Nitrogen legacies in agricultural catchments

An analysis of 170 years of nitrogen legacies data in four Swedish agricultural catchments

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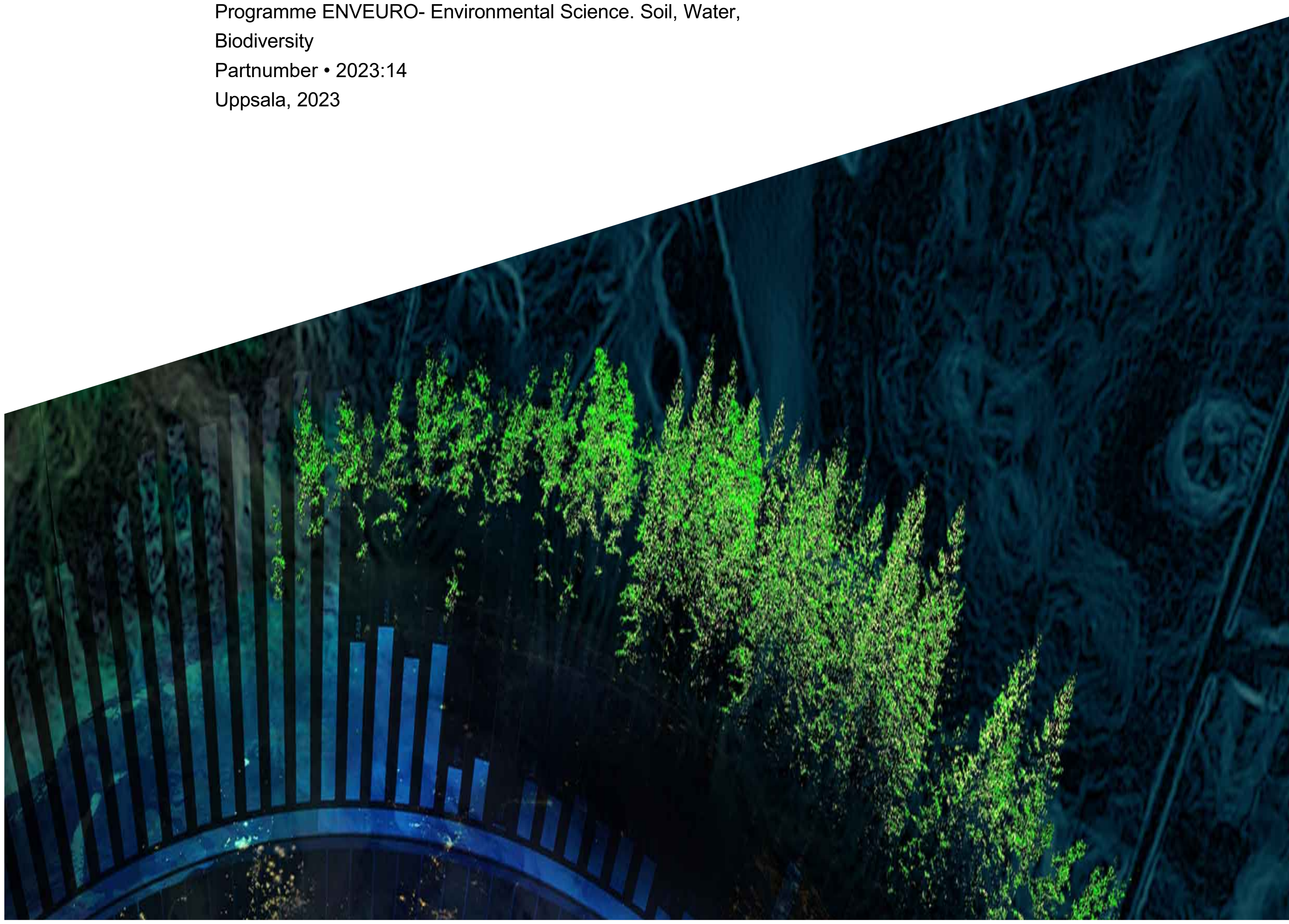

Nitrogen legacies in agricultural catchments.

An analysis of 170 years of nitrogen legacies data in four Swedish agricultural catchments

Nährstoff Anreicherungen in landwirtschaftlichen Einzugsgebieten.

Eine Analyse der Daten von 170 Jahren Stickstoff legacies in vier schwedischen landwirtschaftlichen Einzugsgebieten

Laura-Ainhoa Prischl

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Abstract

Despite the enforcement of legislation and application of measures, nitrogen (N) levels continue to be high in streams. One of the reasons for the low N reduction, is the legacy accumulation of N. Here we evaluate 170 years of N legacies in four Swedish agricultural catchments, by reconstructing land use trajectories, N mass balances, and N surplus over time. It was expected that a high percentage of agricultural land and high N input would lead to a higher N surplus and consequently higher total nitrogen loadings (TN) in the stream. N surplus was expected to create a delayed impact on total nitrogen loading, leading to a hysteresis effect. The long-term analysis showed that in catchments with a high percentage of agricultural land, mineral fertilizer was the main driver of N surplus. A higher N surplus led to high TN loadings in the stream. A positive correlation between N surplus and TN loadings was seen when a one year-shift of TN loadings was applied. No hysteresis effect was seen in any of the catchments. These results suggest that N surplus continues to be high in all catchments, therefore measures focusing on reducing the main drivers of N accumulation are needed. To identify legacies further research considering a longer period of measured TN loadings and variables such as tile drainage density and groundwater travel time is needed.

Keywords: Nitrogen legacies, nitrogen mass balance, nitrogen surplus, land use trajectories, hysteresis effect, water quality, agriculture

Abstract German

Trotz der Durchsetzung von Gesetzen und der Anwendung von Maßnahmen sind die Stickstoffwerte in den Flüssen nach wie vor hoch. Einer der Gründe für diese geringe Reduzierung ist die legacy Anreicherung von Stickstoff. In dieser Studie wurden 170 Jahre Landnutzungs-Entwicklung, Stickstoff Massenbilanzen und Stickstoffüberschuss analysiert, um legacy Anreicherungen in vier schwedischen landwirtschaftlichen Einzugsgebieten zu bestimmen. Die Annahme war, dass ein hoher Anteil an landwirtschaftlicher Fläche und ein hoher N-Eintrag zu einem höheren N-Überschuss und folglich zu einer höheren Gesamtstickstoffbelastung (TN) in den Bächen führen würde. Es wurde davon ausgegangen, dass der N-Überschuss eine verzögerte Auswirkung auf die Gesamtstickstoffbelastung (TN) hat, was zu einem Hysterese-Effekt führt. Die Langzeitanalyse zeigte, dass in Einzugsgebieten mit einem hohen Anteil an landwirtschaftlicher Nutzfläche Mineraldünger der Hauptfaktor für den N-Überschuss war. Ein höherer N-Überschuss führte zu einer hohen TN-Belastung der Bächer. Eine positive Korrelation zwischen N-Überschuss und TN-Belastung wurde mit ein Jahr Verzögerung festgestellt. Ein Hysterese-Effekt wurde in den Einzugsgebieten nicht festgestellt. Diese Ergebnisse deuten darauf hin, dass der N-Überschuss in allen Einzugsgebieten nach wie vor hoch ist, weshalb Maßnahmen erforderlich sind, die sich auf die Verringerung der Hauptfaktoren für die N-Akkumulation konzentrieren. Weitere Analysen, die einen längeren Zeitraum gemessener TN-Belastungen und Variablen wie die Dichte der Flächendrainage und die Grundwasserlaufzeit berücksichtigen, sind notwendig, um legacies zu identifizieren.

Stichworte: Stickstoff-Massenbilanz, Stickstoffüberschuss, Landnutzung-Entwicklung, Hysterese-Effekt, Wasserqualität, Landwirtschaft

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Abbreviations

N	Nitrogen
TN	Total nitrogen
FER	Mineral fertilizer
MAN	Livestock manure
DEP	Atmospheric N deposition
BNF	Biological nitrogen fixation
REM	Removal of N
ET	Evapotranspiration
Eq./eqs.	Equation
NUE	Nutrient use efficiency
NPP	Net primary production
CAP	Common Agricultural Policy
WFD	Water Framework Directive
IGO	Intergovernmental Organisations

1. Introduction

Nitrogen is an essential nutrient needed for the growth of most of organisms (Rütting et al. 2018; Bieroza et al. 2019; Marques et al. 2022). Due to the increasing demand for food production and the intensification of agriculture, changes in the natural nitrogen cycle have been observed (Bieroza et al. 2019; Basu et al. 2022). Anthropogenic activities such as the burning of fossil fuels, high depositions of reactive N to terrestrial, and aquatic systems, and the application of high mineral fertilizers rates, have contributed to increased emissions of reactive N, creating a global challenge for water quality. One of the main drivers for the reduced water quality is the high nutrient input from agriculture. (Zhang 2016; Basu et al. 2022; Marques et al. 2022)

Diffuse pollution of N in agricultural catchments has led to an over-enrichment of N in the aquatic system, and consequently to an excessive growth of algae and plants, leading to eutrophication and hypoxia in surface waters (Smith et al. 1999; Basu et al. 2022; HELCOM 2023). The creation of hypoxic zones has severe environmental impacts such as fish kills, losses in ecosystem services but also economic effects (Smith et al. 1999). For example, in the Baltic Sea, increased nutrient inputs from agriculture have led to the eutrophication of 97% of its area (HELCOM 2023).

To reduce the impacts of agricultural nutrient inputs on surface waters, management strategies combining legislation and management practices in Europe, at country and catchment scales, have been adopted. For instance, in 1991 established Nitrates Directive (91/676/EEG), the 2000 Water Framework Directive (WFD) (2000/60/EC), and the Common Agricultural Policy (CAP). Furthermore, intergovernmental organizations (IGO), such as the Helsinki Commission (HELCOM) have been implemented to manage and stop the deterioration of aquatic systems on a regional scale (Norell & Söderberg 2013; Bieroza et al. 2019; European Commission, Directorate-General for Environment 2019; HELCOM 2023; European Commission n.d.).

In Sweden, despite the enforcement of legislation and application of measures along the pollution continuum, nitrate loads continue to be high in surface waters. Reports from the second River Basin Management Plans (RBMP) showed no improvements in ecological status between the first and second management plans, with increasing deterioration of rivers (24%) and lakes (European Commission 2019).
Additionally, studies conducted in two Swedish agricultural catchments, E23 and Tullstorpsån, showed low improvements in water quality, after the implementation of measures such as structural liming, cover crops, wetlands, buffer zones, and two-

stage ditches. The low improvement in water quality could be attributed to time lags (Bieroza et al. 2019; Tullstorpsån Ekonomisk förening 2019; Hallberg et al. 2022).

The reasons behind the time lags[1] between the application of measures and improvements in water quality are multiple, e.g. the scale of the application of measures, financial and administrative constraints, interpretation of legislation, the readiness of farmers to reduce N inputs, and applying voluntary measures (Voulvoulis et al. 2017; Basu et al. 2022), but also the long-term accumulation of legacy nitrogen in the catchment. Studies analysing legacies in catchments showed that legacy nitrogen was one of the main drivers for the low improvements in water quality (Van Meter & Basu 2015; Van Meter et al. 2017; Basu et al. 2022; Marques et al. 2022).

Legacy stores are long-term accumulations of N in different landscape compartments such as biogeochemical legacies in the organic matter of the soils, hydrological legacies in the subsurface and groundwater, and legacies in sediments in streams and reservoirs (Van Meter & Basu 2015; Van Meter et al. 2017; Basu et al. 2022). They are accumulated due to high nutrient inputs and N surplus over time and can impact the total nitrogen (TN) loading to the stream, typically creating a hysteresis effect as N inputs are delayed in relation to outputs – stream N load. A hysteresis effect describes the non-linearity between N inputs and TN loading, meaning that TN loadings in the stream are not only a function of current but also past inputs (Basu et al. 2022).

1.1 The role of long-term land use trajectories and N surplus in legacy assessment

Estimating long-term nutrient dynamics in agricultural catchments at different times requires an analysis of the current and past land use and nutrient inputs (Van Meter et al. 2017; Basu et al. 2022; Marques et al. 2022).

The development of landscapes at different points in time has a high influence on the nutrient balance in the catchment and therefore has to be assessed. Catchments with long-term agricultural use might have different N fluxes over time than catchments with higher percentages of non-agricultural land (Van Meter et al. 2017). Hence, different impacts on N surplus accumulation and nutrient loadings in the stream are expected.

Long-term nitrogen surplus in a catchment is used as an indicator for the net accumulation and depletion of N (Klages et al. 2020; Batool et al. 2022; HELCOM 2023). Moreover, in the past, studies have shown a correlation between N surplus and total N loadings in the stream, making it a good indicator of water pollution (Billen et al. 2009; Hong et al. 2011; Van Meter et al. 2017).
To estimate N surplus at the catchment scale, land use trajectories and long-term nitrogen mass balances have to be evaluated (Van Meter et al. 2017; Marques et al.

[1] This study defines time lag as the time that passes between an action and a reaction e.g., the time that passes between the application of measures and improvements in water quality or the time that passes between the input of N surplus in the catchments and increasing TN loadings in the stream.

2022). The mass balance consists of N inputs: mineral fertilizer, manure application, biological nitrogen fixation (BNF), and atmospheric N deposition (DEP), while N outputs consist of N removal through the crop, pastureland, and forest production (Zhang 2016; Van Meter et al. 2017).

In the past, several studies analysed N surplus accumulations, offering insights into the depletion and accumulation of nitrogen at the country level (Hong et al. 2011; Savchuk 2018; Zhang et al. 2021b). A recent study by *Batool et al. 2022* calculated long-term soil nitrogen surplus in Europe, at a 5 arcmin resolution, 100 km x 100 km, for the years 1850-2019, using global, country-based, and catchment data. The study provided gridded N surplus data but did not offer an insight into the long-term trend of the different mass balance variables that drive the accumulation of nitrogen. Assessing the different mass balance variables is important to get an insight into the main drivers leading to the accumulation of N surplus.

Gridded long-term N surplus estimates can be used as an input parameter in models such as in the process-based model "*Exploration of Long-tErM Nutrient Trajectories (ELEMeNT)*" to estimate legacies (Van Meter et al. 2017; Basu et al. 2022; Marques et al. 2022). Though, to reduce the time lag and improve water quality at the catchment scale, there is also a need to understand the behavior of the main drivers influencing nitrogen accumulation.
Therefore, having seen that the strategies of the EU policy so far have not led to the desired effects, this study considers that understanding the long-term behavior of nitrogen inputs and outputs in agricultural catchments is an additional pillar to implement adequate measures, for the necessary improvement of water quality. For example, measures that target and reduce pollution at the source level.

In this study we calculated land use trajectories, N inputs, outputs, and N surplus, for four Swedish agricultural catchments: E23, E21, Tullstorpsån, and Silverbäcken, for the years 1850-2019, using the comprehensive approach presented in *Batool et al. 2022*. To further reduce the uncertainty of nitrogen accumulation at catchment scale, we adapted input parameters such as mineral fertilizer rates to catchment- and regional-specific data, offering a better representation of the behavior of nutrient inputs, outputs, and N surplus over time. Additionally, we compared ten years (2010-2019) of reconstructed N inputs, outputs, and N surplus data to measured TN load in the stream to analyze the hysteresis effect and some of the complex interactions that lead to increases in nitrogen loading.

1.1.1 Aims and Hypothesis in the study

This study focuses on the reconstruction of annual, long-term (1850-2019), land-use distribution, N input, and outputs, in four agricultural catchments located in Sweden, to analyse the development of nitrogen nutrient legacies.

The research objectives were to:

1) Reconstruct long-term land use trajectories, nitrogen inputs, outputs, and N surplus and to identify key drivers for N surplus accumulation, while reducing uncertainties associated with calculations of N surplus.

2) Identify interactions between N surplus, total nitrogen (TN) load, and other environmental drivers of TN loading.

3) Evaluate the time lags between N surplus and TN load, to see if there is a hysteresis effect in the studied catchments.

Our hypotheses were:

H1 Catchments with higher agricultural land distribution are expected to have a higher annual average N surplus, due to higher N inputs.

H2 The accumulation of N surplus in the soil layer, over time, is one of the main drivers of TN load.

H3 A time lag between N surplus and total nitrogen load is expected, creating a hysteresis effect in the catchments.

2. Methods and Materials

2.1 Data Sources

2.1.1 Study area

This study focuses on the reconstruction of N surplus and nitrogen mass balance components of four catchments: Tullstorpsån, E23, E21, and Silverbäcken (Figure 1)

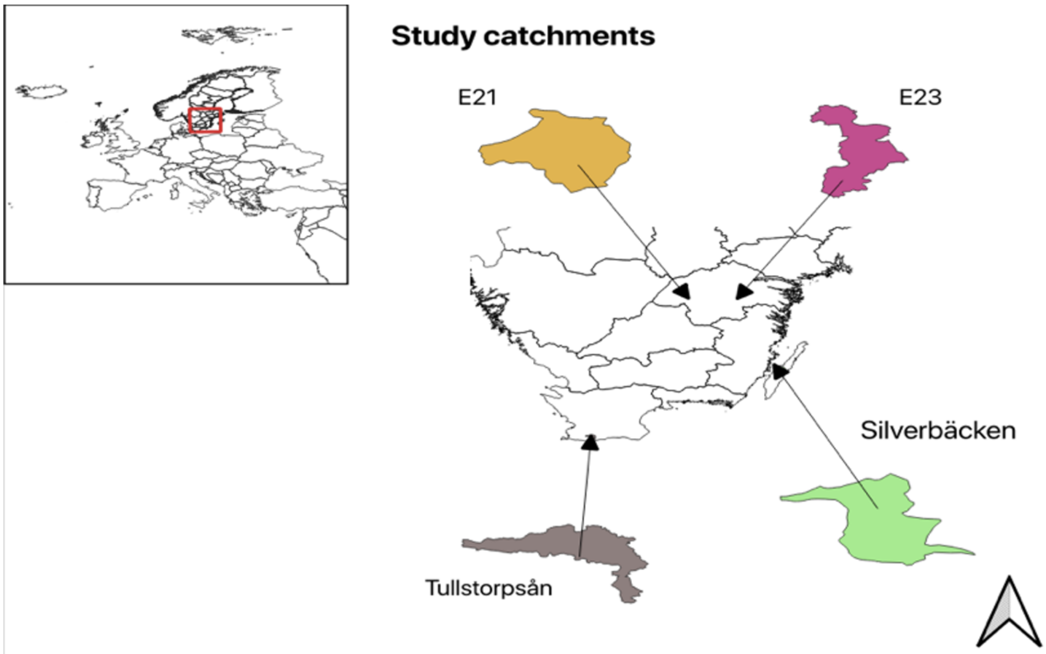


Figure 1 *Location of the four studied catchments. E21 and E23 are located in the Central East, Silverbäcken is located on the Island Öland, South-East, and Tullstorpsån is located in the South of Sweden.*

The studied catchments differ in land use, temperature, precipitation, land use, soil type, flow, N loading, and area (Table 1). The agricultural soils in all study catchments are typically tile-drained. The soil types differ between the catchments with clay (E23) and sandy loam (E21) in the eastern, loam in the southern (Tullstorpsån), and sandy loam in the south-eastern situated catchment

(Silverbäcken). E21 and Tullstorpsån have the highest percentage of agricultural land use as well as the highest measured N mean loading (kg ha^{-1}) in the stream (Bieroza et al. 2018; Havs- och Vattenmyndigheten 2023). The catchments are dominated by different crops such as cereals, ley, and oil seeds (Kyllmar et al. 2014; Bieroza et al. 2018; Hallberg et al. 2022) Highest mean precipitation, mean temperature, and flow were measured in Tullstorpsån, while catchments E23 and E21 have the lowest temperatures and flow. The study catchments are all agricultural headwaters of 1^{st}-3^{th} Strahler order (Strahler 1957), with catchment sizes ranging between 7 and 58 km^2, and are located in the South and Central East of Sweden (Figure 1)
The ecological status of all four catchments was classified as moderate, according to the Vattenkartan from the *"Vatteninformationssystem Sverige (VISS)"* (Vatteninformationssyteme Sverige (VISS) 2023). All four catchments are located in designated nitrate vulnerable zones, according to the 7^{th} report of the European Commission, Nitrates Directive (91/676/EEG), (Vatteninformationssyteme Sverige (VISS) 2023; European Commission n.d.).

Table 1 *Catchment characteristics. Area, mean temperature and mean precipitation (mm), soil type and agricultural land use (Bieroza et al. 2018; Havs- och Vattenmyndigheten 2023; Hallberg et al. 2022)*

Catchment	Area (km^2)	Temperature (ºC)[a]	Precipitation (mm)[a]	Soil type	Agricultural land use (%)[b]	TN load (kg ha^{-1})[c]	Flow (m^3s-1)[a]
Tullstorpsån	57.34	8.24	706	Loam	81	17.72	0.454
E23	7.33	7.01	626	Clay	54	5.37	0.038
E21	16.64	6.63	605	Sandy Loam	89	22.41	0.068
Silverbäcken	33.29	7.51	567	Sandy Loam	61	0.85	0.173

[a] Mean 1961-2022
[b] Agricultural land use 2016
[c] Mean of total N loading 2010-2019

Mitigation measures in the studied catchments

The Tullstorpsån catchment has a long history of agricultural land use, with historical records dating back to 1767. To increase crop yields and the area suitable for agriculture, tile drains, and ditch networks were installed and in the 1900s the Tullstorpsån stream was channelized. From the 1940s onwards the use of fertilizer increased in this catchment. To restore the stream, cooperation between the landowners was established in the year 2009 (Tullstorpsån ekonomisk förening).
Remediation activities, such as constructed wetlands and stream restoration in the form of two-stage ditches (Hallberg et al. 2022) were gradually introduced between 2009-2020, and have led to a moderate improvement in the ecological status

(Tullstorpsån Ekonomisk förening 2019). The yearly "Vattenundersökningar i Tullstorpsån report showed an improvement in phosphorus concentrations, which were still considered "unsatisfactory" according to HVMFS 2019:25 (Tullstorpsån Ekonomisk förening 2019). As for nitrogen concentrations, no improvements have been observed in the stream since the start of the remediation activities in 2009. N concentrations in the stream were classified as "extremely high" according to the Swedish Environmental Protection Agency's assessment criteria (1999). The average (arithmetic annual mean concentrations in manual samples) over the years 2019-2022 was 5.4 mg L-1, with high N concentrations measured when the flow was high (Tullstorpsån Ekonomisk förening 2019; Tullstorpsåprojektet 2023).

E23 is part of the regional Swedish Monitoring Program (Kyllmar et al. 2014), where high-frequency data was also available for the years 2007-2018 (Bieroza et al., 2019). This catchment is more prone to erosion and susceptible to high hydrological flashiness due to the dominance of clay soils in the agricultural areas, than the other studied catchments. As part of the "Focus on Phosphorus Program" (Malgeryd et al. 2015), mitigation measures such as structure liming, lime filter drainage, buffer zones, a 2 km two-stage ditch, and a sedimentation pond were implemented in the catchment in 2014-2016. About 80% of the catchment's water and associated nutrients pass through the sedimentation pond and the two-stage ditch (Bieroza et al., 2019).
The total N load in the catchment is lower compared to the other study catchments. This is due to the clay soils, which are susceptible to erosion, making P the more problematic nutrient in this catchment (Forsberg et al. 2014; Villa et al. 2015; Bieroza et al. 2019). Long-term analysis of hydrological data showed an increase of 13% in NO_3-N concentrations, from 3.0 to 3.4 mg L^{-1} in the stream, and a decrease of P by 15% and SS by 28% since the implementation of the measures (Bieroza et al. 2019).

E21 is part of the national program from the Swedish University of Agriculture (SLU) and the Swedish Environmental Protection Agency (EPA), with long-term monitoring data available for discharge and water quality, 1989-2019. E21 has higher measured TN loads compared to the nearby, E23 (< 100km) (Havs- och Vattenmyndigheten 2023, which is likely due to a higher percentage of cropland. To reduce the impact of nutrient loadings in the catchment, measures like cover crops and buffer zones were applied (Linefur et al. 2021).

In comparison to the other catchments, Silverbäcken has the lowest total nitrogen loading (Table 1), probably due to the low cropland area and high percentage of pasturelands. The sandy loam soil in the area is more prone to nitrate leaching than the heavy clay soil in catchment E23. In Silverbäcken no measures to improve water quality, have been applied.

2.2 Long-term land-use trajectories

To calculate legacies and nutrient dynamics in the study areas, a reconstruction of land use trajectories in the catchments was required (Van Meter et al. 2017; Batool et al. 2022). This study examined gridded land use trajectories for cropland, pastureland, and non-agricultural land in the period 1850-2019. Based on the land use trajectories, it was investigated how nitrogen fluxes (nitrogen inputs and outputs) had developed over time (Van Meter et al. 2017; Chen et al. 2018; Basu et al. 2022; Marques et al. 2022). The software QGIS was applied to retrieve, conduct corrections and increase the resolution of gridded data (QGIS Development Team 2023).

Batool et al. 2022 method with some changes to reduce uncertainty was applied to reconstruct long-term land use trajectories and estimates of N surplus in the study catchments (Figure 2).

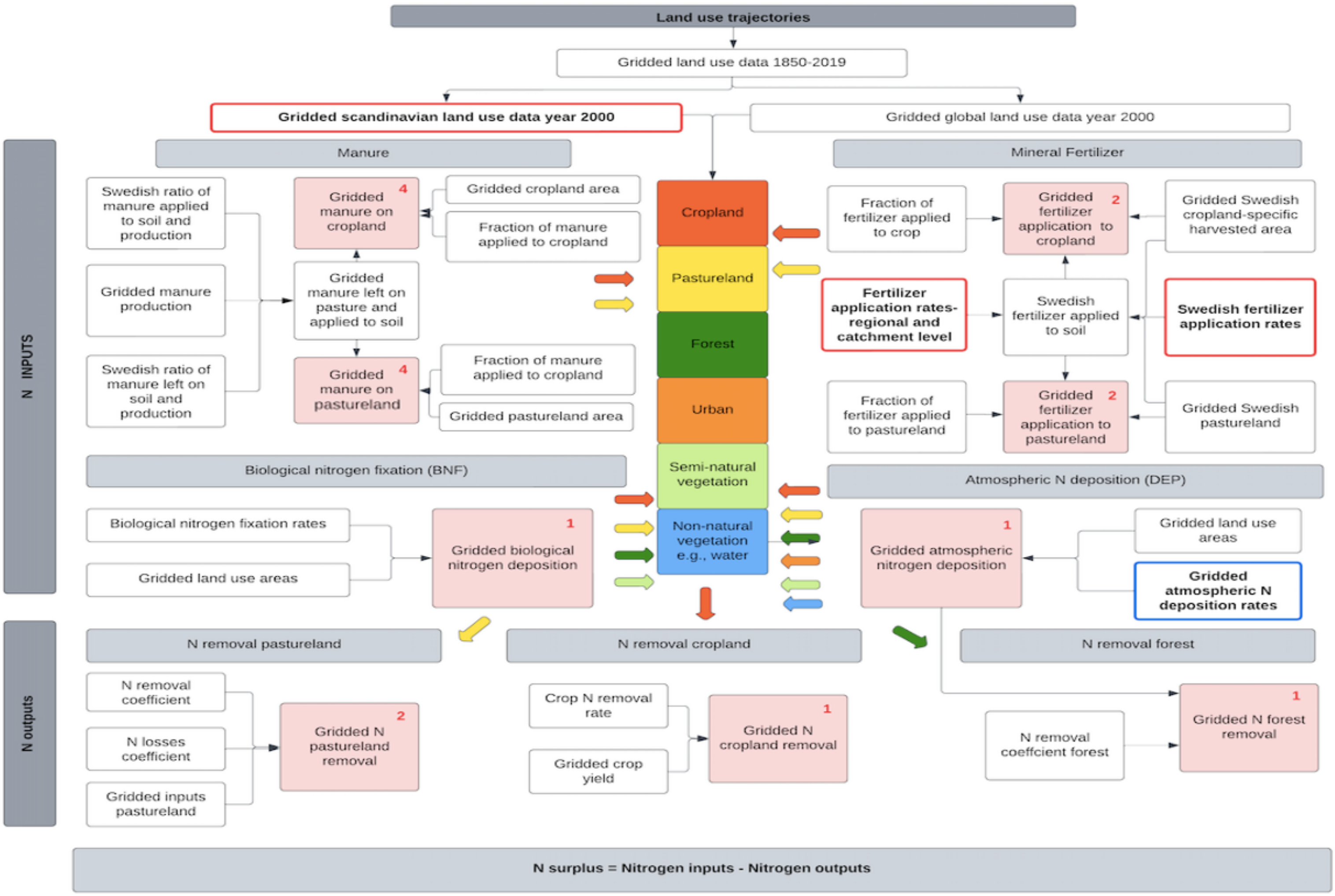


Figure 2 *Flow chart showing the steps applied to calculated land use trajectories, nitrogen inputs, nitrogen outputs, and nitrogen surplus for the years 1850-2019. 48 N surplus (3 methods) estimates were generated using different datasets to reduce the uncertainties. The red numbers represent the number of gridded estimates that were generated. The red boxes represent changes made in the Batool et al. 2022 method to reduce uncertainties at the catchment level. The blue box represents a dataset that increases the uncertainties in the estimates. (Adapted from Batool et al. 2022)*

Since, this research focused on a smaller scale, only the grids, 5 arcmin resolution, that enclosed the catchments, were considered.
The catchment E21 was covered by four grid cells, E23 by two grid cells, Silverbäcken by four grid cells, and Tullstorpsån by five grid cells. The following classification of the grid cells was used (Figure 3):

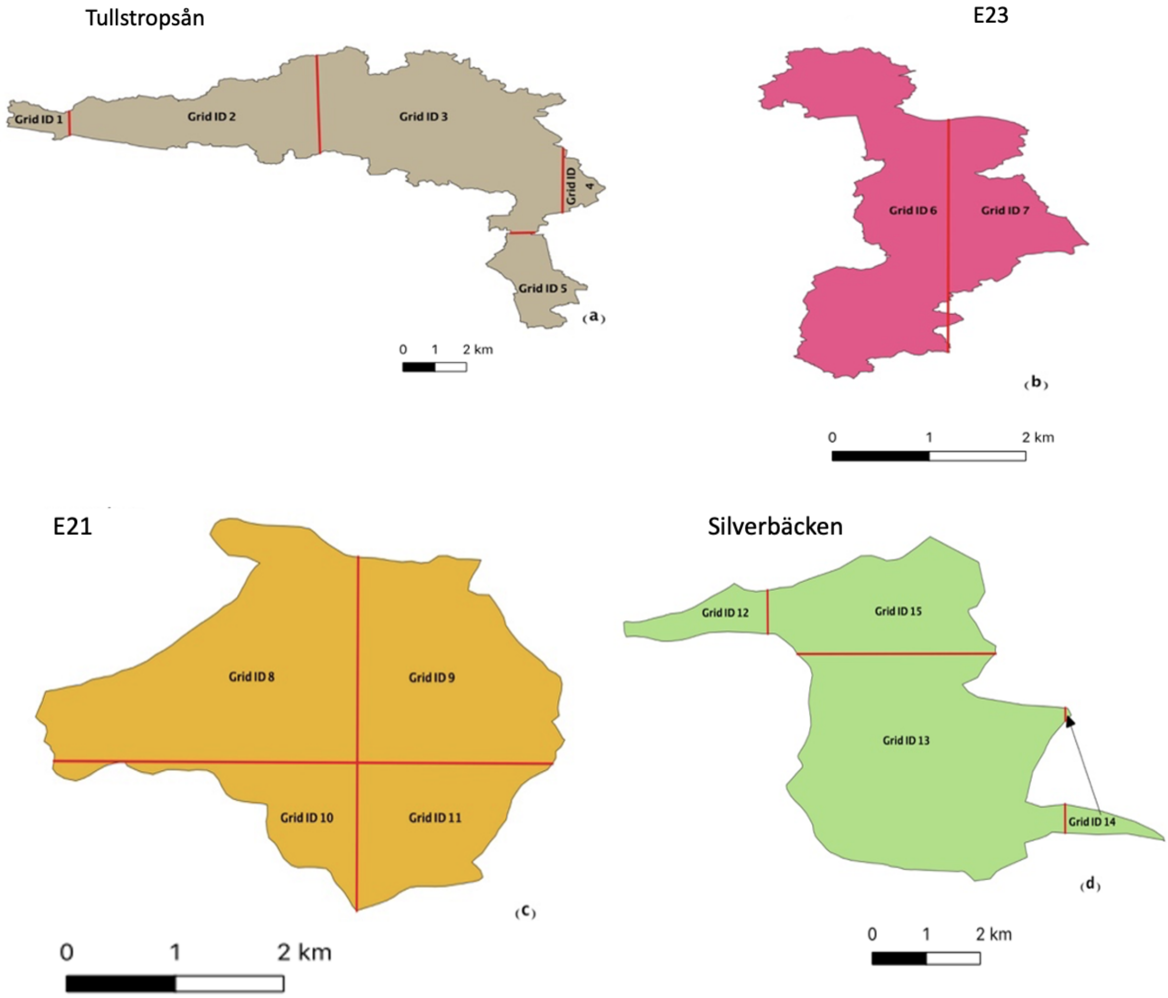


Figure 3 *Classification of catchments according to grid cell coverage, resolution of 5 arcmin: Tullstorpsån covered by five grid cells (a), ID1-ID5; E23, covered by two grid cells ID6, ID7 (b); E21, covered by four grid cell ID 8-ID11 (c) and Silverbäcken, covered by four grid cells ID12-ID15 (d).*

2.2.1 Land use change Method 1

Historical land use change for the period 1850-2019 was calculated by retrieving data from the "*History database of the Global Environment, HYDE 3.2, baseline scenario",* (Klein Goldewijk et al. 2016), and the *"Global spatial distribution of* *cropland and pastureland area"* (Ramankutty et al. 2008), for the year 2000 at a 5 arcmin resolution.

The annual time series, 1850-2019, for land use changes, was calculated by retrieving the gridded, decadal (1850-2000), and annual (2000-2019) values for cropland and pastureland at a resolution of 5 arcmin, an area of 100 km^2, from the HYDE 3.2 database (Klein Goldewijk et al. 2016; Batool et al. 2022). A linear decadal interpolation for the years 1850-2000 was done to complete the dataset. For the years 2018 and 2019, no data was available, therefore the gridded estimates for the year 2017 were applied ($A_{HYDE\text{-}past\text{-}2017}$ (ha)) and ($A_{HYDE\text{-}crop\text{-}2017}$ (ha)).

By referencing the HYDE 3.2 area, 1850-2019, to the year 2000 ($A_{HYDE\text{-}past\text{-}2000}$ (ha)) and ($A_{HYDE\text{-}crop\text{-}2000}$ (ha)), the values were temporally normalized, eqs. (16-17)[2]. Using the dataset from *Ramankutty et al. 2008* the normalized area was referenced to the year 2000 *(A* $_{\text{Ramankutty-crop (i,y 2000)}}$ (ha)) and *(A* $_{\text{Ramankutty-past (i,y 2000)}}$ (ha)), eqs. (18-20). According to *Batool et al. 2022* this allowed to keep a spatial distribution of the areas while taking into account the temporal variability of the HYDE data. The gridded values for grid ID 12 and 14 were missing, therefore the same values for cropland and pastureland, as for ID 15, were assumed.

A comparison of the *Ramankutty et al. 2008* data with the HYDE 3.2 gridded database, showed that the database was incomplete, several grids had missing data values. The *Batool et al. 2022* paper did not mention the incomplete data for Sweden and therefore did not offer a correction method. To correct the data set to match the total HYDE 3.2 gridded data, a spatial joined, nearest neighbour approach was conducted, using the QGIS software. (ESRI 2011; QGIS Development Team 2023).

The FAOSTAT dataset was used to extract the Swedish annual (1961-2019) estimates for "Cropland ($A_{FAOcrop}$ (ha))" and "Land and permanent meadows and pastureland ($A_{FAOpast}$ (ha))" (FAOSTAT 2023c). The data sets were applied to eqs. (22-23), to calculate a Swedish correction ratio, $(R_{A_{cr}}(u,\ y_{1961-2019}))$, $(R_{A_{past}}(u,\ y_{1961-2019}))$. To ensure compliance with FAOSTAT data, the ratios were applied to the in eqs. (18-19) estimated gridded cropland and pastureland area (ha), eqs. (23-26). The physical consistency of the max possible area in one grid cell (i), 100 km^2, was maintained.

2.2.2 Land use change Method 2

When comparing the reconstructed area for cropland and pastureland in method 1, with the catchment-level data from the SMHI database (Havs- och Vattenmyndigheten 2023, it was noticed that the reconstructed gridded area, eqs. (16-26), was underestimated and therefore did not match the SMHI agricultural area for the year 2016. *Ramankutty et al. 2008* dataset overestimated the area of total Swedish cropland, which led to an underestimation of the cropland area at the grid cell level. To correct the reconstructed area, the *Ramankutty et al. 2008* data set was replaced, with the *"Dataset of 1 km cropland cover from 1690 to 1999 in Scandinavia", (Wei et al. 2021).*

[2] The reference to the equations can be found in *Batool et al. 2022* paper. The equations shown in detail are the ones subject to modification or interpretation.

Wei et al. 2021, provided a high-resolution, 1km x 1km, gridded cropland data set for Scandinavia, with estimates until the year 1999. This data set provided a better fit for the total Swedish cropland area and led to a better representation of cropland at the gridded level.

To adapt the gridded values at 1 km x 1 km resolution to the 5 arcmin resolution used in this study, the mean of all grid cells enclosing the grids ID 1-15 was calculated. *Ramankutty et al. 2008* values applied in eqs. (18-19), were replaced by *Wei et al. 2021* cropland area estimates *(*$A_{\text{Wei-crop (i,y 1999)}}$ (ha)).
The Swedish total pastureland data from *Ramankutty et al. 2008*, had a better fit, therefore, no correction was needed.

The physical consistency of the max possible area in one grid cell (i), 100 km^2, was not maintained for grid cell IDs 1-5 and 8-11. Therefore, a redistribution of the area had to be done, see Appendix 1.

2.2.3 Reconstruction of non-agricultural area

Using *Batool et al. 2022* method, eq. (28) non-agricultural area ($C_{A_{other}}$ (ha)) was derived.

Further classification of the non-agricultural area was conducted by using eqs. (29-36). Global land cover (GLC) data for the year 2000, from the *European Commission; Forest Resources and Carbon Emissions (IFORCE), (European Commission 2003)* was used *(European Commission 2003)* to classify the ($C_{A_{other}}$ $(i, y_{1850-2019})(ha)$), the data had a resolution of 300 m.
According to *Batool et al. 2022* method the GLC data for the studied catchments was grouped into four groups: *forest, semi-natural-vegetation, non-vegetation, and urban*. Equations (29-36) were applied to reconstruct the non-agricultural area at a gridded level, for the years 1850-2019 (Batool et al. 2022).

2.2.4 Reconstruction of crop-specific area

Reconstruction of non-fodder-crops area

For the reconstruction of non-fodder crops for the period 1850-2019 the gridded cropland estimates ($C_{A_{cr}}$ $(i, y_{(1850-2019)}(ha)$) and the gridded crop-specific harvested area for the year 2000, at 5 arcmin resolution, provided by *Monfreda et al. 2008* were used. Both estimates were applied to eq. (37) to reconstruct the crop-specific harvested area ($A_{crops}(i, c\ y_{1850-2019})$ (ha)) for the period 1850-2019.
Monfreda et al. 2008 provided a data set for 175 crops (fodder and non-fodder), from which *Batool et al. 2022* selected 17 crops with high N content (N ≥ 1kg of N tonne $^{-1}$).
According to the gridded data from *Monfreda et al. 2008*, eight (wheat, barley, oats, rye, rapeseed, triticale, sugar beet, and potatoes) out of 17 crops were harvested (ha), in the year 2000, in our catchments. The crop-specific harvested area for ID14 was missing, therefore the same value as for ID12 was applied. This study focused

on the reconstruction of the area of eight non-fodder crops using *Batool et al. 2022* presented method.

To comply with the FAOSTAT crop-specific harvested area for Sweden, 1961-2019, a country-level correction was calculated for each crop type, eq. (38) ($R_{A_{non-fodder}}(u, c, y_{1961-2019})$), using FAOSTAT crop-specific harvested area data (FAOSTAT 2023a). This ratio was applied, eqs. (39-40), to the annual gridded crop-specific ($A_{crops}(i, c, y_{1850-2019})(ha)$) estimates calculated in eq. (37), to determine the annual crop-specific harvested area ($C_{A_{crops}}(i, c, y_{1850-2019})$ (ha)).

Reconstruction of fodder crops area

Using *Einarsson et al.* 2021 dataset the fodder crop-specific harvested area was reconstructed. The Swedish country-based data for the harvested area for fodder crops: *temporary grassland, lucerne, other leguminous plants, green maize, plants harvested from arable land,* and *other root crops* (Einarsson et al. 2021), was extracted. The data was used to calculate a country-level correction ratio for the year 1961, eq. (41). The ratio ($R_{A_{fodder}}(u, y_{1961})$) and the in eqs. (23, 25) reconstructed cropland area ($C_{A_{cr}}(i, y_{1961})(ha)$) were applied in eq. (42) to harmonize the estimated Swedish temporal fodder crop specific area for the years 1850-1960 ($A_{fodder}(u, c, y_{1850-1960})$ (ha)).

Batool et al. 2022 did not specify how the downscaling to gridded level, ($A_{fodder}(i, c, y_{1850-2019})$ (ha)) was conducted but stated that the approach was similar to the method used for non-fodder areas.

This study adapted eqs. (38-40) for non-fodder crop-specific harvested areas (Batool et al. 2022), to calculate the fodder estimates [3].
Monfreda et al. 2008 also provided crop-specific harvested areas for the fodder crops: Turnip forage, swede forage, sorghum forage, rye forage, oil seeds forage, mixed
grasses forage, maize forage, legumes forage, grassness forage, fornes forage, clover forage, carrot forage, cabbage forage, beet forage, alfalfa forage and vegetable forage. *Monfreda et al. 2008* fodder crops were classified into six categories, to match the fodder crop classification from *Einarsson et al. 2021,* Appendix 2.

The gridded harvested area (ha) for non-fodder crops (Monfreda et al. 2008) for the year 2000, was retrieved and applied to eq. (37), (Batool et al. 2022) to calculate the annual fodder crop-specific area ($A_{fodder}(i, c, y_{1850-2019})(ha)$). In all four catchments only mixed grasses were harvested as a fodder crop (Monfreda et al. 2008).

Einarsson et al. 2021 Swedish fodder estimates, 1961-2019, were used to ensure that the Swedish country-level fodder-crop estimates are fulfilled, eq. (38 adapt.):

[3] Note of the author: Equations 38-40 adapt. represent an interpretation of the *Batool et al. 2022* method.

$$R_{A_{fodder}}(u,c,y_{1961-2019}) = \frac{A_{fodder_{Einarsson}}(u,c,y_{1961-2019})}{\sum_{i=1}^{n_u} A_{fodder}\,(u,c,\ y_{1961-2019})}$$
(38 adapt.)

The ratio ($R_{A_{fodder}}$) was applied to the in eq. (37) (Batool et al. 2022) calculated gridded annual estimates, to estimate the gridded harmonized fodder-crop-specific area for the studied catchments, ($C_{A_{fodder}}\,(i,c,y_{1850-2019})\,(ha)$), eqs. (39-40 adap.):

$$C_{A_{fodder}}\,(i,c,y_{1961-2019}) = R_{A_{fodder}}(u,c,y_{1961-2019})\ x\ A_{fodder}(i,c,y_{1961-2019})$$
(39 adapt.)

$$C_{A_{fodder}}\,(i,c,y_{1850-1960}) = R_{A_{fodder}}(u,c,y_{1961})\ x\ A_{fodder}(i,c,y_{1850-1960})$$
(40 adapt.)

2.3 N surplus

Nitrogen surplus estimates for the period 1850-2019 were calculated based on eqs. (1-15) in *Batool et al. 2022* (Van Meter et al. 2017; Basu et al. 2022; Batool et al. 2022; Marques et al. 2022). The total surplus for the years 1850-2019 ($Surp_{soil}\,(i,y_{1850-2019})$ was divided into agricultural ($\mathrm{Surp_{agri}}\,(\mathrm{i},y_{1850-2019})$ and non-agriculture ($Surp_{non-agri}\,(i,y_{1850-2019})$. All N surplus was calculated in the unit ($kg\ ha^{-1}\ yr^{-1}$).

Based on the estimated gridded land use area (ha), for pastureland and cropland, the N inputs for the period 1850-2019 were recalculated. Agricultural N surplus was estimated by calculating the N balance: inputs and outputs, for cropland and pastureland.
N inputs in agricultural soils consisted of mineral fertilizer, animal manure, biological nitrogen fixation (BNF), and atmospheric deposition (DEP). N outputs consisted of N removal through animal grazing and crops harvested (Van Meter et al. 2017; Batool et al. 2022; Marques et al. 2022). Annual agricultural N surplus was determined using eqs. (1-8).

Non-agricultural annual N surplus is a function of other-land use area $C_{A_{other}}\,(ha)$. Total $Surp_{non-agri}\,(i,y_{1850-2019})$ consisted of $Surp_{forest}\,(i,y)$, semi-natural surplus $Surp_{NatVeg}\,(i,y)$, $Surp_{Urban}\,(i,y)$ and $Surp_{NonVeg}\,(i,y_{1850-2019})$, eq. (9). Forest N surplus ($Surp_{forest}\,(i,y)$) was calculated by applying the nitrogen mass balance. The balance consisted of N forest inputs: BNF_{forest} and DEP_{forest} and forest outputs: N removal by forests Rem_{forest} (Batool et al. 2022), eqs. (10-12). In the remaining categories, the mass balance considered no outputs ($Rem_{NatVeg,\ Urban,\ NonVeg} = 0$). The surplus was estimated by recalculating DEP_{NatVeg}, DEP_{Urban}, DEP_{NonVeg}, and BNF_{NatVeg}, with eqs. (13-15) (Batool et al. 2022).

2.3.1 Mineral Fertilizer

Annual mineral fertilizer at the gridded level was retrieved by applying the method presented by *Batool et al. 2022*. This method did not use a gridded data but, derived data from four different data sets to calculate gridded N fertilizer inputs on crop and pastureland. The following data sets were used: "*Global coverage of N fertilizer production data"* published by *Holland et al. 2005;* FAOSTAT, "*N fertilizer application for agricultural use (kg*)" (FAOSTAT 2023b); *Einarsson et al. 2021,* "*Synthetic fertilizer application (kg)"* and IFA *"Country-and crop-specific N fertilizer application rates"* (IFA 2022). By using two different data sets on country-level fertilizer application, *Batool et al. 2022*, created two gridded estimates for temporal N fertilizer inputs and reduced the uncertainty of the calculated data.

Country-level mineral fertilizer application rates for the period 1850-1960, were recalculated using fertilizer production data published by *Holland et al. 2005* (Holland et al. 2005; Batool et al. 2022). *Holland et al. 2005* provided data from 1925-1960, therefore a linear interpolation was done for the years 1920-1925. 1920 was the starting point of fertilizer application, therefore an application rate of 0 kg yr^{-1} of N fertilizer was assumed until that year (Batool et al. 2022). The global fertilizer production data was adjusted to Swedish-country-level by relating the *Holland et al. 2005* data for the year 1961, to the FAOSTAT estimates for Sweden 1961 ($N_{fer_{FAO}}(u, y_{1961})(kg)$) (FAOSTAT 2023b), eqs. (43*-44*)[4]. This method was repeated using *Einarsson et al. 2021* estimates ($N_{fer_{Einarsson}}(u, y_{1961})\ (kg)$).

$$R_{N_{fer}}(u, y_{1961}) = \frac{N_{fer_{FAO}}(u,\ y_{1961})}{N_{fer_{Holland}}(y_{1961})} \tag{43*}$$

$$N_{fer_{soil}}(u, y_{1920-1960}) = R_{N_{fer}}(u, y_{1961})\ x\ N_{fer_{Holland}}(y_{1920-1960}) \tag{44*}$$

u, refers to the country, in this study Sweden and *y* to the year.
An annual fertilizer time series for Sweden was created by combining the data from the data sets to $N\,fer_{soil}\ (u,\ y_{1850\text{-}2019})$ (Holland et al. 2005; Einarsson et al. 2021; FAOSTAT 2023b)

N fertilizer in croplands and pasturelands

Fertilizer distribution for croplands and pasturelands were recalculated with eqs. (47-53). Gridded temporal N fertilizer applications were estimated for non-fodder crops, fodder crops, and pastureland using IFA fertilization rates (kg ha^{-1}).
Batool et al. 2022 used an older IFA report 2014-2015, that provided EU-28 estimates and not country level. The new report *"Fertilizer Use by Crop and Country for the 2017-2018 period"* was published in 2022 with country-level fertilizer application rates for thirteen crop group types and grassland (IFA 2022). The values from the report 2017-2018 were used, since the more precise country-level fertilization rates, reduced the uncertainty. The same classification of the

[4] Note of the author: Equations 43 and 44 represent an interpretation of the *Batool et al. 2022* method.

studied crops into IFA's crop groups, as used by *Batool et al. 2022*, was adopted. N fertilization rates for grassland were applied to fodder crops (Batool et al. 2022).

The in eqs. (46-49) estimated values were harmonized to fit country-level fertilizer inputs published by *FAOSTAT,* (FAOSTAT 2023b) and *Einarsson et al. 2021*, eqs. (50-51). The new harmonized crop-specific fertilizer application rates were applied to the reconstructed crop-specific (fodder and non-fodder) and pastureland area, eqs. (52-53). (Batool et al. 2022).

Method II: N fertilizer cropland and pasturelands

IFA's crop fertilization rates were exchanged with regional and catchment-specific data to further reduce the uncertainty of, *Batool et al. 2022* gridded annual N fertilizer estimates, eqs. (46-51). Catchment-specific-temporal fertilization rates for the catchments E23 and E21 and regional-crop-specific temporal fertilization rates for the catchments Tullstropsån and Silverbäcken were applied. For fodder crops and pastureland, the same IFA N fertilization rates (IFA 2022) were used, since no fodder fertilization rates at the catchment or regional level were available and pastureland fertilization rates were incomplete.

Catchment-specific-temporal N fertilization rates were retrieved from *"The National Database on Agriculture land (Datavärdskap Jordbruksmark)"* managed by the Swedish University of Agricultural Science, Department of Soil and Environment, on behalf of the Swedish Environmental Protection Agency (Datavärdskap Jordbruksmark 2023). This database is an environmental monitoring program that works directly with landowners. It provides catchment and year N fertilization rates (kg ha-1 yr^{-1}).

E21 catchment-specific-fertilization rates ($N\ fer_{rate_{E21}}(kg\ ha^{-1})$) were available for the years 1995,1996 and 2002-2019. We assumed the same fertilization rate as in 1995 for the period 1850-1994. For the period 1997-2001, the same fertilization rate as for the year 2002 was applied to calculate the harmonized temporal-specific fertilizer application rates.

E23 fertilization rates were available for the years 2006-2013. The rates for the years 2006 were applied to the period 1850-2005. Moreover, the rates for the years 2014-2019 were assumed the same as for the year 2013, to calculate the harmonized temporal-specific fertilizer application rates. Due to the missing differentiation of rates between crops, the same rate was applied to all crops.

Regional-crop-specific temporal fertilization rates were retrieved, from the Statistics Sweden (SCB) data base *"Tillförsel av kväve efter region, grödgrupp, gödselslag, tabellinnehåll och år, brutna"* 1998/1999-2018/2019, (SCB 2020). This source provided temporal fertilizer data for five crop categories. Fertilizer rates for the category *"cereals"* were applied to the crops: wheat, barley, rye, oats, and triticale. Rates for the category "*all field crops except cereals, grassland, and pasturelands"*, were applied to the crops: potatoes, sugar beet, and rapeseed. Fertilizer application rate for the year 1999 was applied to the years 1850-1998, to calculate the harmonized crop-temporal-specific fertilizer application rates.

By applying the new catchment and regional fertilizer rates to eqs. (46-51), four new crop-specific-harmonized fertilization rates ($C_{Nfer_{crop-rate}}(kg\ ha^{-1})$) and four new grasslands-specific-harmonized fertilization rates ($C_{Nfer_{grass-rate}}(kg\ ha^{-1})$) were estimated (Appendix 4).

2.3.2 Manure

Estimating manure N application rates over time, at 5 arcmin resolution, had a high degree of uncertainty (Meisinger & Randall 2015; Miller et al. 2020; Batool et al. 2022). The general uncertainty is the amount of N per kg of manure, which differs depending on e.g., livestock type, N losses, wet vs. dry manure, climate, and calculation approaches of N excretion rates. (Meisinger & Randall 2015; Batool et al. 2022).

Batool et al. 2022, used two different data sets *FAOSTAT* (FAOSTAT 2023d) and *Einarsson et al. 2021,* to reduce the uncertainties that arise with the calculation of manure N fertilizer application rates. Four gridded manure estimates were calculated for the period 1850-2019, by deriving the manure application values for the categories *"left on pastureland"* and *"applied to soils (N content)"*.
Both databases did not consider N losses through storage and management of manure (N volatilization) but accounted for the reduced N forms through atmospheric deposition (Batool et al. 2022)
Einarsson et al. 2021 data set provided a different classification of manure categories, then the *FAOSTAT* categories "Applied to soil (N content)" and "Left on pastureland". Since *Batool et al. 2022* did not provide a classification, we categorized *Einarsson et al. 2021* livestock manure data to fit the *FAOSTAT* categories, Appendix 5.
To calculate N manure gridded application amounts for cropland and pastureland data from *Zhang et al. 2017, FAOSTAT 2023d*, and *Einarsson et al. 2021* was retrieved and applied to eqs. (54-62).

The second approach used to calculate gridded manure application amounts calculated a new ratio *of "manure applied to cropland and pastureland*" (Einarsson et al. 2021) to *"total applied manure to soil"* (Einarsson et al. 2021; FAOSTAT 2023d). The ratio $R_{man_{appsoil}}(u, y_{1961-2019})$, eq. (62*), was used to adapt the calculated gridded manure amounts, eqs. (60-62). *Batool et al. 2022* suggested applying the new ratios to eqs. (60 and 62). Considering that the ratio was calculated for manure applied to soil and not for manure left on pasture, the new ratio was applied to eqs. (60-61). The results of eq. (61) were applied to eq. (62).

$$R_{man_{appsoil}}(u, y_{1961-2019}) = \frac{Man_{app_{crop}}(u, y_{1961-2019}) + Man_{app_{past}}(u, y_{1961-2019})}{Man_{app_{soil}}(u, y_{1961-2019})} \tag{62*}$$

2.3.3 Biological nitrogen fixation

Biological nitrogen fixation (BNF) of cropland, pastureland, and non-agricultural land is a process in which atmospheric N_2 is converted to reactive nitrogen, by bacteria. BNF was recalculated by applying BNF rates to the estimated crop-specific area (fodder), pastureland area, forest area, and natural vegetation area in eqs. (63-66). BNF of fodder crops was estimated using eqs. (67-68). Eq. (67) calculated BNF rates for different fodder crops, by applying *Einarsson et al. 2021* N fixation (kg yr^{-1}) values of fodder crops to the production of fodder crops (kg yr^{-1}) (Batool et al. 2022).

2.3.4 Atmospheric deposition (DEP)

The calculations to determine ($DEP\ (i, y_{1850-2019})\ (kg\ ha^{-1})$) differed substantially from *Batool et al. 2022.* Due to problems retrieving and resampling the gridded data from the "*National Centre for Atmospheric Research (NCAR), Chemistry-Climate Model Initiative (CCMI*)" we applied a different method.

DEP from 1850-1998 was calculated by extracting gridded data for the years 1860 and 1993, at 50 km x 50 km resolution, from the data set provided by *Deneter 2006* (DENTENER 2006; Van Meter et al. 2017). *Deneter 2006* used an *atmospheric transport model* and calculated estimates for global N, NH_x, and NO_y deposition. Resampling the data to 5 arcmin was not possible, due to problems with the QGIS software, therefore the gridded values at a resolution of 50 km x 50 km were used. The lower resolution increases the uncertainty of the results. A lower resolution is considered to not have a significant impact on the results, since it still captures the Swedish southern-northern trend of atmospheric N deposition, with higher deposition in the southern part of the country (Andersson et al. 2018).

For 1998-2021 period, more detailed data on atmospheric N deposition for the years 1998-2021 was retrieved (SMHI 2023). The gridded DEP data set (kg ha^{-1}) had a resolution of 20 km x 20 km and was downscaled to fit the 5 arcmin resolution. Gridded deposition values for oxidized N (NO_x-N) and reduced N (NH_x-N) were retrieved from the database.
Batool et al. 2022 method, was applied to estimate the DEP (kg yr^{-1}) of cropland, pastureland, forest, semi-natural vegetation, urban and non-vegetation, multiplying the land area for each grid cell (i) by the retrieved $DEP\ (i, y_{1850-2019})\ (kg\ ha^{-1})$).

2.3.5 N removal from cropland

In croplands, N removal was calculated by considering the N removal of harvested crops. Crop yields were multiplied by the crop-specific N content, eq. (69) (Batool et al. 2022).
To derive long-term crop yield data, three data sets: Our World in Data (OWD) (Bayliss-Smith & Wanmali 1984), FAOSTAT non-fodder crop-yield (kg) (FAOSTAT 2023a), and *Einarsson et al. 2021* for fodder crop yield, were applied. Wheat yields for the years 1850, 1911, 1934, 1950, and 1961 were derived from the *OWD* database. Swedish data was only available for the estimates in 1961 (Bayliss-Smith & Wanmali 1984), therefore, the previous year's European averages were

calculated and used as input data (Bayliss-Smith & Wanmali 1984; Batool et al. 2022). From 1961 onwards Swedish estimates for non-fodder crops (FAOSTAT 2023a) and fodder crops (Einarsson et al. 2021) were applied.

Monfreda et al. 2008 offered a gridded data set for crop yields (fodder and non-fodder crops) in the year 2000, the crop-specific estimates for each grid cell (i) were retrieved and applied to eq. (71) to calculate gridded crop yields, 1850-2019. The value for the grid cell ID 14 was missing, therefore the same value as for ID12 was assumed.

2.3.6 N removal pastureland

Estimating N removal from pastureland was done by applying eq. (72). N removal from pastureland consisted of animal grazing and harvested pastureland (Van Meter et al. 2017; Batool et al. 2022; Marques et al. 2022). *Batool et al. 2022* method was applied, which considered N removal from pastureland as an equation of Nitrogen Use Efficiency (NUE), eq. (72). To calculate the total N removal from pastureland, total annual pastureland N inputs, N inputs were calculated with eq. (7), were multiplied with the N removal coefficient of 0.6 and modified with an N loss coefficient of 0.2, to consider N losses.
By adopting the removal and loss coefficients and decreasing the uncertainty of the estimates, two additional N pastureland removal estimates were calculated. In the second scenario, *Batool et al. 2022* differed between Eastern and Western European countries but did not specify how the European countries were classified. We classified Sweden as a Western European country and replaced the removal coefficient with 0.5. Furthermore, the N loss coefficient of 0.2 was set to 0.

2.3.7 N removal forests

Nitrogen removal from forests considered the effect of atmospheric N depositions fertilization on forests, with higher depositions leading to an increase in biomass and to a higher removal effect (Chang et al. 2021). Annual N removal from the forest was calculated by applying eq. (73) which considered, that the removal was a function of atmospheric N depositions in forests and an N forest removal rate of 0.02 (Batool et al. 2022).

2.4 N surplus at catchment level

Using the above-presented method, 48 annual N surplus estimates (Appendix 6) for 15 grid cells using three different methods were generated, for the years 1850-2019. The estimates were compared to the 16 extracted gridded N surplus estimates generated by *Batool et al. 2022*:

1. In the first method, the *Batool et al. 2022* method was applied with minor changes, i.e., changing IFA crop-specific fertilization rates to the new rates from the 2017-2018 report, (IFA 2022) and applying a different method to calculate DEP.

2. In the second method, cropland land use distribution at grid level was recalculated, using the Scandinavian database provided by *Wei et al. 2021.*
3. In the third method, IFA crop-specific fertilization rates (IFA 2022), were exchanged with regional-temporal-crop-specific fertilization rates for Tullstorpsån and Silverbäcken. In catchments E21 and E23, available yearly catchment-specific crop fertilization rates were used.

The different input parameters, used to calculate the 16 annual N surplus rates for the period 1850-2019, can be found in appendix 8.

To estimate catchment-specific N Surplus rates, the mean for the 16 N surplus gridded estimates, IDs 1-15, for each method, were calculated. The mean N surplus in the catchments was estimated by calculating the mean of the grid cells that enclosed the catchment. The same procedure was applied to estimate the N mass balance components: N mineral fertilizer, manure, DEP, BNF, and outputs, at the catchment level.

2.5 Estimation of Time Lag and Statistical Analysis

2.5.1 Time lag and hysteresis effect of N surplus and total nitrogen load

An analysis of the time lag between N surplus and TN load was done to assess the hysteresis effect in the catchments. The analysis was done for the years 2010-2019, since total nitrogen loading data, for the studied catchments, was only available in that period. For catchments E23 and E21, a longer period of data was available, however, we decided to use the same period for all catchments to allow a better comparison between the sites.
The TN load data were extracted from the SMHI model database (Havs- och Vattenmyndigheten 2023) and converted to kg ha $^{-1}$, to allow a better comparison with the N surplus estimates. Using the linear Pearson correlation approach, N surplus was plotted against TN load. The resulting graphs, one for each catchment, were visually analysed to find out if an anticlockwise hysteresis effect was seen. An anticlockwise loop would show a time lag between N surplus input and TN loading, meaning that N surplus would decrease, while TN loading would increase (Van Meter et al. 2017; Basu et al. 2022), suggesting a delay over time.

Since the hysteresis effect is impacted by different variables such as climate, soil type, groundwater travel time, and tile drainage density, we decided to conduct a Principal Component Analysis (PCA), considering drivers of TN load and N surplus. A Principal Component Analysis is a multivariate analysis, that allows a comparison of independent quantitative variables, to assess correlations (Bro & Smilde 2014). This statistical technique provided a better insight into the complex interactions that impact TN loading and legacy accumulation.
The variables; annual precipitation, flow, actual evapotranspiration (ET), and temperature were chosen since measured data could be downloaded from the SMHI

database (Havs- och Vattenmyndigheten 2023; SMHI 2022). Actual evapotranspiration (ET) was estimated by calculating the difference between annual flow (mm) and precipitation and dividing it by annual precipitation (mm). Furthermore, in method 3 estimated N surplus, N inputs, and yield, were also used as input variables in the PCA analysis, to provide a better picture of the interactions at catchment scale. Manure and mineral fertilizer were added to one variable named *"Fertilizer"*. An overview of the included driving variables is given in Table 1.
Variables with an angle > 90-180 degrees represent a negative correlation, variables with an angle < 90 degrees represent a positive correlation, and angles of 90 degrees represent no correlation of the data (Bro & Smilde 2014).

Table 2 *PCA variables and sources of data.*

Variables	Source
N surplus	*Own estimations (method 3)*
DEP	*Own estimations (method 3)*
Fertilizer (Mineral fertilizer and manure)	*Own estimations (method 3)*
Yield	*Own estimations (method 3)*
Flow	*(Havs- och Vattenmyndigheten 2023)*
Precipitation	*(SMHI 2022)*
Temperature	*(SMHI 2022)*
ET	Own estimations
TN load	*(Havs- och Vattenmyndigheten 2023 2021)*

2.5.2 Pearson correlation (R^2) and multivariate correlation analysis

To analyse the correlation between N inputs, outputs, N surplus, and TN loading a linear Pearson correlation was applied. Applying a Pearson correlation allows a statistical comparison of two variables. Correlation coefficients have a value between -1 and +1, zero indicates no correlation between the data. To interpret our results we applied (Schober et al. 2018) interpretation of the correlation values. R^2 between 0.89-0.7 shows a strong correlation and R^2 between 0.4-0.69 a moderate correlation.

Applying the Pearson correlation function in the software program R Studio allows the comparison of only two variables. Therefore, a multivariate correlation analysis of the N input, N output, and N surplus variables, using the R studio packages *"corrr" and "Hmisc",* was done (RStudio Team 2021). The multivariate analysis allowed a better representation of the correlation between the variables.

2.5.3 Mann-Kendall test

The Mann-Kendall test was applied to conduct a long-term trend analysis of the land use trajectories, N inputs, N outputs, N surplus, total nitrogen loading, precipitation, temperature, flow, and actual evapotranspiration. The following hypotheses were tested (Ezzati et al. 2023):

H_0 (null hypothesis) = There is no monotonic linear trend in the data ($p \geq 0.05$)

H_1 (alternative hypothesis) = There is a monotonic linear trend in the data. The trend can be increasing or decreasing over time. ($p \leq 0.05$)

3. Results

3.1 Land use distribution: Comparison between catchments

Over the studied period of 1850-2019, all four catchments showed a similar trend in the land use trajectories, with decreasing cropland and increasing pastureland and non-agricultural land “other land” (Figure 4, Mann-Kendall trend analysis results can be found in Appendix 7).

Cropland

Catchment E21 and Tullstorpsån had the highest percentage of cropland area of all four catchments, with an average of 93% ± 3 (E21) and 85 % ± 2 (Tullstropsån) respectively. The southeast-located catchment Silverbäcken had the lowest cropland area of all four catchments: 26 % ± 3.

The cropland area increased until the 1870s when it reached a plateau until the 1960s, followed by a fast decrease until the 1970s.

From the 1970s until 2019, according to the Mann-Kendall trend analysis, all catchments showed a significant linear decrease ($p \leq 0.05$) in cropland area. E23 showed the highest decrease ($\tau = -0.98$, $p < 0.05$) with 8%. The lowest decrease was seen in Silverbäcken ($\tau = -0.21$, $p = 0.04$) with 1%.

Pastureland

The trend analysis for pastureland showed a significant linear increase ($p \leq 0.05$) for all four catchments.

Silverbäcken had the highest percentage of pastureland with an average distribution of area of 28% ± 1 and the highest significant increase of pastureland ($\tau = 0.82$, $p < 0.05$).

The lowest pastureland area was found in catchment E23 with an average distribution of area of 1 % ± 0.3 and a significant linear increase in time ($\tau = 0.58$, $p < 0.05$).

A comparison of the annual land use results showed that pastureland increased when cropland decreased (Figure 4).

Non-agricultural land

Silverbäcken had the highest percentage of non-agricultural land “other land” with an average distribution of area of 40 % ± 2. The lowest percentage of non-agricultural land was found in E23 with an average of 5 %.
The reconstruction of non-agricultural “other land”, showed a linear increase ($p < 0.05$) for catchments E23, E21, and Silverbäcken. No significant increase or decrease could be proven for Tullstorpsån ($p > 0.05$). Non-agricultural land increased with decreasing cropland.

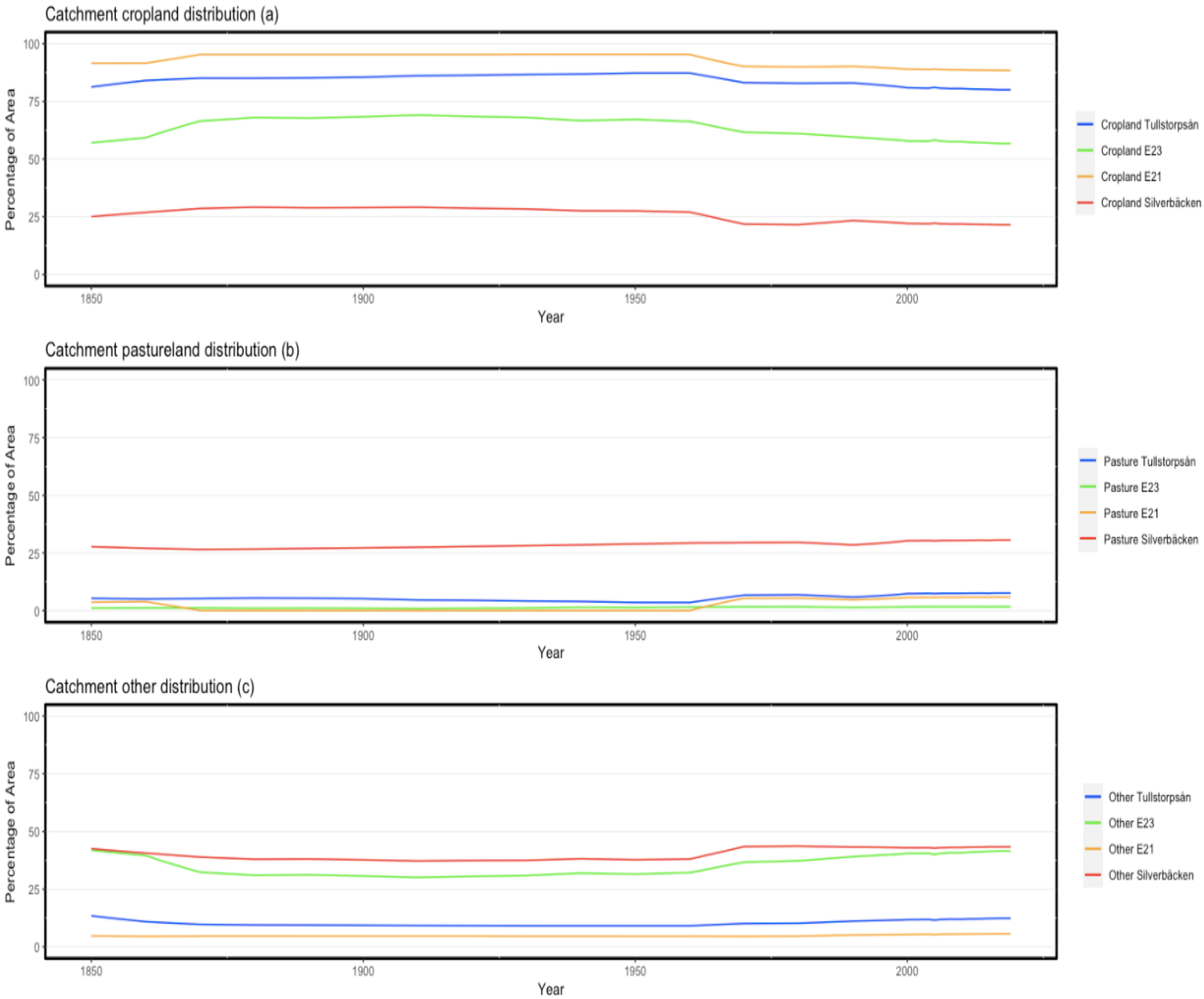


Figure 4 *Land use distribution for the four studied catchments, period 1850-2019. Cropland (a), pastureland (b), and non-agricultural land “other distribution” (c) trajectories (Area (%) were reconstructed. Tullstorpsån (blue), E23 (green), E21(yellow) and Silverbäcken (red).*

3.2 Reconstruction of crop-specific areas

The results showed a linear increase in crop-specific area (ha) in all catchments by 8.11%, until 1960. From 1961 until 2019 the crops rye (τ=-0.58, $p < 0.05$), barley (τ=-0.43, $p < 0.05$), and oats (τ=-0.72 $p < 0.05$) showed a significant monotonic linear decrease. Whereas for wheat a significant linear increase (τ=0.67, $p < 0.05$) was seen. For the crops triticale, sugar beet, and rapeseed, no linear increase or decrease ($p > 0.05$) was seen.

For the studied period 1850-2019, the yield data showed a significant linear increase ($p \leq 0.05$) for all catchments, except for catchment E21, where no significant linear increase or decrease was found for the crop rapeseed.
When comparing the decadal yields from 1961-2019, catchment Tullstorpsån had the highest yields per crop (kg ha^{-1}), followed by E21 and E23. Silverbäcken had the lowest yields of all four catchments. Tullstorpsån and Silverbäcken had high yields of sugar beet, potatoes, and wheat. E23 and E21 had high yields of potatoes and cereals (Appendix 8).
The only fodder crop that was harvested in all four catchments was temporary grassland. The results showed a linear increase in fodder crop yields in the studied period 1850-2019.

3.3 N mass balance

Inputs, outputs, and N surplus were estimated using three different methods; see section 2.4. This section will focus on the N mass balance results (Figure 5) from method 3 since they had the lowest uncertainty.

Mineral fertilizer input

Mineral fertilizer was the highest N input in three (E21, Tullstorpsån, and Silverbäcken) out of the four studied catchments.
Tullstorpsån (66 kg ha^{-1}, 1984) and E21 (56 kg ha^{-1}, 2015) showed the highest mineral fertilization rates. All catchments except E21, followed a similar increasing trend, until the fertilization peak in 1973. A second peak was reached in 1984. In E21 fertilizer input was high in the years 1973 and 1984 but reached a global peak in 2015.
The general trend analysis was conducted for the years 1961-2019. The results showed no significant change in fertilizer rates in the catchments, except for Silverbäcken, where a linear increase in application rates was seen (τ=0.366, $p < 0.05$), table 3.

Manure input, BNF, and DEP

Catchment Tullstropsån had the highest manure rates, with a peak in 1984 (8 kg ha^{-1}), whereas catchment Silverbäcken had the lowest manure rates. BNF was the

highest in catchment E21 with a peak in 2013 (10 kg ha^{-1}). DEP was the highest in the catchments Tullstorpsån (8 ± 3 kg ha^{-}) and Silverbäcken (6 ± 2 kg ha^{-}). DEP values were higher in the southern-located sites (Silverbäcken and Tullstropsån), compared to the more northern-located sites (E23 and E21).
The general trend analysis for the years 1961-2019, showed a linear increase of BNF and manure, in all catchments until the 1960s. A linear increase in DEP was seen until 1998. After that, manure and DEP showed a significant linear decrease, while BNF, showed a significant linear increase ($p < 0.05$) (Table 3).

N Outputs

The highest output values were found in the catchments with the highest mineral fertilizer inputs and yields; Tullstropsån and E21.
The general trend analysis for the years 1961-2019, showed a significant linear increase of output (kg ha^{-1}) for all catchments except for Tullstropsån (Table 3). Due to the low yields of crops, the year 2018 appears with a sharp decrease in N output.

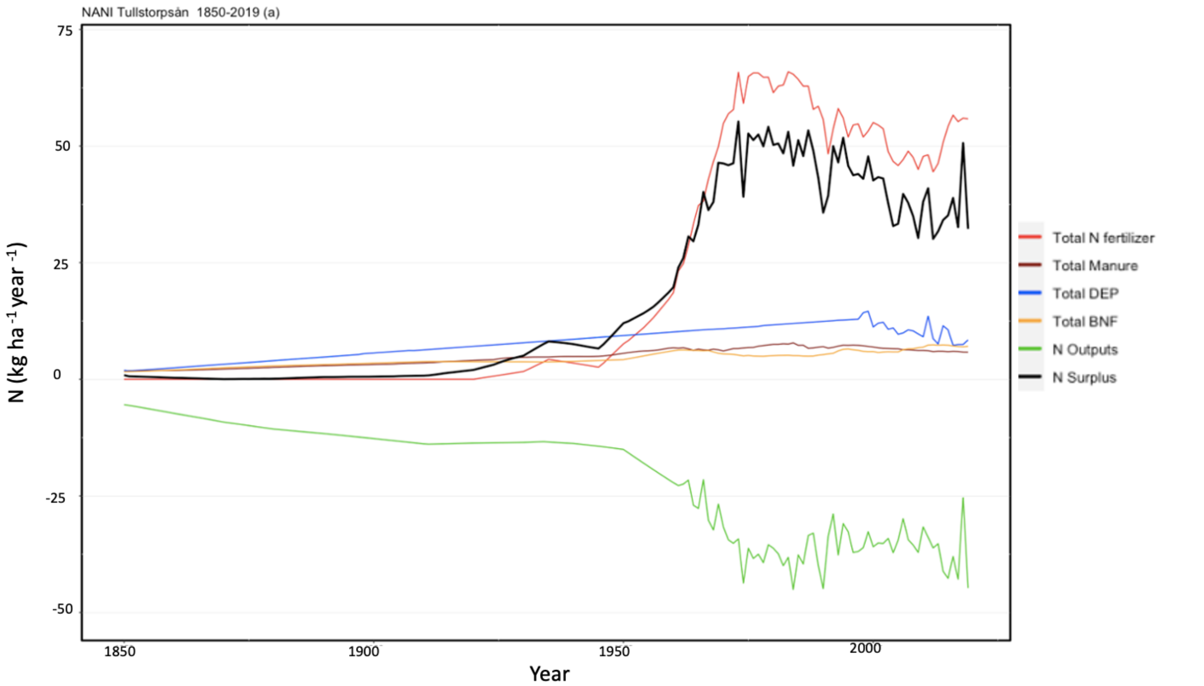
N Surplus

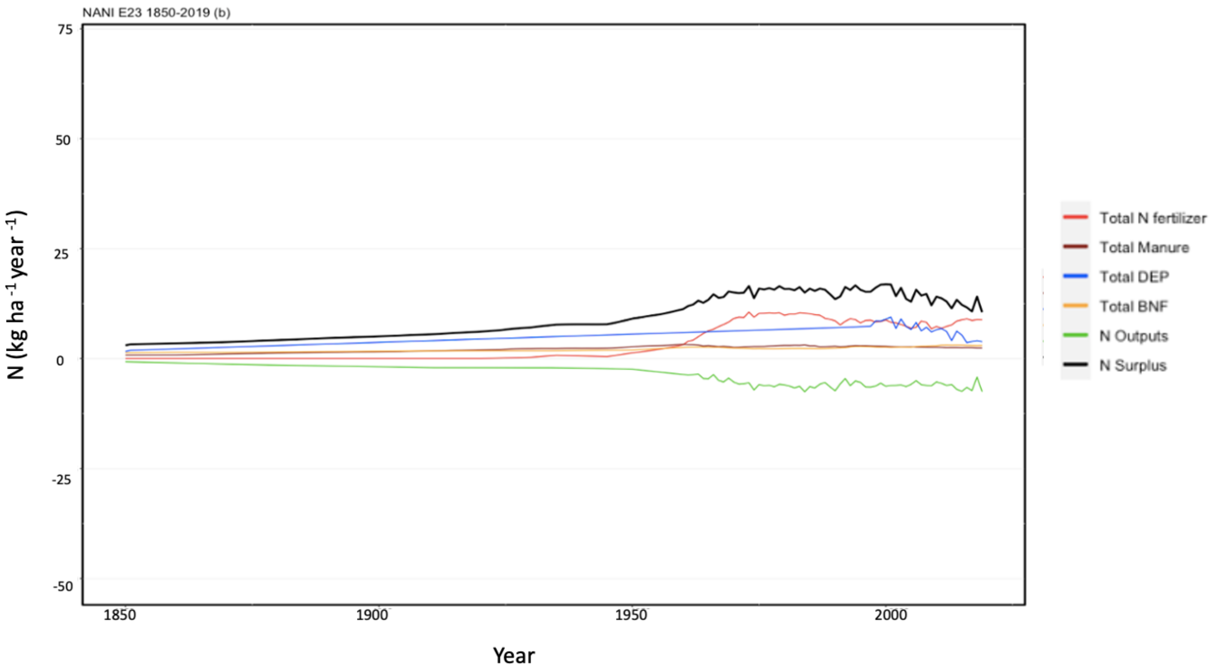
The highest N surplus was found in the catchments Tullstropsån (1973, N surplus 55 kg ha^{-1}) and E21 (2018, N surplus 55 kg ha^{-1}). These catchments had the highest percentage of agricultural land and the highest percentage of mineral fertilizer inputs. In the sites, E23 and Silverbäcken, surplus rates were lower, with a rate of 9 ± 5 kg ha^{-1} and 9 ± 5 kg ha^{-1} respectively.
In the years 1961-2019, mineral fertilizer had the highest significant correlation with N surplus in the catchments E21 (R^2=0.88, $p < 0.05$), Tullstorpsån (R^2=0.82, $p < 0.05$) and Silverbäcken (R^2=0.62, $p < 0.05$), while in catchment E23, DEP had the highest significant correlation with N surplus (R^2=0.74, $p < 0.05$).
The trend analysis for the years 1961-2019, showed no significant decrease or increase of N surplus (Table 3).

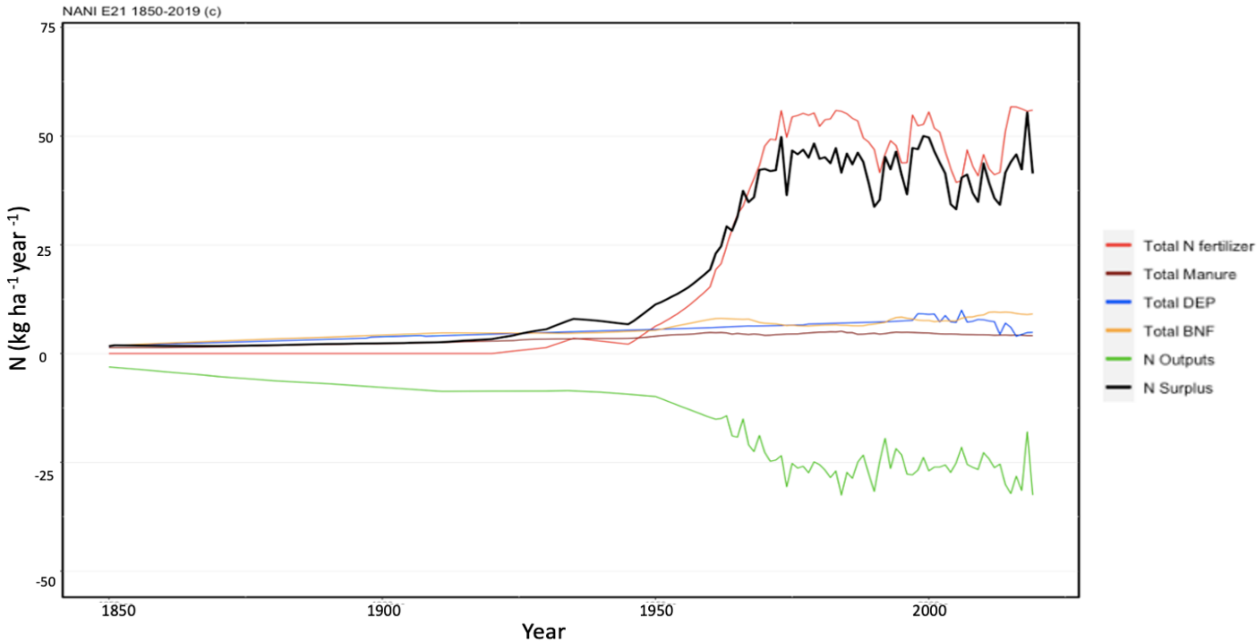
Table 3 *Trend analysis of N mass balance. N inputs and outputs in the period 1961-2019 were analysed for monotonic linear trends with the Mann-Kendall test. Atmospheric N Deposition trend analysis was tested for 1997-2019. BNF stands for Biological Nitrogen Fixation (BNF). A linear increase or decrease is proven when p ≤ 0.05*, p ≤ 0.01** and p ≤ 0.001***.*

Site	DEP (kg ha^{-1})		Fertilizer (kg ha^{-1})		BNF (kg ha^{-1})	
	τ-value	p-value	τ-value	p-value	τ-value	p-value
Tullstropsån	-0.60	< 0.001 ***	0.19	0.26	0.60	0.0002***
E23	-0.71	< 0.001 ***	-0.11	0.20	0.45	< 0.001 ***
E21	-0.31	< 0.001 ***	0.07	0.24	0.19	0.002***
Silverbäcken	-0.58	0.0001***	0.37	< 0.001 ***	0.43	< 0.001 ***

Site	Manure (kg ha^{-1})		Outputs (kg ha^{-1})		N surplus (kg ha^{-1})	
	τ-value	p-value	τ-value	p-value	τ-value	p-value
Tullstropsån	-0.92	< 0.001 ***	0.28	0.09	-0.22	0.18
E23	-0.50	< 0.001 ***	0.31	0.0005 ***	-0.11	0.23
E21	-0.15	0.015*	0.16	0.010**	0.05	0.40
Silverbäcken	-0.70	< 0.001 ***	0.28	0.0016*	0.17	0.05







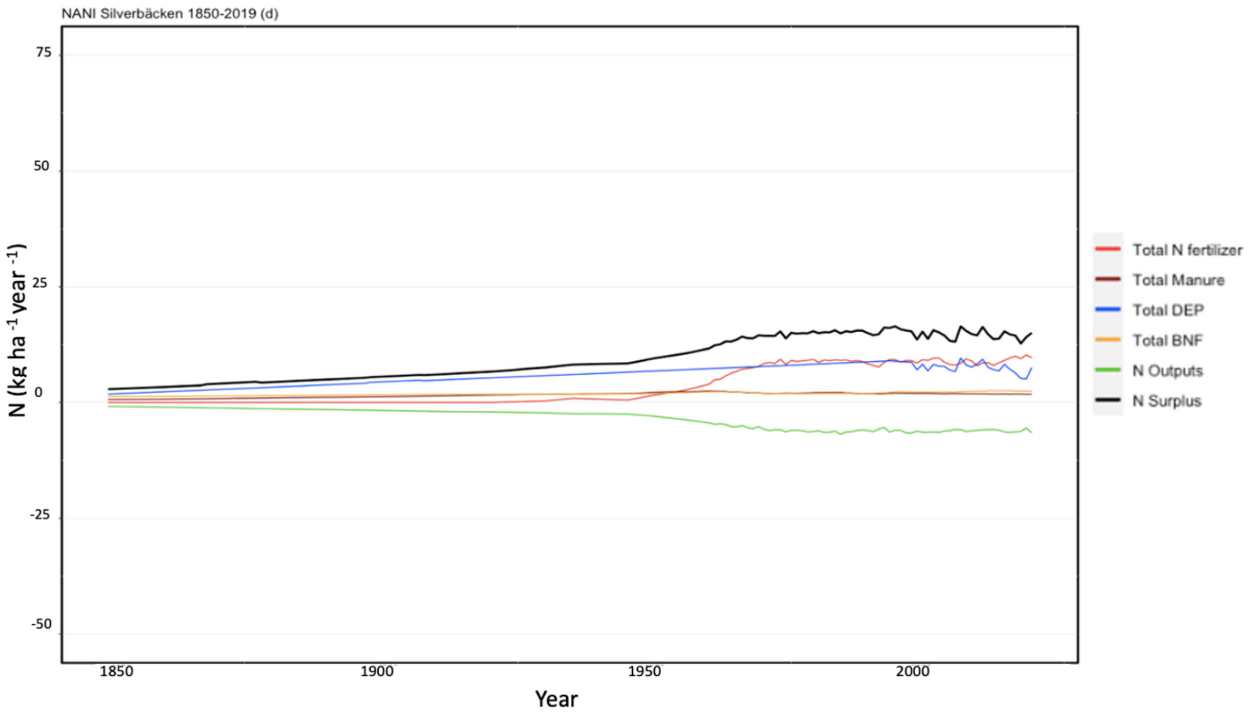


Figure 5 *Trend of nitrogen mass balance in studied catchments, method 3. Inputs, outputs, and N surplus were plotted against time, 1850-2019 for four catchments. Tullstorpsån (a), E23 (b), E21 (c) and Silverbäcken (d). Inputs: Total mineral N fertilizer (red), total manure (brown), total atmospheric N deposition (DEP) (blue), total biological N fixation (BNF) (orange), N outputs (green), and total N surplus (black).*

3.4 Comparison of N surplus estimates methods

The comparison of 48 mean annual N surplus estimates (16 per method, 3 methods) for each studied catchment, with N surplus results *from Batool et al.* 2022, showed a correlation of the trend between all methods (Figure 6). The correlation decreased when a shorter period was considered.

Batool et al. 2022 results showed high negative N surplus estimates for the period 1850-1960 i.e., depletion of N from the soil, due to high crop yields. Our results, methods 1-3, showed a higher N surplus for the first years of the study 1850-1940. From 1961 onwards N surplus was lower in all catchments, than in the study of *Batool et al. 2022*. In the last two years of the studied period, a sharp increase in N surplus in 2018 and a decrease in surplus in 2019 was seen in our results. While *Batool et al. 2022* results showed an increase in surplus in the year 2018, N surplus continued increasing in the year 2019.

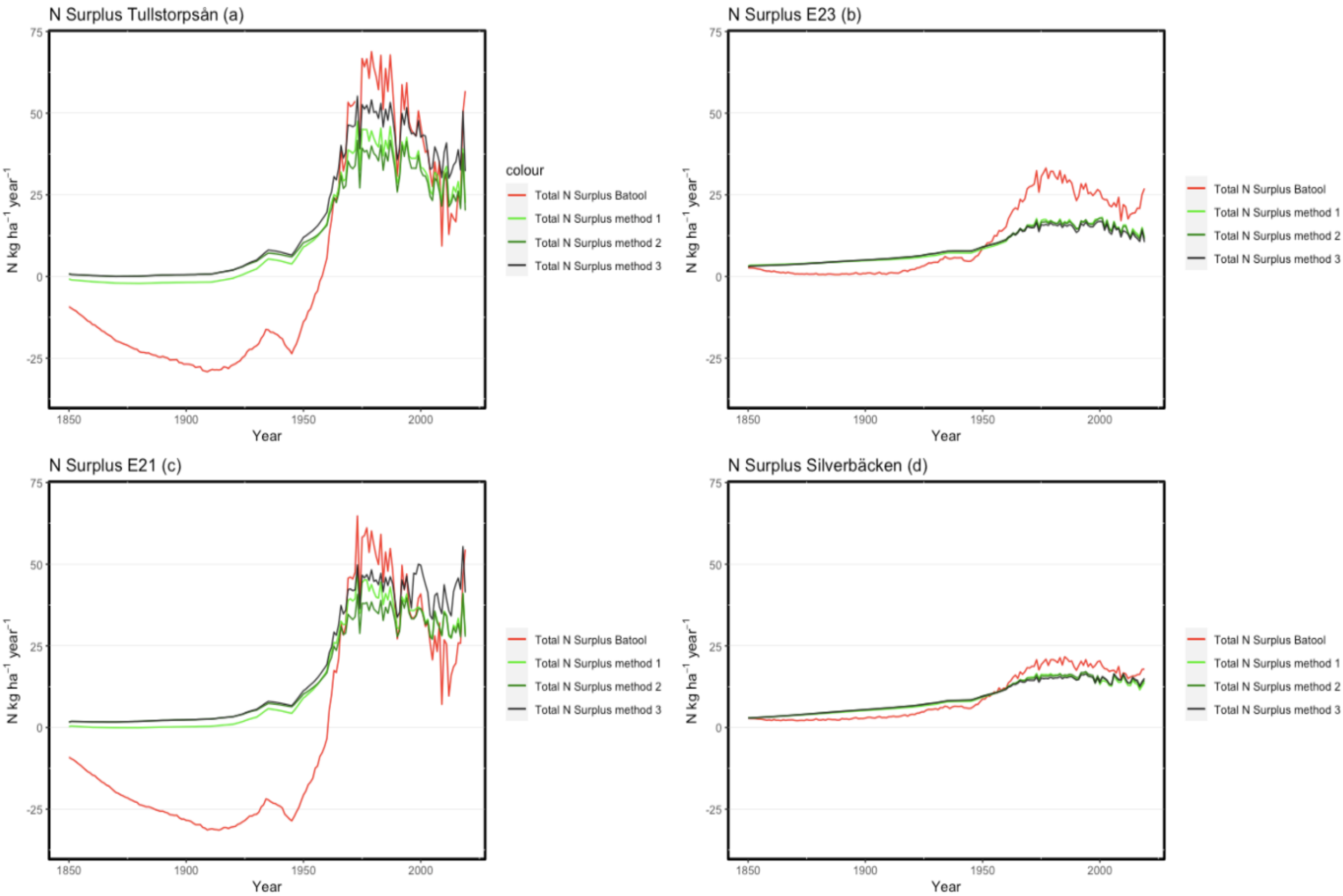


Figure 6 *Comparison of annual average N surplus (kg ha^{-1}) estimates between methods. Three different methods were applied to calculate 48 estimates: Total N Surplus method 1 (green), Total N Surplus method 2 (dark green), and Total N Surplus method 3 (black were compared to the 16 N surplus estimates retrieved from the Batool et al. 2022 dataset (red).*

3.5 Relationship between N surplus and TN loading in streams

In the years 2010-2019, catchment E21 with the highest N surplus (42 ± 6 kg ha^{-1}), had the highest TN loading (23 ± 6 kg ha^{-1}) in the stream, showing a correlation between high N surplus and high TN loading, relating to the first hypothesis that analysed the relation between N surplus and TN loading in the stream.
No hysteresis effect, the time lag between the application of N surplus and TN loading in the stream, was however seen, relating to the second hypothesis. The results showed no clockwise or anticlockwise trend, but a high annual variability of data in the catchments.

The results of the PCA analysis showed a similar correlation between the tested variables in all catchments (Figure 7a-d). The first principal component (PC1), ranging from 40% in Tullstorpsån to 50 % in E23, separated DEP, flow, TN load, and precipitation from temperature, actual evapotranspiration (ET) and fertilizer rates. The second principal component (PC2), ranging from 23% (E21) to 25% (Tullstorpsån), separated N surplus from BNF and yield in all catchments except for catchment Silverbäcken, where PC2 separated N surplus from fertilizer. The two first components explained 65% of the variance for Tullstorpsån, 72% of the variance for E23, 68% of the variance in E21, and 77% of the variance in Silverbäcken. A high positive correlation was found in all catchments between the variables DEP and precipitation; flow and precipitation; temperature and yield; and evapotranspiration and temperature. A negative correlation between N surplus and yield was found for all catchments, except for Silverbäcken, where N surplus did not correlate with yield, but with fertilizer. In all study sites, flow and precipitation showed a negative correlation with temperature with the highest correlation found in Silverbäcken (Figure 7d).

A significant correlation ($p < 0.05$) between TN loading (response variable) and PC1 and PC2 was found for the catchment Silverbäcken. In catchments Tullstorpsån, E23, and E21 a significant correlation was established when DEP and BNF were not considered in the PCA analysis. TN loading had a low positive correlation with N surplus in Silverbäcken, no correlation with surplus in E23, and a negative correlation with N surplus in Tullstorpsån and E21. TN loading showed a positive correlation with precipitation and flow in all catchments.

The Mann-Kendall trend analysis for the year 2010-2019, showed different trends between the catchments. A significant increase ($p < 0.05$) was seen for temperature and yield in Tullstorpsån; for actual evapotranspiration in E23, and fertilizer in all catchments. A negative significant trend was found for flow in E23 and DEP in all catchments, except Tullstorpsån. The other variables did not have a significant change, showing a high annual variation of the data (Appendix 9).

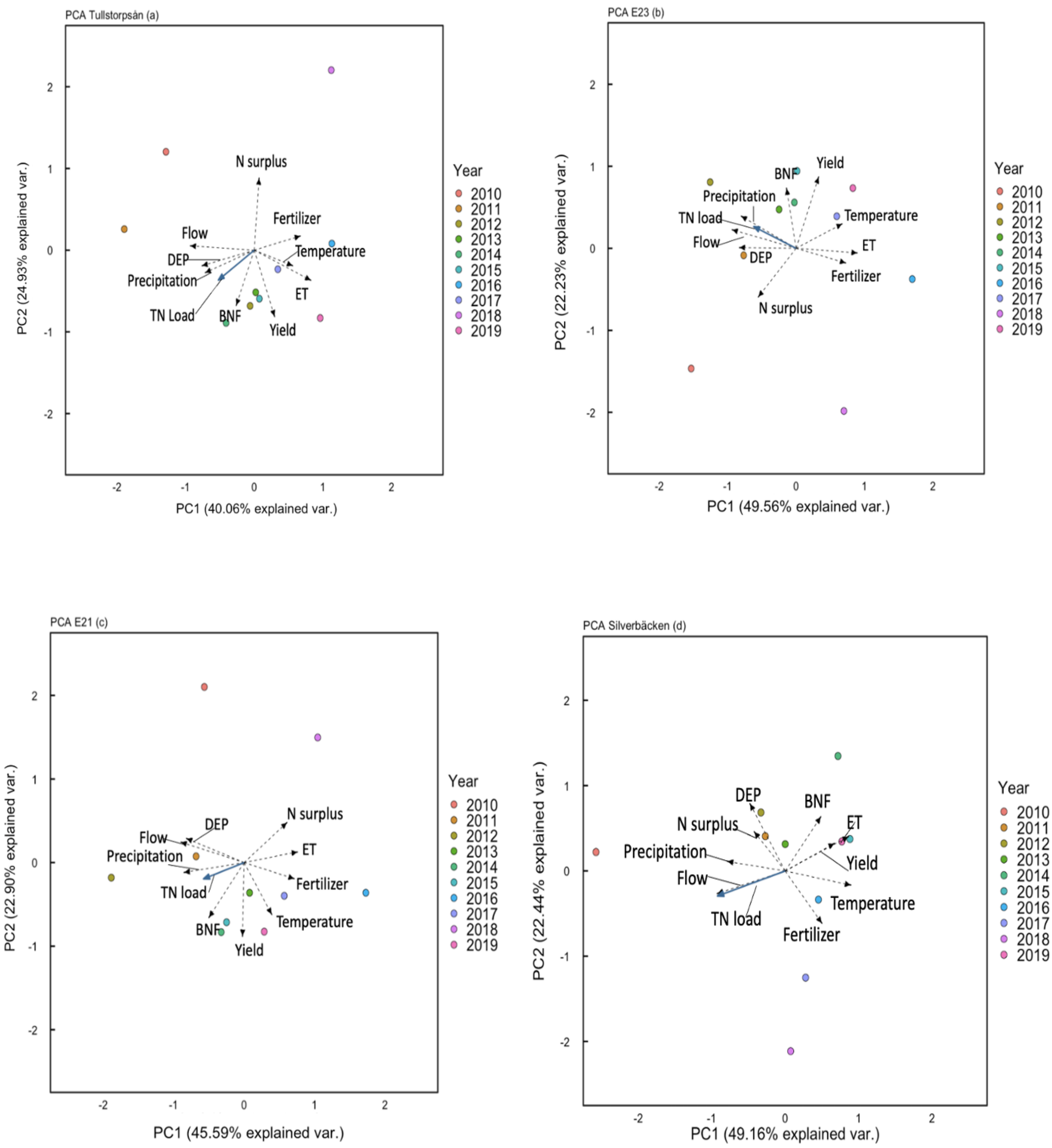


Figure 7 (a-d) *PCA analysis of annual variables, 2010-2019. The plots show inputs, outputs, climate variables, and flow. The circles represent different years. The blue arrow represents the response factor total nitrogen load (kg ha^{-1}). Actual evapotranspiration (ET), Atmospheric N deposition (DEP), Biological N fixation (BNF), Total Nitrogen load (TN load).*

TN load and N surplus

The PCA analysis showed no correlation for E23, a small positive correlation for Silverbäcken, and a negative correlation for E21 and Tullstorpsån. To see if there is a short time lag between the variables, a one-year and two-year shift to the TN loading data was applied (Appendix 10). In the one-year shift, N surplus (year) and TN load (year+1) were compared with each other. In the two-year shift N surplus (year) was compared with TN load (year+2). The results showed a positive correlation in the one-year shift for the catchments Tullstorpsån (R^2= 0.37), E23 (R^2= 0.58), and E21 (R^2= 0.2817). In Silverbäcken, the highest positive correlation (R^2= 0.53) was found when applying a two-year shift of TN loading.

3.6 Uncertainties of results

Uncertainties in N surplus values

The results of the plotted variance of 16 N surplus values, showed a higher deviation in the catchments E21, and Tullstorpsån (Figure 8). These catchments had the highest percentage of agricultural land.
In the period 1850-1970, the standard deviation was higher than in the years 1970-1990, when the estimates approached the annual average N surplus. The uncertainties stayed low until the year 2008/2009, after which an increase was seen for catchments Tullstorpsån and E21.
A higher standard deviation was mainly the result of increased differences in the values between the mineral fertilizer and manure dataset (*FAOSTAT* and *Einarsson et al. 2021).*

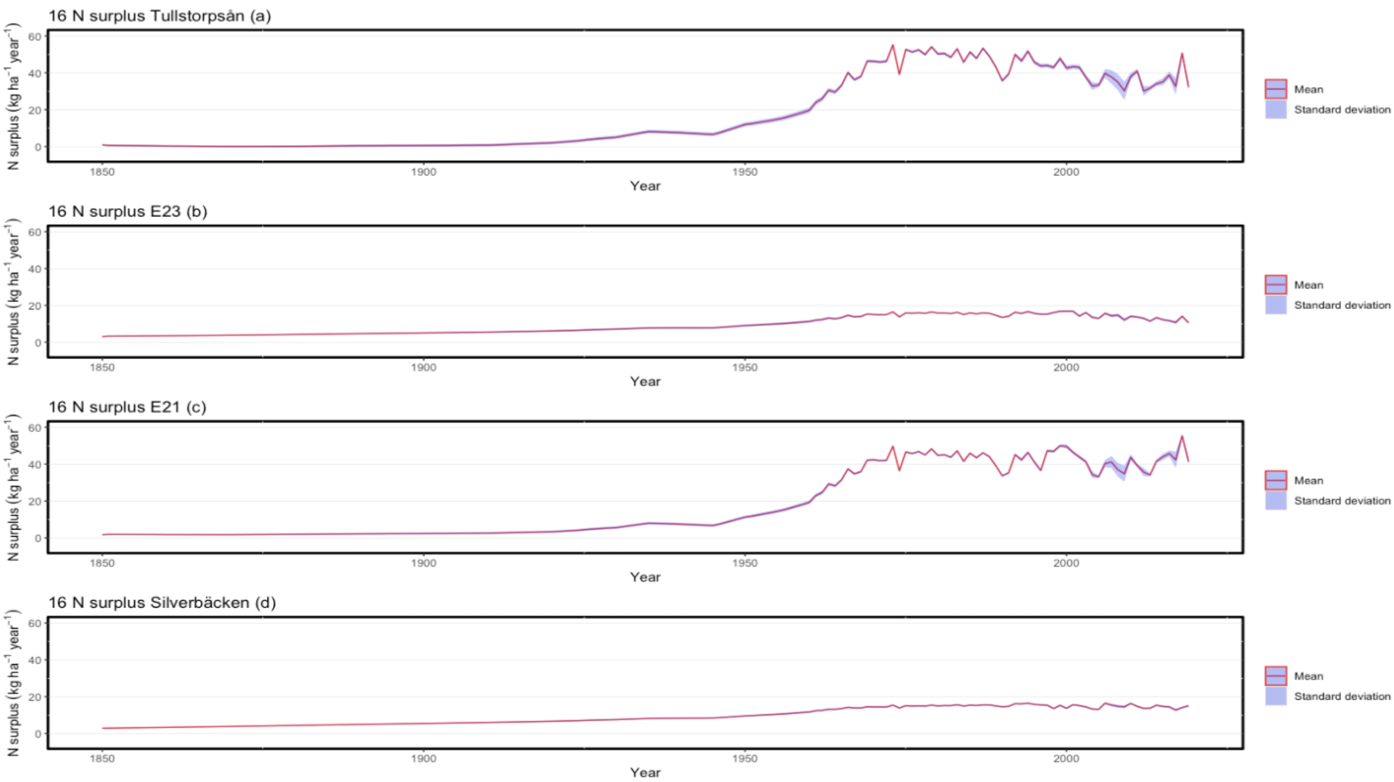


Figure 8 Uncertainties of N Surplus estimates. Annual average N surplus (red) plotted against standard deviation (blue) for the catchments: Tullstorpsån (a), E23 (b), E21 (c), Silverbäcken (d).

4. Discussion

This study aimed to analyse the development of nitrogen legacies in four different agricultural catchments by reconstructing 170 years of land use distribution and nutrient balances to estimate nitrogen surplus.
The first hypothesis related high agricultural land distribution to high nitrogen inputs and consequently higher nitrogen surplus. The results answered the first hypothesis.
The second hypothesis related high N surplus (kg ha^{-1}) to high TN loading (kg ha^{-1}) in the catchment. This study showed a positive effect of N surplus on TN loading, with a higher surplus leading to higher TN loading. The third hypothesis tested the time lag between inputs of N surplus and the effect of TN loading in the stream, expecting to see a hysteresis effect in the catchments. A positive correlation between N surplus (year) and TN loads (year +1) was seen for catchments Tullstorpsån, E23, and E21. For Silverbäcken N surplus correlated with TN loads when a two-year shift was applied (year+2). No exact estimation of the time lag and hysteresis effect was possible.

4.1 Variation of land use distribution and N inputs

Our study showed that in the catchments Tullstorpsån and E21, with the highest percentage of agricultural land use and nitrogen inputs, a higher annual average N surplus was seen.

Van Meter et al. 2017 and *Basu et al. 2022* stated that N surplus is a function of land use trajectories and N inputs over time. Increasing and decreasing cropland i.e., crop harvested area, pastureland, and non-agricultural land, has an impact on the input rate of mineral fertilizer, manure, BNF, and DEP, as well as on the output rate of N through the crop, pastureland, and forest uptake. When N inputs exceed the total N output, a N surplus is accumulated (Batool et al. 2022). In all the studied catchments a N surplus accumulation was seen since 1850. In catchments E21 and Tullstorpsån, with a higher percentage of agricultural land, the accumulation of N surplus was smaller, in the years until 1940, due to less amounts of N mineral fertilizer input.

The interactions between land use, nitrogen inputs, and N surplus were also seen in other studies (Howarth et al. 2012; Han et al. 2014, 2020). For instance, a study by *Han et al. 2020*, analysing global and regional anthropogenic nitrogen inputs, showed that until the 1960s the main driver of N accumulation was atmospheric N deposition. From 1961 onwards mineral fertilizer became the main driver of N surplus, followed by atmospheric N deposition. This pattern was also seen in the studied catchments, except for E23, which had a smaller percentage of agricultural land, where DEP was the main driver for the whole studied period 1850-2019.

In the last 60 years until 2019, a decrease in cropland and crop-harvested areas was seen in all catchments. Moreover, in 1984 a sharp decrease in mineral fertilizer input was seen. This can be attributed to changes in the Swedish agricultural policy. Policies such as the 1984 introduced mineral fertilizer tax, the 1989 Swedish agricultural policy, the Common Agricultural Policy (CAP), the 1991 Nitrate Directive (91/676/EEG), and the 2000 Water Framework Directive (WFD), enforced by Sweden (Daugbjerg 1997; Jordbruksverket 2011; Jordbruksverket. Swedish Board of Agriculture. 2013; European Commission 2019, n.d.).
The reduction of cropland area, for example, could be attributed to measures such as direct support to farmers, through the single payment scheme (CAP reform 2003, in pillar 1) and voluntary agri-environmental climate measures "Eco-schemes" (pillar 2-rural development). This resulted in farmers reducing the area of crop production, due to e.g., the implementation of buffer strips, enhancing biodiversity (Norell & Söderberg 2013; Bieroza et al. 2021).

The decrease in mineral fertilizer in 1984 seen in the results of the study, could be attributed to the 1984 introduced environmental tax on mineral fertilizers, which targeted the reduction of nitrogen leaching to the Baltic Sea and nitrogen in drinking water. The implementation of the tax led to a decrease in mineral fertilizer sales and reductions in fertilizer inputs in the years 1984-1992 (Jordbruksverket 2011; Andersen 2018), reducing N surplus in all catchments. Further decreases in fertilizer could be seen when Sweden joined the European Union in 1995 when European laws such as the CAP but also the Nitrate Directive (91/676/EEG) were enforced (Norell & Söderberg 2013; European Commission n.d.). Though, in the last years of the study the mineral fertilizer inputs increased again. One reason for the increase could be the termination of the mineral fertilizer tax, due to the 2008/2009 financial crisis (Andersen 2018).
Moreover, in Sweden, Nitrogen Vulnerable Zones (NVZs), under this directive (91/676/EEG), were established in areas with a high risk of N pollution (Vatteninformationssystem Sverige (VISS) 2023). All catchments investigated are situated in NVZs, therefore measures such as the timing of fertilizer input, storage of manure, and amount of manure input allowed in a year (Jordbruksverket Swedish Board of Agriculture. 2013) had to be implemented. This study considers that since the Nitrates Directive (91/676/EEG) in Sweden does not target mineral fertilizer input rates, and manure fertilizer rates were low in the catchments in the years between 1850-2019, the impact of the EU directive has a moderate effect on N surplus and therefore does not meet the expectations.

The findings in this study showed that policies and applied measures had an effect on the reduction of mineral fertilizer, but that long-term enforcement of policies and measures is needed.

4.2 N surplus and interactions with TN loading in the catchment

4.2.1 Interactions between climate variables, N inputs, and TN loadings

Our results showed that TN loading in the stream was not only impacted by N surplus but also other parameters such as precipitation, DEP, flow, and temperature.

Precipitation and flow are transport media for nitrogen losses and thus are the main drivers of TN loading in the stream (Deelstra et al. 2014; Stålnacke et al. 2014; Ezzati et al. 2023). In catchment E21, a study by *Ezzati et al. 2023* showed the relationship between TN loading and flow, with increases in TN loading after low flow periods. The drought in Sweden in 2018 led to an increase in water stress for the crop system (Campana et al. 2018) and consequently reduced yields (SCB 2020). The reduction in yields led to lower outputs of N by the crops and increased the N surplus in all catchments. Due to low precipitation and flow conditions, nitrogen was not transported to the stream but accumulated in the soil layer. Hence, when precipitation and consequently flow increased in the year 2019, the TN loading in the stream increased. Similar findings were also seen in our study in the catchment Tullstorpsån (Tullstorpsån Ekonomisk förening 2019).
Moreover, our study showed that precipitation also had an impact on the transport of atmospheric N deposition in the catchments. The atmospheric transport of N_2 and reactive nitrogen and deposition of N is an important part of the global N cycle, making up to 70 Tg N yr^{-1} of input to terrestrial surfaces (Fowler et al. 2013). Nitrogen is deposited as reactive nitrogen from the atmosphere through dry and wet deposition. Wet deposition is transported through precipitation (Fowler et al. 2013; Andersson et al. 2018; Zhang et al. 2021a). Hence, when N is deposited in e.g., the stream, impacts on the total TN loading are expected (Deelstra et al. 2014; Stålnacke et al. 2014; Ezzati et al. 2023).

4.2.2 Interactions between N surplus and TN loading

Although a positive correlation between N surplus and TN loadings in all catchments was seen when applying a one or two-year shift of TN loadings, the linear correlation was generally poor. Additionally, when plotting TN loading against N surplus no hysteresis effect was seen.
One of the reasons behind the non-linearity and missing hysteresis effects could be that N surplus and TN loading were analysed for a short time scale of ten years. The relationship between N surplus and TN loading is very complex and driven by interannual changes, therefore a longer time scale would be needed to get a better insight into the formation of legacies.

Moreover, important interactions, such as climate, soil type, groundwater travel time, and tile drainage density (Basu et al. 2022), which require a modelling approach, were not considered in the analysis due to time constraints. Another reason could be, the high annual variation of N surplus, TN loading, and precipitation, that created a high background noise in the data.

All the studied catchments are tile drained. Drainage systems decrease the denitrification rates and increase the mineralization of N and NO_3^- leaching to groundwater and surface water (Bieroza et al. 2019; Castellano et al. 2019; Hallberg et al. 2022). A study showed a positive impact of tile drainage on the net primary productivity (NPP) of inorganic N (Castellano et al. 2019). The immobilization of inorganic N led to an accumulation of N in the soils as legacy sources. A net accumulation of N in the sub-surface of soils in tile-drained systems was also seen in *Baresel & Destouni 2006* study on the uncertainties behind the accumulation and depletion of N. The direct discharge mobilized N sub-surface pools, such as legacies, which were transported to the groundwater and accumulated in sub-surface pools e.g., stream. Consequently, leading to a redistributing of N from the soil to the stream (Baresel & Destouni 2006).

To quantify N legacies stored in different landscapes, such as in organic matter, groundwater, sediments, reservoirs, and riparian areas (Van Meter et al. 2017; Basu et al. 2022), further research is needed. The process-based modelling approach Exploration of Long-tErM Nutrient Trajectories (ELEMeNT), could be used to further analyze the interactions of N surplus and other parameters such as mineralization rates of N surplus (Van Meter & Basu 2015; Van Meter et al. 2017; Basu et al. 2022) and provide a more detailed insight into the expected hysteresis effect between the input of N surplus and TN loading. Quantifying N legacies and the time lag between measures applied in the catchment and improvements in water quality is important if we want to create realistic time frames for water quality improvements and water quality goals in policy. This allows the application of adequate management measures, that target the whole pollution continuum (Van Meter et al. 2017; Basu et al. 2022).

4.3 Validation of estimates with country and regional-level data

A comparison of the land use distribution in the studied catchments, with former studies analysing historical land use trends in Sweden (Jordbruksverket 2011; Wei et al. 2021), showed similar land use trends for the studied period 1850-2019. Comparing the calculated agricultural land (cropland + pastureland) for the year 2016 with the 2016 SMHI catchment-specific agricultural land data (Havs- och Vattenmyndigheten 2023), showed that in catchments Tullstorpsån and E21, our estimates for agricultural land were higher (<10%). In catchments E23 and Silverbäcken, the results showed a lower agricultural land area (< 10%).
To validate the non-agricultural land distribution, a comparison of the non-agricultural land area with the GLC dataset for the year 2000 (European

Commission 2003; Bartholomé & Belward 2005) was made. Our results showed a higher percentage of non-agricultural land area in the catchments Tullstorpsån (6.3%) and E23 (14.9%). An overestimation of the area of the non-agricultural land could lead to an overestimation of N inputs and N outputs.
Considering that the calculations of N input and N output were based on the crop-specific harvested area, the overestimation or underestimation of land use area has a low impact on the final N surplus results.

Data on the catchment-specific manure application rates was only available for catchment E21 for the years 1995-1996, and 2002-2019 (Datavärdskap Jordbruksmark 2023). The comparison of the manure estimates with the "*Datavärdskap Jordbruksmark"* data, showed an underestimation of manure application rates (kg ha^{-1}) in our calculations. The manure rates in the calculations were half the amount.
An underestimation of manure application rates would mean that the results portrayed an underestimation of N surplus. Though, the underestimation of N surplus is not considered to be significant since manure application was not the main driver of N surplus in this catchment. An adaption of manure application rates to catchment-specific rates should be done to reduce uncertainty.

Lastly, annual yield estimates for the years 2010 and 2019 were compared with regional data (SCB & Jordbruksverket 2018, 2023). The comparison showed that the yield estimate for the crops wheat, rye, oats, barley, and potatoes were lower. Moreover, it was seen that potatoes (E23 and Silverbäcken) and sugar beet (in all catchments) were not harvested in the year 2019. Higher crop yields in the catchments would lead to higher N outputs by the crops. To further reduce uncertainties in the estimation of N output, additional calculations could be done correcting yield estimates with Swedish regional data (1965-2019) (SCB & Jordbruksverket 2018, 2023).

4.4 Uncertainties and limitations of the study

4.4.1 Uncertainties

Applying European and global methods at 5 arcmin resolution to evaluate land use distribution, N inputs, and N surplus at the catchment level, leads to uncertainties in the data.

In general, we can state that uncertainties increased when estimating N surplus (N inputs and N outputs) for the years 1850-1960 since most of the applied databases at the country level started with the year 1961. The calculated ratio for the year 1961 was used for the years 1850-1960, increasing the uncertainty in that period.

Several sources of uncertainties arise when calculating N surplus estimates. First, it has to be considered that *Batool et al. 2022* study calculated long-term N surplus at 5 arcmin for all of Europe, therefore some simplifications in the calculations were made. For example, only one gridded data point for every studied crop for the year

2000 was applied to reference the gridded cropland temporal dynamics to the crop-specific, harvested area (Monfreda et al. 2008), increasing the uncertainty in the results.
Moreover, we assumed in the calculations that all eight analysed crops were harvested every year in the studied catchments. This assumption could lead to an overestimation of the N output from the system, hence an underestimation of N surplus in the calculations.

For the period 1921-1960, the uncertainty of mineral fertilizer input increased, since a global database (Holland et al. 2005) was applied.
Moreover, Swedish statistics on mineral fertilizer input started in 1961 (FAOSTAT 2023b) and 1968 (Jordbruksverket 2011). Considering a later start of mineral fertilizer input would lower N surplus in the years 1921-1960. Therefore, the calculations could portray an overestimation of mineral fertilizer input and N surplus in that period.

High uncertainty in the data for atmospheric N deposition arises for the years 1850-1997, since – due to difficulties with the data extraction – a different method than *Batool et al. 2022* was applied. Gridded global maps of atmospheric deposition for the years 1860, 1993, and 2050, at 50 km x 50 km resolution (Deneter 2006) were used. The higher resolution and the linear interpolation of three gridded values for the years 1860, 1993, and 1997, led to an increase in the uncertainty.
DEP is assumed to linearly increase until 1998. This assumption is incorrect, considering that DEP in Sweden was higher in the years 1984-1992 than in the last years (Andersson et al. 2018). In catchments E23 and Silverbäcken, due to the lower fertilizer rate, DEP had a higher influence on the N surplus balance, therefore increasing the uncertainty of the results for these two catchments.

In general, the results in the catchment Silverbäcken had the highest uncertainty since different databases were missing gridded values. For example, *Monfreda et al. 2008* database on crop-specific harvested area and crop-specific productivity, was missing gridded values, for the grid ID14. Therefore, the same values as for grid ID 12 had to be applied, which increased the uncertainty in the calculations.

The higher standard deviation in Tulllstopsån and E21 was mainly the result of increasing differences in the values between the mineral fertilizer, manure rates (*FAOSTAT* and *Einarsson et al. 2021)*, and the higher percentage of agricultural land. Moreover, the increase of uncertainty in the year 2008/2009, for all catchments, could be related to the 2008/2009 financial world crisis, which had a severe impact on mineral fertilizer prices and sales (Zhang & Broadstock 2020).

Lastly, uncertainties were created due to different interpretations in *the Basu et al. 2022* method. For example, for the adaptation of global mineral fertilizer data, no equation was available, which required an interpretation of the method.

4.4.2 Limitations of the study

This study has potential limitations, that need to be considered when evaluating the results.

Important soil processes such as denitrification, ammonification, and mineralization were not considered in the calculations since they required a modelling approach. These processes have an important role in the mass balance of N and therefore should be considered in the calculations.
Moreover, crop rotations in the catchments were not considered. Not taking rotations into account could lead to an overestimation of N outputs by the crop and hence an underestimation of N surplus.
Additionally, catchment-specific data was not available for all variables. For example, no long-term information on the catchment-specific harvested areas could be found. For that reason, *Monfreda et al. 2008* gridded values could not be adapted to more precise catchment data.
Other limitations arise from the omission of social aspects, such as the influence of wars, economic crises, and pandemics. Such social events have an impact on nutrient inputs and yields, therefore also influencing total N inputs and N surplus.

Lastly, due to time constraints of my master's thesis, it was not possible to conduct further adaptations of the *Batool et al. 2022* method. For example, adjusting calculated annual crop yield data to regional yield data could be done in the future to reduce further uncertainties.

5. Outlook and Implications for future nitrogen management

The increasing pressure on aquatic systems caused by excess nutrient inputs requires accurate estimations of time lags and legacies in agricultural catchments.
To estimate legacies and get an insight into the complex interactions that lead to the formation of nutrient legacies, nutrient inputs, and N surplus have to be traced back in time.
This study reconstructed 170 years of N surplus estimates and reduced uncertainties at catchment level, adapting the *Basu et al. 2022* method. Reconstructing the N surplus estimates allowed insight into the impact of the different input variables; DEP, BNF, manure, mineral fertilizer, and output variables over time. It was seen that regions with a high percentage of agricultural land had the highest N inputs (mineral fertilizer) and N surplus over time. Moreover, it showed the influence of N surplus on TN loadings in the stream.

With increasing fertilizer inputs in the last years and expected increases in DEP at global and European scales until 2100 (Lamarque 2005), N surplus accumulations could rise and impact TN loadings in the stream.
To avoid further accumulation of N surplus in the catchment and increasing TN loadings and legacies, there is an urgent need for change. Change comes in many forms, such as through the implementation of legislation, stakeholder engagement (voluntary), but also the application of adequate measures. Such measures should consider the whole nitrogen transfer continuum, from source to mobilization, to delivery and impact (Haygarth et al. 2005). Measures focusing on increasing NUE and reducing mineral fertilizer inputs in the catchments should be applied. For example, through the application of the “4R Framework” goals, right source, right amount, right timing, and right placement of fertilizer applications (Drechsel et al. 2015).

In the studied catchments E23 and Tullstorpsån, reports showed the low short-term effect of the implementation of measures such as structural liming, cover crops, buffer zones, wetlands, and two-stage ditches on TN loading (Bieroza et al. 2018; Tullstorpsån Ekonomisk förening 2019). A possible reason behind the high nutrient levels could be the increasing fertilizer rates, but also the possible legacy storage.
Estimating legacies also plays an important role in reducing N fertilizer input and N surplus in the catchment. Biogeochemical legacies stored in the organic matter of the unsaturated soil layer could be a source of N for the crops, hence reducing the need for fertilizer application (Basu et al. 2022). This underlines the importance

of applying measures to reduce fertilizer input, but also the necessity to estimate legacies.

Our research provided insight into long-term, N inputs, outputs, and N surplus in the catchments Tullstorpsån, E23, E21, and Silverbäcken, and reduced uncertainties at catchment level. Considering past, current, and future pollution of nutrients and legacies in headwater stream catchments is an important step to reduce the environmental impacts on surface waters.

This study showed that to understand all complex interactions and identify legacies and time lags at catchment scale, further research is needed. For this purpose, our N surplus results could be used as an input parameter in the process-based legacy model ELEMeNT (Van Meter et al. 2017; Basu et al. 2022; Marques et al. 2022).

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Popular science summary

Nitrogen accumulation in agricultural land and streams. How 170 years of nitrogen inputs have impacted water quality

Nitrogen is a nutrient that is used as fertilizer in agriculture since most organisms such as crops need it for growth. In the last century, due to an increase in global food demand and agricultural activity, nitrogen use has increased.
High amounts of nitrogen inputs lead to nitrogen emissions and create a global challenge for water quality. Because of the high nitrogen in the stream, algae grow and consume the oxygen, this is what we call the eutrophication of waters, which causes environmental issues such as fish kills.
In Sweden to reduce pollution and improve water quality, environmental legislation and measures have been implemented. Despite those efforts, emissions continue to be high in the streams. The reasons why the pollution in the stream is still high, are multiple. For example, farmers are not always willing to reduce fertilizer application, which leads to an excess of nitrogen in the soil. Another reason for the low reduction is that nitrogen accumulates in the soil and stream over a long time. This accumulation is also called legacy.
Legacies do not impact the water quality right away but create a delay. Meaning that for example nitrogen inputs from 20 years ago, could be emitted in the environment today, impacting the water quality negatively and creating a time lag. A time lag is the time that passes between an action and a reaction. For example, the time that passes between the input and accumulation of nitrogen excess (action) and the increase in nitrogen emissions in the stream (reaction).

To improve the water quality, we need to understand how big these accumulations (legacies) are. For this, we first have to analyse how much nitrogen, for example how, much fertilizer containing nitrogen is applied by farmers. As a second step, we look at the output, meaning how much nitrogen is taken up by the plant. If we subtract Inputs-Outputs for every year over 170 years, we can say how much nitrogen excess was accumulated in the soil. We then compared the excess of nitrogen to the pollution levels we measure in the stream, to see if there is a time lag.

We looked at four agricultural areas in Sweden and found out, that a high percentage of agricultural land and high nitrogen inputs, led to more accumulation of excess nitrogen. The excess of nitrogen led to higher pollution levels in the stream.
Since the 1960s we can see that the overfertilization is the main reason for the excess and accumulation of nitrogen in the stream.

Over time different measures and policies such as a tax that increased the prices of fertilizer, were applied. The higher prices of fertilizer led to the reduction of fertilizer application by the farmers. When the tax was cancelled, we could see that the fertilizer amounts increased again. Therefore, if we want to improve the water quality of our streams, it is important to work at the source and reduce the amount of fertilizer inputs over a long period.

Moreover, in the studied areas we were able to see that the pollution levels in the stream increased one year after the application of nitrogen excess. For example, the excess of nitrogen input in the year 2018, led to higher pollution in the year 2019. We were not able to see more interactions between excess in nitrogen and emissions in the stream, which could be interpreted as a legacy.
The behaviour of nitrogen is very complex, therefore, to be able to see if there was an accumulation of nitrogen in the past, we need to compare the excess nitrogen with a longer period of pollution measurements and consider factors such as temperature, rain, and soil type.

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7. Appendix

Appendix 1.: Redistribution of cropland area to fit physical consistency of the max possible area in one grid cell (i), 100 km².

The equations (17-28) used in *Batool et al. 2022* were applied to extract the historical land use changes. An adaption of this method was conducted to estimate the area in the catchments that falls under the classification "other". The adaptations are following:

After linear interpolating the extracted values from the HYDE 3.2 database, the value for the category $A_{HYDEother}(i, y_{1850-2019})$ (ha) was calculated applying eq. (1). The max area (A_{max}) in a grid cell equals 100 km^2. The value *i* is grid cell; *y* refers to the year.

$$A_{HYDEother}(i, y_{1850-2019}) = A_{max} - \left(A_{HYDEcrop}(i, y_{1850-2019}) + A_{HYDEpast}(i, y_{1850-2019})\right) \quad (1)$$

In the paper from *Batool et al. 2022,* the temporal variability for cropland and pastureland is derived in eqs (16-17). To be able to derive the temporal variability $R_{HYDE\text{-}other\ (i,y\ 1850\text{-}2019)}$ for the category "other" the same equation was applied, adapting to calculate the normalised values for the area "other.

$$R_{HYDEother}\ (i, y_{1850-2019}) = \frac{A_{HYDEother}\ (i, y_{2000})}{A_{HYDEother}\ (i, y_{1850-2019})} \quad (2)$$

The normalised values for the category "other" eq. (2), were harmonised using *Wei et al. 2021* "other" estimates for the year 2000. To estimate the proportion of land belonging to the category "other" and harmonising the normalised values to gridded other-land area, eqs. (3-4) were used:

$$A_{Wei\_ramankutty-other}\ (i, y_{2000}) = A_{max} - A_{Wei-crop}\ (i, y_{2000}) + A_{Ramankutty-past}\ (i, y_{2000}) \quad (3)$$

$$A_{other}\ (i, y_{1850-2019}) = A_{Ramankutty-other}\ (i, y_{2000})\ x\ R_{HYDEother}(i, y_{1850-2019}) \quad (4)$$

To calculate the correction ratio for "other" the same method as presented in *Batool et al. 2022* method was applied. FAOSTAT did not provide a dataset for the for the non-agricultural area, therefore the non-agricultural area for Sweden was calculates using eq. (5

$$A_{FAOother}\,(u, y_{1961-2019}) = A_{FAO-Land\,use}\,(u, y_{1961-2019)}) - \left(A_{FAOcrop}\,(u, y_{1961-2019}) + A_{FAOpast}\,(u, y_{1961-2019})\right)$$

(5)

the calculated $R_{A_{other}}(u, y_{1961-2019)})$ was applied to *Batool et al. 2022* eqs. (23-26), to estimate gridded other-land area.

$$C_{A_{total}}(i, y_{1850-2019}) = C_{A_{other\_corrected}}(i, y_{1850-2019}) + C_{A_{crop\_corrected}}(i, y_{1850-2019}) + C_{A_{past_{corrected}}}(i, y_{1850-2019})$$
(6)

The physical consistency A_{max} of one grid cell i was not maintained. The resulting values from $C_{A_{total}}(i, y_{1850-2019})$, eq. (6), either exceeded or were below the 100 km^2, therefore a redistribution of the values was conducted, by normalising the total calculated area with A_{max}. The normalised values were applied to the gridded cropland, pastureland, and other-land estimates.

Appendix 2.: Classification of Monfreda et al. 2008 crops into Einarsson et al. 2021 categories (Table 1)

Table 1. Classification of fodder crops into Einarsson et al. 2021 categories (temporary grassland, lucerne, other leguminous plants, green maize, plants harvested from arable land, and other root crops) (Monfreda et al. 2008; Einarsson et al. 2021).

Einarsson et al. 2021	*Monfreda et al. 2008*
Temporary grassland	Grassness, mixed grass
Lucerne	Alfalfa
Other leguminous plants	Clover, legumes, fornes
Green maize	Maize
Plants harvested from arable land	Oil seed, rye, sorghum, vegetable fodder
Other root crops	Beet, turnip, carrot, swede, cabbage

Appendix 3.: N crop-specific fertilization rates (Table 2)

Table 2. N crop-specific fertilization rates (IFA 2022).

Crop type	N fertilization rate [kg ha^{-1}]
Wheat	125
Rye	75
Barley	75
Oats	75
Triticale	75
Rapeseed	139
Sugar beet	95
Potatoes	92
Grassland	31

Appendix 4.: Fertilizer application rates catchment-temporal and regional-temporal rates (Table 3(a-d))

Table 3. Fertilizer application rates fat catchment level. Tullstorpsån (a), E23 (b), E21 (c) and Silverbäcken (d). (-) signalises that fertilisation rates were not available for those years.

Year	Cereal	Other crops	Pasture	Temporary grassland
1999	125	120	(-)	31
2000	125	120	(-)	31
2001	135	123	(-)	31
2002	135	123	(-)	31
2003	128	121	(-)	31
2004	128	121	(-)	31
2005	137	127	(-)	31
2006	137	127	(-)	31
2007	130	125	(-)	31
2008	130	125	(-)	31
2009	130	135	(-)	31
2010	130	135	62	31
2011	132	136	62	31
2012	132	136	64	31
2013	130	140	64	31
2014	130	140	67	31
2015	144	135	67	31
2016	144	135	67	31
2017	148	122	68	31
2018	148	122	68	31
2019	148	122	68	31

(a)

Year	Cereal	Other crops	Pasture	Temporary grassland
2006	84.80	84.81	31	31
2007	97.31	97.31	31	31
2008	95.18	95.18	31	31
2009	82.15	82.15	31	31
2010	83.01	83.01	31	31
2011	76.42	76.42	31	31
2012	93.36	93.36	31	31
2013	93.84	93.84	31	31

(b)

Year	Cereal	Other crops	Pasture	Temporary grassland
1995	113.89	113.89	31	31
1996	104.49	104.49	31	31
1997	134.74	134.74	31	31
1998	134.74	134.74	31	31
1999	134.74	134.74	31	31
2000	134.74	134.74	31	31
2001	134.74	134.74	31	31
2002	127.11	134.74	31	31
2003	121.95	127.11	31	31
2004	122.52	121.95	31	31
2005	119.06	122.52	31	31
2006	131.61	119.06	31	31
2007	120.55	131.61	31	31
2008	125.93	120.55	31	31
2009	134.74	125.93	31	31
2010	122.16	134.74	31	31
2011	129.23	122.16	31	31
2012	125.24	129.23	31	31
2013	140.33	125.24	31	31
2014	152.06	140.33	31	31
2015	147.43	152.06	31	31
2016	152.52	147.43	31	31
2017	148.69	152.52	31	31
2018	149.50	148.69	31	31
2019	0.00	149.50	31	31

(c)

Year	Cereal	Other crops	Pasture	Temporary grassland
1999	70	55	(-)	31
2000	70	55	(-)	31
2001	96	88	(-)	31
2002	96	88	(-)	31
2003	86	94	(-)	31
2004	86	94	(-)	31
2005	91	68	(-)	31
2006	91	68	(-)	31
2007	103	77	(-)	31
2008	103	77	(-)	31
2009	88	77	(-)	31
2010	88	77	57	31
2011	93	94	57	31
2012	93	94	(-)	31
2013	99	94	(-)	31
2014	99	94	54	31
2015	112	104	54	31
2016	112	104	54	31
2017	116	96	65	31
2018	116	96	65	31
2019	116	96	65	31

(d)

Appendix 5.: Classification of Einarsson et al. 2021 provided livestock manure data

Table 4. Classification of FAOSTAT categories "Applied to soil (N content)" and "Left on pastureland", according to Einarsson et al. 2021 provided livestock manure data.

FAOSTAT	Einarsson et al. 2021
Applied to soils (N content)	Applied to cropland
	Applied to permanent land
	Excreted grazing on cropland
Left on pastureland	Excreted grazing on permanent grassland

Appendix 6.: Input data 16 annual N surplus estimates.

Table 5. Input data for estimation of 16 annual N surplus estimates. Two different fertilizer input rates, four manure input rates and two removal rates were applied.

	N input		N output (Rem_{past})			
N_surplus _ID	Fertilizer source	Manure source	Inp_{past}	Manure source	$C_{Rem\ past}$	N_{losses}
N_surplus _1	FAOSTAT	FAOSTAT	FAOSTAT FAOSTAT	FAOSTAT	0.6	0.2
N_surplus _2	FAOSTAT	Einarsson et al.	FAOSTAT Einarsson et al.	Einarsson et al.	0.6	0.2
N_surplus _3	Einarsson et al.	FAOSTAT	Einarsson et al. FAOSTAT.	FAOSTAT	0.6	0.2
N_surplus _4	Einarsson et al.	Einarsson et al.	Einarsson et al. Einarsson et al.	Einarsson et al.	0.6	0.2
N_surplus _5	FAOSTAT	FAOSTAT_ adapted	FAOSTAT FAOSTAT_a dapted	FAOSTAT_ad apted	0.6	0.2
N_surplus _6	FAOSTAT	Einarsson et al. adapted	FAOSTAT Einarsson et al. adapted	Einarsson et al. _adapted	0.6	0.2
N_surplus _7	Einarsson et al.	FAOSTAT_ adapted	Einarsson et al. FAOSTAT_a dapted	FAOSTAT_ad apted	0.6	0.2
N_surplus _8	Einarsson et al.	Einarsson et al. adapted	Einarsson et al. Einarsson et al. adapted	Einarsson et al. _adapted	0.6	0.2
N_surplus _9	FAOSTAT	FAOSTAT	FAOSTAT FAOSTAT	FAOSTAT	0.5	0
N_surplus _10	FAOSTAT	Einarsson et al.	FAOSTAT Einarsson et al.	Einarsson et al.	0.5	0
N_surplus _11	Einarsson et al.	FAOSTAT	Einarsson et al. FAOSTAT.	FAOSTAT	0.5	0
N_surplus _12	Einarsson et al.	Einarsson et al.	Einarsson et al. Einarsson et al.	Einarsson et al.	0.5	0
N_surplus _13	FAOSTAT	FAOSTAT_ adapted	FAOSTAT FAOSTAT_a dapted	FAOSTAT_ad apted	0.5	0
N_surplus _14	FAOSTAT	Einarsson et al. adapted	FAO Einarsson et al. adapted	Einarsson et al. _adapted	0.5	0
N_surplus _15	Einarsson et al.	FAOSTAT_ adapted	Einarsson et al. FAOSTAT_a dapted	FAOSTAT_ad apted	0.5	0

N_surplus _16	Einarsson et al.	Einarsson et al. adapted	Einarsson et al. Einarsson et al. adapted	Einarsson et al. _adapted	0.5	0

Appendix 7.: Data land use distribution

Table 6. Trend analysis of cropland pastureland and other land and mean land use distribution of studied catchments for the period 1850-2019. A significant value corresponds to p≤0.05. Tau portrays the degree of the trend. A linear increase or decrease is proven when p ≤ 0.05* p ≤ 0.01** and p ≤ 0.001***.

Site	Land use	Mean	Standard deviation	Tau-value	p-value
Tullstorpsån	Cropland	84.50	2.17	-0.07	0.16
	Pasture	5.36	1.21	0.18	< 0.001***
	Other land	10.14	1.12	0.05	0.32
E23	Cropland	64.12	4.29	-0.98	< 0.001***
	Pasture	1.30	0.27	0.57	< 0.001***
	Other land	34.58	4.11	0.40	< 0.001***
E21	Cropland	93.11	2.69	-0.36	< 0.001***
	Pasture	2.10	2.46	0.27	< 0.001***
	Other land	4.79	0.33	0.27	< 0.001***
Silverbäcken	Cropland	26.16	2.91	-0.57	< 0.001***
	Pasture	28.34	1.23	0.82	< 0.001***
	Other land	39.93	2.46	0.26	< 0.001***

Appendix 8.: Yield (kg ha^{-1}) of studied catchments

Table 8. Estimated decadal, 1961-2019 yield (kg ha^{-1}) amounts for the crops wheat, rye, rapeseed, oats, barley, triticale, sugar beet and potatoes in the studied catchments.

Year/ Site	Crop							
2019	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	6521.25	5785.90	4849.87	4683.09	6272.61	3228.93	0.00	27227.09
E23	6598.39	5854.34	4907.25	4738.49	6346.81	3267.13	0.00	27549.17
Silverbäcken	3378.46	2664.83	2412.67	2408.36	3225.80	1720.11	36513.67	16228.66
Tullstorpsån	8815.62	7811.11	6491.33	4698.12	6292.74	4009.17	74185.55	39527.19
2010	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	4881.57	4291.91	3732.51	3573.74	4471.38	2353.95	0.00	23525.59
E23	4886.10	4295.90	3735.97	3577.06	4475.54	2356.14	0.00	23547.43
Silverbäcken	2497.83	1952.38	1833.93	1815.21	2271.15	1238.54	25825.06	13849.59
Tullstorpsån	6579.73	5777.23	4981.16	3574.71	4472.60	2914.20	52968.37	34053.50
2000	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	5282.24	4660.93	3650.13	3849.51	4544.78	2284.76	0.00	23000.40
E23	5282.24	4660.93	3650.13	3849.51	4544.78	2284.76	0.00	23000.40
Silverbäcken	2660.65	2087.15	1765.45	1924.76	2272.39	1183.36	22634.91	13329.01
Tullstorpsån	7117.85	6272.26	4869.90	3849.51	4544.78	2827.77	47148.74	33284.24
1990	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	6189.73	4428.52	4525.48	4617.45	0.00	2443.15	0.00	28840.18
E23	6111.50	4372.55	4468.29	4559.10	0.00	2412.27	0.00	28475.72
Silverbäcken	2934.58	1866.57	2060.24	2173.09	0.00	1191.06	27093.30	15731.33
Tullstorpsån	8275.48	5912.90	5990.56	4581.35	0.00	3000.17	59489.44	41408.82
1980	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	4182.08	3287.59	3513.65	3914.30	0.00	1893.34	0.00	23173.38
E23	4018.33	3158.87	3376.08	3761.04	0.00	1819.21	0.00	22266.03
Silverbäcken	1984.61	1386.98	1601.11	1843.90	0.00	923.89	23029.67	12652.18
Tullstorpsån	5579.07	4379.94	4640.98	3875.19	0.00	2319.91	50408.58	33199.50
1970	Wheat	Rye	Barley	Oats	Triticale	Rapeseed	Sugar beet	Potatoes
E21	3745.65	2867.95	3343.42	3814.43	0.00	2131.39	0.00	24806.26
E23	3574.30	2736.75	3190.47	3639.93	0.00	2033.89	0.00	23671.45
Silverbäcken	1753.89	1193.87	1503.30	1772.99	0.00	1026.23	20418.40	13363.81
Tullstorpsån	4995.43	3819.78	4414.87	3775.24	0.00	2610.85	45281.60	35528.74

1961								
E21	3708.24	2698.63	3353.79	3418.71	0.00	2715.98	0.00	17300.68
E23	3493.26	2542.18	3159.36	3220.51	0.00	2558.52	0.00	16297.67
Silverbäcken	1701.07	1100.54	1477.30	1556.74	0.00	1281.11	22427.77	9130.81
Tullstorpsån	4992.21	3628.18	4470.36	3415.52	0.00	3358.34	51249.34	25012.74

Appendix 9.: Mann-Kendall trend analysis (2010-2019)

Table 10: Mann-Kendall trend test results showing p and tau values of annual climatic drivers (precipitation and temperature), annual total N load and N surplus, atmospheric N deposition (DEP), biological N fixation (BNF), fertilizer input, evapotranspiration (ET), flow and mean annual yield. The highlighted values show a significant trend ($p \leq 0.05$). The τ value portrays the trend. A linear increase or decrease is proven when p ≤ 0.05* p ≤ 0.01** and p ≤ 0.001***.

Site	TN load (kg ha^{-1})				N surplus kg ha^{-1})			Precipitation (mm)			
	τ-value	p-value	Mean	SD	τ-value	p-value	Mean ± SD	τ-value	p-value	Mean	SD
Tullstropsån	-0.27	0.32	17.87	3.93	0.08	0.86	36.44 ± 6.07	-0.38	0.15	823.65	124.50
E23	-0.11	0.72	5.37	2.57	-0.47	0.07	12.48 ± 1.35	-0.33	0.21	646.15	114.07
E21	0.24	0.37	22.88	6.14	0.29	0.28	42.35 ± 5.85	-0.24	0.37	631.69	104.85
Silverbäcken	-0.38	0.15	0.85	0.37	-0.47	0.07	14.51 ± 0.99	-0.20	0.47	588.85	98.67

Site	Temperature (°C))				Sum Yield (kg ha^{-1})			DEP (kg ha^{-1})			
	τ-value	p-value	Mean	SD	τ-value	p-value	Mean ± SD	τ-value	p-value	Mean	SD
Tullstropsån	0.56	0.03*	8.86	0.90	0.51	0.05*	131806.75 ± 11761.62	-0.37	0.15	9.16	2.08
E23	0.41	0.13	7.76	0.98	0.47	0.07	53123.80± 5381.18	-0.64	0.01**	5.07	1.29
E21	0.45	0.08	7.37	1.01	0.47	0.07	52697.345 ± 5258.73	-0.56	0.03*	5.81	1.49
Silverbäcken	0.56	0.03*	8.24	0.85	0.51	0.05*	59061.00 ± 5378.64	-0.51	0.05*	7.06	1.30

Site	Fertilizer (kg ha^{-1})				BNF (kg ha^{-1})			ET (mm)			
	τ-value	p-value	Mean	SD	τ-value	p-value	Mean ± SD	τ-value	p-value	Mean	SD
Tullstropsån	0.64	0.01**	57.48	4.52	-0.38	0.15	7.18 ±	0.42	0.11	0.68	0.04

							0.19				
E23	0.56	0.03*	10.89	0.70	-0.29	0.28	3.01 ± 0.06	0.64	0.01*	0.73	0.06
E21	0.51	0.05*	39.18	3.29	-0.38	0.15	9.27 ± 0.25	-0.16	0.59	0.41	0.17
Silverbäcken	0.64	0.01**	11.04	0.73	-0.38	0.15	2.50 ± 0.04	0.20	0.474	0.97	0.01

Site	Flow ($m^3 s^{-1}$)			
	τ-value	p-value	Mean	SD
Tullstropsån	-0.42	0.11	0.47	0.11
E23	-0.63	0.02*	0.04	0.01
E21	-0.47	0.07	0.07	0.01
Silverbäcken	-0.33	0.21	0.17	0.06

Appendix 10.: PCA analysis with one year shift of TN load

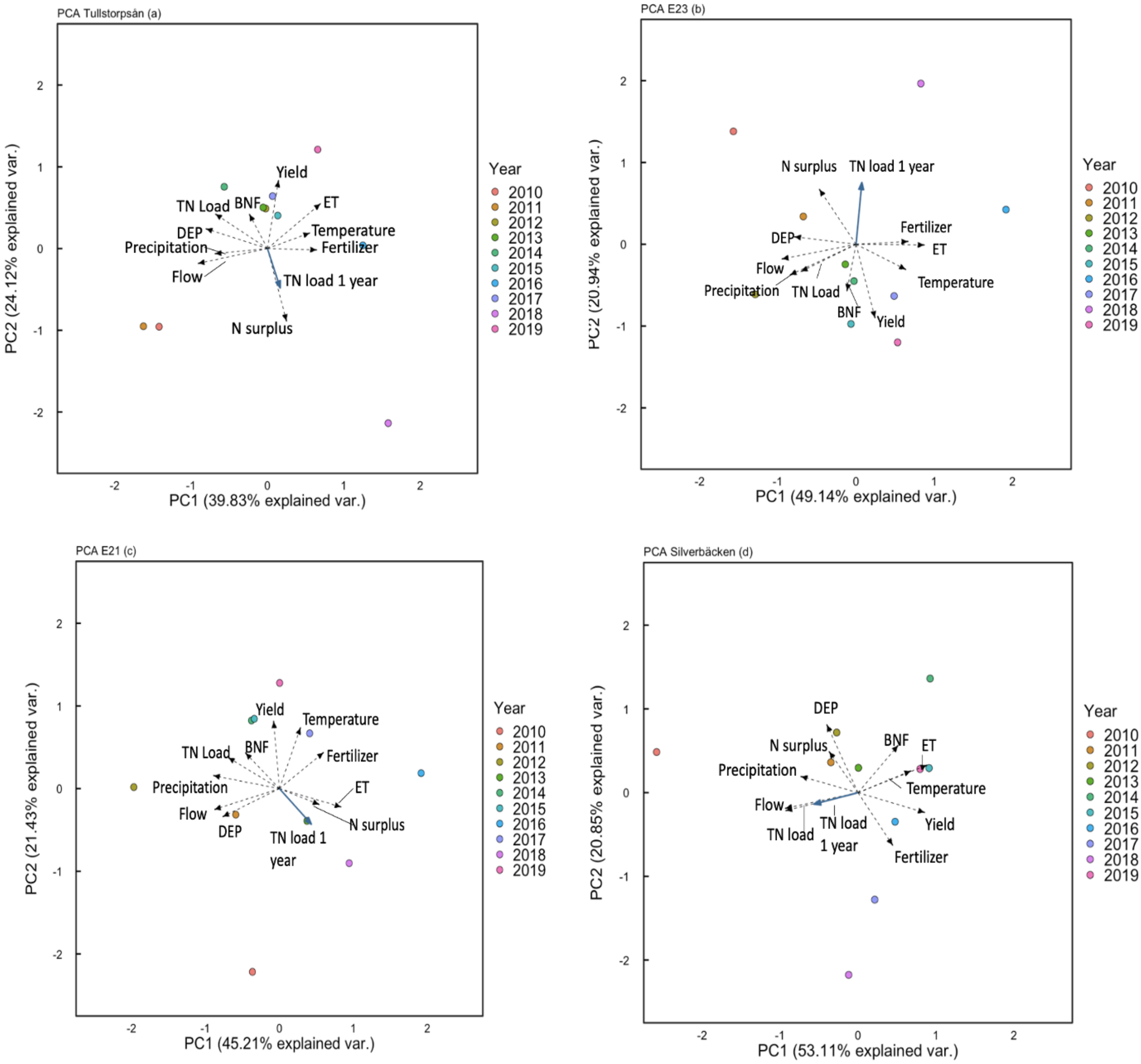

Figure 1 (a-d) *PCA analysis of annual variables with one-year total nitrogen shift, 2010-2019. The plots show inputs, outputs, climate variables, and flow. The circles represent different years. The blue arrow represents the response factor total nitrogen load (kg ha^{-1}). Actual evapotranspiration (ET), Atmospheric N deposition (DEP), Biological N fixation (BNF), Total Nitrogen load (TN load).*

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