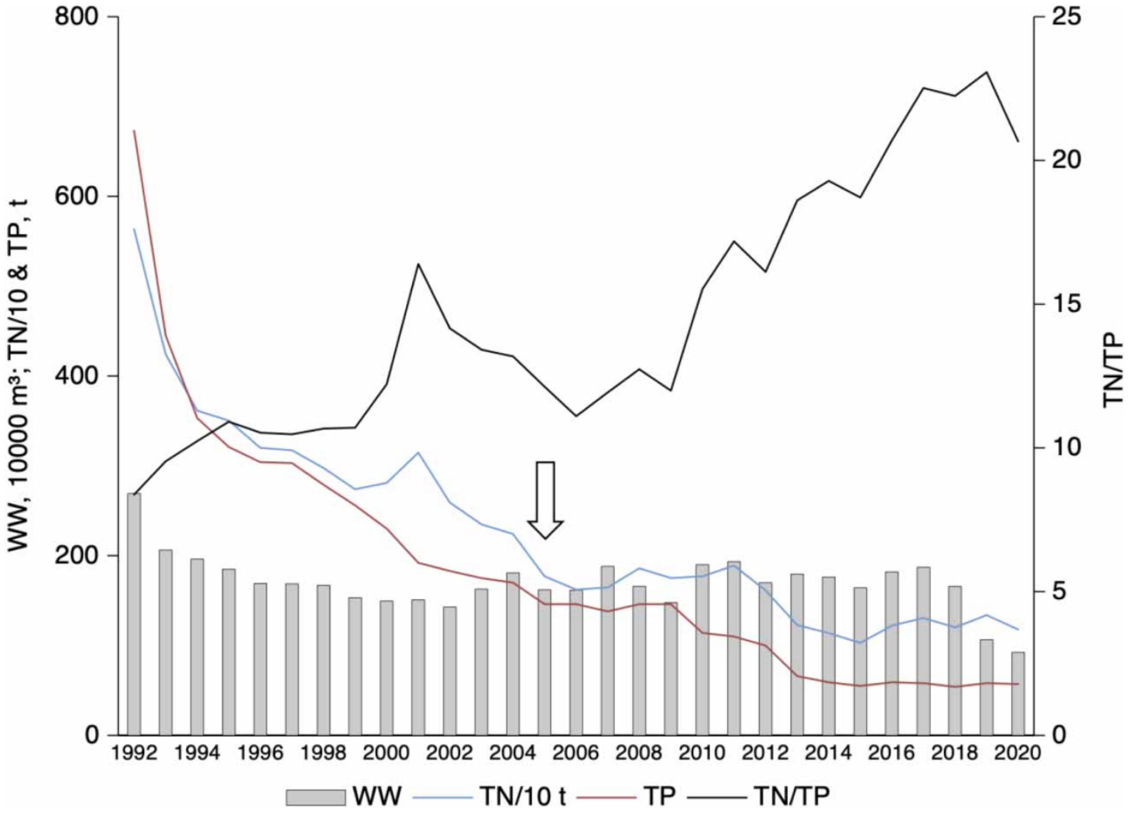



**Figure 3** | Nutrient load to Estonian surface water bodies with discharged wastewater (WW); arrow indicates the step change from 2004 to 2005 ($p < 0.01$) occurring in both nutrient series.

### Natural vs. anthropogenic river types

The WFD-compliant river typology in Estonia in mainly based on two natural characteristics: catchment size and permanganate oxygen demand as a proxy for the content of humic substances or water colour (Pinnaveekogumite 2020). However, water quality and nutrient dynamics are largely controlled by anthropogenic impacts, such as long-term population density and land use in the catchment. For the study period, both parameters can be considered static and used for typifying the rivers. We entered these catchment parameters into cluster analysis to categorise the selected river sections by their degree of anthropogenic influence (although catchment size was also included). Three river clusters were identified, differing significantly by percentages of forest and non-cultivated agricultural land in their catchment areas (Figure 4). Additional variables, such as area, population density, percentages of wetlands and cultivated agricultural lands contributed to distinguishing clusters from each other although not all differences were significant.

Cluster 1 consisted of seven WFD type 2 rivers characterised by low population density, highest percentages of wetlands and forests, and lowest percentages of agricultural lands in their catchments (Table 1). All four humic water rivers were included in Cluster 1. Cluster 2 consisted of five rivers, including all three WFD type 3 rivers. These rivers exhibited highest average population density and percentage of pasture and grasslands, and lowest wetland percentage, in their catchments, whereas proportions of forests and cultivated lands were average. Cluster 3 included the three WFD type 1 rivers of the NVZ, plus R. Keila. This cluster had the highest proportion of cultivated lands and the lowest proportion of forests in their catchments (Figure 4).

### Changes in river water quality

Mean concentrations of TN and TP, and the TN/TP ratio, differed ($p < 0.01$) between the clusters (Table 2). The least impacted rivers (Cluster 1) had the lowest TP, rivers in densely populated watersheds (Cluster 2) had the lowest TN and

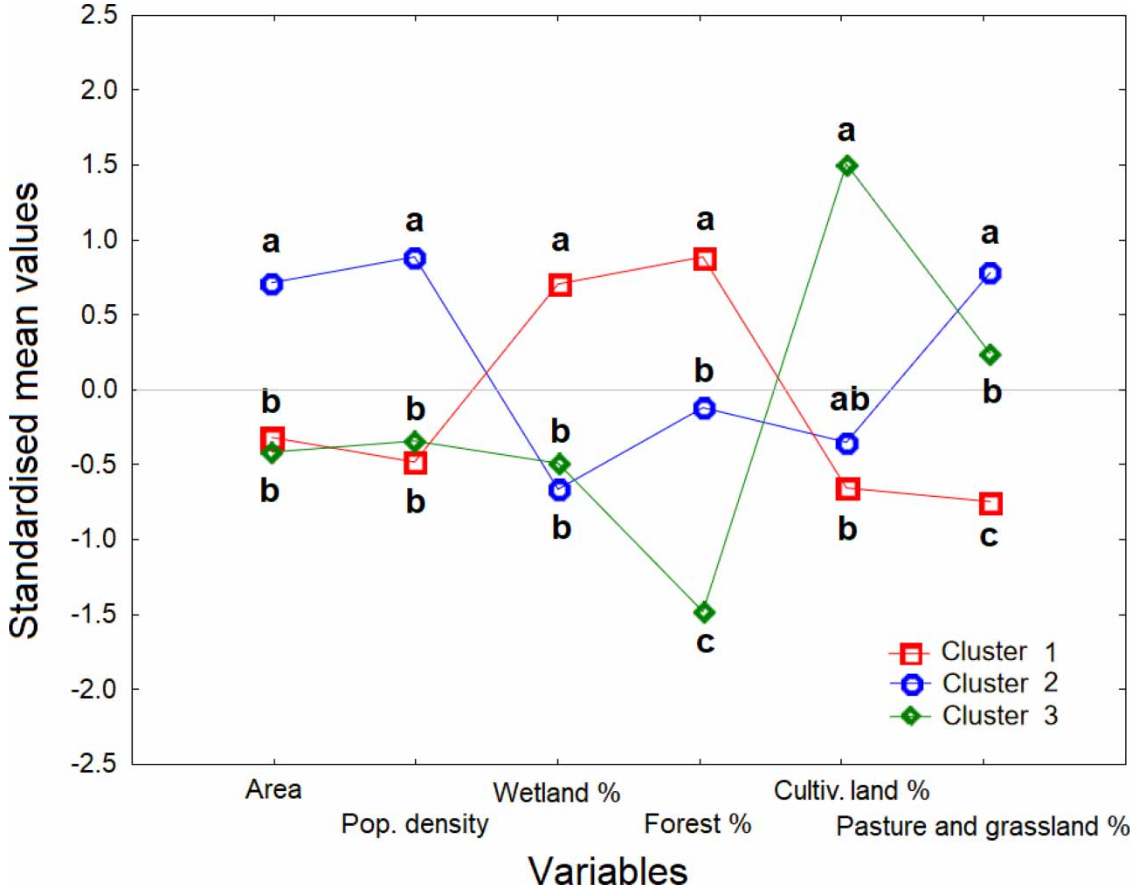



**Figure 4** | Standardised mean values of river characteristics used for clustering. For each variable, different letters denote differences between groups determined using the Fischer LSD test at 95% confidence. Variables are in arbitrary order and connected with lines to better visualise the contrasting patterns.

**Table 2** | Concentrations of TN, TP, and the TN/TP ratio in three river clusters (mean $\pm$ standard error)

| Cluster | TN (mg $L^{-1}$) | TP ($\mu$g $L^{-1}$) | TN/TP |
|---|---|---|---|
| 1 | 2.18 $\pm$ 0.03 | 36 $\pm$ 1 | 83 $\pm$ 2 |
| 2 | 2.01 $\pm$ 0.03 | 98 $\pm$ 3 | 33 $\pm$ 1 |
| 3 | 3.50 $\pm$ 0.05 | 54 $\pm$ 2 | 199 $\pm$ 12 |

TN/TP ratio, but the highest TP, whereas rivers in the most agricultural watersheds (Cluster 3), including the NVZ rivers, had the highest TN and TN/TP ratio.

These findings are in line with the different sources and pathways of these elements in the catchments. N and P are synanthropic elements, as their concentrations in surface waters tend to increase with population densities in the catchment (Chen *et al.* 2016). The capacity of agricultural soils to retain excess N is low, and leaching losses from common grain-production systems typically range from 10 to 30% of total N inputs (Meisinger & Delgado 2002). P forms complexes with iron, aluminium, calcium, and organic matter and is well-bound to a variety of soil types. Leaching of P to surface waters increases only after exceeding soil saturation levels (Djodjic *et al.* 2004). P loading may also be associated with discharges of untreated wastewaters (Edwards & Withers 2007) and, hence, can be considerably reduced by wastewater treatment.

TP concentrations declined in all river clusters from 1992 to 2020, especially in Cluster 2, where TN concentrations also declined (Figure 5). In the other two clusters, TN increased marginally (Cluster 1, $p = 0.152$; Cluster 3, $p = 0.052$), coinciding

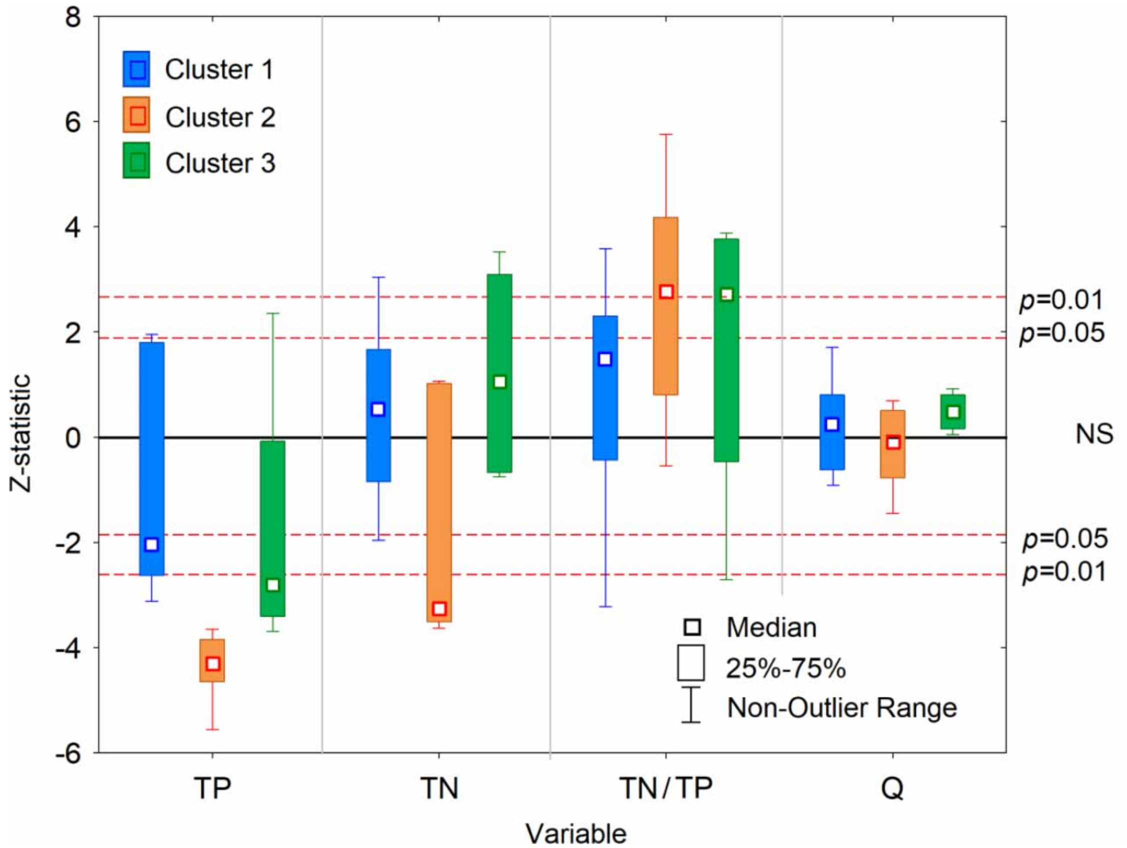



**Figure 5** | Mann–Kendall trends in river nutrient concentrations (TP and TN), nutrient ratio (N/P), and discharge (Q) from 1992 to 2020. Dashed red lines denote strength of the trends, with the NS range indicating no trend. Please refer to the online version of this paper to see this figure in colour: http://dx.doi.org/10.2166/nh.2022.057.

with increased mineral N fertiliser use. As a result, TN/TP ratio increased in most cases, and most strongly in Clusters 2 and 3. River discharges showed no trend in any of the clusters.

Step changes of nutrient concentrations occurred in several rivers (Figure 6). Most of the downward step changes in both TN and TP occurred in Cluster 2 between 2003 and 2010, whereas upward step changes were frequently observed in Cluster 3. No significant step change was found for the discharge of any river (not shown).

Observed changes in river water quality reflect sensible outcomes of measures applied in the watersheds. Some improvements in river water quality were evident, with major changes observed in rivers draining catchments with high population and livestock densities (Cluster 2). Livestock densities were indicated by the percentage of non-cultivated agricultural lands (i.e., pastures and grasslands). Both high population and livestock densities suggest a major role for point sources, such as runoff from municipal settlements and cattle farms (Kiedrzyńska *et al.* 2014), which are theoretically easier to mitigate (Bowes *et al.* 2008). In this cluster, TP declined, but TN step changes also occurred in the mid-2000s, coinciding with construction of wastewater treatment plants (Supplementary material, Figure 4).

The use of organic fertilisers did not change remarkably during the study period, while that of mineral N fertilisers has increased since the late 2000s (Figure 2). Simultaneously, TN concentrations increased in river water (Figure 6), especially in Cluster 3, with the highest proportion of cultivated lands and lowest proportion of forests in their catchments, including rivers in the NVZ (Figure 4).

Despite these efforts in fertiliser and wastewater management, the main objective of the WFD, to achieve good water quality status by 2015, was not met (Tsakiris 2015). HELCOM (2021) pointed out that nutrient loading, dominated by mineral fertilisers inputs, still exceeds the goals of the Baltic Sea Action Plan, and a further load reduction has been agreed upon (ICES Advice 2019). From 2000 to 2010, the use of mineral fertilisers around the Baltic Sea has increased by 68% for N

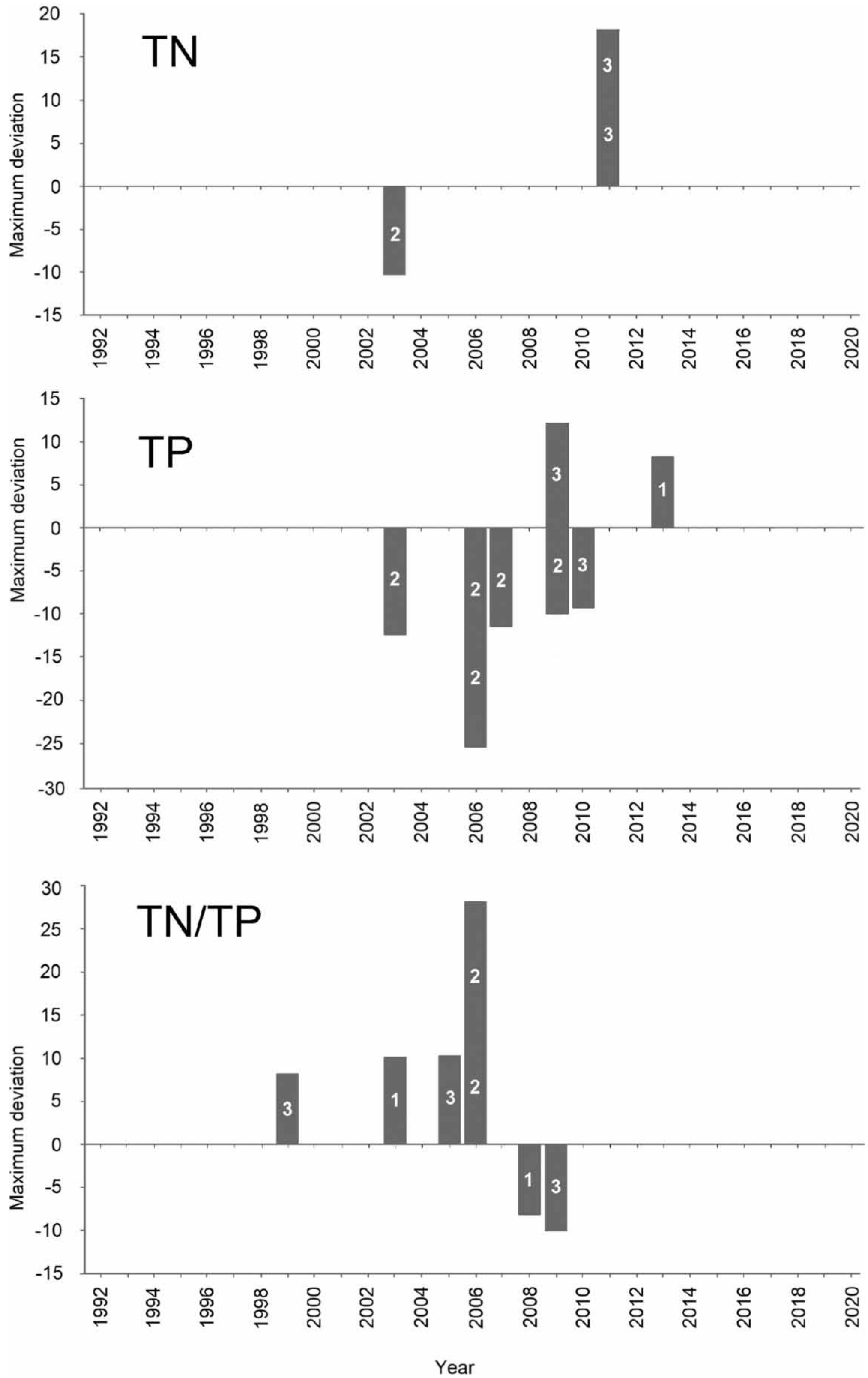



**Figure 6** | Step changes ($p < 0.05$) in nutrient concentrations and their ratios in rivers from 1992 to 2020. Number on the bars denote cluster numbers of the individual rivers, and two numbers on one bar mean two rivers. Height of the bar denotes strength of the change.

and by 70% for P (Hong *et al.* 2017), despite efforts of conservationists and governments to reduce impacts on water resources. No decreasing trends have been observed, regardless of new agro-requirements, such as keeping field books, fertilisation plans, compliance with fertilisation restrictions in the NVZ, requirement for winter vegetation cover, etc.

In a study of 53 Estonian rivers over 1984–2006, Iital *et al.* (2010) found declining trends for TN in 18 rivers and for TP in 13 rivers. Increasing trends occurred in two rivers for TN and six rivers for TP. Annual N losses from agricultural land ranged from 10 to 40 kg $ha^{-1}$ (Iital *et al.* 2014), but variability was greater for P, ranging from 0.09 to 7.5 kg $ha^{-1}$ (Pengerud *et al.* 2015). Bechmann *et al.* (2014) studied trends in N balance and its impact on N concentrations in streams. In four of 14 long-term time series in streams, Bechmann *et al.* (2014) found positive relationships between annual N balance in the catchment and TN concentrations.

The above studies apply to mineral fertilisers, but the role of organic fertilisers has not been studied as intensively (see Supplementary material, background information). However, to reverse large-scale eutrophication of water bodies, both N and P from mineral and organic fertilisers and wastewaters need to be reduced (Haque 2021; Löw *et al.* 2021; Sabo *et al.* 2021). Nutrient losses could also be reduced by partial replacement of mineral fertilisers with effective manure application (Hong *et al.* 2017). In Estonia, the Water Act allows 175 kg N and 25 kg P to be applied per hectare; for manure, the resulting N/P ratio is 7. The actual N/P ratio is 5, so it is recommended to calculate manure applications according to P requirements to avoid P over-application. In practice, the calculations are often based on N. P over-application may be acceptable in some cases, but it should be strictly monitored to avoid P losses to surface waters. According to the European Nitrate Directive (Council Directive 91/676/EEC 1991), the maximum allowed amount of N in mineral fertilisers in the NVZ is 100 kg per hectare of the area under cultivation, and the use of organic fertilisers is restricted by allowing not more than 1.5 LSU per hectare of the area under cultivation. The threshold $NO_3$ concentration in ground and surface water is 50 mg $L^{-1}$.

## CONCLUSIONS

The collapse of soviet-era agriculture in Estonia in the early 1990s contributed to declines in N and P concentrations in rivers. Adoption of EU legislation on water use and protection created a good basis for further improvement of river water quality. Success was achieved in reducing point source pollution from large farms and municipalities, supporting our hypothesis that regulatory measures would improve water quality. However, despite more strict governance on fertiliser use, the decline of nutrients in river water ceased after Estonia joined the EU in 2004, as the financial means to buy fertilisers improved. From 1996 to 2020, the use of mineral N and P fertilisers has increased in Estonia by factors of 2.5 and 3.7, respectively. A particular concern is increased N concentrations in rivers within the NVZ. Among three river clusters, changes were minimal in Cluster 1, representing rivers least impacted by humans, but median TP declined in this cluster. The largest changes were observed in rivers draining catchments with high population and livestock densities; i.e., with a high proportion of point source pollution, thus supporting our second hypothesis. The mitigation of point source pollution resulted in declines in both N and P concentrations, whereas step changes coincided with or followed construction and upgrades of new and existing wastewater treatment plants, respectively, in the early 2000s. We found highest N concentrations and strongest increasing trends in Cluster 3, comprising rivers draining the most agricultural catchments, including those within the NVZ. These results indicate that the rules set by the EU Nitrates Directive (Council Directive 91/676/EEC 1991) to protect surface and groundwater quality in karst areas have been insufficient to reduce nutrient concentrations in surface waters within these watersheds.

## ACKNOWLEDGEMENTS

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## AUTHOR CONTRIBUTIONS

T.N. and P.N. conceptualised the study; S.V. and P.N. studied methodology and did formal analysis; S.V. did investigation and data curation, and wrote original draft; T.N., P.N., M.M. and M.T. wrote, reviewed and edited the article; S.V., M.T. and P.N.

visualised the study; T.N. was involved in project administration. All authors have read and agreed to the published version of the manuscript.

## DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

## CONFLICT OF INTEREST

The authors declare there is no conflict.

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