ESTIMATION OF NITROGEN CONCENTRATIONS FROM ROOT ZONE TO MARINE AREAS AROUND THE YEAR 1900

Scientific Report from DCE – Danish Centre for Environment and Energy No. 241 2017

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Data sheet

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Abstract: Factors affecting the nitrogen concentration from root zone to the coast around the year 1900 have been investigated. The aim of this investigation is to establish an estimate of the nitrogen concentration in the water discharged into marine areas around the year 1900. Besides this, the national and international literature have been consulted to find references to nitrogen concentrations around the year 1900. The best estimate of the nitrogen concentration back in 1900 is within the range of 1-2 mg N/l.

Keywords: Reference condition, nitrogen, agricultural practice year 1900, nitrogen retention, nitrogen deposition year 1900, water discharge year 1900

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Contents

Preface

Denmark decided to use ecological conditions in the year 1900 as a reference point defining high ecological status in coastal waters, based on which an acceptable deviation defines 'good ecological status' – the status necessary in order to comply with the EU Water Framework Directive. One of the parameters that affect the ecological status is the nitrogen loads from e.g. agricultural activities, such as fertilization and soil cultivation, and point sources, such as households. As part of environmental management, it is therefore a goal to reduce nitrogen loads to a level that ensures good ecological status.

In order to use the year 1900 as a reference point, an estimation of the actual concentration and the water discharge that year is needed. However, lack of data from that period complicates the estimation.

Aarhus University decided to undertake an in-depth analysis of the main factors that may have influenced the entire nitrogen cycle, i.e. land use and agricultural practices affecting nitrogen losses from the root zone, as well as landscape features affecting nitrogen retention in groundwater and lakes and streams around the year 1900 and, thus, influenced the level of nitrogen concentration in the stream water that reached the coast. This study should enable a coherent description of the nitrogen cycle from the soil to the coast going back in time.

The overall aim of the analysis is to improve the scientific basis, including uncertainty assessments for estimation of nitrogen concentration and amounts discharged to the coastal zone around the year 1900.

The work was organized internally at Aarhus University, led by a steering group:

Niels Halberg, DCA – Danish Centre for Agriculture and Food
Hanne Bach, DCE – Danish Centre for Environment and Energy
Poul Nordemann Jensen, DCE – Danish Centre for Environment and Energy
Jørgen Eriksen, Department of Agroecology
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Brian Kronvang, Department of Bioscience
Jørgen Windolf, Department of Bioscience

The different chapters have been written by researchers from the departments Bioscience, Environmental Science and Agroecology, and the individual chapters have been quality assured by researchers not involved in the project.

The preliminary results were presented and discussed at an international workshop together with presentation of experiences from Germany, Sweden and Holland in November, 2016. At the workshop, a number of organizations were present (representing e.g. farmers and nature conservation interests) together with representatives from the Ministry of Environment and Food.

A draft report was presented and discussed at a national workshop in April, 2017, with the participation of the above-mentioned organizations and the Ministry.

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We want to thank Bo Gustavson, Stockholm University, Alena Bartosova, SMHI, Flemming Vejen, DMI, Markus Venohr, IG-Berlin, Birgitte Hansen, GEUS and Erwin van Boekel, Wageningen University for presentations at the national work shop in December 2016.

We especially want to thank the two reviewers, Markus Venohr, IGB, Germany and Lars Bergström, SLU, Sweden for their effort and very useful comments.

Introduction

A determination of the reference conditions for the various water types (rivers, lakes and coastal areas) is a central element in the Water Framework Directive (WFD), as it sets the starting point for determining the boundaries between the quality classes, especially the boundary between good and moderate quality, as this line in general defines whether or not a certain water body fulfils the objectives. This is illustrated in Fig. 1.

The reference condition in the WFD is defined as high status, see Box 1.

Box 1:

WFD, annex II.3: : «The specific biological reference conditions shall be established – for that surface water body type at high ecological status»

General definition of high status, Annex V:
There are no, or only very minor, anthropogenic alterations to the values of the physico-chemical and hydromorphological quality elements for the surface water body type from those normally associated with that type under undisturbed conditions. The values of the biological quality elements for the surface water body reflect those normally associated with that type under undisturbed conditions, and show no, or only very minor, evidence of distortion. These are the type-specific conditions and communities.

The guideline (WFD CIS) outlines a number of methods for setting reference conditions – as a prioritized approach:

- An existing undisturbed site or a site with only minor disturbance
- Historical data and information
- Models
- Expert judgement.

When it comes to coastal areas, there are no sites with a high status in Denmark or in areas comparable to Danish coastal areas. Therefore, option A is not applicable and reference conditions must be established using one of the remaining methods.

In general, the marine reference situation in the Baltic area, including Denmark, is defined as the ecological status corresponding to a period around the year 1900 (+/- 15 years), where human impact was supposedly limited (Kaas 2015).

For the ecological quality element - eelgrass - it has been possible to establish a reference condition around 1900 for Danish coastal areas from historical information. Therefore, the boundary between good and moderate for this element is a percentage (26 %) deviation from the reference = the 1900 occurrence of eelgrass.

For other quality elements like chlorophyll a, it has been necessary to use models to determine the reference concentration. A reference value for load of nutrients, including nitrogen, is needed to be able to run the models and establish a reference situation around the year 1900 for chlorophyll a.

It is stated in Kaas, 2015:

”The overall procedure to establish a reference condition for chlorophyll and the corresponding boundaries between quality classes for coastal areas is as follows:

- Establish input data that represents the conditions around 1900. Input data is used for model simulations of a reference situation (reference scenario).
- For all coastal areas covered by either a statistical or mechanical model, a reference scenario is established using input data that represents the conditions around 1900, and the chlorophyll values in a reference situation are estimated.”

The reference situation/concentration established using the models is the starting point for setting the boundaries between quality classes, including the important boundary between good and moderate status – see Fig. 1.

Figure 1. The 5 quality classes. The arrow shows the boundary between fulfilment (above) and not fulfilment (below) of the objective.

One approach presented in Bøgestrand (2014a+b), was to use the concentrations from small water courses in catchments with only minor human impact – both from diffuse and point sources as a proxy for background concentrations and further on as a proxy for the nitrogen concentrations around the year 1900.

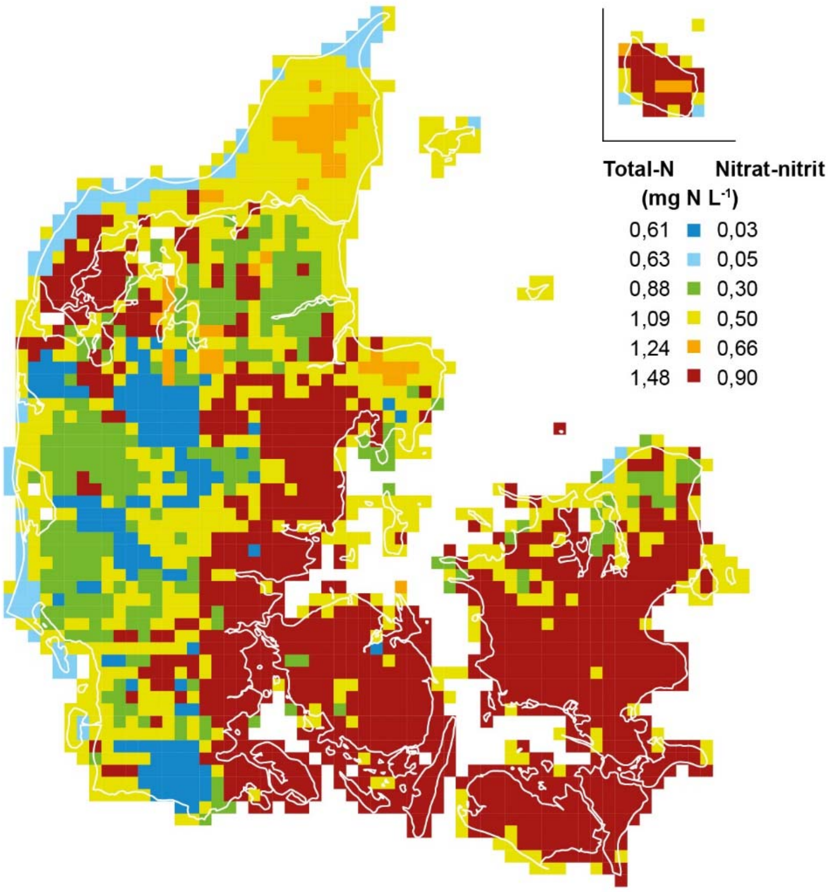


Figure 2. Nitrogen concentrations as total N and nitrate in a reference situation according to Bøgestrand (2014a+b).

As small watercourses are distributed over most of the country, it was possible to establish a geographically distributed estimate of the nitrogen concentration from the data basis, as shown in Fig. 2. In general, the concentration of total N was low, < 1 mg N/l, in most of the western and northern parts of Jutland (mainly sandy areas), whereas the concentrations in the remaining part of Denmark were in the range 1-1.5 mg N/l (mainly loamy soil).

The overall aim of the present analysis is to improve the scientific basis, including uncertainty assessments for estimation of nitrogen concentration in water discharged to the coastal zone around the year 1900. So other possible pressures on the marine environment will not be discussed in this report.

It is important to emphasize, that very little historical data is available for nitrogen concentrations in rivers – be it Danish or European – around the year 1900. Therefore, apart from this restricted information, the estimation of the reference concentration is based on a number of indications, which together with the few actual observations hopefully will provide a picture of the reference concentration. It is obvious that the individual estimates are quite uncertain, an uncertainty which cannot be quantified. It is assumed that if the majority of the indications point in the same direction (within the same range of N-concentration), this will strengthen the final estimate.

To get a coherent view of the situation around 1900, it is necessary to look at a number of important factors that might influence nitrogen content from the field to the sea. These factors are shown in Fig. 3.

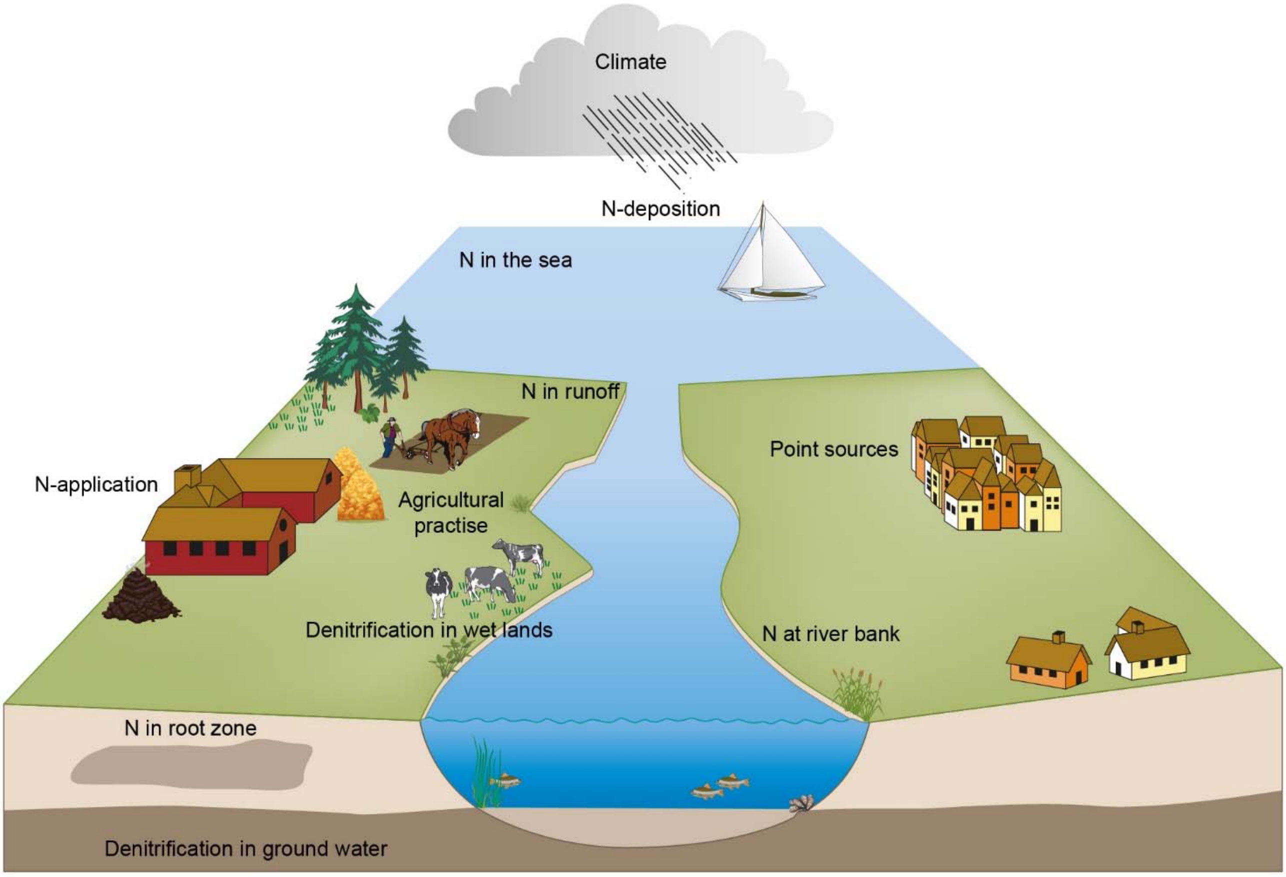


Figure 3. Conceptual illustration of the most important sources and elements influencing the nitrogen concentration around the year 1900.

These factors are further described and discussed in the following chapters:

- Chapter 1 – Describes the climate and hydrological conditions
- Chapter 2 – Agricultural practices and the resulting concentration of nitrogen in the root zone
- Chapter 3 – Deposition of N mainly on land
- Chapter 4 – Point sources
- Chapter 5 – Historical, models etc. providing estimates of the N-concentrations in rivers based on different sources / methods
- Chapter 6 – Nitrogen-retention / denitrification mainly in surface areas like wetlands, lakes etc. using models providing an estimate of the combined retention from root zone to the seashore.
- Chapter 7 – A synthesis, where the findings from the previous chapters are compiled. It is discussed whether the different elements described point in a certain direction and whether there is coherence in the information.

The most important factor in relation to nitrogen discharge into coastal areas is the land use. The overall land use around 1900 (and 2000) as reported in Levin & Normander (2008) is shown in Table 1:

Table 1. Land use in 1888 and 2000. Numbers are rounded compared to Levin & Normander (2008).

Type	Year 1888	Year 2000
Arable land	66%	57 %
Forrest	6 %	13 %
Permanent grass types (meadows, marshes)	9 %	4 %
Heath, dunes and bogs	15%	5 %
Urban areas including infrastructure	3 %	10 %
Fresh water areas	No data	2 %
Other types	1 %	9 %

It should be noted that the total Danish area was smaller in 1888 compared to 2000 due to foreign occupation of the southern part of Jutland until 1920. It is assumed, that the relative distribution between the area types was the same also in the occupied part of Denmark in 1888.

Summary

Determining the reference conditions for the various water types (rivers, lakes and coastal areas) is a central element in the Water Framework Directive (WFD), as it sets the starting point for determining the boundaries between the quality classes, especially the boundary between good and moderate quality, as this line in general defines, whether or not a certain water body fulfils the objective.

Aarhus University has decided to undertake an in-depth analysis of the main factors, that may have influenced the entire nitrogen cycle and to investigate the factors that may have influenced the annual mean nitrogen (N) concentrations from source to sea around the year 1900 in Denmark in order to estimate the N-concentration at that time. Fulfilling such an aim require, that a range of parameters known to influence the N cycle, such as climate, hydrology, land use, agricultural practices, drainage, landscape, etc. , are described for the period around the year 1900, as very few measurements of N concentrations in streams and rivers are available from that time.

A number of different methods, that could assist in estimating and / or finding indications of the N concentrations around the year 1900 in soil water, groundwater and surface waters, are discussed – such as agricultural statistics, trends in climate and hydrology, review of historical measurements found in the international and Danish literature or use of model estimates. The aim of the analysis is to conduct an assessment of, what this range of indicators in combination shows about, what might have been the N-concentration in the water discharging into marine areas around the year 1900.

The main findings of the report are listed below.

- The runoff from the Danish area was app. 25% lower in the year 1900 than today.
- The temperature has increased app. 1.5 °C since the year 1900
- Around the year 1900, 67% of the land was cropped, 8% was bare fallow and 25% was nature areas.
- For cropped land it has been estimated, that the nitrate concentration in the leachate of the root zone would have been 12 mg N / l around the year 1900, which is in line with today's organic cash crop farming.
- Approximately 22% of the area used for agriculture was tile drained around the year 1900 (16% of the total area), as compared with today where app. 50% of the agricultural area is tile drained
- The atmospheric deposition on land has been estimated to 4 kg N / ha in the year 1900 – about one third of today's deposition level.
- Around the year 1900 point sources (cities, industry) may have had an impact locally, but the point source load of nitrogen was too low to have influenced the overall national nitrogen budget.
- Model calculations have shown, that the total retention (from root zone to the coast) was considerably higher than today – in the range of 76 – 87% around the year 1900.
- Routing of the estimated nitrogen concentration in the root zone combined with the estimated nitrogen removal in ground and surface water around the year 1900 results in a concentration range of 1-2 mg N / l.

- Empirical models, measurements from the time around the year 1900 and similar international analyses all show N-concentrations close to or within the range of 1-2 mg N/l some without accounting for full retention in surface water.

In this attempt to estimate an N-concentration in water running to the sea around the year 1900, many assumptions have been made for nearly all elements included in the analysis. This means that the result of the individual element is probably associated with relatively high uncertainty, but nearly all elements point in the same direction (same range).

Overall conclusion

From the indications etc. described in the chapters and synthesis above, the best estimate of the nitrogen concentration in the water running into Danish coastal areas at the time around the year 1900 is within the range of 1-2 mg N/l.

Udvidet sammenfatning

Der findes kun meget få og spredte målinger af kvælstofindholdet i f. eks. danske vandløb omkring år 1900. For at kunne estimere en koncentration af kvælstof i det vand, der løb til de danske kystområder omkring år 1900, er det derfor nødvendigt at anvende andre supplerende metoder.

Formålet med denne rapport har været at belyse de faktorer, som potentielt kan have haft indflydelse på koncentrationen af kvælstof i det vand, der løb ud i de danske kystnære områder omkring år 1900. For at nå dette formål er der beskrevet en række relevante parametre som klima, vandafstrømning, landbrugspraksis, landskabselementer, punktkilder m.m. med udgangspunkt i år 1900.

Der indgår ligeledes i rapporten en opsamling af de få danske og udenlandske målinger af kvælstof i vandløb fra den tid samt andre relevante sammenstillinger og modelleringer.

Resultaterne fra denne analyse har vist, at kvælstoftabet fra landbrugsarealerne var den dominerende kilde omkring år 1900, hvorfor der er fokuseret på denne kilde.

Dansk landbrug og landskab omkring år 1900

Dansk landbrug omkring år 1900 var grundlæggende forskelligt fra nutidens, men dækkede rent faktisk en større del af det samlede danske areal end tilfældet er i dag. De vigtigste forskelle ift. nutidens landbrugsdrift er:

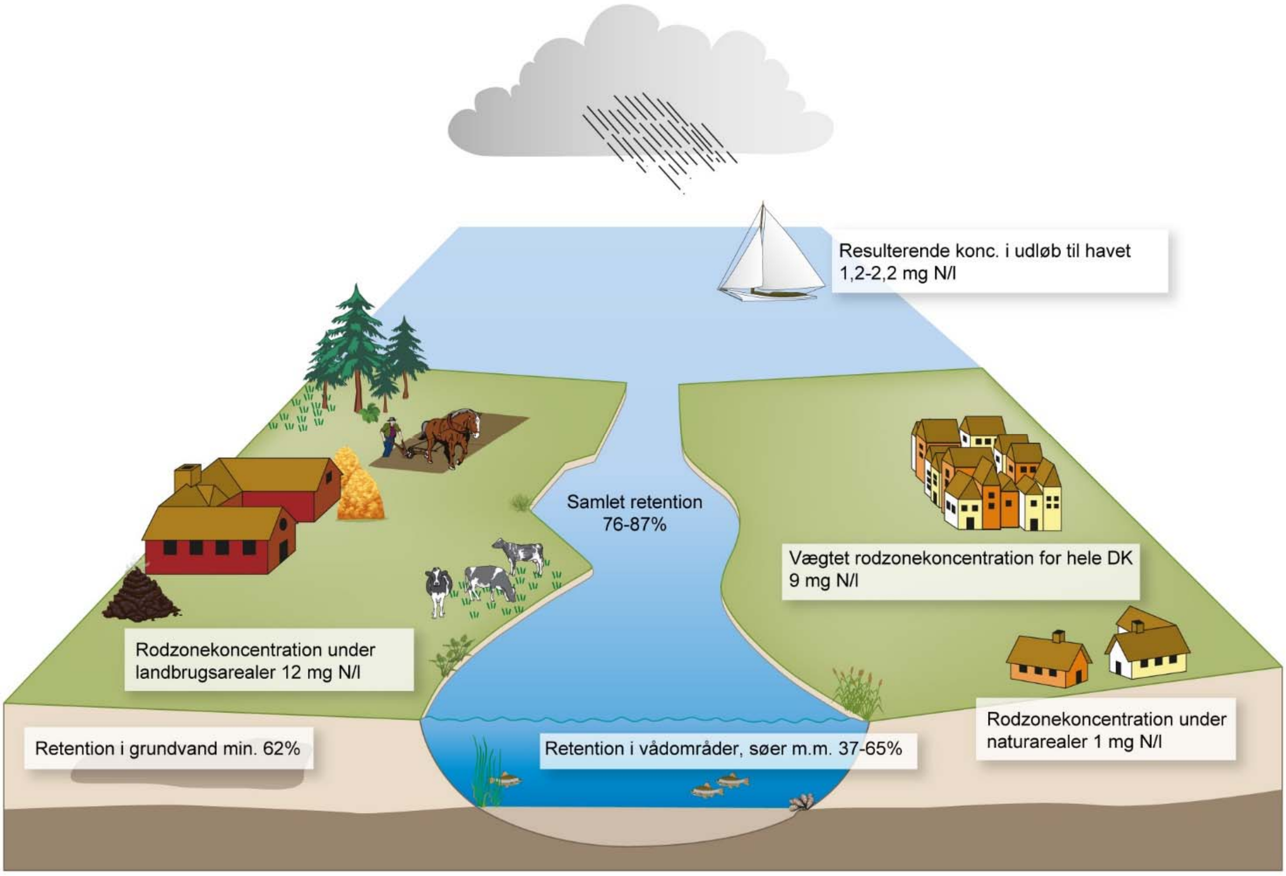
- Bedriftsstørrelse – der var langt flere men mindre landbrug
- Husdyrhold – mere differentieret med flere dyrearter på samme ejendom
- Kun brug husdyrgødning (ingen handelsgødning).
- Ingen brug af kemiske plantebeskyttelsesmidler
- Lav mekaniseringsgrad ("simple" maskiner)
- Lavtydende afgrøder
- Rørdræning mindre udbredt.

Praksis var også på mange områder anderledes omkring år 1900 med f.eks. braklægning af marker (ukrudtsbekæmpelse) og udbringning af gødning om efteråret – begge dele kan have betydet relativt høje kvælstoftab. Derudover har opdyrkning, herunder rørdræning, af ny landbrugsjord i den sidste halvdel af 1800-tallet kunnet medføre et øget N-tab i en årrække derefter som følge af omsætning af kvælstof i jordens organiske pulje.

Den ekstensive dyrkning var også årsagen til, at landskabet så meget anderledes ud omkring år 1900 med en meget højere andel af græsarealer, ikke-drænede arealer, vådområder og vandløb, som ikke var blevet udrettede. En anden faktor, som er ændret igennem de seneste ca. 100 år, er klimaet, hvor der omkring år 1900 var lavere temperatur, mindre regn og dermed en lavere vandafstrømning i vandløbene.

Kvælstoffets vej fra marken til kystvandene

Koncentrationen af kvælstof reduceres væsentligt på sin vej fra marken / rodzonen til kystvandene som følge af en kvælstofreduktion (denitrifikation) i grundvand og overfladevandsområder (inkl. vådområder), som illustreret i figur S.1.



Figur S.1. Diffus kvælstofkoncentration samt retention fra rodzone til kystområder på nationalt niveau.

Kvælstofkoncentration i rodzonen

Koncentrationen af kvælstof i det vand, der sivede ud fra de dyrkede marker omkring år 1900, er estimeret til typisk at have ligget i området 5-15 mg N / l afhængig af afgrødetype og landbrugspraksis. Koncentrationen har været højere fra områder med en forlænget græsningsperiode, områder med efterårsudbringning af husdyrgødning og braklagte områder og lavere under permanente græsarealer. I nutidige forsøg med økologisk landbrug (og som har sammenlignelige forhold mht. gødningstyper som omkring år 1900) er det estimeret, at koncentrationen af kvælstof i gennemsnit er 12 mg N / l – med lavere koncentration under permanente græsområder og højere under brakområder uden bevoksning.

For at finde en vægtet gennemsnitskoncentration for hele landet omkring år 1900 er der anvendt en koncentration på 12 mg N / l under dyrkede arealer og 1 mg N / l under ikke-dyrkede arealer. Det er videre antaget, at ca. 75% af det daværende danske areal var anvendt til landbrug i år 1900, og at de resterende 25% var naturarealer dvs. arealer som ikke evt. kun meget ekstensivt har været anvendt til landbrugsformål. Det er i denne sammenhæng antaget, at veje, byer m.m. kun optog en meget lille del af arealet.

Ved at kombinere disse antagelser kan der estimeres en vægtet kvælstofkoncentration på 9 mg N/l i det vand, der sivede ud fra jorden (rodzonen) omkring år 1900.

Kvælstoffjernelse – denitrifikation

Landskabet i år 1900 var meget vigtigt for, hvad der skete med den kvælstof, der forlod rodzonen. Kvælstof kan fjernes (omsættes til luftformigt kvælstof) via den proces som kaldes denitrifikation (også kaldet N-retention) i grundvand. Da det drænede areal omkring år 1900 var betydeligt mindre end i dag, sivede en større del af vandet ned til grundvandet og ikke via dræn direkte ud til vandløbene. Hertil kommer, at et mere tørt og koldt klima omkring år 1900 har forøget denne retention i grundvandet. Det betyder, at den generelle kvælstoffjernelse i grundvand omkring år 1900 var højere end de 62%, som er fundet med de nuværende forhold. Det har dog ikke været muligt at fastsætte fjernelsen omkring år 1900 nærmere, hvorfor de 62% er anvendt i de videre estimater som et absolut minimum.

Det fremgår af kortmateriale fra omkring år 1900, at vådområderne langs vandløb og søer var betydeligt større omkring år 1900 sammenlignet med i dag. Dette betyder, at den samlede N-fjernelsen i disse vådområder var større, end den er i dag. I 3 forskellige områder i Danmark er der fundet en fjernelse på 37-65% med de vådområder, der var omkring år 1900. Da disse 3 områder repræsenterer forskellige landsdele af det daværende danske område er det antaget, at dette interval er et rimeligt estimat for hele det danske område omkring år 1900. Den samlede kvælstoffjernelse fra rodzone til kystvand (grundvand + overflade) kan derfor estimeres til 76-87% - sammenlignet med en fjernelse i dag på 65-70%.

Punktkilder og kvælstofafsætning fra luften

Det er estimeret, at der i gennemsnit blev afsat ca. 4 kg N/ha fra luften (sammenlignet med i dag ca. 12 kg N/ha). Der er antaget, at denne kvælstofafsætning er indregnet i den estimerede rodzoneudvaskning – både på landbrugsarealer og naturarealer.

En direkte kvælstoftilførsel kunne ske fra punktkilder (byspildevand, industrier), og lokalt har det givet været en væsentlig kilde. Det er dog antaget, at på nationalt plan var bidraget fra punktkilder ubetydeligt omkring år 1900.

Resulterende kvælstofkoncentration i afstrømningen til kystvande omkring år 1900

Ud fra de beskrevne tilgange og antagelser m.m. kan det estimeres, at den resulterende kvælstofkoncentration i det vand, der strømmede til kystområderne omkring år 1900 har ligget i intervallet 1-2 mg N/l – som afrundede værdier. Det skal igen understreges, at der her er tale om et nationalt estimat, og at der har været betydelige geografiske forskelle.

Supplerende estimater af kvælstofkoncentrationen i overfladevand omkring år 1900

I rapporten er der også opsamlet såvel danske som internationale informationer, data m.m. for estimater af kvælstofkoncentrationen omkring år 1900. Hovedresultaterne er vist i tabel S.1, og alle koncentrationer refererer til overfladevand (vandløb). Det er vigtigt at understrege, at koncentrationerne vist i tabel S.1 ikke alle er umiddelbart sammenlignelige, da de dels repræsenterer forskellige former for kvælstof (total kvælstof, nitrat osv.), dels er målt, modelleret m.m. forskellige steder i kredsløbet (primært med / uden fuld fjernelse i overfladevandsområder).

Tabel S.1. Estimater og malinger af kvælstofkoncentration omkring år 1900.

	N form	Koncentration mg N/l	Kommentarer
		Målinger omkring år 1900	
Seks danske vandløb Kapitel 5	NO_3-N + Org. N	0.7 – 2.7	Domineret af org. N. Der er formentlig ikke indregnet fuld overfladeretention.
Større europæiske floder Kapitel 5	NO_3-N	0.1-0.5	Kun oplande med mere end 25 % dyrket areal medtaget. Der er formentlig ikke indregnet fuld overfladeretention
Themsen Kapitel 5	NO_3-N	2	Sandsynligvis lav grundvandsretention og der er formentlig ikke indregnet fuld overfladeretention.
		Modellering	
Baseret på iltet grundvand Kapitel 5	NO_3-N	2	Modelestimat baseret på det nuværende klima og retention – et maksimum estimate.
Modellering med MONERIS Kapitel 5	Total N	0.7	Gennemsnitskoncentration for det tyske opland til Østersøen omkring år 1880.
		Estimater baseret på nutidige data.	
Extrapolation af data 1990-2014 Kapitel 5	Total N	2	Modelestimat baseret på det nuværende klima og retention – et maksimum estimate..
Bøgestrand et al. (2014)	Total N	0.6-1.5	Baseret på data fra 1990-2012. Der er ikke indregnet fuld overfladeretention.
Naturarealer Kapitel 5	Total N	0.1-0.2	Estimeret til 1 mg N/l i rodzonen.

De få konkrete målinger omkring år 1900 i vandløb tyder på, at næsten alt nitrat var fjernet (denitrificeret i grundvand og overfladevand og mest udpræget i sandjordsområder) så det totale indhold af kvælstof primært bestod af organisk bundet N (alger m.m.).

To empirisk baserede modeller samt to estimater baseret på nutidige data indikerer alle en kvælstofkoncentration på max. 2 mg N / l.

Resultaterne præsenteret i tabel S.1 er generelt lavere end eller i den lave ende af intervallet 1-2 mg N / l – hertil skal det tages i betragtning, at en del af resultaterne ikke indregner en fuld fjernelse i overfladevandet.

Geografisk fordeling

Kvælstofkoncentrationen i det vand, der afstrømmede til kystområder omkring år 1900, var formentlig regionalt forskellig afhængig af oplandsfaktorer som landbrugspraksis, jordtype (dvs. grundvandsretention), forskelle i dræningsgrad, forskelle i overfladevandsretention eller forskelle i nedbør. Disse forhold bidrager formentlig til de forskelle i koncentration, der ses i tabel S.1. I Bøgestrand (2014b) var baggrundskoncentrationen geografisk forskellig med generelt lave koncentrationer (< 1 mg N / l) i de vestlige sandede områder af landet og de højeste (1-1,5 mg N / l) i den østlige del af landet.

Usikkerheder

Der er foretaget en række ekspertvurderinger, antagelser m.m. for at komme frem til et estimat for kvælstofkoncentrationen i det vand, der omkring år 1900 strømmede af til kysten. De enkelte elementer kan være behæftet med en relativ stor usikkerhed, men det er vigtigt at have for øje, at næsten alle tilgange til at finde en resulterende koncentration peger i den samme retning (ligger indenfor det samme interval).

Konklusioner

- Vandafstrømningen i år 1900 var ca. 25% lavere end i dag.
- Temperaturen er steget ca. 1.5 °C siden år 1900
- Omkring år 1900 anvendtes ca. 75% af det danske areal til landbrug – ca. 67% i omdrift og ca. 8% som brak. 25% af arealet var naturarealer.
- For det dyrkede areal er det estimeret, at gennemsnitskoncentrationen i rodzonen var ca. 12 mg N/l – svarende til koncentrationen man finder ved nutidig økologisk produktion.
- Ca. 22% af landbrugsarealet var rør-drænet omkring år 1900 sammenlignet med ca. 50% i dag.
- Kvælstofnedfaldet fra luften omkring år 1900 er estimeret til ca. 4 kg N/ha – svarende til ca. 1/3 af det nuværende nedfald.
- Punktkilder (byspildevand, industri m.m.) kan have bidraget væsentlig til kvælstoftilførslen til et lokalt kystområde, men anses på landplan ikke at have været en betydende kilde omkring år 1900.
- Modelberegninger m.m. har vist at den samlede kvælstoffjernelse (retention) fra rodzone til kysten var større i år 1900 end i dag – i størrelsesordenen 76-87%.
- En sammenstilling af de forskellige estimater, antagelser m.m. for kvælstoftab og omsætning har resulteret i en koncentration på 1-2 mg N/l i det vand, der omkring år 1900 løb ud til kystområderne i Danmark.
- Ingen af de empiriske modeller, målinger fra omkring år 1900 m.m. har resulteret i kvælstofkoncentrationer, som ligger væsentligt udenfor intervallet 1-2 mg N/l – uden at der for alle estimater er indregnet den fulde retention i overfladevand.

Dette studie har i forløbet været præsenteret ved to work shops for Miljø- og Fødevareministeriet og interessenter, og et udkast har været til ekstern review hos Markus Vernohr, IGB-Berlin, Tyskland og Lars Bergström, SLU, Sverige.

Overordnet konklusion

Den helt overordnede konklusion på denne analyse er, at det bedste estimat for kvælstofkoncentrationen i det vand, der strømmede af til danske kystområder omkring år 1900, er 1-2 mg N/l - som et nationalt gennemsnit med betydelige geografiske forskelle.

Perspektivering

I rapporten har vi givet nogle indikationer på vigtigheden af de to væsentligste faktorer for en lavere kvælstofkoncentration omkring år 1900 – klimaet og landskabet – men kun for 3 udvalgte oplande i Danmark. For fuldt ud at forstå hvordan de to faktorer agerede sammen for at gøre landskabet mere robust overfor kvælstoftab omkring år 1900 er det nødvendigt med en mere integreret modelanalyse i udvalgte repræsentative oplande i Danmark. En sådan fuldt integreret regional modelanalyse vil ikke kun give et historisk overblik over effekten af kvælstofforurening af grundvand og overfaldevand ved at bidrage til at fastsætte geografisk differentierede referencemål for økosystemer. Den vil også kunne vejlede ved fremtidig administration af landskabet, ikke mindst i lyset af behov for klimatilpasning.

1 Climate and hydrology

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Abstract

Purpose: The purpose is to analyse the trends in precipitation and runoff in Danish streams over a period of nearly 100 years and to provide estimates of stream runoff for different Danish locations around the year 1900.

Materials and methods: Data from 18 stream hydrometric stations with at least 80 years' of runoff data available and five climate stations with daily precipitation and maximum and minimum temperature were analysed. Based on these data series, a statistical test was conducted to investigate the development in stream runoff and precipitation and to predict runoff around the year 1900. The analysis was supported by a numerical rainfall-runoff model (NAM), set up and calibrated for three selected stations based on observed data of runoff, temperature and precipitation.

Results and discussion: A trend of increasing annual runoff was observed at all stream / river hydrometric stations over the past 80 years, with the most pronounced increase being ca. 165 mm (33%) in annual runoff supported by similar trends in annual precipitation. The statistical trend analyses of measured runoff estimated the runoff level in 1900 to have been 5 to 48% lower than at present. The hydrological model NAM estimated a 25 to 37% lower runoff around year 1900. Hence, despite the challenge of quantifying the exact uncertainty of the runoff estimates around year 1900, it is reasonable to conclude that runoff was significantly lower compared to present day conditions.

Conclusions: It was concluded that stream / river runoff and precipitation have generally increased across the country. The largest increase, both in precipitation and runoff, was observed in the southwestern part of Jutland. Based on both measured data, the statistical models and the NAM model, the stream / river runoff around the year 1900 was estimated to have been 5 to 48% lower than at present. Yearly runoff as a total for the entire country is estimated to have been 24% lower than at present.

1.1 Background and aims

An important driver for nutrient emissions from agricultural areas to the sea is the net precipitation and the resulting stream and river runoff. Stream and river runoff, together with the nutrient concentration, are decisive for estimating the nutrient transport. Therefore, knowledge about changes in precipitation and surface runoff over time is important for calculating changes in nutrient emissions to coastal waters.

The analysis of stream runoff focused on the long-time series of river discharge that exists for several streams and rivers across Denmark and similar long time-series on precipitation and temperature. No direct analysis of evapotranspiration was included in this report. However, previous work by Clark et al., (1992) documented a decreasing trend in mean annual “basin evapotranspiration” for 9 Danish catchments through the period 1910 to 1990. This analysis did, however, not include the effects of changes in crop cover, crop types and irrigation on evapotranspiration. Hence, the analysis conducted in this chapter has not included other parameters influencing stream and river runoff changes, such as land use and draining.

The aim of this chapter is to conduct a statistical trend analysis of monthly and yearly runoff in Danish streams and rivers having long time series. In addition, the aim is to compare the trends in stream runoff with the trends in precipitation and temperature data to give an estimate of the stream runoff at different locations in Denmark around 1900 and as a total for the entire country.

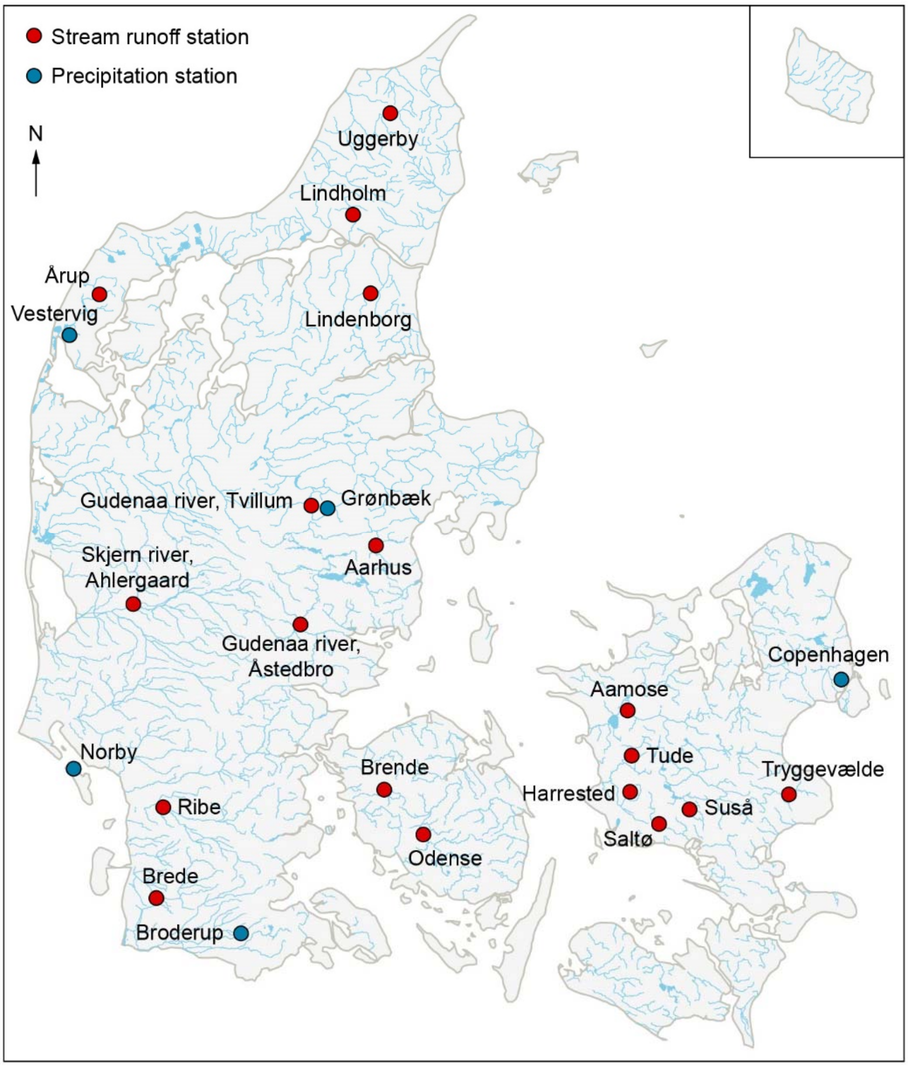
1.2 Data used in the analyses

The first hydrometric stations in Denmark were established around 1917, and in the following years, a number of hydrometric stations was established cross the country (Larsen et al., 2001). Therefore, 18 hydrometric stations exist with time series of daily river runoff from at least the past 80 years. Runoff data has been quality checked through ongoing evaluations throughout the period of measuring. The operations of hydrometric stations is documented through a series of reports and during the “more recent period” through the hydrometric data analysis and database system “HYMER” (DDH, 1923; DDH, 1925;. DDH, 1929; DDH, 1933; DDH, 1940; DDH, 1946; DDH, 1954; DDH, 1960; DDH, 1968; DDH, 1978; Ovesen et al., 2000). No further quality check or time series harmonization has been applied through this project. These 18 stations have been used for the analyses of the trend in runoff through the past 80 to 100 years. The stations used in the analyses are listed in Table 1.1, and the location of the stations can be seen in Fig. 1.1.

Table 1.1. Hydrometric stations used for the analyses, where runoff has been measured continuously for at least the past 80 years.

Name of hydrometric station	Year of establishment	Catchment area (km^2)
Uggerby stream, Astedbro	1917	153
Lindholm stream	1918	106
Årup stream	1936	105
Lindenborg stream	1925	214
Gudenaa river, Tvilumbro	1917	1282
Gudenaa river, Åstedbro	1917	184
Skjern river, Ahlergård	1920	1055
Aarhus stream	1919	119
Ribe stream	1933	675
Brede stream	1922	290
Odense stream	1917	302
Brende stream	1918	71
Åmose stream	1920	292
Harrested stream	1921	16
Tude stream	1932	148
Saltø stream	1918	64
Suså stream	1934	763
Tryggevælde stream	1917	129

Figure 1.1. Overview of the locations of stream hydrometric and precipitation stations across Denmark.



Monitoring climatic parameters, such as precipitation, temperature, etc., was initiated prior to river discharge monitoring in Denmark. Already from 1874 (some even earlier, several climate stations existed across the country (Cappelen et al., 2016)). Since precipitation is one of the main explanatory variables for changes in stream and river runoff, long time series of climate data from five of the climate stations were used to conduct a supplementary analysis of trends in precipitation the past more than 100 years (Table 1.2). For the selected climate stations, precipitation data series have been constructed by the Danish Meteorological Institute DMI (Cappelen et al., 2016), based on daily precipitation observations collected since 1874 for four of the stations and since 1920 for one of the stations (Table 1.2). The locations of the five climate stations can be seen in Figure 1.1. Precipitation represents observed data and has not undergone any test for homogeneity or advanced corrections for wind speed, location etc. However, data has been quality checked and corrected accordingly, mainly visually by Cappelen et al. (2016).

Table 1.2. Climate stations used in the analyses. Data from the Danish Meteorological Institute DMI (after Cappelen et al., (2016)).

Climate station	Dataseries start
Grønbæk	1874
Nordby	1874
Vestervig	1874
Broderup/Store Jyndevad*	1920
København	1874

*Data series constructed by merging data from the two stations, see Cappelen et al. (2016).

1.3 The statistical analysis

Based on the selected stream runoff and precipitation data series (Table 1.1 and 1.2), a statistical trend analysis was used. Overall, the statistical analyses of the stream runoff were conducted on three different types of time series: yearly runoff, monthly runoff and yearly maximum and minimum runoff. The statistical analyses of the precipitation were conducted on yearly and monthly time series. The analyses of the yearly time series were conducted in order to investigate the overall development in runoff and precipitation since the onset of the measurements. The analyses of the monthly trends were conducted to investigate whether there have been any seasonal changes in runoff and precipitation. For the stream runoff, changes in maximum and minimum runoff across the years were also analysed.

For all-time series of precipitation and runoff, the statistical non-parametric Kendall's trend test (Hirsch et al., 1982) was conducted to investigate trends in runoff and precipitation across the measurement years. The test is based on Kendall's tau (Kendall, 1975) and tests whether the development in the time series can be described by a monotone trend. Under the assumption of a linear trend, the yearly change in the time series was estimated by the estimator described in Theil (1950) and Sen (1968). A 95% confidence interval for the estimate of the yearly change was estimated by a non-parametric method, which did result in a non-symmetric confidence interval around the estimate (Gilbert, R.O. 1987).

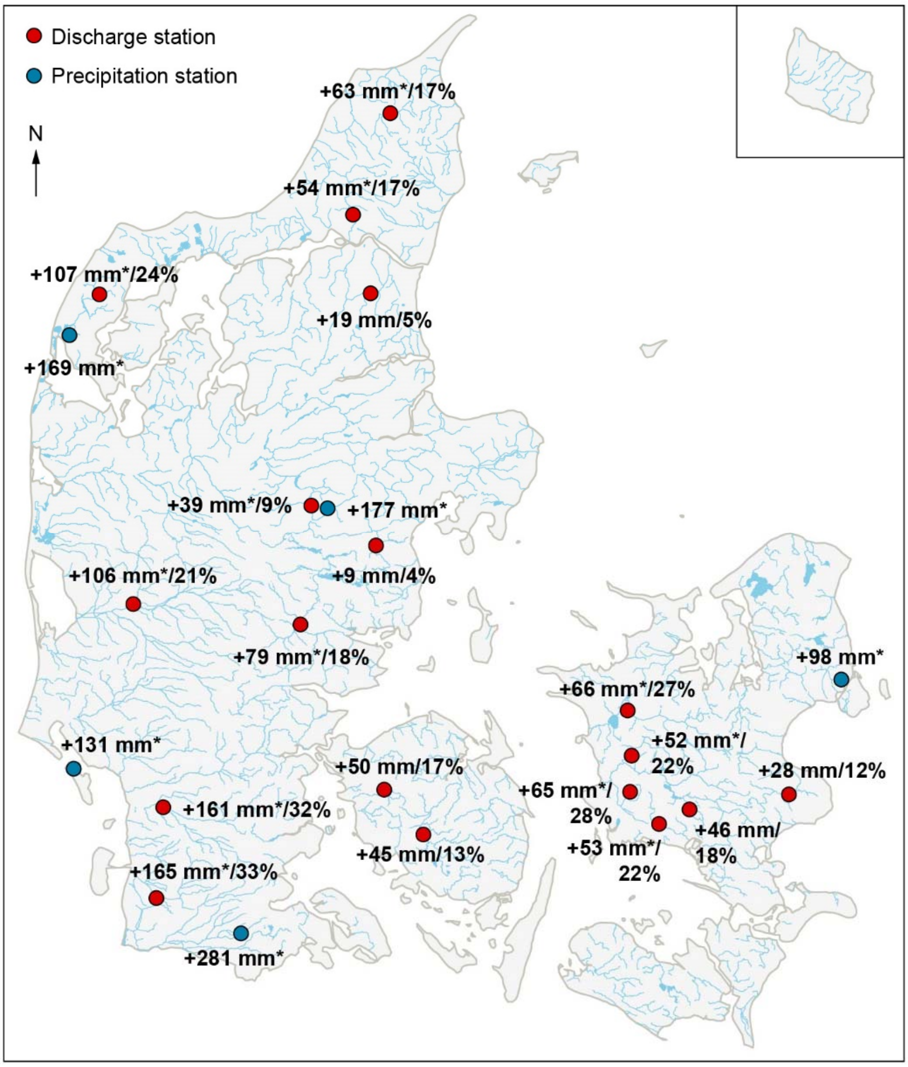
1.4 Trends in stream runoff and precipitation

In the following sections, the results from the statistical trend tests will be shown for both stream runoff and precipitation. In Appendix 1, the results of all statistical trend analysis on both discharge and precipitation are shown, and in Appendix 2, all stations and their mean yearly runoff as well as the statistical trend lines are shown.

1.4.1 Changes in yearly stream runoff

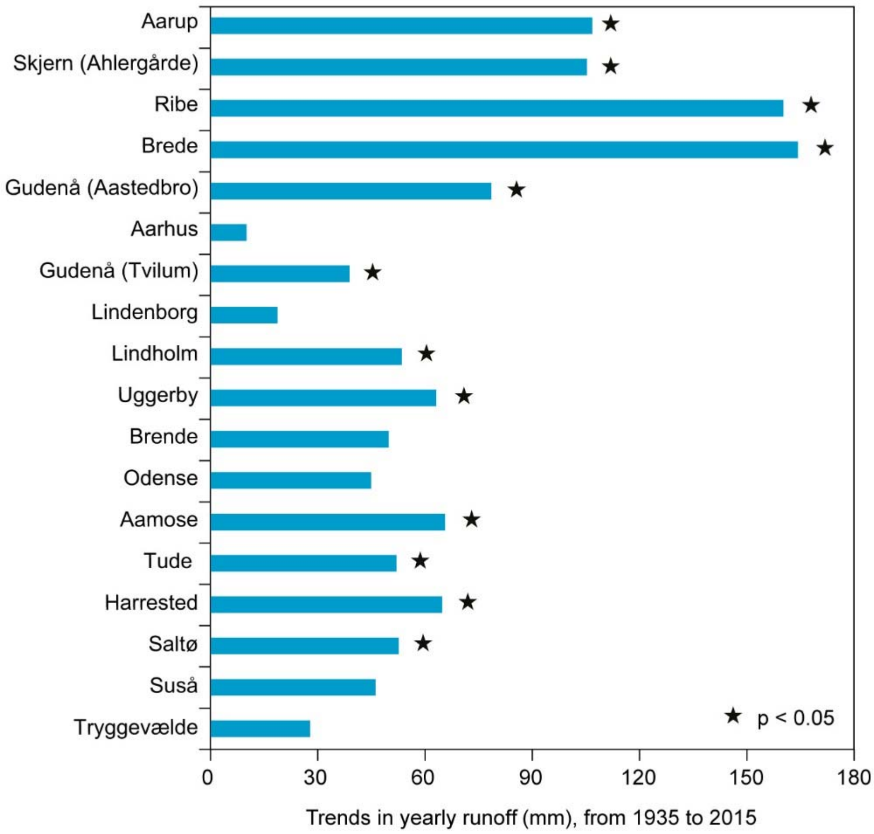
As can be seen in Table 1.1, all discharge and climate stations have data from at least the past 80 years. In Fig. 1.2, the changes in yearly runoff during the period from 1935 to 2015 is shown. For comparison, the results of the trend analysis of precipitation at the five climate stations are also shown.

Figure 1.2. Overview of changes in runoff (mm/%) and precipitation (mm) in the period 1935 to 2015 (for Aarup: 1937 to 2015). Significant trends (p<0.05) are marked with a star. Trend analyses are conducted on the full length of the data series, see Table 1.1 (for runoff) and Table 1.2 (for precipitation).



Stream runoff increased at all monitored hydrometric stations, and at 12 out of the 18 hydrometric stations, the increasing trends in runoff were significant (Fig. 1.3). Generally, the largest increase in stream runoff was seen in the western part of the country, in particular in the southwestern part of Jutland, where the two stations in Brede and Ribe streams saw the largest increases in runoff of 165 (33%) and 161 mm (32%), respectively, in the period 1935 to 2015 (Fig. 1.1, Fig. 1.3). Karlsson et al., (2014) found comparable changes in runoff for the river Skjern at the Ahlergårde hydrometric station, when analyzing the period 1920 – 2007.

Figure 1.3. Trends in yearly stream runoff in the period 1935 to 2015. Significant trends are marked with a star. Trend analysis were conducted on the full length of the data series, see Table 1.1.

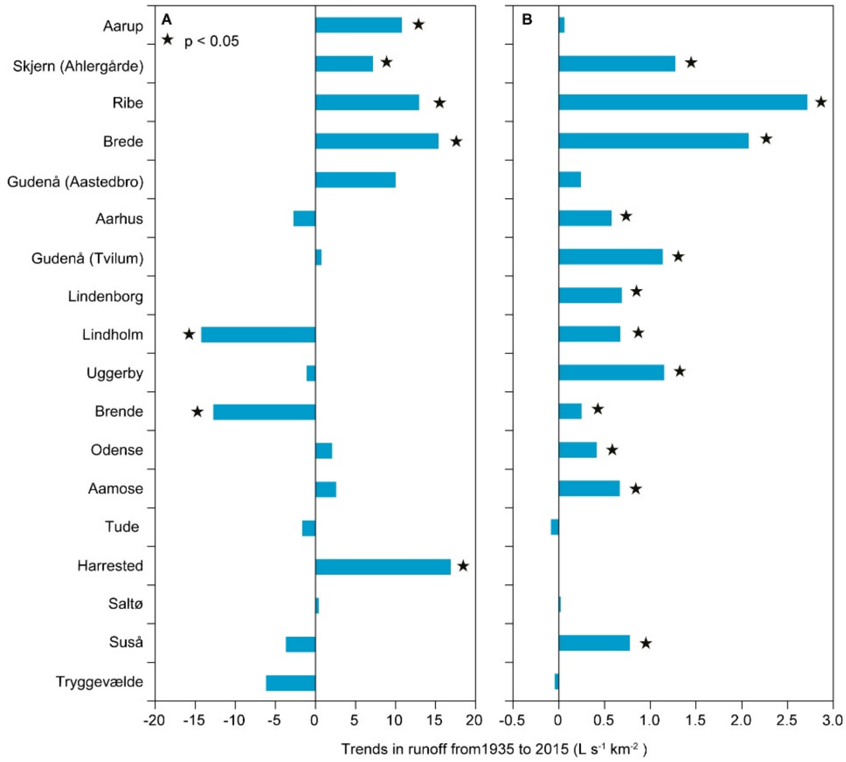


1.4.2 Changes in maximum and minimum stream runoff

For the majority of the hydrometric stations, a positive trend was detected for both the maximum and the minimum daily runoff (Fig. 1.4). For the daily maximum runoff per year, 10 out of 18 hydrometric stations showed an increasing trend, where five were significant (Fig. 1.4a). Seven out of the 18 hydrometric stations showed a decreasing trend, two of which are significant (Fig. 1.4a). The largest increase in the daily maximum runoff in the period 1935 to 2015 was found for the Harrested hydrometric station, amounting to an increase of 17 l s^{-1} km^{-2}. The largest decrease in daily maximum runoff per year was found at the Lindholm and Brende hydrometric stations, both with a decrease of approx. 15 l s^{-1} km^{-2} (Fig. 1.4a).

For the minimum runoff per year, 14 out of the 18 hydrometric stations showed an increasing trend, of which 12 were significant (Fig. 1.4 b) for the period 1935 to 2015. Only two stations showed a decreasing trend (Tude and Tryggevælde streams), but none were significant. The largest increase in daily minimum runoff per year was seen for Ribe stream, with an increase of nearly 3 l s^{-1} km^{-2} (Fig. 1.4 b).

Figure 1.4. Trends in maximum (a) and minimum (b) runoff from 1935 to 2015. Each column represents the change in runoff for the one day per year with maximum (a) or minimum (b) runoff. Significant trends are marked with a star. Trend analyses were conducted on the full length of the data series, see Table 1.1.



1.4.3 Changes in monthly runoff

An overall development in the pattern of seasonality change was seen in the monthly stream runoff in the period 1935 to 2015, with the largest increase during the winter months (Fig. 1.5). This was found using both seasonal Mann-Kendall test and by analyzing the months separately. Around 35 % of the monthly runoff estimates showed a significant increasing trend, and only a few stations showed a (non-significant) decreasing monthly trend (see Appendix 1 for an overview of all monthly data). The Brede hydrometric station showed the most pronounced monthly increase, with the largest increase in runoff in November and December. This is in agreement with Brede stream showing the largest yearly increase in runoff as well (Fig. 1.3).

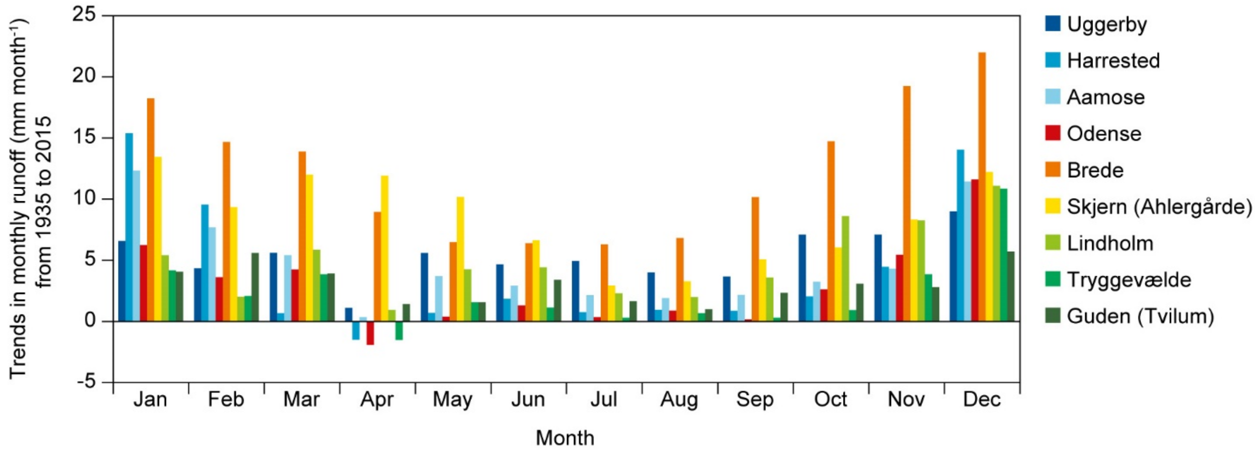
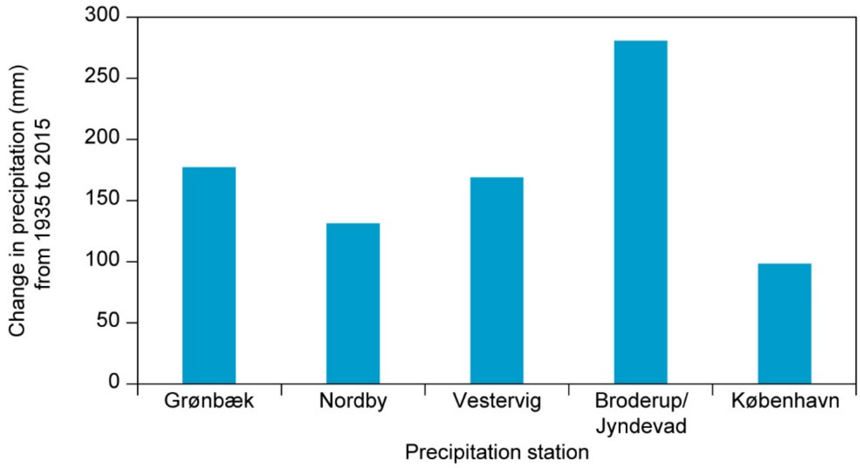


Figure 1.5. Trend in runoff from 1935 to 2015. Trend analyses, based on the full data series (see Table 1.1). Approximately 35 % of the analysed data had a significant trend ($p<0.05$) (see App. 1 for further details).

1.4.4 Changes in yearly and monthly precipitation

In order to conduct a firsthand comparison of trends in runoff and precipitation, data from the five climate stations with long time series were analysed. The analyses of data were conducted in the same way as for the runoff data (see section 3). Overall, data from the five climate stations showed a significant increasing trend, with the largest increase in precipitation observed at Broderup / Jyndevad (Fig. 1.6).

Figure 1.6. Trends in precipitation from 1935 to 2015. Trend analyses are based on data from 1874 to 2015, except for Broderup/Jyndevad, which is based on data from 1920 to 2015 (see Table 1.2). All trends are significant (p<0.05).



The most pronounced increasing trends for the monthly precipitation were seen in the winter months and in June during the period 1935 to 2015 (Fig. 1.7). A decreasing trend in precipitation in July and August was found for the Grønbæk, Vestervig and København climate stations, although none of them was significant trends. Overall, the general trends in precipitation were in accordance with the trends seen for the stream / river runoff, where the largest increases in monthly runoff were also seen in the winter months.

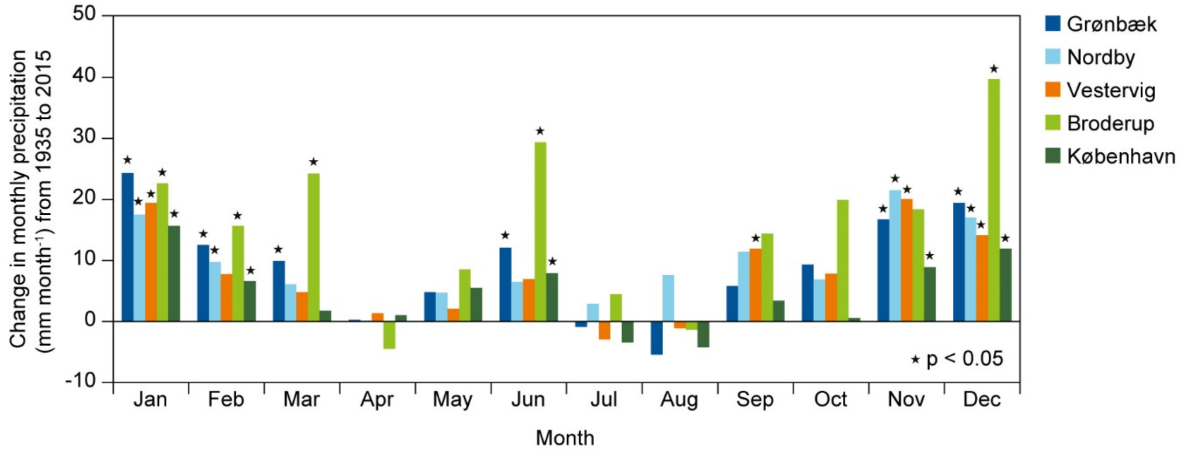


Figure 1.7. Monthly trends in precipitation from 1935 to 2015. Trend analyses are based on data from 1874 to 2015, except for Broderup/Jyndevad, which is based on data from 1920 to 2015 (see Table 1.2). Significant trends (p<0.05) are marked with a star.

It should be noted that since the precipitation data used for the analysis were not corrected e.g. in terms of wind speed, location of rain gauge, type of rain gauge etc., the exact amount of mm the different years should be interpreted with caution. However, it is expected that this plays a minor role in terms of the overall trend in data. Hence, the trend analysis conducted on the precipitation data for the past 80 years is expected to give a good estimate of the overall development in precipitation during this period.

1.5 Estimation of stream runoff around the year 1900

Since stream runoff data before ca. 1920 is sparse, it was not possible to estimate the stream runoff around the year 1900 through actual monitored data. However, based on the detailed time series from the 18 hydrometric stations for the past 80 years, one approach is to use the developed statistical trends from the existing data series to extrapolate back in time. In this way, it was possible to calculate an estimate of the stream runoff at the 18 stations, based on the assumption that the linear trends developed for the data series are also valid back in time. Since the majority of the stations (71 %) showed a significant linear trend for the 80 – 90 years with data in terms of yearly runoff, it is reasonable to assume that this trend can also be continued twenty years back. However, it should be noted that the assumption of a linear trend in yearly runoff back in time could not be verified, since the majority of the stations neither have stream nor rainfall data to support the trend. Hence, the estimates of the runoff in year 1900 based on back-writing the linear statistical trends are an estimate that should be interpreted with some caution.

However, another approach to estimate the runoff around the year1900 is to use the available information about temperature and precipitation from the climate stations to set up a numerical rainfall-runoff model (NAM). This is possible since the climate stations also have data about maximum and minimum temperature along with the precipitation data (Cappelen et al., 2016). Thereby, a rainfall-runoff model can be set up and calibrated against the at least 80 years of stream discharge data and can then be used to estimate the runoff back in time. This was done for three of the streams; Tryggevælde, Aarup and Ribe, which are within reasonable proximity to the climate stations København, Vestervig and Nordby (see Fig. 1 for locations). The advantage of this approach is that the numerical model is able to estimate a more dynamic trend than the linear statistical trend, since the model also includes temperature data.

In order to be able to compare the NAM estimates of runoff around the year 1900 with the estimates based on the statistical model for the measured data, the same type of statistical trend model as used for the measured stream runoff was applied to the NAM estimates of runoff. As seen in the measured runoff (Appendix 2), major year-to-year differences occur despite the significant overall trends in runoff levels. Therefore, when evaluating the change in runoff between year 1900 and the present (2015 used as reference), the estimated values from the statistical models were also used to ensure more comparable values. In the two following sections, the results from the statistical and the numerical models are presented.

1.5.1 Statistical model

Based on the long time series from the 18 hydrometric stations, the linear trends in yearly runoff developed from the statistical analysis were written back to estimate the yearly runoff in 1900. An example of the procedure is seen in Fig. 1.8 on data from the Ahlergårde hydrometric station in Skjern River. Based on the statistically estimated trend, the runoff level in 1900 was estimated to have been 30% lower than the estimated runoff level in 2015.

The results of the trend estimates of runoff around year 1900 showed a consistent pattern across all hydrometric stations of a lower level of runoff in year 1900 compared to 2015 (Table 1.3). It is important to note that the 95% confidence interval of the estimated trends (example in Fig. 1.8) only gives the certainty of the trend value, whereas it does not represent the uncertainty of the estimate of the actual runoff precisely in the year 1900. The reason for this is the large climate driven year-to-year variations in runoff, which occur consistently all through the 80 years of measurements on top of the overall trend.

Table 1.3. Estimated stream runoff in year 1900 by back-writing the statistical trend models; and estimated runoff in year 2015 from the statistical trend models. Average yearly runoff and 95% confidence interval of the statistical trend models are given. RMSE is given for the yearly runoff as well as the 95% confidence interval of the estimated value in year 1900. For comparison, the measured runoff in 2015 at the hydrometric stations is also shown.

Hydrometric station	Measured runoff 2015	Estimated runoff 2015	Estimated runoff 1900	95% conf. interval of estimated trend, min.	95% confidence interval of estimated trend, max.	RMSE of yearly runoff / 95% conf. interval of year 1900 estimate	Change estimated runoff, 1900 relative to 2015
	(mm yr^{-1})	(mm yr^{-1})	(mm yr^{-1})	(%)	(%)	(mm/%)	(%)
Uggerby stream	471	363	274	-3	3	±134/±49	-25
Tude stream	269	215	140	-15	14	±132/±94	-35
Harrested stream	274	229	136	-9	9	±139/±98	-41
Aamose stream	303	243	149	-7	8	±137/±92	-39
Brende stream	363	302	230	-4	5	±161/±70	-24
Odense stream	379	335	271	-4	4	±155/±57	-19
Brede stream	635	492	255	-6	6	±193/±76	-48
Ribe stream	555	503	274	-11	11	±198/±72	-46
Aarhus stream	396	274	260	-4	5	±155/±60	-5
Skjern river, Ahlergaarde	591	502	350	-3	7	±137/±39	-30
Gudenaa river, Aastedbro	640	426	314	-3	4	±170/±54	-26
Gudenaa river, Tvilum	485	425	369	-2	2	±111/±30	-13
Lindenborg stream	417	364	337	-2	3	±89/±26	-7
Aarup stream	632	444	292	-3	11	±167/±57	-34
Lindholm stream	433	319	242	-4	4	±154/±64	-24
Saltø stream	291	239	163	-6	7	±171/±105	-32
Suså river	304	252	186	-13	12	±157/±84	-26
Tryggevælde stream	274	226	185	-5	5	±142/±77	-18

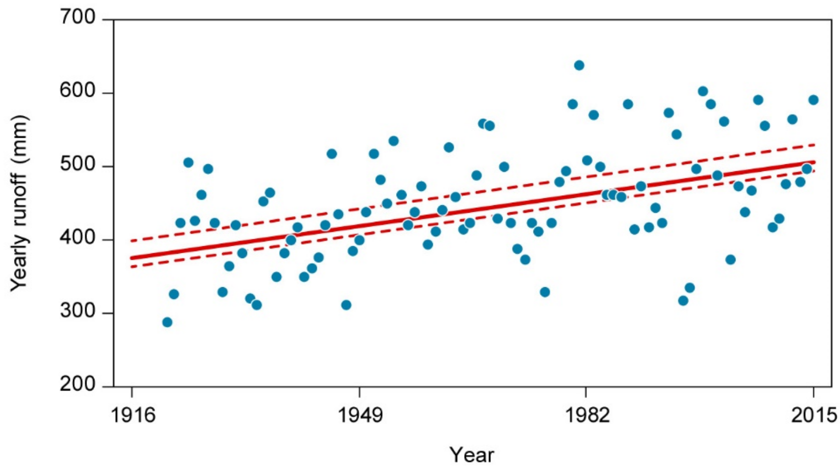


Figure 1.8. Yearly measured stream runoff from 1920 to 2015 (blue dots), the estimated linear trend in yearly runoff continued to year 1900 (red line) and 95% confidence interval for the trend model (dotted red lines). Example from hydrometric station Ahlergårde in Skjern River.

Assuming that the climatic year to year variations were similar around year 1900, they must be added to the trend value in order to give an interval that represents the most likely runoff around the year 1900. This interval was calculated as the 95% confidence interval from the root mean square value (RMSE) on the time series and are, hence, an expression of the year-to-year variation in runoff based on the pattern observed from the measured data. The 95% confidence interval of the estimated runoff in 1900 varied between

±26 (Lindenborg stream) and ±105% (Saltø stream) (Table 1.3), whereas the 95% confidence intervals on the trend estimate in year 1900 varied between +14% and -15% (non symmetrical) (Table 1.3).

1.5.2 Rainfall-runoff model

The hydrological/Rainfall-runoff model NAM by DHI (DHI, 2004a) was applied for modelling the stream runoff for the period around year 1900. NAM is a lumped conceptual hydrological model that describes the hydrological system through three linear reservoirs (Chow et al. 1988, DHI 2004a). The linear reservoirs are placed in series: (1) surface storage, (2) root zone storage and (3) groundwater reservoir. Routing of water to the watercourse occurs from the surface as overland flow, from the surface storage as interflow, and from the groundwater storage as base flow. Water from the additional snow storage can only be transported to the surface storage.

Evapotranspiration occurs from the surface and the root zone storages if water is available in the surface storage and/or root zone storage (DHI 2004a). Input climate data to the NAM model is daily accumulated precipitation (P), daily reference evapotranspiration (Ep) and daily mean air temperature (T). The only direct use of temperature in NAM is to determine whether precipitation falls as rain or snow, where 0°C was chosen as the discriminating temperature. The degree-day snowmelt model included in NAM was used and a value of 2 mm d^{-1} $°C^{-1}$ was chosen (DHI 2004b; Thodsen 2007). This value determines the rate of snowmelt and thereby influences the magnitude of peak flows associated with snowmelt. Realistic NAM parameter ranges were determined before calibration to ensure that the model parameters reflected the physical conditions of the catchments (Wagener 2007). Typical global values of the root zone storage are 50–300 mm; dealing with sandy catchments, a range of 50–150 mm was chosen (DHI 2004b). Madsen et al. (2002) used root zone storage capacity values between 290 mm and 390 mm for a very clayey catchment elsewhere in Denmark. According to DHI (2004a), as a rule the surface storage capacity can be set to 10% of the root zone storage capacity. Surface storage capacity values were chosen according to this rule.

As mentioned previously, the precipitation data used were not corrected (for gauge under catch). Some model parameters would have been set slightly differently if the data used were corrected. However, since the NAM model was calibrated against long time series of runoff, the uncorrected precipitation data was not expected to disturb the overall NAM estimates of runoff.

Three different NAM models were setup and calibrated for Aarup stream, Ribe stream and Tryggevælde stream, all stations with long time series of observed runoff. The hydrometric stations were chosen because their climate stations with time series have existed since 1874 (complete from 1875) relatively close by, and because the catchments have approximately the same land use throughout the modelling period and are not subjected to major groundwater abstractions. For Aarup stream, the climate station Vestervig was used, for Ribe stream the climate station Nordby was used and for Tryggevælde stream the climate station København was used (see Fig. 1.1 for locations). There is a relatively small difference between the precipitation and the potential evaporation at these sites and in the entire catchment. The difference is partly because the station/point is outside the catchment area, and because a station/point never fully represents a relatively large catchment area. The Vestervig climate station is located very close to the Aarup stream catchment. The Hargreaves method was used to calculate the solar radiation and the

Makkink method was used to calculate the potential evaporation (Allen et al., 1998). The daily precipitation available from the three long time series was uncorrected for gauge under catch. The temperatures available from the same sites are daily minimum and maximum values (Capellen et al., 2016).

The calibration was conducted using very long time series (72 - 97 years) (Fig. 1.9) and the auto calibration module in NAM. Between 9.000 and 12.000 simulations were run to finish the auto calibration procedure and find the optimal parameter set. The Water balance errors were between 0.0% and 2.8%. The Nash-Sutcliffe values were between 0.61 and 0.78 (Fig. 1.9). The observed and simulated discharge time series shown in Fig. 1.9 revealed no general biases over the very long time periods, which implies that it is reasonable to assume that the models can be used for modelling the stream discharge back to 1875.

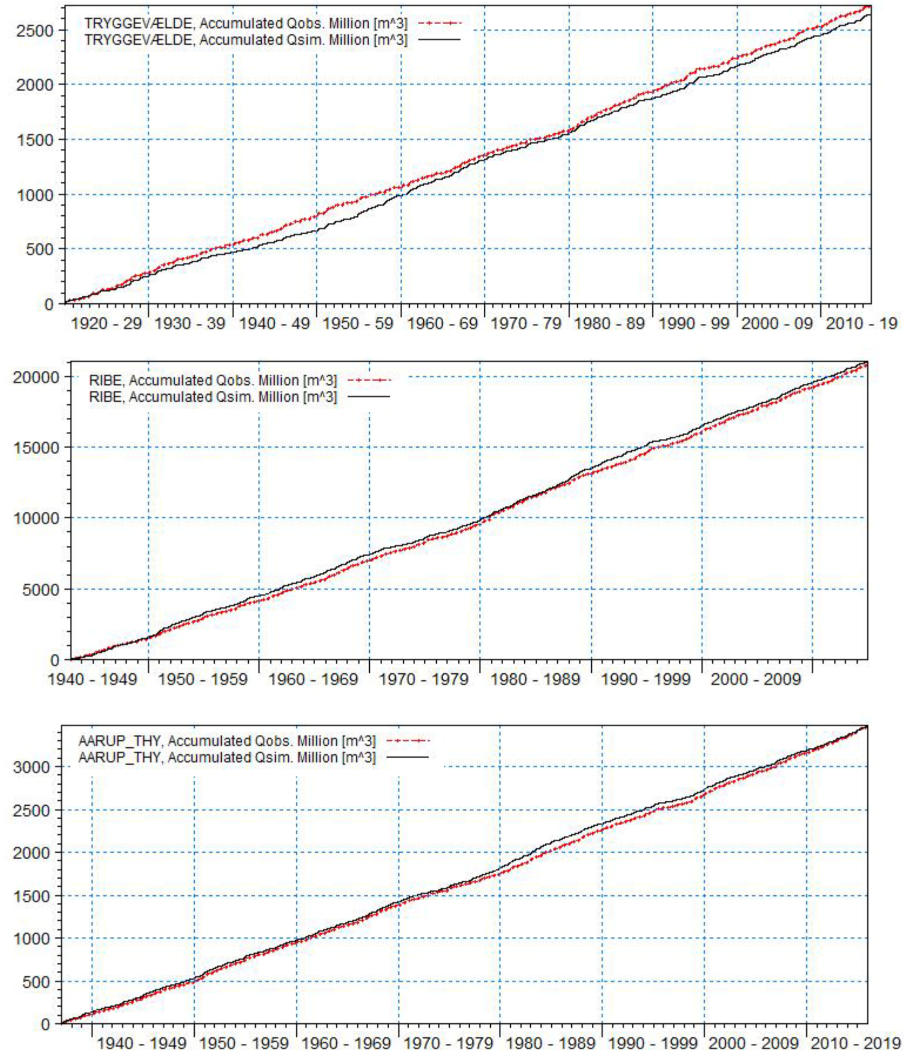


Figure 1.9. Accumulated observed and -simulated river discharge for the three calibration sites and periods. Top: Tryggevælde stream, middle: Ribe stream and bottom: Aarup stream. Red dots are observations. Black lines are simulated values.

The NAM models were run for the period 1875-2015. The linear trend models on NAM data as well as the yearly NAM estimated runoff at the three hydrometric stations are seen in Fig. 1.10. All three streams showed a significant increasing trend (all have $p<0.0001$) in runoff from 1900 until 2015. The runoff in 2015 was estimated to have increased 34% and 59% compared to year 1900 (for the year 1900 runoff was estimated to have been 25 to 37% lower than the estimated runoff level in 2015) (Table 1.4). The estimated trend lines for both NAM data and measured data (Fig. 1.10) showed similar trends, which strengthens the conclusion of significantly lower stream runoff around year

1900. However, the NAM estimated level of runoff around the year 1900 was higher for the streams Ribe and Aarup (9 and 11% respectively) and lower at Tryggevælde stream (18%) compared to the trend estimates based on measured data (Table 1.4).

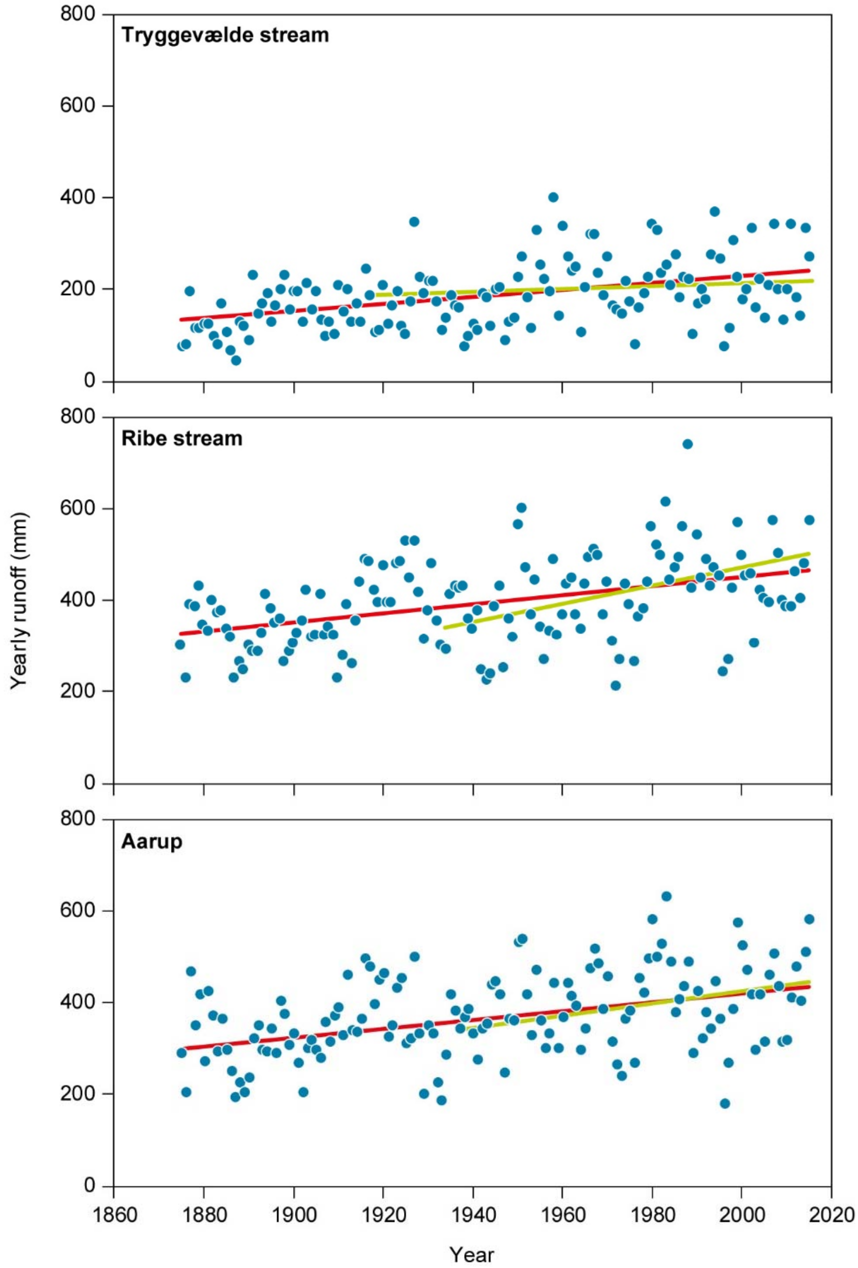


Figure 1.10. Yearly NAM modelled runoff (blue dots) and the statistical trendlines (red line). For comparison, statistical trendlines from the measured data are shown (green lines). a) Tryggevælde stream ($p<0.0001$), b) Ribe stream ($p<0.0001$ and c) Aarup stream ($p<0.0001$).

Table 1.4. River runoff in year 1900 and in 2015 calculated from the statistical trends models for NAM data. % difference of 1900 compared to 2015 is shown. For comparison, the estimated runoff in 1900 from back writing the statistical model based on measured data is repeated in this table.

	Estimated runoff 1900 (Statistical model for NAM data)	Estimated runoff 2015 (Statistical model for NAM data)	95% confidence interval of estimated trend, min.	95% confidence interval of estimated trend, max.	Change 2015 relative to 1900	Estimated runoff 1900 (statistical model for measured runoff)
	(mm yr-1)	(mm yr-1)			(%)	(mm yr-1)
Ribe stream	354	467	-3	3	32	274
Aarup stream	324	433	-3	3	34	292
Tryggevælde stream	152	242	-5	5	59	185

1.5.3 Comparison of trend analysis: NAM modelled and measured data

As can be seen in Fig. 1.10, Ribe and Aarup streams both had a steeper trend line when the statistical trend analyses was based on the measured data than when they were based on the NAM modelled data. This is also reflected in the trend estimates of the level of runoff around the year 1900, which were lower when estimated based on the trend from the measured data compared with NAM data. In addition, the steeper slopes entail that the overall estimated difference in runoff level in 1900 compared to 2015 is larger when it is based on back-writing the statistical trend for the measured data (-46 and -34% for Ribe and Aarup, respectively (Table 1.3)), compared to the NAM data (-24 and -25% for Ribe and Aarup, respectively (Table 1.4)). The opposite is the case for Tryggevælde stream, where the overall difference in runoff level in 1900 compared to 2015 was estimated to be -18% when the trend is based on measured data (Table 1.3) and -37% when the trend is based on NAM data (Table 1.4).

The exact reasons for this difference in trend estimates are difficult to clarify. The NAM model was setup and run with temperature and precipitation data for a period starting in 1874, which means that the statistical trend analysis for NAM modelled runoff was also based on this long period. This provides a certain robustness in terms of capturing the stream runoff dynamics. The estimates from 1900, based on back-writing the statistical trend for the measured data, have the weakness, that it is assumed that the trend observed from around 1920 to 2015 also is valid the 20 years back in time to 1900, which cannot be verified precisely for these 18 hydrometric stations. Hence, the combined estimate from measured and NAM modelled runoff level being in the range of 5 to 48% lower in the period around year 1900 with an uncertainty of the trend being -15 to 14% (non symmetrical 95 % confidence interval) is the best estimate that can be given based on the chosen approach.

1.5.4 Total runoff from the entire country around year 1900

In addition to estimates of the nutrient emission as a total for the entire country around 1900, it is also necessary to have some estimate of the runoff as a total for the entire country. The 18 hydrometric stations are located across the country, as seen in Fig. 1.2. Hence, it is reasonable to investigate whether the 18 stations are representative of the entire country. In order to do this, additional data were used consisting of estimated yearly freshwater runoff values from the entire country, which are calculated every year under the national monitoring program NOVANA (e.g. Wiberg-Larsen et al. 2015). The freshwater runoff estimates are based on monitoring data and model data for the ungauged catchments as described in Windolf et al., (2011).

When the average of yearly runoff at the 18 hydrometric stations was compared with the estimated yearly runoff for the entire country in the period 1990 – 2014, a good correlation (R^2 = 0.98) was found between the two (Fig. 1.11). This suggests that it is reasonable to use the 18 hydrometric stations as a proxy for the "whole country" runoff, back in time.

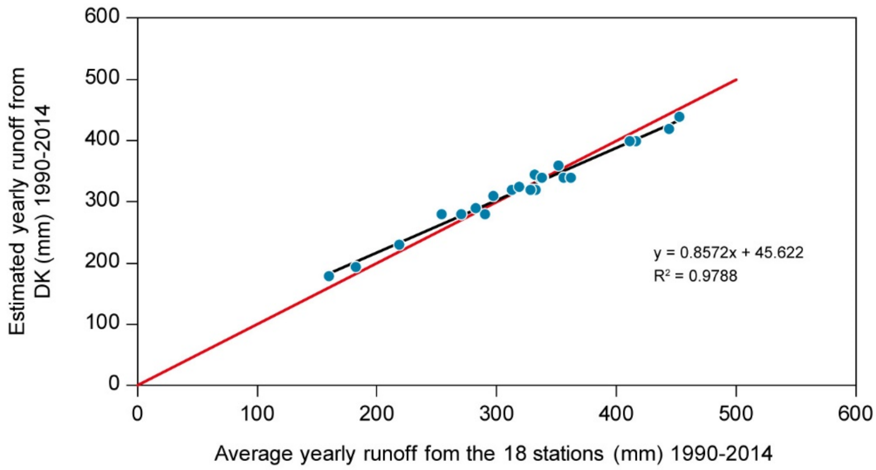


Figure 1.11. Comparison of average yearly runoff calculated as an average of the 18 stations and yearly runoff estimates from the entire country from Wiberg-Larsen et al. (2015). Trend line is shown in black and the 1:1 line is shown in red.

If the estimates of runoff from the statistical trend models for measured data (see section 1.5.1) were used, the average runoff from the 18 stations in 1900 amounted to 246 mm. Thus, when the relationship described in Fig. 1.11 was used, the total runoff from Denmark in year 1900 was calculated to have been 256 mm. The average of the trend estimates of the runoff from the 18 stations in 2015 was 342 mm, and when used together with the equation described in Fig. 1.11, the total runoff for the country in 2015 was estimated to be 339 mm. This means that based on the described approach, the runoff for the entire country in year 1900 would have been 24% lower than in 2015.

It should be noted that the 256 mm runoff for the entire country in 1900 is an estimate, and the exact uncertainty cannot be quantified from the given data and methods. The relation between average runoff at the 18 stream and river stations and the runoff from the entire country is well described, which is also reflected in the high R^2 value. However, in order to get the average runoff at the 18 stations in the year 1900, the statistical trend models are used and, as described in section 1.5.1 and 1.5.3 the trend models merely provide the most likely runoff level in year 1900. In addition, there is a station specific interval of runoff due to the climatic year-to-year variations. They were quantified to be from 26 to 105% (Table 1.3) for the 18 stream and river stations in year 1900. Therefore, the estimate of the total runoff from the entire country in 1900 of 256 mm is expected to have an uncertainty at least within that range of 26 to 105%. However, in this context it is also important to distinguish between the uncertainty of the exact runoff in year 1900 and the trend in the runoff from 1900 to 2015, where the latter is significant for 12 out of the 18 stations. Therefore, it is also expected that the trend of a 24% lower total runoff from the entire country, calculated from the average of these 18 stations, is a reasonable estimate.

1.6 Development in temperature and NAM-modelled snow cover

An additional analysis, on the number of "frost days" (days with maximum temperature < 0ºC) and snow cover (from the NAM model) have been included in this chapter (related to chapter 6 *Modelling nitrogen retention around year 1900*).

From the temperature data measured since 1890 at the three stations København, Nordby and Vestervig, the number of frost days around year 1900 was compared with the present situation. This is relevant in a more general analysis of nutrient emission, since a frozen ground prevents nutrients to leached through the upper soil, and this could influence the general dynamics of nutrient emission. The analysis showed that the number of days where the daily maximum temperature was below zero was 30 – 40% higher in the period around the year 1900 compared with the period 1995-2015 (Table 1.5).

Table 1.5. The NAM modelled estimates of the number of days with temperatures below zero in the period 1890-1910 and 1995-2015 at the three climate stations.

	Number of days per year where max. temp. < 0 °C (average of the period 1890-1910)	Number of days per year where max. temp. < 0 °C (average of the period 1995-2015)	Change from period 1890-1910 to the period 1995-2015 (%)
København	23	15	-36
Nordby	17	11	-34
Vestervig	21	14	-32

Precipitation falling as snow can also be an important player in terms of nutrient emissions during the time of snow cover and in situations with a quick thaw event, where most snow disappears as surface or near surface runoff. Therefore, it is also relevant to investigate the difference in snow cover around year 1900 compared with the present climate. Along with precipitation, the NAM model also estimates the snow accumulation and, thereby, it is possible to calculate the number of days with snow cover. Therefore, based on the NAM model and precipitation and temperature data from the three stations it was estimated how many days snow were present (calculated as days with more than 2 mm of water equivalent modelled by the NAM model) in the period 1890-1910 and 1995-2015 (Table 1.6), respectively. For the two stations København and Vestervig, a significant decrease in number of days with snow cover was estimated, whereas a small increase was modelled at the Nordby location.

Again, it should be mentioned that the precipitation data used for the NAM model was not corrected for precipitation gauge under catch (Allerup & Madsen 1980; Halldin 1988). This must be taken into account as a not quantified uncertainty of the NAM estimated changes in snow cover days, since potential changes in the precipitation input would also influence the modelled number of days with snow cover. The simulated number of days with snow cover would be higher if corrected precipitation had been used. This is due to the amount of precipitation falling in periods with freezing conditions would have been larger, and therefore it would have taken more time to melt the snow, thereby increasing the period with snow cover.

Table 1.6. NAM modelled estimates of the average number of days per year with snow cover (>2mm snow as water equivalents) in the period 1890-1910 and 1995-2015 at the three climate stations.

	Average number of days per year with snow cover (>2mm) for the period 1890-1910	Average number of days per year with snow cover (>2mm) for the period 1995-2015	Change compared to 1995-2015 (%)
København	32	23	-29
Nordby	20	22	8
Vestervig	35	21	-38

1.7 Conclusion

Based on the stream discharge time series from the 18 different hydrometric stations, it was found that yearly stream runoff has increased at stations over the past 80 years, and at 12 out of 18 hydrometric stations the increasing trend in runoff is significant. Generally, the largest increase in stream runoff was seen in the western part of the country, in particular in the south western part of Jutland, where the two stations in Brede stream and Ribe stream had the largest increase in runoff of 165 (33%) and 161 mm (32%), respectively, in the period 1935 to 2015. This is supported by the trends seen in precipitation at the five investigated climate stations.

For the majority of the hydrometric stations, an increasing trend was found for both the maximum and the minimum daily runoff. For the daily maximum runoff per year, 10 out of 18 hydrometric stations showed an increasing trend, five of them being significant. For the minimum runoff per year, 14 out of the 18 hydrometric stations showed an increasing trend, and 12 of them were significant in the period 1935 to 2015. Both stream runoff and precipitation showed a seasonal trend, where the most pronounced increase in both precipitation and stream runoff occurred during the winter months.

The statistical trend analyses on data from the 18 hydrometric stations were applied to write back the runoff trend to the year 1900. For all stations, lower yearly runoff was predicted by the trend for year 1900, with a 95% confidence interval on the trend ranging from -15 to 14% (non symmetrical). The station specific runoff in year 1900 was estimated to have been between 5 and 48% lower compared to the predicted runoff level in 2015, with an uncertainty inherent from year-to-year changes in the climate at ±26 to ±105% in 1900. The NAM model also predicted increasing runoff between 1874 and 2015 at Ribe, Tryggevælde and Aarup streams. Based on the statistical trend analysis of the NAM modelled runoff, the stream runoff for the three streams is estimated to have been 25 to 37 % lower in 1900 compared to 2015, with a 95% confidence interval for the trend in 1900 varying from ±5 to ±3%. The change in yearly runoff from the entire country is estimated to have increased with 24% since 1900, with an R^2 value of 0.98 on the relationship between yearly runoff for the entire country and the yearly average of runoff at the 18 hydrometric stations.

2 Year 1900: Agriculture and leaching of nitrogen from the root zone

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Abstract

Purpose: The purpose of this chapter is to provide an average estimate of the concentration of N in water leaving the root zone of land under agricultural use around the year 1900. At that time, agricultural management and land use differed significantly from current practices.

Materials and methods: The chapter relies on literature addressing agricultural management around the year 1900, on official agricultural statistics, and on contemporary studies of agricultural soils with a year 1900 relevant management. The applicability of farm- and field-gate balances and surplus is analysed with respect to their feasibility in estimating N leaching from the root zone around the year 1900.

Results and discussion: Estimates of N leaching losses from agricultural land around the year 1900 cannot be deduced by extrapolation of national farm-gate or field N balances and N surpluses. Land subject to agricultural activity and to tile-drainage increased substantially during the second half of the 19th century and induced a large and long-lasting additional loss of N stored in agricultural soils. While soil N mineralized during the growth period has contributed to crop growth and N uptake, mineralization outside the growth period has contributed significantly to the N leaching loss. Around 1900, leaching of soil N released due to historic changes in land use may have added 10 to 100 kg N / ha annually to the loss associated with the ongoing agricultural activity. Fallow was an integrated element in land under rotation and associated with substantial losses of soil N; concentrations of N in water leaving the root zone of fallow land most likely exceeded 20 mg N / l. Grazed grassland and spring-sown cereals dominated agriculture around year 1900, with most of the animal manure applied in the autumn and winter period. This provided inferior crop yields and was conductive for substantial losses of N by leaching.

Conclusions: Concentrations of N in leachates from soils carrying a crop typically ranged between 5 and 15 mg N / l, depending on management and crop. Concentrations were higher in leachates from bare fallow and land subject to prolonged grazing periods, and land with manure application in the autumn and left bare after ploughing. We estimate that the concentration of N in root zone leachates averaged 12 mg N / l from land under agricultural use.

2.1 Background and aims

The ecological status in coastal waters depends on several factors, including the N load. For year 1900, measurements of N concentrations in streams are extremely few (Westermann, 1898) and their representativeness is uncertain. This is also true for the retention of N in landscape elements. Consequently, the N load of coastal waters in the year 1900 currently remains unknown. Bøgestrand et al. (2014a) attempted to deduce the N status in water bodies around the year 1900 applying measurements of current N concentrations in streams from minimally disturbed catchments (Kronvang et al., 2015; Bøgestrand et al., 2014b) and national N surplus for agriculture (Kyllingsbæk, 2008). It was concluded that the estimated background concentrations reported by Bøgestrand et al. (2014b) provided an indication of N concentrations in streams around year 1900.

The calculations made by Bøgestrand et al. (2014a) rest on some key assumptions. These are:

- The national N surplus reported by Kyllingsbæk (2008) can be used to estimate N leaching from the agricultural area.
- Changes in agricultural land use and management do not affect the relative losses of N via ammonia volatilisation, denitrification and leaching, and the amount of N in the soil organic matter pool.
- The minimal agricultural activity associated with areas reported as natural catchments represents the activity practised on the total agricultural area in year 1900.

This chapter addresses these assumptions and attempts to provide estimates of the concentration of N in water leaving the root zone of land under agricultural use around the year 1900. In most aspects, the agricultural management and land use at that time differ significantly from current practices. This affects the interpretation of farm N balances and N surpluses, including the proportion of the N surplus susceptible to leaching.

2.2 Nitrogen balances and surplus

The N balance and surplus for Danish agriculture derived by Kyllingsbæk (2008) applies a farm-gate approach with Denmark being considered as one big "farm" (Fig. 2.1). This farm-gate approach does not reflect internal N flows in Danish agriculture. Consequently, Kyllingsbæk (2008) included only half of the estimated atmospheric N deposition in the balance; the other half was assumed to originate from agriculture itself.

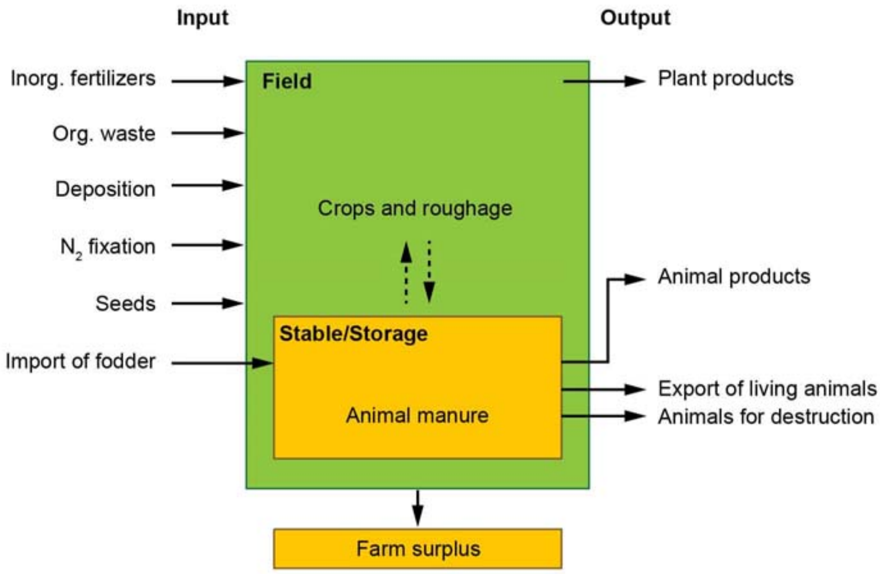

Figure 2.1. The national "farm-gate" N balance approach adopted by Kyllingsbæk (2008).

The balance includes atmospheric deposition of N, N added by biological N_2 fixation, N imported in mineral fertiliser and animal feed, and N exported in various agricultural products. The balance does not reflect the crop use efficiency of N in manure and of N provided by biological N_2 fixation or any management-derived impact on the allocation of surplus N to leaching, ammonia volatilisation, denitrification and changes in the soil N pool (Fig. 2.2). The distribution of the N surplus among these fluxes and the soil N pool is affected, for example, by changes in crop species and management, livestock composition and housing, in the storage and use of animal manure and in the ratio between homegrown and imported animal feed.

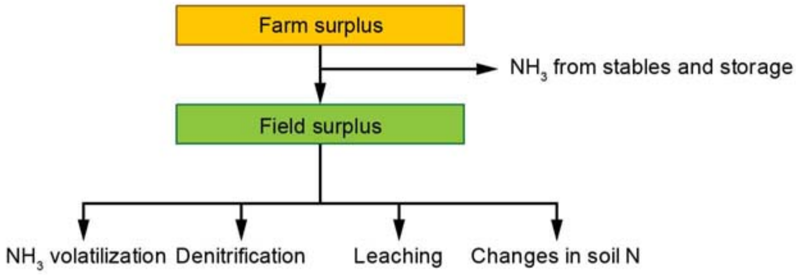

Figure 2.2. Distribution of farm-gate N surplus on ammonia volatilization from stables and during manure storage, and the potential distribution of the resulting field N surplus.

Setting up a traditional field balance requires knowledge of inputs and outputs of N at the field level. For cultivated land, the input consists of atmospheric deposition, mineral fertiliser and animal manure (N in mineral and organic form) and biological fixation of atmospheric N_2. Output is N in harvested plant material and removed during grazing. The difference between inputs and outputs represents the field N surplus, which covers N losses by ammonia volatilisation, denitrification and leaching, and changes in the soil N pool (Fig. 2.2).

Determination of field N surplus and measurements of the associated N leaching losses from the root zone show little or no correlation between surplus and leaching (Blicher-Mathiesen et al., 2014; Eriksen et al., 2015; Hansen et al., 2015). In an analysis of 39 streams from catchments with between 0 and 90% of the area in agricultural use, Kronvang et al. (2015) found a good correlation between the percentage of land in agriculture and the flow-weighted N concentration in streams without incorporating the field N surplus. Similar results were found in a Canadian study (Chambers et al., 2012).

The lack of correlation between N surplus and N leaching may reflect that crop rotation, soil tillage and other management practices have a larger impact on leaching than the size of the field surplus and that:

- The soil N pool is not constant over time
- The fertilizer N is mainly supplied in animal manure
- Crop N supply is dominated by biological N_2 fixation.

When estimating the N load of water bodies around year 1900, Bøgestrand et al. (2014a) assumed that the N leaching loss associated with a given N surplus is the same for current agriculture and year 1900 agriculture and, thereby, that farm management practices are the same for these two periods. However, agriculture has been subject to a dramatic change since year 1900. Differences relate to plant production (e.g. crop varieties, weed pressure, plant protection, fertilisation, drainage, irrigation, liming, tillage and other cultivation techniques) and animal production (e.g., livestock composition, feed quality and rate of feeding, housing conditions, grazing intensity and periods, and storage of manure). Moreover, the balance between exports of plant and animal products were different. Therefore, a similar distribution in year 1900 and today of a given farm N surplus between denitrification, ammonia volatilization, leaching and soil N storage is very unlikely. For the situation in year 1900, the N leaching would likely have constituted a higher proportion of the N surplus than what is the case under current conditions. Factors that contributed to this were decline in soil organic N content from recently cultivated land and management factors such as use of bare fallow and autumn application of manure that enhanced N leaching losses compared to other loss pathways.

2.3 Land use and management around the year 1900

The area of land in rotational cropping generally increased until about 1960, but the largest increase occurred in the second half of the 19th century. In the period 1861-1896, the total area in agricultural use increased from 2.4 to 2.9 million ha and accounted for 75% of the Danish area in 1896. The area in rotation increased by 546,000 ha (Jensen, 1988; Levin & Normander, 2008). The size of the area with heathland and sand dunes was reduced by 656,000 ha in the period 1850-1907 (Mortensen, 1969), and from 1861 to 1888 the area under rough grazing and common grazing fell by 70,000 ha (Levin & Normander, 2008). Currently, the farmed area covers almost 60% of the land, but in the year 1900 land use history was very different from that of today. Large areas of land had been included in rotational cropping in the preceding decades and there was widespread use of bare fallow. Moreover, much of the land had recently been tile-drained, and animal manure management differed greatly from current practices. These differences are essential when interpreting the N surplus and estimating the N leaching from the root zone of land used for agriculture.

2.3.1 Agricultural statistics

The area subject to agricultural use in 1900 accounted for 2.94 million ha, of which 86% was in rotation and 14% in permanent grassland, meadows and pastures (Danmarks Statistik, 1968). The area used for cereals, grassland in rotation, fallow and root crops accounted for 39%, 30%, 9% and 8%, respectively. Spring sown cereals occupied 73% of the cereal area. The dominating spring sown cereal was oats, while cereal rye made up 89% of the area with autumn sown cereals. For comparison, cereals and oil-seed rape occupied 63% of the agricultural land in 2015; of this, autumn sown crops made up 65% (Statistikbanken.dk). In 2015, the area with rotational grassland was 10% and that outside rotation accounted for another 10%, and the area with spring-sown annuals for silage production (mainly silage-maize) was 9%. Clearly, there are major differences between year 1900 and today in terms of agricultural areas allocated to cereals and grassland. For cereals, there has been a marked shift from spring-sown crops, leaving the soil vegetation-free during autumn and winter, to autumn sown crops leaving the soil with a plant cover over-winter.

Converted into livestock units, the agricultural sector included 2.6 million units in 1898 (Danmarks Statistik, 1969). The livestock units were distributed with 54% cattle, 16% pigs, 15% horses, 8% poultry, and 7% sheep and goats. Today, horses are not used in agriculture. The number of cattle is slightly below that in 1900, but the productivity per number of livestock unit has increased 3- to 4-fold (Kristensen et al., 2015). The number of sheep and goats has reduced to very low numbers, whereas pig production has increased dramatically from 1900 until today (Statistikbanken.dk). In 1900, grass ingested in fresh condition was the dominating source of ruminant forage (49%), while root crops, hay and cereal straw accounted for 19%, 18%, and 13%, respectively (Danmarks Statistik, 1968). In terms of digestible protein (the source of N ending up in animal manure), grass accounted for 67% and hay for 24% of the home-grown forage.

Compared with today, the number of livestock and its distribution among different animal categories was very different in year 1900. This was also true for the farm structure. Most farms were small in terms of acreage and production volume and relied on livestock that was diverse with respect to animal categories. This also meant that most farms across the country relied on similar plant production systems dominated by spring-sown cereals and grasslands.

2.3.2 Land use change and the soil N pool

It is well known that the conversion of natural areas and other areas with permanent plant cover into arable rotation reduces the content of organic matter in soils (Rasmussen et al., 1998; Girma et al., 2007; Johnston et al., 2009; Poeplau et al., 2011; Oberholzer et al., 2014). The loss of soil organic matter induced by cultivation may continue for several decades (Fig. 2.3). Oberholzer et al. (2014) found an average annual loss of soil C from 0-20 cm, ranging from 100 to 250 kg / ha during the first 60 years after cultivation of extensively managed grassland, and after 60 years of cultivation the pool of soil C had still not reached a new equilibrium. Assuming a C / N ratio of 10 (Thomsen et al., 2008), the cultivation derived loss of soil organic matter corresponds to an annual loss of 10 to 25 kg N / ha.

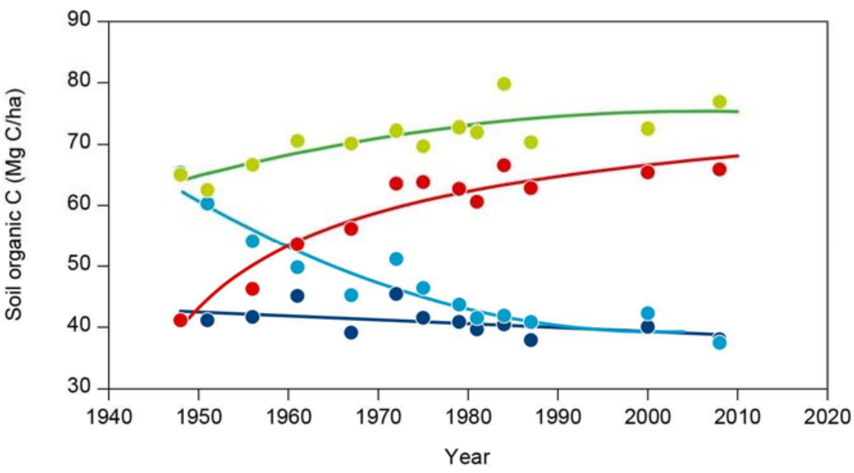

Figure 2.3. Changes in soil C (0-23 cm depth) in land under long-term permanent grass (green circles), permanent grassland converted to arable rotation (light blue circles), permanent arable rotation (dark blue circles), and arable rotation converted into permanent grassland (red circles). From Johnston et al. (2009).

Whitmore et al. (1992) found that ploughing of permanent grassland reduced the soil N pool by up to 40% over a period of 20 years. Howden et al. (2010) found similar results for the Thames river basin, where a change of the cultivated area from about 30% to 60% in the period from 1940 to 1945 more than tripled the nitrate concentration in the Thames. Conversely, introduction of grassland may increase the soil organic matter pool by more than 1000 kg C / ha / year (Christensen et al., 2009; Taghizadeh-Toosi et al., 2014). A field experiment initiated at Rothamsted Experimental Station in 1949 shows large and long-lasting changes in soil organic matter contents when old grassland is ploughed out and converted to arable rotation and when land under long-term rotation is converted into permanent grassland (Fig. 2.3; Johnston et al., 2009).

Results from the Danish Square Grid (Kvadratnettet) show that significant changes in soil organic matter may follow the general development in the structure of agricultural production (Fig. 2.4). In the period 1986-2009, 200 kg C / ha was lost annually from the 0-100 cm layer as an average of all soil types (Taghizadeh-Toosi et al., 2014), equivalent to 20 kg N / ha. This loss, however, hides an annual increase of approximately 500 kg C / ha on sandy soils (JB 1 and 2 in the Danish soil classification system), while on more clayey soils (JB 5, 6 and 7) the annual loss was between 500 and 1200 kg C / ha, equivalent to 50 and 120 kg N / ha. The increase in the N pool on sandy soil was attributed to a concentration of farms based on milk production and associated forage production (including grass-clover crops in rotation), whereas cereal-intensive farms based on pig production dominated on clayey soils. In areas with intensive cereal cultivation and no livestock, part of the mineralised soil N is taken up by the crop and exported with grain and straw.

There is little doubt that in year 1900 significant losses in soil organic N occurred on areas that had been brought into cultivation in the previous decades. The growing crops most likely recovered part of the mineralized soil N, but a substantial part of the cultivation-induced decline in soil N was lost by leaching due to mineralisation of organic N outside the growing season. This loss of N adds to the N leaching losses associated with the plant production practiced around the year 1900.

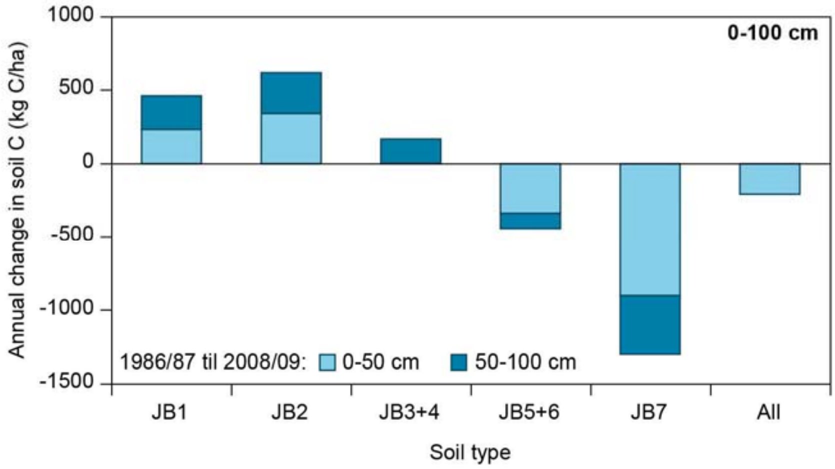

Figure 2.4. National Square Grid: Annual changes in soil C storage during 1986/87 to 2008/09 on sites under agriculture (Taghizadeh-Toosi et al., 2014). JB 1 = Coarse sand; JB 2 = Fine sand; JB 3+4 = Loamy sand; JB 5+6 = Sandy loam; JB 7 = Loam. See text for further explanation.

2.3.3 The impact of soil drainage

Today, roughly half of the agricultural area is tile-drained, corresponding to about 1.4 million ha (SEGES, 2015). Around the year 1900, the size of the drained area was 650,000 ha, most of which had been drained in the period from 1860 to 1900 (Jensen, 1988; Olesen, 2009). Drainage of waterlogged soil increases the turnover of soil organic matter and, thus, promotes loss of N from the soil pool. Van Wesemael et al. (2010) found that draining a grassland with a high groundwater table led to an annual average loss of 500 to 1000 kg soil C / ha for the next 30-45 years. With a C / N ratio of between 10 and 15, this corresponds to an annual loss of between 30 and 100 kg N / ha.

Although part of the mineralised N most probably became assimilated by increased plant productivity, the draining of temporarily waterlogged soils and soils with high groundwater tables during the preceding decades have accomplished an additional N leaching loss derived from N mineralised outside the growing season, but also a lower loss of N via denitrification under anoxic conditions in the soil.

2.3.4 Use of fallow

Today, vegetation-free fallow is no longer used, but in the period around the year 1900 fallow was an integrated part of the plant production system. Around 1900, the area in fallow accounted for 259.000 ha or 9% of the cultivated area (Danmarks Statistik, 1968), of which 60% was bare fallow. In bare fallow, the land remained vegetation-free throughout an entire year.

The purpose of fallowing was to reduce weed infestations (especially perennial weeds) and increase the availability of plant nutrients (Christensen, 1898; Madsen-Mygdal, 1919). With respect to nutrient availability, the main goal was to stimulate mineralisation of soil organic N and thereby accumulate plant-available N for the subsequent crop. Most often, ploughing of rotational grassland in early August initiated the bare fallow period, and during the subsequent fallow year the land was exposed to intensive tillage (ploughed up to four times with frequent harrowing in-between). After one year of bare fallow, farmyard manure was applied and ploughed-under before planting of autumn sown cereals (typically cereal rye). Similarly, intensive tillage with ploughing and harrowing characterized the semi-fallow management in which the fallow period began in May / June and ended in the autumn with the application of animal manure, ploughing and planting of cereals.

Several studies have shown significant leaching losses of N from soil under bare fallow. Measurement of N leaching in lysimetric experiments with undisturbed soil columns, established in 1870 at Rothamsted Experimental Station, showed an average annual loss of 45 kg N / ha during the first seven years under permanent fallow (Addiscott, 1988). In a lysimetric experiment at Askov Experimental Station, the annual average N leaching over a four year period corresponded to 104 kg N / ha for unmanured bare fallow (Thomsen et al., 1993). In this experiment, the average percolation during the experimental period was 504 mm / year and the concentration of nitrate in the leachate was 21 mg N / l.

For bare fallow established after ploughing an area that for the past 14 years had carried permanent grass-clover, Fraser et al. (2013) measured annual leaching losses of approximately 40 kg N / ha. Losses from the area with permanent grass-clover remained at <5 kg N / ha. During 1956-1986, two trials were conducted with permanent bare fallow at Askov Experimental Station (Christensen, 1988, 1990). In the 30 years of continuous fallow, the soil N content decreased 35-38%. In the field experiment, the average loss was 53 kg N / ha / year, while the corresponding loss from soil under continuous rotation was 23 kg N / ha / year. Thus, the additional loss due to bare fallow was 30 kg N / ha / year.

In 1900, the use of fallow was associated with substantial N leaching losses because of the lack of plant cover and, thus, no plant N uptake, and because of the intensive tillage during the fallow period. Although contemporary textbooks for farmers and agricultural advisors (Christensen, 1898; Madsen-Mygdal, 1919) stressed the need for fallowing to combat weeds, it was recognized that bare fallow led to substantial losses of nutrients from the fallowed soil.

2.3.5 N inputs, plant production, and potential N losses

Plant production around 1900 differed significantly from that of common Danish agriculture today for virtually all growth factors: inferior crop varieties, lack of crop protection, inferior mechanization of field operations, low nutrient supply, and manure management practices that resulted in substantial losses of manure N, both during storage and after field application.

Around 1900, there was very little use of mineral N fertiliser, and the cultivated area only received N in animal manure (solid farmyard manure and liquid manure), N_2 fixation, N applied with irrigation and atmospheric deposition.

The amounts and use efficiency of N supplied to farmland via N_2 fixation are difficult to quantify because N_2 fixation depends not only on choice of crop, but also on its growing conditions. The balance between the N_2-fixing crop component and the companion non-fixing plant depends on plant-availability of soil nutrients, competition between species and the climatic conditions during the growing season. The proportions of clover in pastures and green manures grown around year 1900 remain unknown. However, Kyllingsbæk (2008) estimated the N supply from N_2 fixation to account for 55% of the total N input in Danish agriculture around the year 1900, the corresponding value for 2005 being 8%.

The annual average (1900-1904) input of N with animal manure has been estimated to 21 kg N / ha when corrected for 15% loss of N during feeding and 25% loss of N during manure storage (Danmarks Statistik, 1968). This statistic does not account for subsequent losses of N during and after field application. Losses of manure N during the autumn / winter period reduced the N fertiliser value of the applied manure leading to poor crop yields. In 1900, most of the animal manure was applied and ploughed under during the autumn and winter period, due partly to soil tillage requirements, the availability of farm labour and the lack of proper storage capacity. Autumn applications of animal manure lead to poor N use efficiency and substantial leaching losses of mineral N applied with the manure and mineral N derived from mineralisation of organically bound N outside the growing season. As an illustration, Hoffmann et al. (2000) calculated a leaching loss of 1 kg N per kg harvested N for Swedish agriculture around 1900. The corresponding figure in 1985 was 0.4 kg N.

Crop yields around the year 1900 were very low when compared with yields obtained today. The average grain yields for oats, barley, rye and wheat were 14, 18, 20 and 28 hkg / ha, respectively (Iversen, 1942; Danmarks Statistik, 1968). These cereal grain yields are comparable to those achieved in the period 1894-1904 in the Askov long-term experiments (Christensen et al., 2006) and to yields currently obtained in plots kept unmanured for more than 100 years (Christensen and Thomsen, 2014). These yields are slightly lower than yields of cereal crops grown under unmanured conditions in ongoing organic farming experiments (Olesen et al., 2002).

As for cereals, the productivity of grassland was also very small around year the 1900. For hay produced on rotational and permanent grassland (incl. meadows), the yield was 24 and 27 hkg / ha, respectively (Danmarks Statistik, 1968). Even though contemporary textbooks prescribed generous use of liquid manure (urine) to meadows, permanent grasslands and grass-clover crops in rotation in late autumn and again in the spring, the yield level of grasslands around the year 1900 was below that obtained currently for rotational grass-clover grown under unmanured conditions (Christensen and Thomsen, 2014).

2.4 Year 1900: Indications of N concentrations in leachates from the root zone

Although there is a lack of nation-wide measurements of N concentrations in leachates from the root zone of land under agricultural use in 1900, a number of subsequent studies may provide indications of the most likely range of N concentrations at that time. For drains without retention, these concentrations also apply to water that reached local streams.

During autumn 1922 to spring / early summer 1923, 69 and 44 samples of drainage water were collected at Askov Experimental Station from fields under rotation and from a neighbouring field under first-year grass-clover, respectively (Hansen, 1926). For the land in rotation, the concentration of nitrate-N in drainage water varied between 7.6 and 14.4 mg N / l, while the concentration in drains under grassland ranged between 0.7 and 3.8 mg N / l.

For an unmanured crop rotation with spring barley, grass used for cutting, and winter wheat grown in lysimeters from 1985 to 1989, the N leaching loss ranged from 15 kg N / ha for the grass crop to 40 kg N / ha following spring barley (Thomsen et al., 1993). The crop yields were similar to those reported for comparable crops in year 1900. The average percolation during the four-year experimental period was 504 mm / year, and the concentration of nitrate in the leachate was 3 and 8 mg N / l, respectively.

The fate of N supplied with residues of N_2-fixing crop remains uncertain because the N bound in the residue has to be mineralised before being available to subsequent crops. Mineralisation outside the growing season provides a substantial potential for N leaching. Following unfertilized grassland, terminated in the spring and then seeded to spring barley, Eriksen et al. (2008) found an annual flow-weighted nitrate concentration of 36 mg N / l in drainage collected during the leaching period that followed the barley crop. Grasslands terminated in the autumn lead to higher leaching losses of N than grasslands terminated in the spring (Djurhuus & Olsen, 1997).

Leaching of N may also be substantial from grazed grasslands, in particular grass-clover swards subject to long grazing periods. Around year 1900, liquid manure was often added to grasslands in the early spring, and after having delivered a first cut of hay they were grazed until late autumn (Christensen, 1898). Urination by grazing animals creates very high inputs of mobile N locally. For a 4-year old grass-clover field subject to grazing from late April to late October, Hansen et al. (2012) estimated that the one-third of the area was affected by urination and that the N concentration in percolate from this area was 23 mg N / l. When first exposed to a grass cut in the spring and then subjected to grazing, the N concentration was 19 mg N / l.

Around year 1900, the use efficiency of N in liquid manure and farmyard manure applied in the autumn was substantially lower than that achieved today. Compared to N leaching from soil with farmyard manure applied and ploughed under in December and March, manure applied in September increased the N leaching loss with 30 kg N / ha (Thomsen, 2005). The first-year loss corresponds to 6-10% of the N added, with manure and the loss cumulated over the subsequent three winters accounting for 21% of the N applied with farmyard manure in September.

A study of different organic farming practices was initiated in 1997 at three sites varying in climate and soil type (Jyndevad, sand; Foulum, loamy sand;

Flakkebjerg, sandy loam; see Olesen et al., 2000). One four-year crop rotation included grass-clover ley, winter cereals, spring cereals, and either a potato or a grain legume crop. The rotations were unmanured or animal manure applied at an average rate of 70 kg total-N / ha / year. Nitrate leaching was measured using porous ceramic suction cups situated at the bottom of the root zone (Jyndevad, 60 cm; Foulum and Flakkebjerg, 100 cm; Askegaard et al., 2011). The average flow-weighted nitrate-N concentration of leachates from unmanured and manured rotations was similar and remarkably constant across the different sites and the three rotation cycles of the period 1997 to 2008. The nitrate-N concentration averaged 11.7 mg N / l, with a variation from 7.4 to 14.9 mg N / l (J. E. Olesen, pers. comm.). Percolation was 637, 362 and 238 mm at Jyndevad, Foulum and Flakkebjerg, respectively, providing considerable differences between sites in amount of N lost by leaching. It is recognized that present-day organic crop production is not fully comparable to crop production around the year 1900. Present-day organic production relies on modern cultivars, more efficient tillage and seeding equipment, and on manure storage and spreading facilities that provide a higher overall N use efficiency.

2.5 Conclusions

Determination of N leaching losses from the root zone of agricultural land around the year 1900 cannot be deduced directly from national farm-gate or field N balances and N surpluses.

Land subjected to agricultural activity and to tile-drainage increased substantially during the last half of the 19th century. This historical development induced a large and long-lasting decrease in N stored in agricultural soils. Soil N mineralized during the growth period contributed to crop growth and N uptake, but mineralization outside the growth period contributed significantly to the N leaching loss from the root zone. The leaching of soil N released due to historic changes in land use may have ranged from 10 to 100 kg N / ha / year. This loss adds to the N leaching losses associated with the agricultural management practiced around the year 1900.

Bare fallow was an integrated element in land under rotation, and bare fallow induced substantial annual losses of soil N, with N concentrations in water leaving the root zone exceeding 20 mg N / l.

Grazed grassland and spring-sown cereals dominated the agricultural land use around year 1900, with most of the animal manure applied in the autumn and winter period. This management was conductive for loss of N by leaching. Based on contemporary studies of agricultural soils with a year 1900 relevant management, concentrations of N in leachates from the root zone for most fields ranged between 5 and 15 mg N / l, depending on management and crop. Concentrations were higher in leachates from bare fallow and land subject to prolonged grazing periods and land with manure applications in the autumn and left bare after ploughing. The average concentration of N in leachates from the root zone is estimated to 12 mg N / l from land under agricultural use.

3 Deposition of nitrogen to fresh and marine waters in Denmark in year 1900

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3.1 Introduction

The aim of this chapter is to provide a short review on the historical deposition of nitrogen to land areas and to fresh and marine waters in Denmark around the year 1900. The review presents results from measurements of wet deposition of nitrogen in Denmark in the period around 1900 and model calculations of the total deposition (wet plus dry deposition). The model part is based on results for Denmark from our own model (DEHM) and on results from three international studies, which describe the historical trends of the deposition of nitrogen at a regional level, e.g. all EU or the entire Baltic Sea.

3.2 Measurements of wet deposition in Denmark around 1900

Two older Danish studies of the wet deposition of nitrogen from the period around 1900 exist. In both studies, simple bulk precipitation samples were collected and subsequently chemically analyzed for the content of ammonium and nitrate.

In the first study, measurements of the wet deposition of nitrogen were carried out at Landbohøjskolen in Copenhagen in 1880-1885 (Tuxen 1890). Average annual wet deposition was determined to 13.6 kg N / ha, with 11 and 2.6 kg N / ha coming from ammonium and nitrate, respectively. This is much higher than measured both in 1990 and 2015 at the Danish measurement stations (Ellermann et al., 2016; Table 1). The wet deposition in 1990 is the highest measured wet deposition in the current monitoring program. The results from Landbohøjskolen vary considerably between the years, with a factor of nearly two between the annual wet deposition of ammonia (from 7.5 to 13.5 kg N / ha) and a factor of nearly 5 for nitrate (from 1.3 to 6.1 kg N / ha) (Tuxen 1890). Today, the variation between the years is often only about 20-30% for both ammonia and nitrate (Ellermann et al., 2016). The wet deposition in Copenhagen was of similar size as those measured for a number of other European sites close to larger cities (e.g. Montsouris, Paris 12 kgN / ha, Tuxen 1890) and was considerably higher than those measured at more remote locations such as Kurchen in Germany (2.5 kg N / ha), Rothamsted in England (3.9 kg N / ha, Tuxen, 1890) and Jönköping in Sweden (3.8 kg N / ha; Hansen, 1931)).

In the second study, the wet deposition of nitrogen in Denmark was determined at four research stations in Denmark in the period from 1923 to 1927 (Hansen, 1931). The average annual wet depositions at these sites are listed in table 3.1.

The measured wet deposition both in 1880-1885 (Tuxen, 1890) and 1923-1927 (Hansen, 1931) is high compared to the expectations based on the current knowledge on the long term trends of the Danish emissions (see section 3 and 4) and on the levels measured today (Ellermann et al., 2016). The reason for this is discussed in section 5.

Table 3.1. Annual wet deposition of ammonium and nitrate measured at Danish measurement stations. The results from the DCE-stations are average for six Danish measurement stations.

Location*	Ammonium kg N/ha d	Nitrate kg N/ha	Total Kg N/ha
Copenhagen (1880-1885)	11.0	2.6	13.6
Blangsted (1925-1926)	6.4	3.4	9.8
Spangsbjerg (1925-1926	6.4	4.4	10.8
Hornum (1925-1926)	3.7	2.6	6.3
Askov (1923-1927)	5.3	2.7	8.0
DCE-stations (1990)	5.8	4.5	10.3
DCE-stations (2015)	4.2	3.5	7.8

*Copenhagen here refers to Landbohøjskolen 3 km west of Copenhagen centre. Blangsted is located 1-2 km east-south-east from Odense city centre. Spangsbjerg is located very close to and north-east of Esbjerg. Hornum is placed 15 km east of Livø Bredning, Limfjorden and about 40 km west of the nearest larger city (Aalborg). Askov is in southern Jutland and approximately mid between Esbjerg and Kolding. The term DCE-stations here refers to the average of the wet deposition at Anholt, Keldsnor, Lindet, Ulborg, Sepstrup and Pedersker.

It is mentioned in chapter 2, that the loss of nitrogen including volatilization from storage etc. may have been high around year 1900. If this is correct, it may have led to a higher deposition than the average close to the storage etc. However, the modeled deposition of 4 kg N/ha is the best estimate of the general deposition for the entire Danish area.

3.3 Model calculations of deposition in 1900 using DEHM

The average deposition of nitrogen to Danish land surface, fresh and marine waters in 1900 and 2000 has been calculated using the Danish Eulerian Hemispheric Model (DEHM), which is used for the annual model calculations of deposition in connection with the Danish Air Quality Monitoring Program under NOVANA (the National Monitoring and Assessment Program for the Aquatic and Terrestrial Environment).

DEHM (version 5.0) is an Eulerian model, where emissions, atmospheric transport, chemical reactions as well as dry and wet depositions of air pollutants are calculated in a 3D grid covering the northern hemisphere with a resolution of 150 km x 150 km. The model includes a two-way nesting capability, which makes it possible to obtain higher resolution over limited areas. Three nested domains are used in the model runs under NOVANA, where the first domain covers Europe with a resolution of 50 km x 50 km. The second domain covers Northern Europe with a resolution of 16.7 km x 16.7 km. The calculations of air quality in Denmark are carried out in a third domain with a horizontal resolution of 5.6 km x 5.6 km. Results from the second domain are used in this study. In the vertical direction, the model is divided into 29 layers covering the lowest 15 km of the atmosphere. Of these, the lowest layers are relatively thin (20 m), while the upper layers are relatively thick (2000 m). The model includes a comprehensive chemical scheme designed for calculating the chemical reactions in the lower part of the atmosphere. Further details about DEHM can be found in Brandt et al. (2012 or under http://www.au.dk/thor). The meteorological data used as input for DEHM is calculated using the meteorological model MM5v7 (Christensen, 1997; Brandt et al., 2012).

Two different emission inventories are used as input to the model in this study. The first emission inventory is the reconstructed emissions covering 1900 and 2000 that are available in the Representative Concentration Pathways (RCP) database (Lamarque et al., 2010). The primary purpose of this inventory is to provide consistent gridded emissions of reactive gases and aerosols for use in chemistry model simulations needed by climate models for the Climate Model Intercomparison Program #5 (CMIP5) in support of the Intergovernmental Panel on Climate Change (IPCC) Fifth Assessment report (AR5). The emission data are available at http://tntcat.iiasa.ac.at/RcpDb/dsd?Action=htmlpage&page=welcome.

The second emission inventory (called NOVANA) consists of the standard emission data used in connection with the Danish Air Quality Monitoring Program. This emission inventory is only used in order to be able to compare the model results using RCP with measurements and the standard model calculations carried out as part of the monitoring program. Only data for 2000 is used in this part of the study. This NOVANA inventory has a geographical resolution of 1 km x 1 km for Denmark that subsequently is transformed into the 16.7 km x 16.7 km resolution domain. For the area outside Denmark, an international inventory with resolution of 50 km x 50 km is used. The emissions are based on Danish national emission inventories compiled by DCE – Danish Centre for Environment and Energy (http://envs.au.dk/en/knowledge/air/emissions/) and international emission inventories collected and distributed by EMEP (www.emep.int).

Table 3.1 shows the results from model calculations of the annual average nitrogen deposition to Danish land surface, marine and fresh waters for 2000 using the two different sets of emission inventories and meteorological data for 2000. As seen, there is good agreement between the results using the two different emission inventories.

The annual deposition of nitrogen is also determined based on the measurements in the monitoring program (Ellermann et al., 2015). The wet precipitation and air concentrations of the main inorganic nitrogen compounds are all measured at four measurement stations in Denmark (Figure 3.1). Dry deposition is subsequently calculated based on the measured air concentrations. By addition of this dry deposition and the measured wet deposition, we can obtain a value for the total nitrogen deposition at the four measurement stations that, to a large extent, is based on measurements and is independent of the model used and the choice of emission inventories. On average for the four stations, this semi measured deposition is 13.5 kg N/hectare for deposition to land surface. Deposition to the marine waters is only calculated for the two coastal measurement stations (Keldsnor and Anholt), and the average deposition is 10.8 kg N/hectare for the annual deposition to water surfaces. The semi measured depositions and the model calculated depositions (Table 3.1) are therefore in good agreement.

Table 3.2. Average deposition of nitrogen to Danish land surface, marine and fresh waters in average for 2000 using meteorology for 2000.

	Marine waters kgN/hectare	**Fresh waters kgN/hectare**	**Land surface kgN/hectare**
NOVANA emissions	9.5	11.8	16.5
RCP emissions	10.5	10.1	13.7
Difference	-9 %	13 %	17 %

Figure 3.1. Map showing the position of the Danish measurement stations used in this study. The measurement stations at Tange and Sepstrup are regarded as one station in this context.

Table 3.3 presents the results from the model calculations of the historic depositions in 1900 together with the results calculated for 2000. We have calculated the deposition for an average of 12 years in order to eliminate the influence of the annual variations due to the natural variations in the meteorological conditions from year to year. Hence, we have carried out the model calculations for 12 years using meteorology from 2000 to 2012 using RCP emissions for 1900 and 2000. The results show that the depositions in 1900 were from 26 % to 30 % of the depositions in 2000. The higher depositions to land surfaces in both 1900 and 2000 compared to marine and fresh waters are mainly due to a lower deposition rate because of lower roughness of the surface and because certain compounds do not deposit on water surfaces (nitrogen dioxide). In addition, marine waters are on average further away from the sources than fresh waters and land surface.

Table 3.3. Average deposition of nitrogen to Danish land surface, marine and fresh waters on average for 1900 and 2000, respectively. The values are average for the meteorological conditions in the period 2000-2012 and are based on the RCP emissions.

	Marine waters kgN/hectare	Fresh waters kgN/hectare	Land surface kgN/hectare
1900	2.5	2.7	3.9
2000	9.7	9.8	13.1
1900 in % of 2000	26 %	28 %	30%

3.4 Model results from literature

In 1998, Alveteg et al. published results from a study in which they reconstructed historic atmospheric depositions of nitrogen in relation to nutrient uptake in forests for different forest sites throughout Europe. They estimated the long-term trends between 1800 and 2000 for non-marine deposition of nitrate and ammonia separately. From the model calculations in the Danish Monitoring Program (Ellermann et al., 2007), it is known that the depositions of reduced and oxidized nitrogen account for about 43 % and 57 % of the total deposition in Denmark, respectively. Combining this ratio with the trend from Alveteg et al. (1998), it is possible to calculate the relative trend for nitrogen in the period from 1900 to 2000 (Figure 3.2). The deposition peaks in 1980 and decreases towards 2000 due to the international efforts taken to reduce the emission of nitrogen to the atmosphere. The deposition in 1900 is about 21 % of the value in 2000.

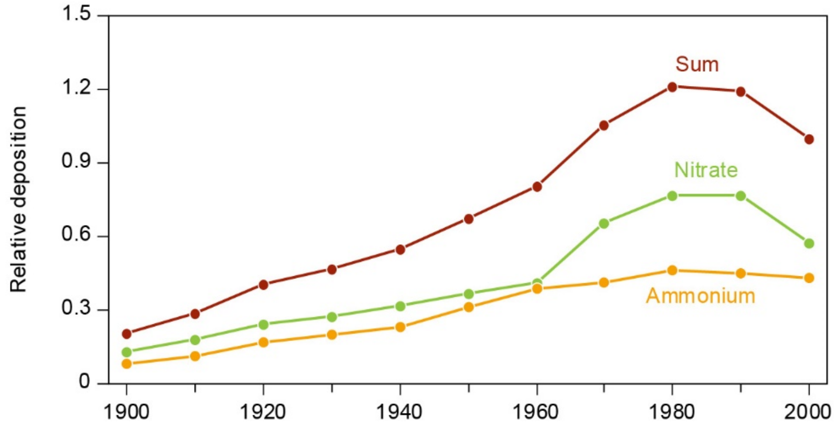

Figure 3.2. Relative time series of the non-marine deposition of nitrate, ammonium and the sum of nitrogen to Danish land surface estimated from Alveteg et al. (1998). The ratio between nitrate and ammonium is estimated from Ellermann et al. (2007). The sum is scaled to 1 in 2000.

Ruoho-Airola et al. (2012) carried out a study of the atmospheric nutrient input to the Baltic Sea from 1850 to 2006, and as part of this work they compiled a long time series for the deposition of nitrogen to the entire Baltic Sea. The time series was made by combining deposition data from the Baltic Nest Institute from 1970 to 2006, historical monitoring data and the RCP emission inventory. The deposition is given as the total deposition to the entire Baltic Sea and, hence, is not directly comparable to our own results. For this reason, we have calculated a relative time series based on their data (Figure 3.3). In general, the pattern of the time series is in good agreement with the data from Alveteg et al. (1998), although the increase in 1950-1970 and the decrease from 1980-2000 are steeper than determined by Alveteg et al. (1998). The deposition in 1900 is about 20 % of the value in 2000.

Rouho-Airol et al. (2012) based part of their long time series on actual measurements of nitrate and ammonium in precipitation at Rothamsted (UK) from 1853 to 1984. These data showed approximately a 3 and 4 fold increase in the wet deposition of ammonium and nitrate, respectively, between 1900 and 1980. These data are generally seen as one of the most extensive and valuable observation series of deposition of nitrogen in Europe.

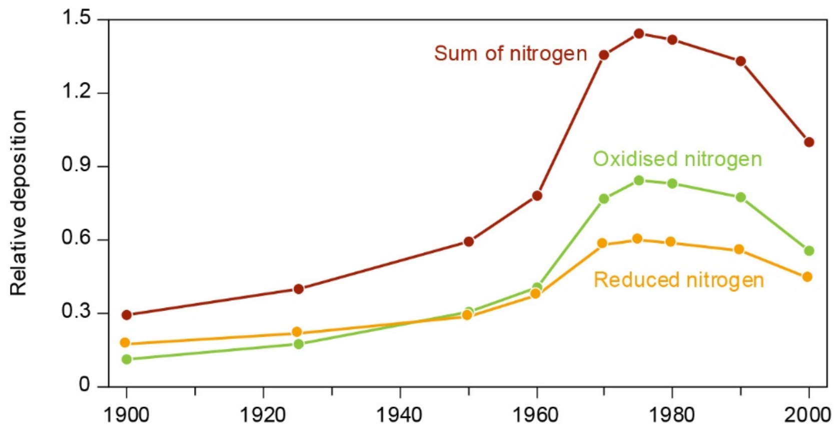

Figure 3.3. Relative time series of the deposition of oxidized nitrogen, reduced nitrogen and sum of nitrogen to the entire Baltic Sea. The relative time series are calculated based on the deposition data shown in Figure 3 in Ruoho-Airola et al. (2012). The deposition of the sum of nitrogen is set to 1 in 2000 and this corresponds to a total deposition of inorganic nitrogen to the Baltic Sea of about 240.000 tons of nitrogen annually.

A large consortium consisting of more than on hundred leading sceintists on atmospheric nitrogen has recently published their final report (Sutton et al., 2015), and their work included model calculations of the historical time series of deposition of nitrogen to EU28, including Norway and Switzerland. The model calculations were carried out using the EMEP model from the Meteorological Synthezising Centre West and the Swedish MATCH model that used the same set of emision data. Emission data were based on data for 2005 from IIASA that were extended back to 1900 based on EMEP emissions for 1990 and RCP data for the period from 1990 back to 1900 (see further details in Sutton et al., 2015). The resulting time series using the two different models are shown in Figure 3.4. The two models are in general in good agreement, with MATCH being 16-23 % higher than EMEP (Figure 3.4 above), and the relative trends are nearly identical (Figure 3.4, below). A difference between two models of about 20% is regarded as a small difference in this context, since the uncertainties connected to estimation of the emissions of nitrogen in 1900 are much larger than 20 %.

Again, the time series generally show the same pattern as obtained by Alveteg et al (1998) and Rouho-Airol et al. (2012). Moreover, the deposition of nitrogen in 1900 is about 31 % of the deposition in 2000, and this is also close to the results presented above. The absolute values of the deposition are as an average for the two models about 3.1 and 9.8 kg N / hectare as average for EU28 plus Norway and Switzerland for 1900 and 2000, respectively (Sutton et al., 2015). These values are also very close to the results for Denmark, estimated in this study (3.8 and 13.1 kg N / hectare for 1900 and 2000, see table 3.3). The higher depositions in Denmark in 2000 compared to the average of EU28 plus Norway and Switzerland is due to the high emission in Denmark and the neighbouring countries south of Denmark compared to the average of EU28 plus Norway and Switzerland.

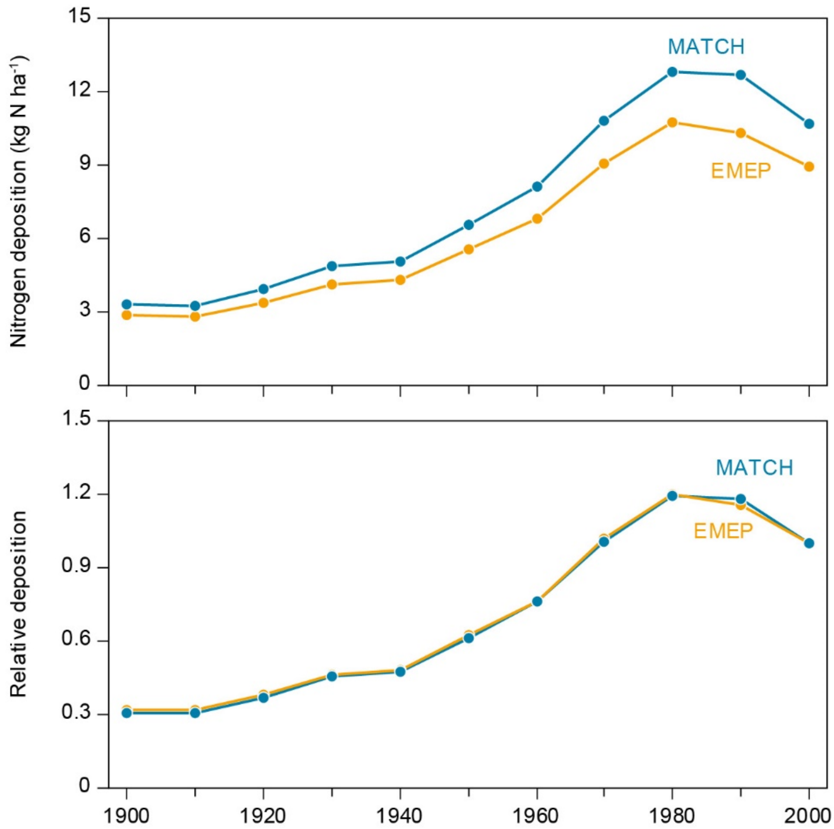

Figure 3.4. Above: Average nitrogen deposition to EU28 + Norway and Switzerland calculated using the EMEP-model and the MATCH-model (Sutton et al., 2015). Below: Relative time series of the deposition of nitrogen based on the data shown in the figure above. The deposition of the sum of nitrogen is set to 1 in 2000.

3.5 Discussion

The high wet depositions of nitrogen measured in both 1880-1885 (Tuxen, 1890) and 1923-1927 (Hansen, 1931) are, as explained in section 2, not in line with the current expectations. Mostly, ammonium is much higher than expected, with the wet deposition in 1890 being higher than ever measured in the current monitoring program by a factor of approximately 2. Moreover, the ratio between ammonium and nitrate deviates considerably from the current expectations. In 1880-1885, the ratio was 4:1, in 1923-1927 2:1 and in 1990 and 2015 1:1. In addition, there were large inter-annual variations in the measured wet depositions. All in all, this indicates two main reasons for the large depositions measured in the two historic data sets:

- The equipment used to sample precipitation was contaminated with ammonium and perhaps, to some extent, with nitrate. Even in the current measurement program, it is very difficult to avoid considerable contamination of the samples, unless precautionary actions are taken in order to avoid this problem. One of the main problems is bird droppings directly in the sample or on the sample equipment. In the current measurement program, double sampling is carried out at all stations, and a thorough data quality check is carried out in order to reject samples that are contaminated. Neither Tuxen (1890) nor Hansen (1931) mention anything about this problem or how they handle contamination of the samples. Therefore, it may not have been realized at that time that contamination of precipitation samples may cause big problems.

- Both Tuxen (1890) and Hansen (1931) explain large wet depositions with proximity to larger cities. Again, this is not in line with current knowledge. Nowadays, the main source of wet deposition of nitrogen is long-range transboundary transport, and local sources are, in general, regarded as minor sources. However, it might have been different in cities about 100 years ago, where modern sanitary and waste disposal systems were absent. The emissions of ammonia in the cities were most likely much higher than today, and large local contributions to the wet deposition can therefore not be ruled out.

It is believed that one or both of these reasons explain why the wet deposition is much higher than expected. Moreover, these historical data are therefore not relevant in connection to the nitrogen deposition to Danish background land areas, fresh and marine waters in 1900.

In general, the long time series determined by Alveteg et al. (1998), Rouho-Airol et al. (2012), and Sutton et al. (2015) showed the same general pattern, despite the fact that the time series were calculated for different areas (e.g. Baltic Sea and EU28 plus Norway and Switzerland). From the low deposition in 1900, there is a relatively rapid increase in the deposition, which peaks in 1970-1980 at a level that is roughly 4-5 fold higher than in 1900. From 1970-1980, there is a decrease of about 20-30 % towards 2000. This similarity in the patterns is, of course, not that unexpected due to three main reasons. Primarily, on a broad scale all of Europe has been through the same kind of development with an increasing growth of the economies that lead to ever increasing emissions until the 1970s and1980s. At that time, it was realized that regulating the emissions of air pollutants on a European level was necessary. Through international cooperation and national efforts, the emissions were decreased during the following decades and, hence, the depositions decreased from 1970-1980 to 2000. Secondly, the amount of historic data on nitrogen emissions and depositions back in time to 1900 are limited, and the three different studies therefore more or less rely on the same set of data (e.g. the RCP emission data are used in several of the studies). Thirdly, nitrogen is transported long range, and this most likely leads to similar relative time series for different parts of the European region, as long range transport evens out the spatial differences.

Table 3.4 summarizes the results for the deposition in 1900 from the various studies. As seen, there is good agreement between the results from the different studies, with the relative change lying between 20% and 32% and with good agreement between the absolute depositions determined by Sutton et al. (2015) and this work. The international studies therefore support the results from our model calculations.

Table 3.4. Deposition in 1900 and 2000 and relative change between 1900 and 2000 based on this work, Alveteg et al. (1998), Ruoho-Airola et al. (2012), and Sutton et al. (2015).

	This work			Alveteg	Ruoho-Airola	Sutton
	DK Marine waters	DK Fresh waters	DK land surface	Forest Europe	Baltic Sea	EU28+Norway and Switzerland
Depositon 1900 (kgN/hectare)	2.5	2.8	3.9	-	-	3.1
Depositon 2000 (kg N/hectare)	9.7	9.8	13.1	-	-	9.8
Deposition in 1900 in percentage of 2000	26%	28%	30%	21%	20%	32%

The uncertainties in the calculation of historical depositions are quite high. In the work of Sutton et al. (2015), it was shown that use of two different models resulted in a difference of up to 23%, even in model calculations for the period 1980-2000. However, this difference between the models is small compared to uncertainties on the emission inventories for the period around 1900, due to the limited amount of historical data on emissions around 1900. This can be illustrated by comparing emission data for 1990 from RCP and the NOVANA dataset. Figure 3.5 shows the ammonia emissions from Denmark and surrounding countries. The total emission of ammonia for this area differs with nearly a factor of two, even though the emissions are for 1990, where relatively good data are availabel for emission inventories. Moreover, the spatial distrubution differs significantly between the two inventories, where NOVANA emissions are most correctly distributed with high emissions in Jutland and RCP emissions incorrectly have the highest emissions in Zeeland. Similar data for emissions of nitrogenoxides are in much better agreement both with respect to the size and spatial distribution of the emissions (Figur 3.6), even though there still is a difference of about 20% between the total emissions. Figure 3.7 shows the spatial distribution of the emissions of ammonia and nitrogenoxides in 1900 from the RCP–emissions. It can be seen that the spatial variations are small compared with 1990. However, it is questionable whether or not the highest emissions of ammonia would take place in the most densely populated areas like Copenhagen.

It has not been possible within this work to make a thorough determination of the uncertainty on the depositions calculated for 1900. However, based on the illustrated uncertainties on the emission inventoreis, it is estimated that the uncertainty is at least a factor of 2 for the average deposition to Denmark. Due to the incorrect spatial distributions of the emissions, we have not made any attempt to determine the spatial variations in Denmark.

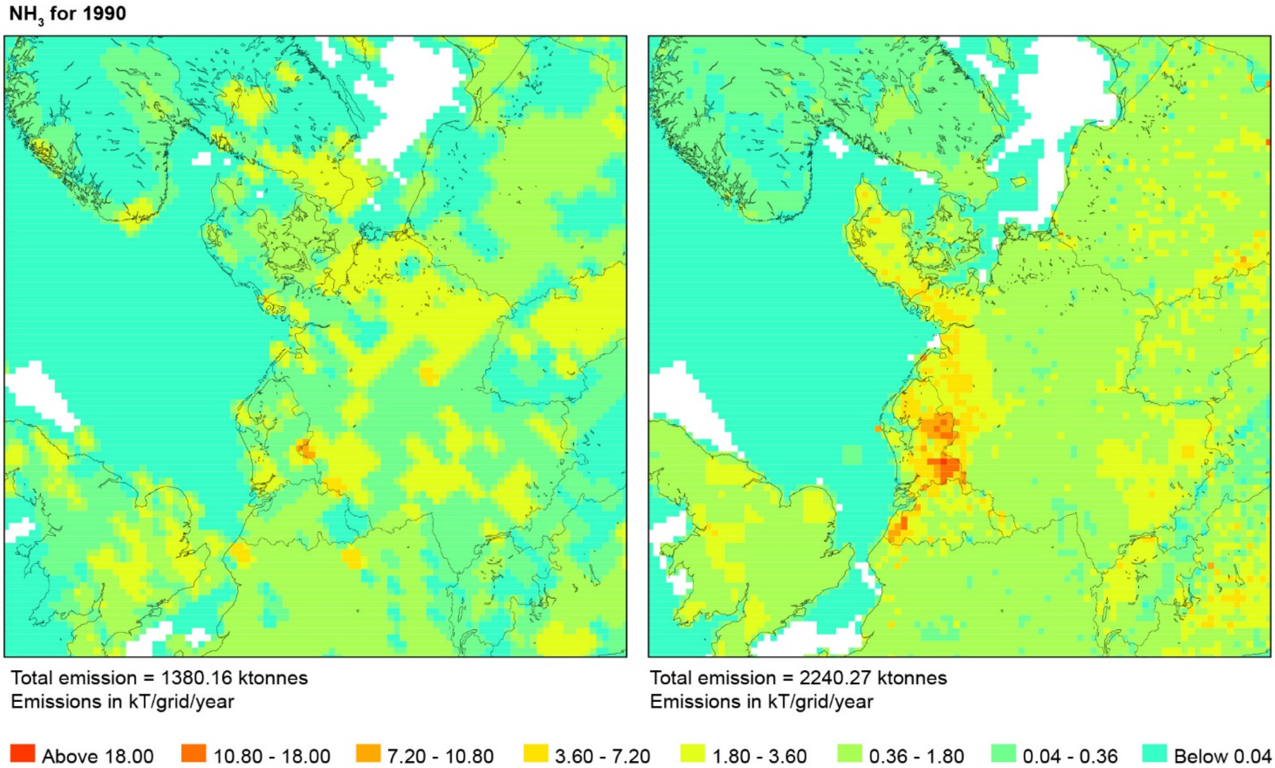

Figure 3.5. Spatial distribution of ammonia emissions in 1990 from RCP (left) and NOVANA (right). Total annual emissions to the area are 1380 ktonnes and 2240 ktonnes for RCP and NOVANA, respectively. The NOVANA dataset relies on the official national emission inventory and is regared as the best available emission inventory for Denmark.

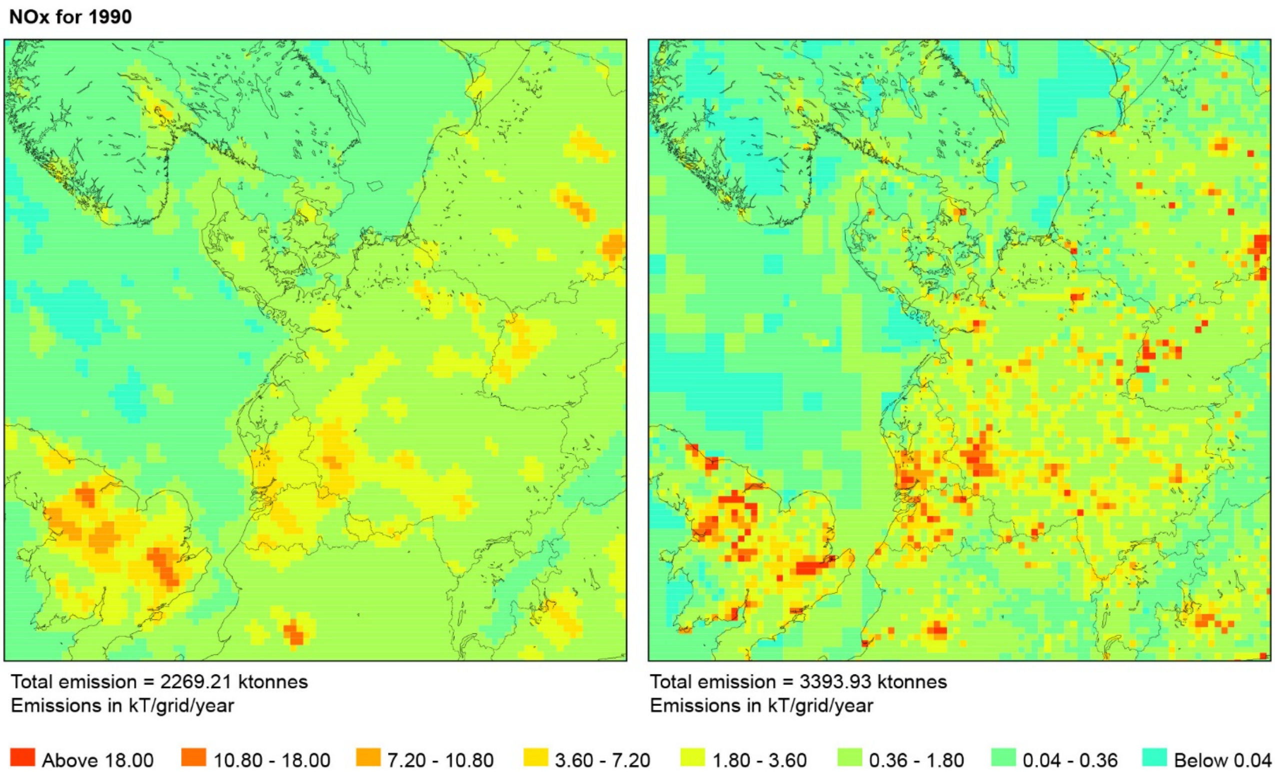

Figure 3.6. Spatial distribution of nitrogenoxide emissions in 1990 from RCP (left) and NOVANA (right). Total annual emissions to the area are 2670 ktonnes and 3390 ktonnes for RCP and NOVANA, respectively.

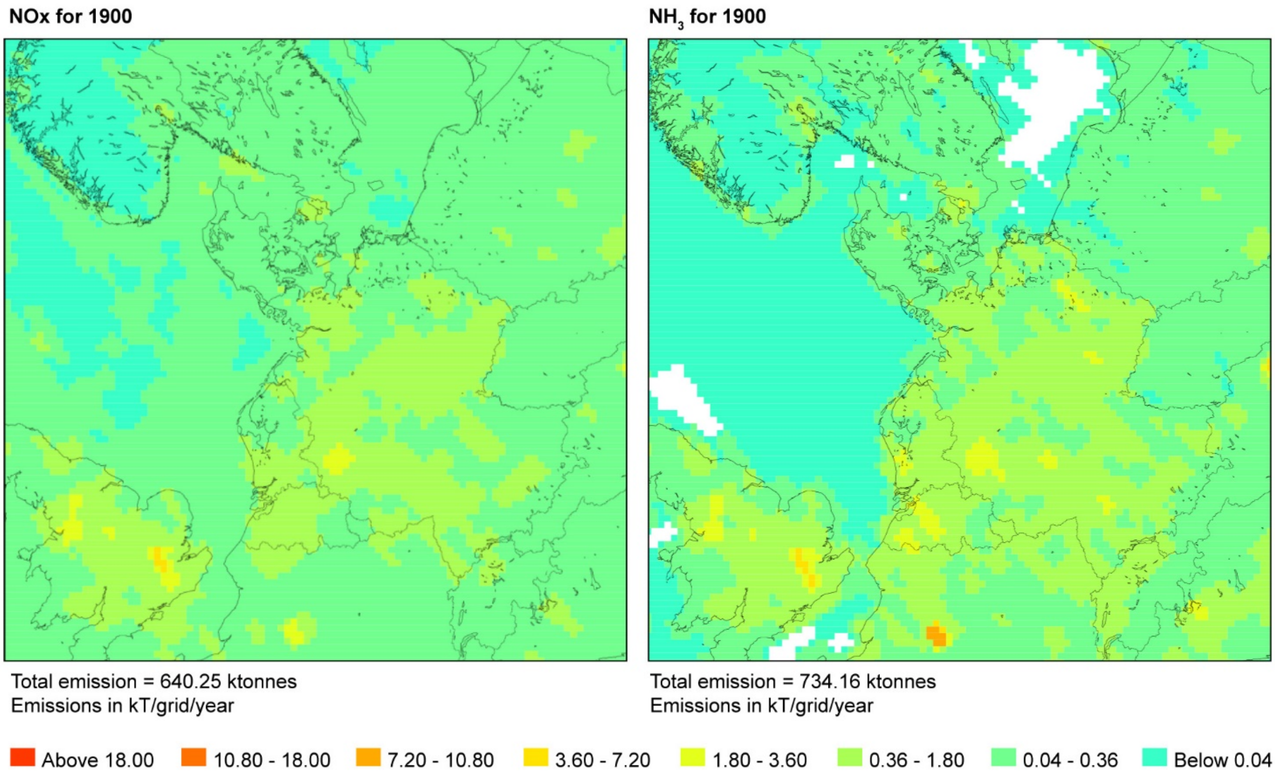

Figure 3.7. Spatial distribution of emissions of nitrogenoxide (left) and ammonia (right) in 1900 from RCP-emissions. Total annual emissions to the area are 640 ktonnes and 730 ktonnes for nitrogenoxides and ammonia, respectively.

3.6 Summary

The aim of this chapter is to provide a short review on the historical deposition of nitrogen to land areas and to fresh and marine waters in Denmark around the year 1900.

The first part of the document presents historic data on wet deposition of nitrogen measured at Danish research stations around 1900. Due to potential problems with contamination of samples and sample equipment and influence from local sources, these data are not regarded as relevant in connection with determination of the nitrogen deposition to Danish background land areas, fresh and marine waters around the year 1900.

The second part presents results from model calculations of the deposition of nitrogen to Danish land areas, fresh and marine waters in 1900 and 2000. These data are compared with three international studies that also focus on the historical trends of nitrogen de-position, although these studies present average depositions on a regional level, e.g. all EU or the entire Baltic Sea. Finally, a brief discussion of the main uncertainties related to estimation of depositions around the year 1900 is presented.

The depositions of nitrogen in 1900 to Danish marine and fresh waters are about 3 kg N / hectare, respectively. The deposition to Danish land surface is about 4 kg N / hectare. The nitrogen deposition in 1900 was 26-32% of the deposition in 2000. The uncertainty of these estimates for 1900 is quite high due to the limited amount of historical data on emissions to the atmosphere.

The results from other model calculations are supported by results from international studies on the historical development in nitrogen deposition in Europe. These studies have been carried out by a large number of leading scientists on this field in Europe.

4 Point Sources around year 1900

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Abstract

Purpose: The aim of this chapter is to provide a brief overview of the nitrogen point sources around the year 1900.

Materials and methods: The present point sources are in reporting divided into the following categories, wastewater treatment plants (mostly treating urban residential wastewater), industrial wastewater and fish farms. This grouping is also used in this chapter. The information used in this chapter is of historical origin. A point source is defined as a single identifiable source of water.

Results and discussion: It was found that there were very few wastewater sewers outside Copenhagen around the year 1900 and Copenhagen was by no means fully equipped. Most urban waste water containing human faeces would have been used as agricultural manure and therefore not to be regarded as a point source. The presence of farm animals in towns would have added to the amount of nitrogen lost from urban areas around the year 1900. It is difficult to estimate the nitrogen load from households / urban areas, and no attempts have been made to quantify this. Some industries would have directed wastewater into fresh and marine waters, as proximity to a recipient was an important location factor in some cases. No attempts have been made to quantify the load from industrial sources.

Conclusions: The nitrogen load from urban point sources is expected to have been small around the year 1900, as there was only a very limited amount of sewers. The presence of large farm animals in towns would have added to the amount of nitrogen rich wastewater from urban areas compared to a situation where urban areas were free of farm animals. Most human and animal waste (excrements) would around the year 1900 not have been directed to aquatic recipients through point sources but will have been used as fertilizers. The amount of industry was limited, but there was no wastewater treatment. In some cases, the N and P load could have been considerable and above modern day levels locally, and in other cases considerably lower. The N load from fish farms would have been very limited.

4.1 Urban waste (humans and animals)

This is a brief summary of the nitrogen point source situation in Denmark around the year 1900. Further work and a deeper look into historical material is needed to get a quantitative estimate of the loads at the time around the year 1900.

Sewers from domestic households were almost non-existing in Denmark around the year 1900. The population was around 2.45 million people in 1900 of which 43% lived in some kind of urban area (Danish statistics). Sewers were largely installed in cities and larger towns only after 1900. Pictures exist from sewer installation earth work in Odense around 1908 (1905-1920) (Figure 4.1).

According to Conradsen, 2004 (in Hofmeister (Ed.), 2004), a few areas in Copenhagen had sewers installed as early as the 1860s. The main sewers were installed in the period 1892-1903 but far from all households were connected or had water closets installed.

Figure 4.1 Sewers are installed in Odense. Pictures from around 1908 and 1905-1920

In general the sanitary installations (human waste) consisted of "buckets" that were emptied by the "night men". Generally the human waste were deposited outside the city limit and used as manure in agricultural fields. There were about 30.000 buckets in Copenhagen around the year 1900 reduced to 14.000 in 1925 (http://www.kloakviden.dk/historie). The "bucket system" would have yielded a relatively high retention compared to the period with sewers and no waste water treatment, where the retention/cleaning would have been very close to 0%.

If all human excrement from the urban population, 857.160-1.041.854 persons (Danish statistics, for 1901 depending on including "suburbs" and "small towns" with "larger towns" (købstæder) or not), was delivered directly to the aquatic environment, assuming 4 kg N pr. person pr. year (Hong et al., 2012), it would amount to 3400 to 4200 tons pr. Year. This is of course a large overestimation of the actual input, as excrements generally were not dumped directly into the aquatic environment.

The nitrogen load from point sources as we know it today did almost not exists as the amount of sewers was very limited. This does not mean that urban areas did not affect the aquatic environment. Probably, much street garbage, including manure from urban living animals (Horses, Cows, Pigs, Sheep etc.), ended up in rivers, lakes and harbours, especially during heavy rain events (Figure 4.2). There would have been around 20.000 horses in Danish towns around the year 1900, assuming that the fraction of 4% "urban horses", as found in 1941, applies to the period around the year 1900 as well. Of these, about 50% would have been for agricultural use (http://www.dst.dk/Site/Dst/Udgivelser/GetPubFile.aspx?id=19920&sid=landbbd2). In Copenhagen, there would have been at least 500 horses just for taxi driving (http://www.b.dk/kultur/taxi-krigen-begyndte-med-500-heste) around the year 1900. In street photographs from the period, animals are commonly seen (and manure as well).

It is not possible to give a robust quantitative estimate of the nitrogen load from urban point sources based on the available data material.

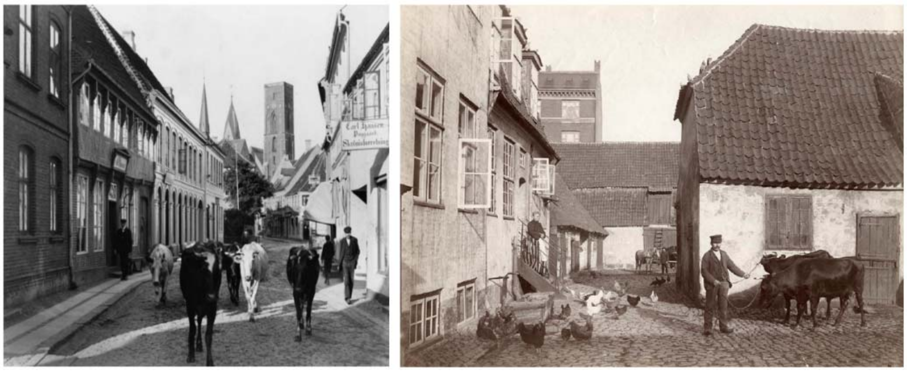

Figure 4.2. Left: Cows in the high street of Ribe, 1900-1920. Right: Cows, horses and hens in the back yard of Allégade 22, Copenhagen/Frederiksberg around year 1900.

4.2 Industrial point sources

The level of industrial production in Denmark was small compared to modern levels. On the other hand, the losses of Nitrogen would in most cases have been larger (pr. produced unit) and there would have been no wastewater treatment. Below are listed some industries that were active at the time (Table 4.1).

Table 4.1. Overview of industries delivering waste water around the year 1900.

Corporative Dairy plants (1067 units in 1901*) (Fig. 4.3)	• 1871 1060 mill. Kg milk (808.000 cows) • 1914 3600 mill. Kg milk (1.310.000 cows)
Corporative Butcheries (26 corporative industrial units in 1900, 50% of all animals butchered)*	• 1871 442.000 pigs • 1900 675.000 pigs* • 1914 2.497.000 pigs
Power plants	• 10 smaller units • Cph 3, Odense (first one), Køge, Aalborg, Vejle, Hjørring, Nakskov, Kolding
Gasworks	• First one in Odense 1853 • 2% N in rock-coal (NH4, cyan, rhodan or into tar)
Fish/Seafood industries	• Apparently not on industrial scale
Sugar factories	• 1873 2 factories (Nakskov and Odense) • 1912 9 factories
Breweries	• Industrialisation starts in 1847, some industries in 1880s • Replaces local/home brewing
Soya oil	• Production of oil and fodder in Aarhus since 1871
Distilleries	• Danske Spritfabrikker is founded in 1881, industrial scale from that period onwards • Replaces small scale production • The waste from small-scale production (also Breweries) fed to pigs was one of the main reasons to the relatively large amounts of pigs in towns in the mid 18-hundreds (Jørgen Mikkelsen, Landbohistorisk Tidsskrift 2010:2). But probably the small scale distilleries largely stopped before 1900 due to regulations of alcohol production
Paper mills	• About 6 in 1900 • (Silkeborg, Dalum, KBH, Næstved)
Tanneries	• Both local and smaller industry tanneries. Tanning was done in various ways but “fat tanning” and “urine tanning” have been used and would have produced nitrogen rich wastewater. From industrial sized tanneries the amount of waste could have been considerable

*Thomsen, 1965

Figure 4.3. Coorporative dairy plants in 1890 (from Bjørn, 1988).

It is difficult to estimate the nitrogen load from these industries around the year 1900, as we have no information about how much sewage water was produced or how much nitrogen it contained.

4.3 Fish farms

The first fish farms were established in 1895 in Vejle municipality. The large expansion in the numbers of fish farms took place between 1950 and 1970. The nitrogen load from fish farms around the year 1900 would therefore have been very small.

4.4 Farm point sources

Farm point sources are direct losses from farms to surface waters. The most important sources were manure heaps and liquid manure containers. In cases where manure heaps were located in proximity to surface waters, loses of N to surface waters could have occurred in periods with heavy rain. It is estimated that the large majority of manure heaps around year 1900 did not have paved (water tight) floors and collection of the potentially overflowing fluid parts in tanks. As in most cases the manure heap would have been located directly on top of the soil, It would have been a spot of very high N leaching. No attempt has been made to quantify the farm point source contribution.

4.5 Conclusions

The nitrogen load from urban point sources is expected to have been small around year 1900, as there was only a very limited amount of sewers and the urban population was much lower than at present. The presence of large farm animals in towns would have increased the amount of waste, but most of the manure is believed to have been used as fertilizer. The amount of industry around year 1900 was limited, on the other hand there was no wastewater treatment. The load could, in some cases, have been considerable and above modern levels and, in other cases, would have been considerably lower. There were almost no fish farms around the year 1900, therefore, the N load from these would have been very limited.

Quantification of the nitrogen load from point sources would ideally be based on input from historians and wastewater treatment engineers.

5 Historical data, models and review of nitrogen concentrations in streams and rivers

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Abstract
Purpose: The aims of this chapter are to retrieve and analyze existing longer-term Danish datasets from streams and rivers regarding measured nitrogen (N) concentrations. Furthermore, two existing statistical models are applied in an attempt to predict possible concentration of total nitrogen or nitrate nitrogen in Danish streams around the year 1900. Finally, we review existing international literature on measured N concentrations in rivers around the year 1900.

Materials and methods: A database retrieved from the Geological Survey of Greenland and Denmark (GEUS) with old and longer-term measurements of N concentrations in Danish streams was investigated and relevant data further analyzed. Three existing statistical models for relationships between N concentrations in oxic groundwater and streams were re-analyzed and used for a prediction of the N concentration in streams around the year 1900. Finally, existing international literature was reviewed for findings of land use and N concentrations in rivers around the year 1900.

Results and discussion: Longer-term N concentrations measured in four large Danish streams show an increase from 1965 to 1990, followed by a decreasing trend from around 1990 to 2015. Measured N concentrations in the River Odense tightly follows the trend in N surplus in Danish agriculture. Early measurements from the River Suså show that the nitrate-N concentration in 1939/40 from diffuse sources amounted to ca. 2.1 mg N/L. Generally, nitrate-N concentrations in the few water samples analyzed from streams around year 1900 are very low, with a clear increase in pattern from western to eastern Denmark. Application of two existing statistical models to predict possible N concentrations in Danish streams around the year 1900 showed concentrations to be around 2 mg N/L, although values of present day groundwater and surface water retention were used. The predictions with these simple models are, however, associated with a high degree of uncertainty. A review of the international literature concerning measured N concentration data in rivers draining catchments with low impacts from point sources, generally showed very low N concentrations (< 0.6 mg NO_3-N/L). However, many of the rivers drained large river basins with a relatively lower proportion of agriculture in the river basins around the year 1900 compared with the situation in Denmark.

Conclusions: Based on the analysis of both historical monitoring data, application of three different statistical models and review of the international literature, the concentration of nitrogen in streams and rivers around the year 1900 was most probably between 1-2 mg N/L. This is considerably lower than the present day concentration in Danish streams, which, on average, is around 4-5 mg N/L.

5.1 Background and aims

The reference concentration of nitrogen (N) in streams and rivers and the reference N loading to coastal waters are important parts of the EU Water Framework Directive when assessing present day boundary between moderate and good conditions for water chemistry and ecology in water bodies. The agreement in Denmark and internationally is that the historical N concentrations and loadings around the year 1900 should be considered the reference situation for coastal waters. The aim of this chapter is threefold: i) to find and analyse existing long-term N concentration data from streams and rivers in Denmark and internationally; ii) to apply statistical models to assist in analysing the N concentrations in streams and rivers around the year 1900; iii) to conduct a review of the international literature for historically measured N concentrations in streams and rivers.

5.2 Materials and Methods

Long-term historical N concentration data were retrieved from the Geological Survey of Denmark (GEUS) and Greenland and from the Danish national database (ODA). Moreover, statistical models were developed between N concentrations and N surplus based on observations from the NOVANA monitoring stations. An empirical model developed by GEUS for the historical concentration of nitrate in oxic groundwater and agricultural application of N was also tested and applied for simulation of the resulting nitrate concentration in surface waters utilizing a general modelled N retention in groundwater (Hansen & Larsen, 2016; Højbjerg et al., 2015a+b). Finally, a review of the international literature for historically measured N concentrations in rivers was conducted.

5.3 Danish historical data

The first Danish nitrogen concentration data was sampled in six major Danish rivers in 1889 and 1892 (Westermann, 1898). Water sampling was conducted upstream major towns, and sampling was done in November, 1889, and again in June and August, 1892. Such a sampling program with water samples taken during 3 months of the year most surely results in biased estimates of the mean annual N concentration. A statistical analysis of the impact of sampling the same three months, as in the Westermann (1898) described sampling program in the period 1990-2000, reveals that such sampling underestimates the mean annual concentration with a factor of 1.03-1.76, the highest deviation being for sampling in the River Gudenaa and the lowest in the River Odense.

All water samples were brought to the Danish Agricultural University in Copenhagen and analysed for nutrient concentrations (Westermann, 1898). Storage of the waters during transport and the quality of the chemical analysis from that period may be critical, and it is therefore not possible to determine the uncertainties involved. The results might show some tendencies and patterns that could help interpreting the concentration level of N in Danish rivers nearly 130 years ago.

The concentration of nitrate-N, which is a key indicator for losses of N from agricultural fields, was very low in the 6 rivers sampled. The nitrate-N concentration was highest in the two rivers draining loamy soils on Zealand and Funen (Table 5.1). Only trace concentrations of nitrate-N were found in all the sampled rivers in Jutland (Table 5.1). This pattern is similar to the pattern found today, with highest nitrate-N concentrations in streams draining loamy soils and lower concentrations in streams draining sandy soils. The mean concentration of organic N measured in the 6 rivers was generally higher than for nitrate-N (Table 5.1), which is opposite to what is found today in Danish rivers. The measured mean concentrations of organic N in rivers sampled in 1889-1892 may be higher because two of the samples were sampled during low flow conditions (June and August 1892), as also described by Westermann (1898). However, higher organic matter concentrations in rivers may have occurred during the period of sampling because of greater abundance of organic soils and wetlands at yhe time. The extremely high concentrations of ammonia-N reported in Westermann (1898) (Table 5.1) in all rivers found to be > 1 mg N/L seems unrealistically high, given that the rivers sampled must have been nearly without point source inputs. An analysis of the ammonium-N concentration in the same rivers during a period with high sewage inputs from point sources (1978-1987) shows that the concentration of ammonium was highest in the River Suså (0.60 mg NH4-N/L) and lower in both the River Odense (0.39 mg NH4-N/L), the River Skjern (0.23 mg NH4-N/L) and River Gudenå (0.12 mg NH4-N/L). Therefore, the reported ammonia concentrations from Westermann (1898) are judged not to representing realistic ammonium concentrations in the rivers.

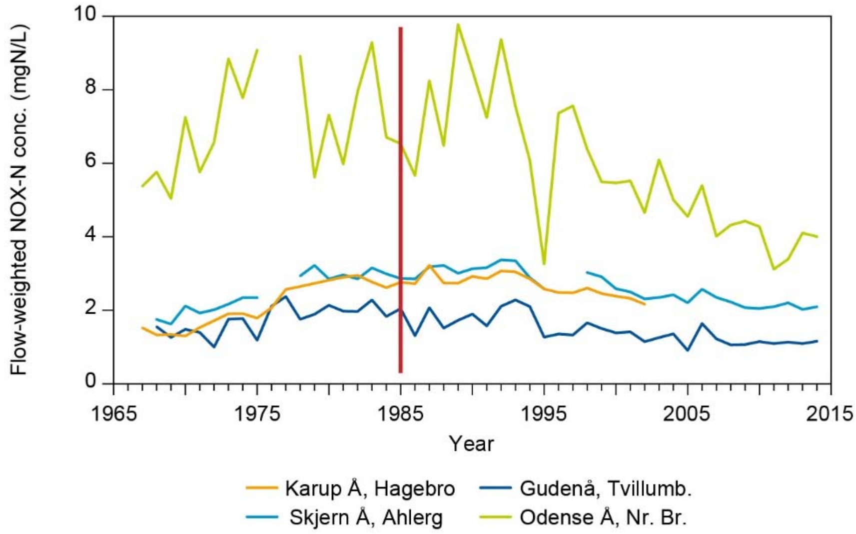
Table 5.1. Nitrogen concentrations from 6 major Danish rivers sampled during 1889-1992.

River sampled	Nitrate-N (mg N/L)	Organic N (mg N/L)	Ammonia N (mg N/L)
Susåen, Broby Vesterskov	0.96	1.78	1.21
Odense Å, upstream Odense	0.62	1.21	1.22
Storåen, upstream Holstebro	0.10 (traces)	0.67	1.22
Skjern Å, Hesselvig	Traces	0.92	1.46
Kongeåen, upstream mouth	Traces	1.01	0.95
Gudenåen, upstream Silkeborg	Traces	0.64	1.48

We only found time series of N data back to the 1960s for some larger Danish rivers sampled as part of the first Hydrological Decade (UNESCO, 1965-1975) and later for reporting to OECD and as part of the NOVANA program. One monitoring station in Suså having data from 1939-40 was also found in the GEUS database. Other older sites typically only had 1-2 measurements of N concentrations in streams and rivers, which are too few for further use in this chapter.

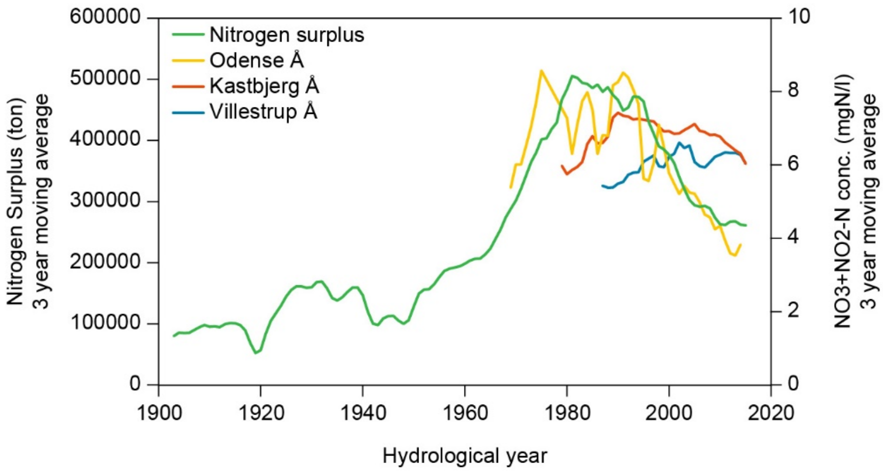
Annual flow-weighted nitrate-N concentration data from four large Danish rivers are presented in Figure 5.1. The first measurements date back to the mid-1960s and the nitrate-N concentrations are generally lowest in the 1960s and increasing towards the 1980s, after which the concentration again decreases due to the Danish Nutrient Action Plans that started in 1985 (Figure 5.1). The concentrations of nitrate-N are much higher in River Odense than in the three other rivers, possibly caused by the combination of intensive agricultural production and a major part of soils being tile drained. The west nitrate-N concentration are measured in the River Gudenå throughout the period because the monitoring station is situated downstream a series of larger lakes, where a large amount of the inflowing N is removed. The N concentrations in River Karup and River Skjern are also generally low, as both catchments have predominantly sandy soils with a low proportion of tile drainage and the rivers are predominantly groundwater fed. The annual flow-weighted concentration of nitrate-N was below 2 mg N/L in River Skjern, Karup and Gudenaa in the 1960s, whereas it amounted to 5-6 mg N/L in River Odense (Figure 5.1). Today, the concentration of N is lower than in the mid 1960s in the River Odense and River Gudenaa (Figure 5.1).

Figure 5.1. Annual flow-weighted concentrations of nitrate-N in four large Danish rivers during the period 1965-2015. First NPO Action Plan indicated with red line.



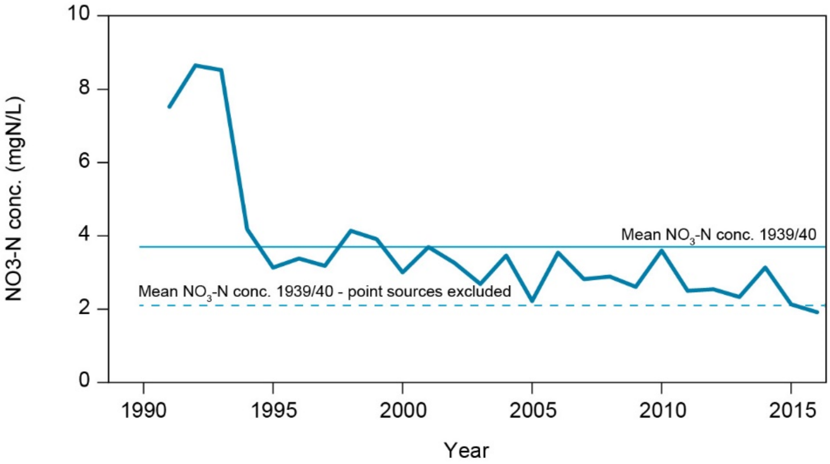
The annual flow- weighted concentration of nitrate-N in River Odense has been coupled to the surplus of N in the catchment throughout the nearly 50-year monitoring period (Figure 5.2). The nitrate-N concentration in river Odense generally closely follows the development in the national N surplus in Denmark during the nearly 50-year observation period. The only exception is in very dry years, such as 1973/74 and 1995/96, where very little water percolated through the root zone from the fields. The continuing lowering of the nitrate-N concentration in river Odense after 2005 might be due to the large investments in restored wetlands (Windolf et al., 2016). It is striking that the nitrate-N concentration in river Kastbjerg and river Villestrup also follows the national N surplus, but is delayed with 20 and 40 years, respectively. The latter is probably related to catchment specific long travel times for N in oxic groundwater, which, of course, also delays responses of the Action Plans implemented in these catchments. River Kastbjerg now seems to respond to the Action Plans like River Odense, but with a ca. 20-year delay, whereas the effects of the Action Plans till now are not observed in River Villestrup (Figure 5.2).

Figure 5.2. National N surplus (from Kyllingsbæk (2008) and Vinther and Olsen (2016)) from agriculture in Denmark (national farmgate balance) and the annual flow-weighted concentrations of nitrate-N from three rivers.



Measurements of the nitrate-N concentration in River Suså upstream the lakes from 1939-1940 were extracted from a database. The measurements did not cover all months, and therefore the mean nitrate-N concentration from the NOVANA monitoring program was calculated for the same months as sampled in 1939-40 (a total of 8 months). The observed concentration of nitrate-N in year 1939-40 was nearly 50% of the observed concentrations in the early 1990s (Figure 5.3). After reduction of the measured nitrate-N concentration for impacts from major industrial and urban point source outlets by performing a source apportionment subtracting the discharges of N from point sources from the calculated transport of nitrate-N, the nitrate-N concentration was estimated to be around 2 mg N/L in 1939-40 in the River Suså (Figure 5.3).

Figure 5.3. Concentration of nitrate-N as observed in the River Suså near Næsby in the two years 1939-1940 and from 1990 to 2015 from the NOVANA monitoring program.



5.4 Utilization of models to estimate historical N concentrations

Another way of assessing the concentration of N in streams is by utilizing a relationship between concentrations of nitrate-N in oxic groundwater at the time of groundwater recharge from dating of groundwater, against total application of N in Danish agriculture at the time of application based on data from 1946 – 1975 (Hansen and Larsen, 2016). The resulting nitrate-N concentrations in Danish streams have been calculated and inserted in this groundwater model based on a total N retention in groundwater amounting to 62% (Højbjerg et al., 2015a+b). The application of N in Danish agriculture around year 1900 was around 100,000 tons N (Kyllingsbæk, 2008). Utilizing the model developed for nitrate concentrations in oxic ground, this results in a nitrate-N concentration in streams around 1.1 mg N/L around the year 1900 (Figure 5.4). However, the number of sampled oxic groundwater wells was low in the first decade of the time series (1946-56: N < 10). Therefore, a new analysis of the relationship was conducted utilizing data from 1957-75 only, where the number of samples was > 10 per year. The nitrate-N concentration in streams utilizing this relationship equalled around 2 mg N/L.

This method for assessing the concentration of N in streams around year 1900 is associated with a high degree of uncertainty mainly due to:

- The relationship is used outside its validity range (1946 – 1975), where different agricultural practices, soil N pools, climatical conditions etc. might have resulted in another relationship between agricultural N application and the resulting nitrate concentrations in oxic groundwater.
- The relationship is based on a varying amount of groundwater samples from each year varying from 2 analyses in 1946 to 118 in 1975. This means that the relationship is weak in the beginning of the period. Therefore, a new model was established for the period 1957-75, where the number of samples was > 10 per year.
- The total N retention (average of 62% applied in the analysis thus being assumed constant in time) in groundwater shows a large spatial variation in Denmark and is combined with a relatively high uncertainty (Højbjerg et al., 2015a+b).

An attempt was made to validate the results. Inserting the application of N in agriculture around year 1939/40, the model reveals a nitrate-N concentration of around 2-3 mg N/L, which is close to the average nitrate-N concentration observed in River Suså during 1939/40 (see Figure 5.3). Moreover, if inserting the N application in the model in 2014/15, it forecasts a nitrate-N concentration of around 4 mg N/L in streams, and the observed concentration in streams as derived from diffuse sources was on average 3.9 mg N/L. These two 'validation' examples seem to confirm that the method applied is very robust, at least within the range covered by age dating of oxic groundwater.

Another way of analysing the reference state for N in Danish streams is to develop a statistical relationship between estimated flow-weighted total N (TN) concentrations in the loadings from Danish rivers to coastal waters from diffuse sources against the national N surplus in Danish agriculture during the two periods 1990-1999 and 2000-2014, respectively (Figure 5.5). The first period is chosen because of a large change in the handling, application and, therefore, use of N in manure that effectively reduced the chemical fertilizer use resulting a lowering of the N-surplus and N-leaching in Danish agriculture during the period 1990-1999. The second period is a period with moderate changes in fertilizer use and N-leaching. However, the established relationships cannot be taken as a correct mirror of the N-leaching situation in Danish agriculture around the year 1900, as described in chapter 2. Moreover, the relationship includes current day N retention in both groundwater and surface waters, which are expected to have been higher around the year 1900 (see chapter 6). The established relationships can, therefore, only be used as an estimation of what present days climate and agriculture would yield of resulting flow weighted N concentration in riverine waters if the N surplus was lowered to a near reference situation with no N surplus.

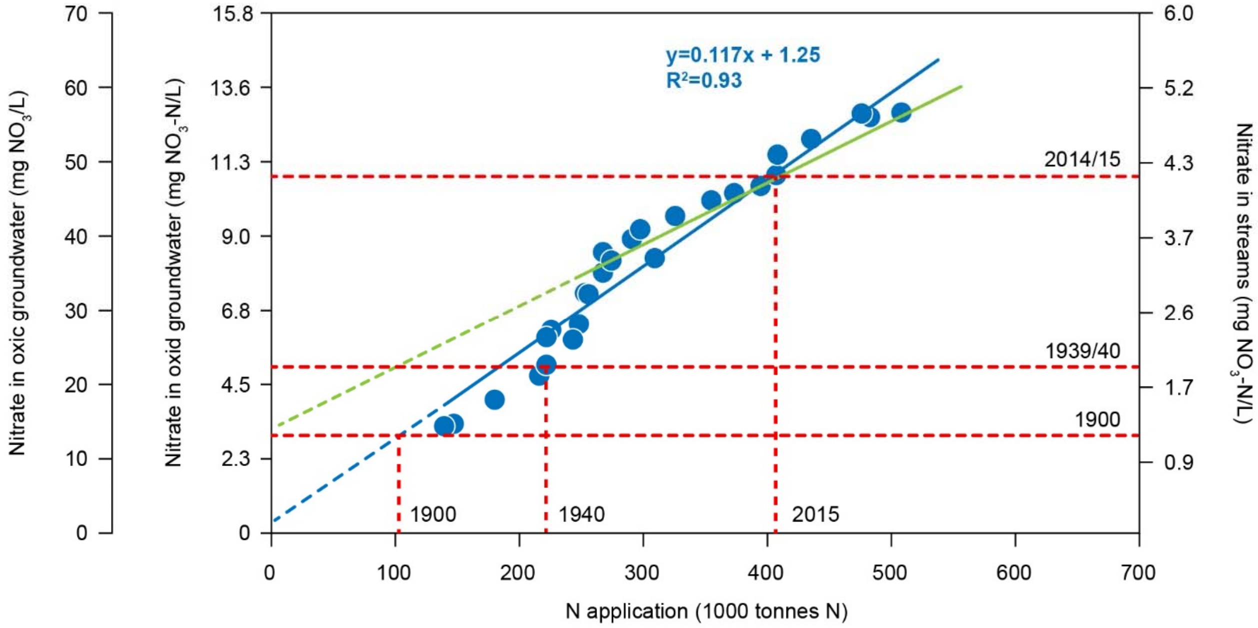


Figure 5.4. Nitrate-N in oxic groundwater (1st y-axis as nitrate and nitrate-N), concentration in stream water (2nd y-axis as nitrate-N) based on an average 62% reduction of N in groundwater and N application to Danish agricultural land (redrawn from Hansen and Larsen, 2016). Shown is also the N concentration in oxic groundwater and streams at the N application in year 1900, year 1939/40 and 2014/15.

The two established relationships explain a major part of the variance in the observations, especially if the dry year 1995/96 is omitted from the relationship established from 1990-1999 (Figure 5.5). In this extremely dry year, nearly no water percolated through the root zone on fields in Denmark. The N surplus around year 1900 (1900-1903) amounted to between 69,000-87,000 tonnes N (Kyllingsbæk, 2008). The established relationship for the period 2000-2014 shows that near natural conditions related to N concentration in riverine waters can be estimated to amount to 1.5 mg N/L in a situation where the N surplus equals zero (Figure 5.5). At an N surplus as around the year 1900, the established model predicts the concentration of N to be around 2.0-2.2 mg N/L with present days N retention in groundwater and surface waters. However, extrapolation of this kind of statistical relationships outside their range of observations is always uncertain, and the outcome should be considered to involve a high degree of uncertainty.

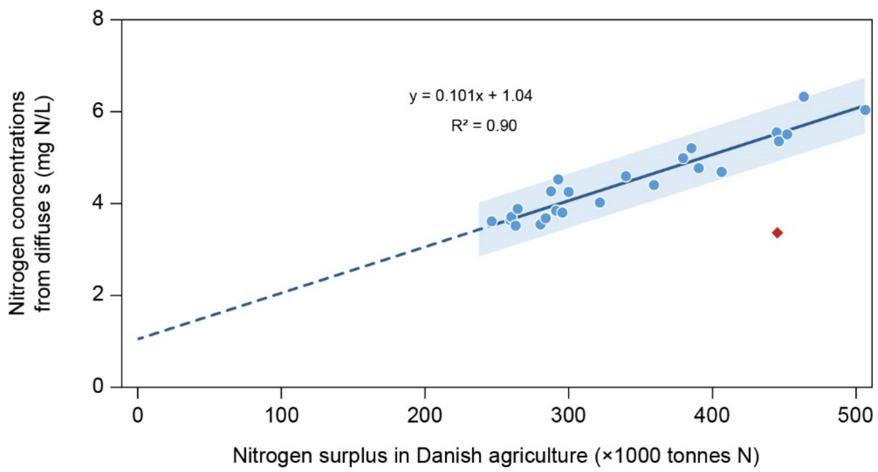


Figure 5.5. Relationship between N surplus in Danish agriculture and flow-weighted total N concentration from diffuse sources in Denmark during the two periods 1990-2001 and 2002-2014. The extremely dry year 1995/96 (red dot) has been omitted from the analysis, as there was nearly no percolation of water through the root zone. The 95% confidence interval for the model predictions is shown in grey.

5.5 Historical data from rivers in UK

The only long-term time-series of river nitrate concentration data that dates back to the period before, around and after the year 1900 is from the River Thames upstream London (Howden et al., 2010). The observed nitrate-N concentration is lowest in the beginning of the time series around 1860 and has increased a little around the year 1900 (Figure 5.6). The nitrate-N concentration in the river Thames around the year 1900 was approximately 2 mg N/L. The geology in the River Thames catchment is chalk, and therefore the N retention in groundwater might be low as in similar Danish catchment with chalk aquifers. The nitrate-N concentration in the River Thames is, therefore, probably more realistic for similar hydrogeological conditions in Denmark, but too high in general for other Danish hydrogeological settings and rivers at that time – maybe by a factor of two. The trends in the concentrations of nitrate-N in the river Thames also generally seems to follow the development in the use of N within the catchment, especially during the latest period 1970 and onward and to a much lesser degree during the period 1940-1960 (Figure 5.5). The main increases in concentrations of nitrate-N in the river Thames were during the 2nd World War, where grassland was converted into arable land, and again during the 1970s, where land drainage support schemes together with the use of N fertilizer increased dramatically (Figure 5.6). The increases in nitrate concentrations in the River Thames are, therefore, not only driven by a higher N surplus to agricultural land, but also by changes in cropping systems and drainage of the landscape (Howden et al., 2010).

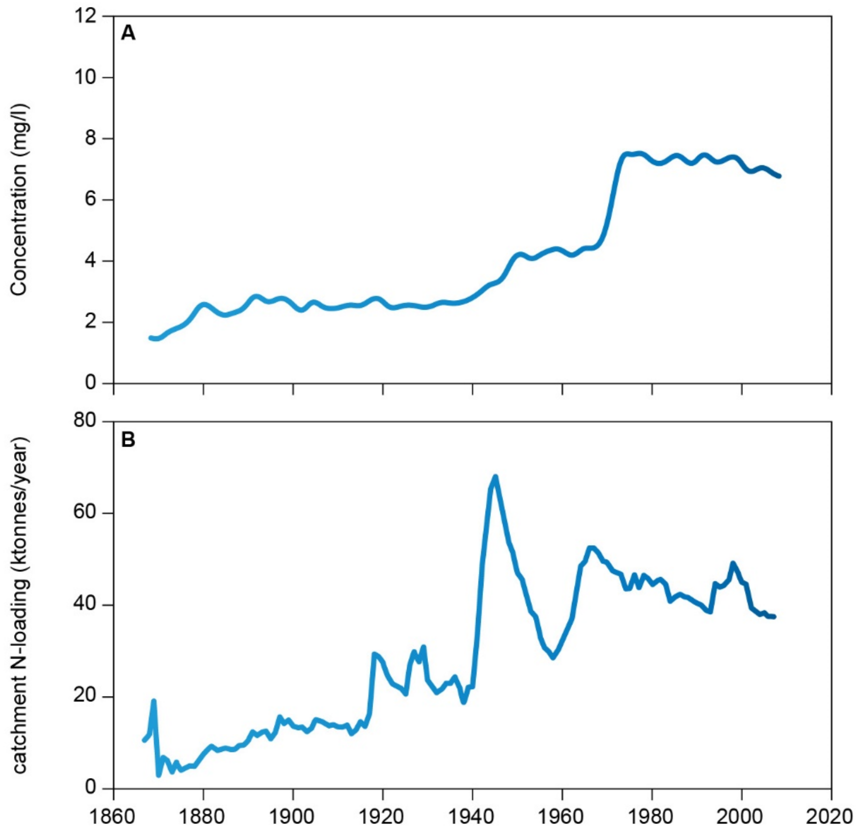


Figure 5.6: Annual net input, including mineralization from ploughing of N within the catchment the mean annual concentration of nitrate-N in the River Thames during the period 1860-2010 (from Howden et al., 2010).

5.6 Review of historical nitrogen data from the international literature

Data on measurements of N concentrations in rivers are very sparse around the year 1900, and only a few European rivers, such as the Thames and the Seine, which were highly polluted by untreated domestic sewage, were monitored (Meybeck and Helmer, 1989). However, these early monitoring programs were restricted to only a few variables, such as dissolved oxygen, pH, and faecal coliforms. Given the sparse measurements of N in rivers, other methods, such as expert judgement, recent measurements of unpolluted rivers or calculated pristine N concentrations, have been used to assess the reference condition.

Recently, Hirt et al. (2014) conducted a comprehensive literature review based on references collected by Topcu et al. (2011) and a literature survey. Data were collected from catchments in the temperate climate zone, especially for rivers affecting the German Bight, supplemented by data from unpolluted rivers in northern Europe and other temperate areas. The literature review showed that total N (TN) concentrations ranged from 0.210-1.316 mg N L^{-1} (mean 0.447 mg N L^{-1}) in lowland rivers with low urban influences, and that nitrate-N ranged from 0.161-2.821 mg nitrate-N L^{-1} (mean 0.643 mg nitrate-N L^{-1}). Hirt et al. (2014) simulated a scenario representing the year 1880 with the model MONERIS (Venohr et al., 2011) and found that the resulting mean concentration of TN and nitrate-N was 0.699 mg N L^{-1} and 0.034 mg N L^{-1}, respectively, in the German Baltic Sea catchments.

In the report 'The data of geochemistry' by Clarke (1916), a tremendous amount of chemical data for the lake and river waters of the world is compiled. The sampling in US rivers and streams was typically conducted as part of a systematic sampling program of the Water Resources Division of the United States Geological Survey (USGS) and has been published in a series of Water-Supply Papers (number 236, 237, 239, 339, 363). These analyses were made in the laboratories of USGS. The aim of these programs was to reflect the average composition of the river for an entire year, as a general rule the samples of river water were taken daily and combined into composite samples of seven to ten samples. The samples were mailed to a designated laboratory as soon as possible after collection, where the phenolsulphonic acid method was used for determination of nitrate. In the Water-Supply Papers, the concentrations are reported as parts per million of the nitrate radicle.

With respect to European rivers, analysis of nitrate-N concentrations in the Rhine, Elbe, and Danube and their tributaries published by Hanaman (1894, 1898), Egger (1887, 1908, 1909), and Schwager (1893) are included in Clarke (1916) together with analysis of Rhone and its tributaries, Thames, Seinen, Fyris, and Ljusnan (Lossier, 1878; Graham et al., 1850; Hofman-Bang, 1905). Hanaman (1894, 1898) used the method of measuring nitric acid in the water samples, which has been converted to nitrate in Clarke (1916). Streams and rivers in urban influenced areas showed high levels of nitrate-N already during the final decades of the nineteenth century. Values reported from cities such as Dayton (Miami river) and Bercy (Seinen) ranged from 1.9 to 2.6 mg nitrate-N L^{-1}, probably due to untreated sewage led directly into the rivers. Thus, the water samples stated as collected directly in cities were not included in Table 5.1.

Land cover of European countries around the year 1900 are withdrawn from the Historic Land Dynamics Assessment (HILDA), which is a reconstruction of historic land cover based on data from multiple sources such as remote sensing products, national inventories, aerial photographs, land cover statistics, old encyclopaedias and historic land cover maps.(Fuchs et al., 2013, Fuchs et al., 2015a, Fuchs et al., 2015b). In the table, land cover is given per country for the European rivers. Land cover in U. S. around 1900 is derived from the U. S. Bureau of the Census (1952). In this report, the U. S. is divided into the following regions: Northeast, North central, South, and West. Furthermore, the calculations presented in Table 5.1 are based on the assumption that cropland accounts for 38 % of land in farms in each region based on the relationship between cropland and land in farms for total land area in 1900. Therefore, land cover in 1950 is also reported as the land cover percentage for each state reported in Wooten (1954). Around 1900, the irrigated and drained land is estimated to account for 1 and 3 % of total land area in U. S, respectively (U. S. Bureau of the Census, 1952).

According to Clarke (1916), the median concentration of the analysed European streams and rivers around 1848-1905 was 0.18 mg nitrate-N L^{-1}, ranging from 0.03 to 2.90 mg nitrate-N L^{-1} (Table 5.2). The median of the riverine nitrate-N concentration in the United States around 1905-1912 was 0.29 mg nitrate-N L^{-1} and ranged from 0.06 to 0.66 mg nitrate-N L^{-1}.

Table 5.2. Median, min, and max of NO3⁻-N concentrations, sample size (n) of locations from Clarke (1916) and cropland in percentage of total land in year 1900 and 1950.

Location	n	Median (min-max)	Cropland/total land in 1900	Cropland/total land in 1950
		NO_3^- -N (mg/L)	%	%
Rhinen+tributaries	4	0.47 (0.31-0.65)	45	
Elben+tributaries	4	0.11 (0.04-0.32)	45-50	
Danube+tributaries	7	0.06 (0.04-0.12)	25-45	
Rhone+tributaries	2	0.13 (0.12-0.14)	10-60	
Sweden	2	0.04 (0.03-0.05)	10	
Mississippi+tributaries	5	0.41 (0.16-0.64)	25	31-60
Tributaries to Ohio River	1	0.27	25	25
Tributaries to St. Lawrence	1	0.43	25	31
Red river	1	0.09	25	13
Rivers of California	5	0.22 (0.13-0.41)	6	10
Columbia+tributaries	3	0.14 (0.13-0.25)	6	7-16
Rivers in Oregon and Washington	3	0.07 (0.06-0.15)	6	7-16
Missisouri river+tributaries	3	0.52 (0.41-0.66)	25	31-46

5.7 Methodological problems with handling of water samples

The historical measurements must be interpreted with caution, as it has yet to be outlined how the samples were preserved and stabilized for transport and storage prior to laboratory analysis. Within the period from sampling up to analysis, the composition of the water sample can be significantly altered due to the chemical, physical, and biological reactions it undergoes. For instance, the concentration of nitrate-N in water samples can both increase or decrease due to the variety of processes the sample can undergo, such as nitrification (+), cell lysis (+), denitrification (-), microbial uptake (-), absorption and adsorption (-) onto bottle surfaces (Heron, 1962; Latterell et al., 1974).

The effect of preservation techniques on TN, nitrate-N, ammonium-N, and nitrous oxide-N concentrations in stream and lake water samples has been investigated by the Danish research institute 'Danmarks Miljøundersøgelser (DMU)'. The water samples were stored at 4 °C and analysed after 0, 1, 2, and 7 days, respectively. The concentration of TN and nitrate-N was relatively stable, although the concentration of nitrate-N increased after 7 days, which presumably was due to a systematic error in the analysis procedure.

In Kotlash and Chessman (1998), the effect of no preservation and different preservation techniques on nitrate-N in water samples was investigated. In the case of unpreserved samples stored for 6 days, the study showed very little difference in nitrate-N concentrations compared with the initial concentration at four of the six sites. At one site, the nitrate-N concentration was 50 % lower after 6 days, which was suggested to be related to a very low initial concentration (0.04 mg nitrate-N L^{-1}). In contrast to this, the nitrate-N concentration rose sharply after 6 days at the last site, due to nitrification.

A more recent study by Moore and Locke (2013) suggested that surface water samples stored at 23°C (24 h), 4°C (7 days), and −20°C (7 days), respectively, resulted in 6.3 % ± 4.1 %, 38 % ± 4.7 %, and 13± 6.7 % of the resulting nitrate-N concentrations being significantly different from controls, which were analysed immediately upon collection. The difference both encompassed mean nitrate-N concentrations being lower and higher than control concentrations.

These studies highlight the fact that the shorter the time between collection of a sample and the analysis, the more reliable the results will be. However, this is not always possible, hence, preservation of the samples is necessary. It is difficult to outline how the preservation technique, if any, might have affected the historical samples. The measurements conducted by USGS were analysed ten days to eight weeks after the samples had been collected, which is sufficient time for marked changes in the nitrate-N concentration of the water sample to be introduced. Thus, the reported historical N concentrations might merely reflect the concentration of nitrate-N in the water sample at the time of analysis rather than the nitrate-N concentration of the stream water when the water sample was collected. However, Clarke (1916) validated the water analysis by conducting anion-cation balances to distinguish good from bad analysis results. In an accurate analysis, the anions and cations reported in the analyses should be equivalent, as the major ions usually represent most of the dissolved ions in water. This validation, however, requires that all major ion concentrations be measured.

5.8 Conclusions

Historical data on N concentrations in Danish rivers are sparse. The N concentration in 6 major Danish rivers sampled during the period 1889-1892 showed very low nitrate-N concentrations, but relatively high concentrations of organic N. The spatial pattern in the observed nitrate-N concentration was similar to present day patterns. The N concentrations in four main Danish rivers show an increase from the time the first sampling was initiated in the mid 1960s to the mid 1980s, after which the N concentration began to decrease as a consequence of the Danish Action Plans. In most cases, the N concentration in Danish rivers very nicely followed the development in the N surplus in Danish agriculture. However, two streams in a region of Denmark with chalk in the underground showed a clear delay in the N concentration response. The findings from Danish historical observations on N concentrations in streams and rivers were somewhat corroborated by a long time series of N concentration data from the River Thames in UK.

Empirical models were established for the relationships between flow-weighted TN concentrations and N surplus from NOVANA monitoring stations (coastal catchments) during two periods, 1990-1999 and 2000-2014. The established model for the recent period 2000-2014 predicts low TN concentrations in Danish streams when using as input the N surplus around the year 1900 (around 2 mg N/L). It is important to be aware that these model estimates of N concentrations assume current climatic conditions and retention of N in surface waters.

Utilising an established groundwater model between nitrate concentrations in oxic groundwater and total N use in Danish agriculture from 1957 – 1975 showed a concentration of nitrate-N around 2 mg N/L for the period around the year 1900, when the nitrate concentration in oxic groundwater was recalculated to stream water nitrate concentrations utilizing present day retention of nitrate in groundwater.

A review of N concentration data from streams and rivers from around the world in the international literature showed that measured nitrate concentrations around the year 1900 were very low in most rivers. However, many of the catchments from where sampling was done had a much lower proportion of agricultural land than was the case in Denmark around the year 1900. Moreover, the review also showed that care should be taken when interpreting these results, as storage of water samples in most cases happened for several days to weeks prior to analysis. Research on the effect of storing water samples on the analysis of nitrate concentrations in most cases shows that storage results in a systematic error in the actual nitrate concentration.

6 Modelling nitrogen retention around year 1900

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Abstract

Purpose: The purpose of this chapter is to analyse the difference in landscape nitrogen (N) retention between the current period and the period around the year 1900. This is done by analysing the major elements of nitrogen retention during both periods.

Materials and methods: Data on wetlands around the year 1900 has been digitized for the River Odense catchment at the Kratholm monitoring station (NOVANA No. 450003), River Skjern catchment at Ahlergaarde monitoring station (NOVANA No. 250082) and River Suseå at Næsby bro (NOVANA No. 570050) from the historical maps (højemålebordsblade) drawn as close to the year 1900 as possible. Two existing models – the SWAT model and the National Nitrogen Model 'NNM' - have been applied running different scenarios for the landscape and climate around the year 1900 using the present day landscape and climate as comparison.

Results and discussion: Reconstruction of the landscape back in time to around the year 1900 showed the importance of tile drainage and wetlands for N retention and transport in streams. Inserting the old wet riparian areas around the watercourses in the River Odense catchment into the model increased the N retention in surface waters from the present 28% to 37% around the year 1900. For the River Skjern catchment, the N retention around year 1900 as compared to present day increases N retention in surface waters from 41% to 58%. For the River Suseå catchment, the total surface water N-retention increased from 60% to 65%. Extending the length of watercourses and the number of ponds in the River Odense catchment back to around the year 1900 only marginally increased the N retention. The importance of tile drainage was analysed with the SWAT model, and a scenario with removal of the tile drainage that at present is installed at around 50% of the agricultural area in the River Odense catchment was conducted. The model predicted a 33% reduction in riverine N loadings as an output of this extreme scenario. Historical tile drainage records show that the amount of tile drainage on Funen probably was about 30% of the agricultural area around the year 1900. This would result in a 13% lower riverine N loading and, hence, an increase in groundwater N retention if the SWAT model results were extrapolated linearly to such a tile drainage level. A SWAT scenario comparing nitrate-N loads for River Odense at present and with a climate similar to around the year 1900 showed that runoff was 26% lower and nitrate-N loads 12% lower around the year 1900 than at present. The dryer climate around the year 1900 is, therefore, also expected to mean an increase in N retention in groundwater.

Conclusions: The national nitrogen model has shown that landscape N retention around the year 1900 was much higher than at present, but also that the N retention in surface waters showed a high regional variation in Danish catchments, depending on the amount of wet riparian areas such as wet meadows and wetlands surrounding the watercourses around the year 1900.

The N retention in surface waters has been shown to increase by 5-17%, totaling 37% to 65% around the year 1900 in the wetland scenarios conducted for the River Odense, River Skjern and River Suså catchments using the national N model. Moreover, a colder and dryer climate and less tile drainage around year 1900 than at present have also been shown, respectively, to lower N loads and increase N-retention in especially groundwater from present day's national average of 62%.

The differences found in N retention around the year 1900 is heavily dependent on local landscape changes from the year 1900 until the landscape and draining characteristics of today. A better spatial knowledge of this will require a modeling of more catchments in different regions of Denmark. Moreover, estimates of the expected increase in N retention in groundwater due to higher percolation to groundwater aquifers without tile drainages around the year 1900 would require use of 3D groundwater models together with best estimates of the depth to the redox zone in areas where drainage has changed.

6.1 Background and aims

The retention of nitrogen (N) in the landscape around the year 1900 was higher than today, as shown by Andersson and Arheimer (2003) in their analysis of a Swedish catchment. The retention of nitrogen was higher due to less intensive draining (less tile drainage) and a higher length and number of streams, lakes and wetlands in the landscape. Therefore, the aim of this chapter is to conduct a model analysis of the retention of N around the year 1900 in three Danish catchments (River Odense, River Skjern and River Suså) by recreating the historical landscape with less drainage and larger numbers of stream channels, lakes and wetlands.

6.2 Material and Methods

Data on streams, wetlands and lakes around year 1900 has been digitized for the catchment to the Kratholm monitoring station (NOVANA No. 450003) (Catchment area 486 km^2) in River Odense, the Ahlergaarde monitoring station (NOVANA No. 250082) (Catchment area 1052 km^2)in the River Skjern and River Suså at Næsby bro (NOVANA No. 570050) (catchment area 610 km^2). The catchments upstream these monitoring stations in river Odense, River Skjern and River Suså have been used as test catchments for the model. Data on streams, wetlands and lakes has been linked to the newly developed National Nitrogen Model 'NNM' (Højberg et al., 2015a+b). This model was developed on a MIKE-SHE hydrological model core (500x500m grid) linked to sub-models for nitrate leaching (NLES4), several surface water retention models and inputs of nitrogen from point sources and organic nitrogen. The nitrogen model was calibrated on a scale of ID15 catchments (ca. 15 km^2) (ca. 3100 polygons in Denmark) (Høbjerg et al., 2015a+b). The SWAT model, set up for the Odense fjord catchment (Thodsen et al., 2015; Trolle et al., 2015), was used for analyzing the effect of tile draining and the effect of changed climate. The lake nitrogen retention model, described in Windolf et al., (2011), was used for analysing the effect on nitrogen retention in larger lakes.

6.3 Nitrogen retention in wetlands around year 1900

When the location and area of present day wetlands are compared with wetlands marked on historical maps, it becomes clear that the estimated potential wetland areas were larger around the year 1900 than today (Figure 6.1). The

NNM was run using historical maps (høje målebordsblade) as a hind cast scenario, where the baseline is the present day wetlands existing on the AIS land use map as well as restored wetlands (Nielsen et al, 2000). The NNM uses modern day nitrogen leaching and discharges from point sources and other terrestrial sources (1990 – 2010) (Højbjerg et al., 2015a+b). The maps showing the estimated historical wetland areas in the river Odense, river Skjern and river Suså (Fig. 6.1) have been constructed by constructing a riparian buffer along all stream / rivers overlaid with the wetland areas digitized from the historical maps. The buffer around the watercourses in the River Odense, Skjern and Suså catchments is scaled according to stream channel width (stream typology), mimicking the river / stream valley width for different river / stream sizes.

Thus, in the scenario runs performed with the NNM model only wetlands located close to water courses are considered, as they are considered the main removers of nitrogen. Only wetlands located within the buffers (both sides of the watercourse) shown below are considered:

- 50m buffer (streams <2m width + stream of unknown width class)
- 150m (streams between 2m and 12m width)
- 300m (rivers >12m width).

The buffer is related to stream width because, in general, it has been shown that the river valley increases in width with increase in stream width (Sand-Jensen et al., 2006).

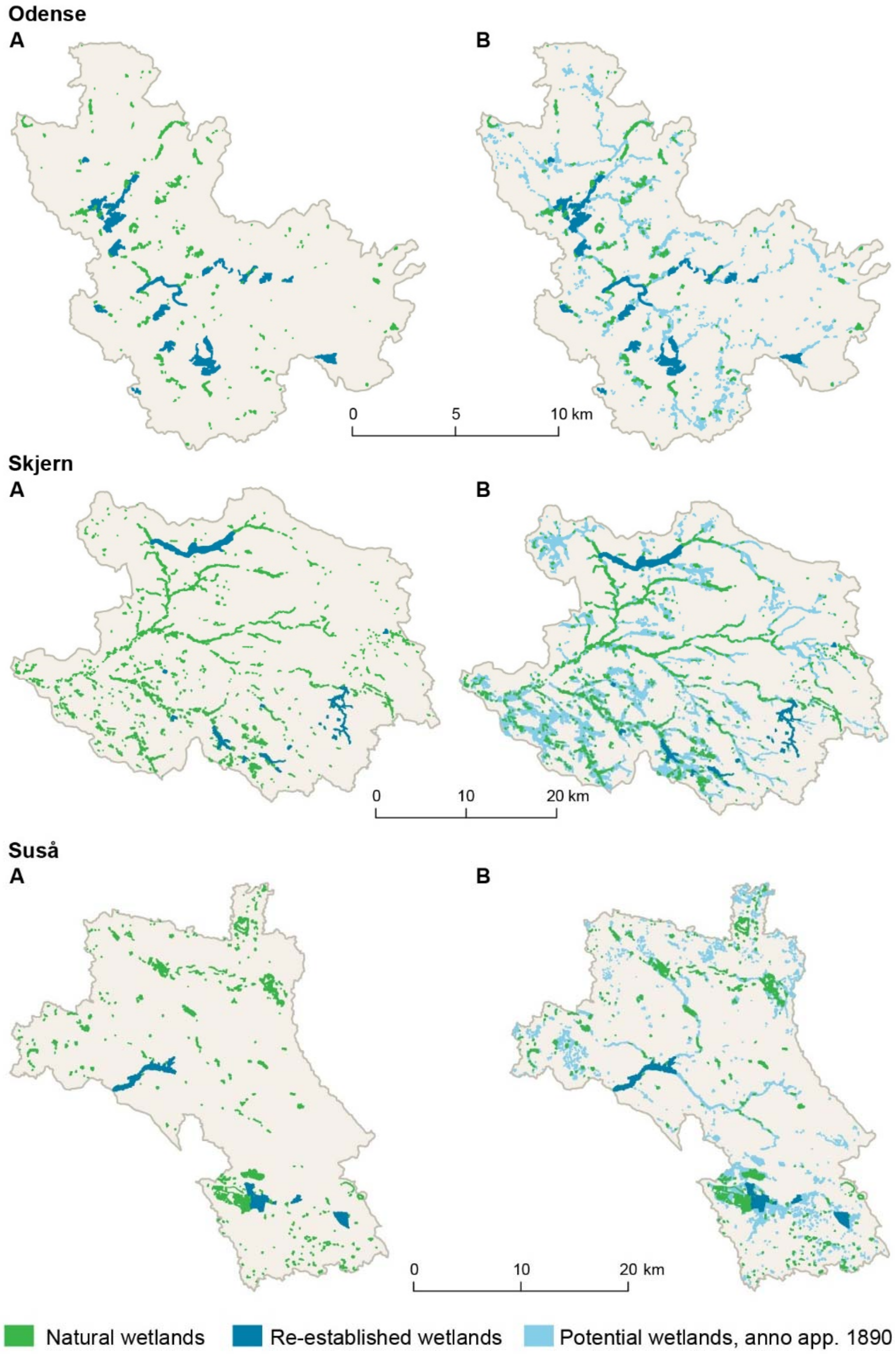
The model runs with NNM include a baseline for the current situation with restored and natural wetlands that is compared to a model scenario run with all wetlands mapped in the buffer along streams around the year 1900 recorded on the old maps (Figure 6.1).

Reestablishing all mapped wetlands around the year 1900 in the hind cast scenario is believed to produce a maximum estimate for N retention. In reality the N loading from local upstream areas towards the recreated wetland areas may not be high enough for the wetland to deliver the expected retention rates. Also another reason why the estimated wetland retention may be too high is because no cascade effects are included in the model.

The original NNM is run / setup with only recreated wetlands and not all wetland actually present in the landscape. In this chapter, we intend to compare the wetlands present in the AIS maps from the 1990s (Baseline) with wetlands present in the historical maps from around the 1890s (Scenario). Therefore, the AIS map results (Baseline) are normalized to yield the same results as the original NNM setup. The same normalization (difference in percent) was applied to the historical map scenario. In this way, the historical map scenario is comparable to the original NNMsetup results (Table 6.1).

The scenario effects of reconstructing wetlands in the river Odense are shown in Table 6.1. The wetland N-retention increases from a baseline of 150 ton N / yr when the existing wetlands are included in the model to 290 ton N / yr (93%) when the estimated and mapped historical wetlands are included in the scenario model run. The wetland area in the river Odense catchment around year 1900 was 185% higher than today (Table 6.1). The N-retention in surface water is shown to increase from 400 to 520 ton N / yr (30%). Thus, the model results show that reinstalling / constructing wetlands has a large effect on the N retention in surface waters (Table 6.1). For normalized values, the total surface water N-retention increases from 28% to 37%.

Figure 6.1: Baseline and scenarios run with the national nitrogen model: A: Present day conditions with present day wetlands in the three catchments. B: The wetland areas around the year 1900 within buffer zones along watercourses.



The wetland N-retention in river Skjern increases in the scenario from a baseline of 350 ton N / yr when the existing wetlands are included in the model to 840 tonnes N / yr. (140%) when the historical wetlands are included in the landscape. The wetland area in the river Skjern catchment around the year 1900 was 279% higher than today (Table 6.1).The N-retention in surface water was found to increase from 1150 to 1530 tonnes N / yr (33%). Thus, the model results show that reinstalling / constructing wetlands has a large effect on the N retention in surface waters (Table 6.1). For normalized values, the total surface water N-retention increases from 41% to 58%.

Table 6.1. Wetland areas and modelled nitrogen retention around the year 1900 and today with the national nitrogen model (NNM) in the three catchments.

Catchment	Scenario	Total wetland area Ha	N-retention wetlands Ton	Total N-retention surface water Ton (%)	Normalized Total N-retention surface water %
River Odense, Kratholm	original NNM	710	90	360 (28%)	-
	Baseline	910	150	400 (32%)	28
	Scenario	2.590	290	520 (41%)	37
River Skjern, Alergårde	original NNM	850	80	940 (41%)	-
	Baseline	2.920	350	1.150 (51%)	41
	Scenario	11.070	840	1.530 (68%)	58
River Suså, Næsby Bro	original NNM	570	50	820 (60%)	-
	Baseline	1.680	160	850 (62%)	60
	Scenario	4.070	310	920 (67%)	65

The wetland N-retention in river Suså increases in the scenario, from a baseline of 160 ton N / yr when the existing wetlands are included in the model to 310 tonnes N / yr. (94%) when the historical wetlands are included in the landscape. The wetland area in the river Suså catchment around the year 1900 was 142% higher than today (Table 6.1).The N-retention in surface water was found to increase from 850 to 920 tonnes N / yr (8%) (Table 6.1). For normalized values, the total surface water N-retention increases from 60% to 65%.

The change in wetland areas and the influence this landscape change was shown to have on modelled nitrogen retention in surface waters (5-17% increase) are believed to cover the range found around the year 1900 for landscapes in Western Jutland, Eastern Jutland and the Islands, except for the conditions on the extremely low-lying areas of Lolland / Falster and Bornholm. Landscapes in Southern Jutland that were under German territory from 1864 to 1918, the marches along the Wadden Sea and the low-lying landscapes in Northern Jutland might have had other drainage conditions around the year 1900 than is the case for the three catchments investigated in this chapter.

6.4 Nitrogen retention in watercourses around the year 1900

Small streams fall within the stream / river category width class <2m. Small streams are considered as small tributaries to larger streams that cross the ID15 catchment (about 3100 catchments on average 15 km^2 in size) boundaries that make up the NNM (Højberg et al., 2015a+b).

It is assumed that because tile drainage was less common around the year 1900 than at present (Hofmeister et al., 2004), there would have been more draining ditches at that time. In addition, the effort of piping small streams and ditches is believed to generally have taken place later than 1900, therefore, these streams would largely have been open around the year 1900. The NNM is run with a 20% increase in stream length, which corresponds to an increase in sinuosity to 1.2 from a straightened stream channels in all ID15 catchments, which, in turn, increases stream N-retention by 10% (Table 6.2). The total N-retention in surface waters is found only to be slightly affected, because N retention in smaller watercourses is of limited importance compared to wetlands and larger lakes.

Table 6.2. Model analysis of the effects of changing the length of watercourses in the river Odense for nitrogen retention.

Scenario	Watercourses (km)	N-retention in smaller streams Tonnes N/yr
Baseline: present day conditions	310	100
Scenario: smaller streams length extended with 20%	370	110

6.5 Nitrogen retention in small lakes around the year 1900

By comparing historical maps (målebordsblade) to the latest "Geo Danmark" GIS map showing lakes, it becomes clear that in many parts of the country a lot of smaller lakes / ponds have disappeared (Figure 6.2). Many ponds / small lakes in the agricultural landscape have been drained or filled with soil material over the past 200 years, to enable crop growing.

The disappearance of ponds / lakes has affected the N-retention in a decreasing direction. Estimates on how large a share of the ponds / small lakes that have disappeared since 1990 are given e.g by. Lauge Madsen, 2004 (in Hofmeister (Ed.), 2004), who without providing any reference states that 2 / 3 of smaller lakes have disappeared.

Most ponds / small lakes are considered of natural origin, but many ponds / lakes have both disappeared and appeared for different reasons during historical times.

Lakes have appeared in the landscape because of several factors:

- Peat excavation in peat bogs (– 1950)
- Marl pits (1800 – 1960)
- Sand and gravel pits (some constructed along major construction works)
- Mill ponds (few hydropower lakes) (1100 – 1960)
- Garden ponds (– present)
- Golf courses
- Delay / sedimentation ponds along major roads and urban areas (1980–present)
- Moats (1000 – 1900)
- Freshwater fish farms (1895 –)
- Horseshoe lakes created from straightening rivers (1800 – 1970).

Many small lakes / ponds have also disappeared because of factors such as:

- Filling
- Draining
- Removal of dams and weirs (since 1980 often to create free fish passage through rivers)
- Over growing.

A model scenario was conducted with the national nitrogen model, in which the scenario included a doubling of the area of smaller lakes (Table 6.3). We chose to double rather than triple (Lauge Madsen suggested that 2 / 3 of all small lakes had disappeared) the area, as it is believed that it is the smallest lakes that have disappeared before 1900. However, doubling the area of small lakes only proved to influence on the resulting nitrogen retention in the river Odense catchment to a limited degree (Table 6.3).

Figure 6.2. Top: Map of agricultural landscape on south Tåsinge. Lakes/ponds present in the newest Geo Danmark GIS map are shown in read, one is circled. Many more ponds are found in the old map. Historical map, Høje målebordblade (around 1900). Bottom: The same landscape as above July 10th 2014 (Google Earth). Red areas are the same ponds/lakes as above.

Table 6.3. Scenario results from the NNM with doubled area of smaller lakes in the river Odensecatchment.

Scenario	**Small-lake area**	**N-retention in smaller lakes**
	Ha	**Tonnes N/yr**
Baseline: Present day conditions	290	14
Scenario: Doubling of area with smaller lakes and baseline retention	580	21

6.6 Importance of changes in runoff for nitrogen retention in larger lakes

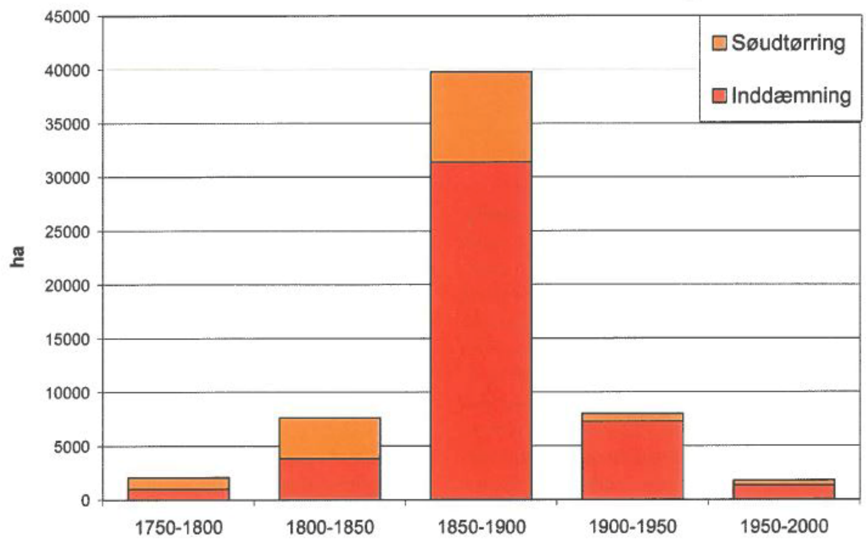
N-retention in large lakes around the year 1900 is estimated using the "lake retention" module in the DK QNP model complex, used for calculating the annual load of water, nitrogen and phosphorous to Danish coastal waters (Windolf, 2011). The model estimates an annual N-retention percentage based on lake water residence time and average lake depth. The model is an updated version of the model presented in Jensen et al., (1994). The model is based on lake N-mass balances for a high load period, (1989-1992).

The estimation of an average lake N-retention for the time around the year 1900 is done by using the model with an annual runoff similar to the runoff around the year 1900. The period around the year 1900 was dryer than at present (see chapter 1). The runoff year 2009 was chosen among the years 1990 – 2013 to represent the period 1895-1905 the best. The year 2009 was, then, compared to the average of the period 1990 – 2013 for 591 lakes existing for the entire modelling period (Table 6.4).

Table 6.4. Average lake N-retention at present and at the year 1900.

Period	N-retention %	% change
1990 – 2013	39.6	-
1895 – 1905 (represented by 2009)	42.1	6.3

Figure 6.3. Danish lake draining and land reclaiming (sea) between 1750 and 2000 (Hofmeister (Ed.), 2004)



Some large lakes have been drained during the past 200 years, but the majority of these lakes were drained before the year 1900 (Fig. 6.3) (Hofmeister et al., 2004).

Over time, lakes have also been re-established, and several larger lakes have been reestablished during the period 1998 to 2015. Almost 2000 ha of new lakes have been included in the annual calculation of lake N-retention in the national N-loading model (DK-QNP), which equals an area increase of 4.6% and an N-retention increase of 3.6%. Most of these new lakes have been constructed as part of the wetland restoration program. Only a few small-scale hydropower lakes have disappeared (or no longer receives water from major inflows) in recent years.

6.7 Importance of point sources for nitrogen retention

Town sewers were not yet implemented in Denmark around the year 1900 (see chapter 4). Day and night waste from towns and cities was brought to local dump sites outside the cities. However, smaller amounts of sewage must have been dumped into streams and marine areas from cities. Moreover, there were many animals, such as horses and pigs, in towns and cities, and part of their manure would also have been flushed from streets to streams and marine waters during rain events. Generally, point source emissions around the year 1900 can be considered to be small in many inland catchments in Denmark.

A model run with the NNM without point source inputs as an end-point was conducted (Table 6.5). It appears to have little effect on the total N-retention (Table 6.5).

Table 6.5. A scenario with the importance of point sources for nitrogen retention in surface waters in the River Odense catchment.

Scenario	Emission from point sources Tonnes N/yr.	Total N-retention in surface waters Tonnes N/yr.
Baseline: Present day situation with point source emissions	24	340 (27%)
Scenario: No point sources	0	330 (27%)

6.8 Importance of tile drainage for nitrogen losses to surface waters

The amount of tile drained agricultural land has increased since 1900. Hofmeister et al. (2004) estimated that in 1881 about 20% of the agricultural area was tile drained, primarily on loamy soils in the eastern part of the country. The tile drained area in 1900 is estimated to 30%. The introduction of drainpipes means that a fraction of the water presently flowing to the river through drainpipes would around the year 1900 have percolated through the soil into the groundwater. Groundwater percolation generally involve major N-retention, while drainpipes bypass the reduced environment in groundwater, where N retention takes place.

The SWAT model has been setup for the Odense fjord catchment, including a detailed description of the agricultural practice and simulating the effect of tile draining (Arnold and Srinivasan, 1998; Thodsen et al., 2015; Trolle et al., 2015). In the model tile drains are present in about 50% of the agricultural area in the model setup. SWAT was run with about 50% tile drainage as a baseline and without tile drainage as a scenario. The total nitrogen load through the river decreased by 33% when comparing the baseline and the scenario model runs. Assuming that, the rate of removing the tile drainage in SWAT shows a linear response in river N-load reduction, the N-load reduction at a level of 30% tile drainage in the catchment, will amount to a 13% decrease in N-load. In chapter 2, a different reference (Olesen, 2009) was used in estimating the percentage tile drained agricultural area around the year 1900, yielding an estimate of 22%. Assuming 22% of the agricultural area being tile drained, the N-load reduction would amount to 18%.

The finding from the model runs with the SWAT model that including less tile drainage than today reduces the N load is a strong indication for a higher N retention in groundwater, which is believed to have been higher around the year 1900 than the national average of 62% today, as calculated by the NNM model (Højbjerg et al., 2015a+b). However, it is impossible to predict the average N retention in groundwater around the year 1900 without utilising a 3D groundwater model with knowledge about the depth to the redox zone.

6.9 Effect of changed climate

The SWAT model was used for running a climate scenario and comparing the runoff and nitrate-N loads in the River Odense, Kratholm station for the period around the year 1900 with the present period. Average monthly precipitation and temperature values were calculated for the periods 1890 to 1910 (representing the period around 1900) and 1995 – 2015 (representing the present period). SWAT ran on the same modern day (2005) agricultural management data for both time periods in order to present the isolated climate change effect (Thodsen et al., 2015). Agricultural management includes crop rotations, fertilizer (chemical and manure) inputs and field operation dates (plowing / sowing / fertilizer applications / harvesting), therefore the N-leaching is different for the two periods, as it depends on climate. Results should be seen as a scenario: "What would the runoff and NO_3 river load be comparing present day climate with the climate around the year 1900?"

For precipitation, the relative change, and for temperature, the absolute monthly change between the two periods, were calculated as the mean change of the Nordby (Fanø) and the Copenhagen climate stations, as no stations with data from the 1890-1910 period exist for a precipitation station closer by the catchment (Table 6.6) (See chapter 2).

Table 6.6. Average change in mean monthly precipitation and temperature, for the Nordby and Copenhagen climate stations from the present period (1995-2015) to the year 1900 period (1890-1910)

Month	Change Precipitation %	Change Temperature °C
January	-28	-1.2
February	-27	-1.5
March	7	-1.3
April	23	-2.2
May	-13	-1.2
June	-12	-0.5
July	-13	-1.5
August	6	-2.1
September	-14	-1.4
October	-15	-1.0
November	-28	-1.4
December	-28	-1.0
Whole period	-13	-1,4

The climate scenario run used the "delta change" approach and utilized the mean monthly changes in precipitation and temperature (Table 6.6) through the inbuilt climate scenario facility in SWAT (Graham et al., 2007a; Graham et al., 2007b; Thodsen, 2007; Trolle et al., 2015). SWAT was run for the 9-year period, 1997-2005, and a 2-year warmup period.

The modelled effect of the altered climate between the present and the year 1900 periods yielded a mean annual change in runoff of -26% (the flow was 26% lower during the year 1900 period, than at present). The NO_3 river load was modelled to be 12% lower during the year 1900 period than at present. In the SWAT setup, there was 10 sub basins of varying size upstream the Kratholm station, the range in runoff change was -25% to -30%, the range in NO_3 load change was -6% to -17%.

As the runoff was modelled to have increased more between the year 1900 and the present period than the NO_3 load the $NO_{3,}$ concentration was found to be 18% higher during the year 1900 scenario period than at present, assuming present day agricultural management in both periods. This is in line with the findings of Trolle et al. (2015). The reduction in NO3 is despite a 2% lower modelled nitrogen plant uptake, caused by higher water and temperature stresses during the year 1900 period than during the present period.

6.10 Meadow irrigation

Irrigation of near river meadows has been practiced in Denmark since at least the early 1600s. But the largest expansion of the meadow irrigation systems was performed in the second half of the 19^{th} century (1850-1900) and was considered finished around the year 1900 (Rasmussen, 1964). However, meadow irrigation as found around the year 1900 is essentially not in use today.

Hedeselskabet constructed 410 km of irrigation canals for 7400 ha of irrigated land. A total of approximatly 600 km of irrigation canals were constructed in Denmark. Assuming an average irrigation area of 18 ha/km (channel) (same as Hedeselskabet), around 10.000 ha of irrigated meadows existed around the year 1900 (Rasmussen, 1964).

Meadow irrigation was mainly used in western part of Denmark with high natural summer river water discharge and in areas with relatively broad and relatively steep river valleys, wherein such valleys edge canals could be constructed with sufficient slope conveying the water. The preferred areas were the sandy glacial outwash plains in Jutland (Fig 5 from Rasmussen (1964)). The amount of water used in meadow irrigation could be considerable. Rasmussen (1964) reports that the "Store Skjern Å canal" had a capacity of conveying 4 m^3/s (width 11m depth 1m). The amount of water needed to irrigate one hectare was reported to be very uncertain, but an estimation was done anyway by Rasmussen (1964).

1 cubic feet (0.0283 m3)/s will irrigate 2-3 ha (2.5 ha)

5 months of growing season then gives app. 150.000 m^3/ha/year.

For 10.000 ha, this amounts to app. 110 m^3/s for a five month growing season. The irrigation is, in most cases, not continuous through 5 months, but adds up to five months in a year.

This is reported to be 100 times more than needed with modern (1964) equipment (150 mm) to ensure the water supply. But it should be remembered that there were more purposes than applying water, e.g. applying nutrients, which would mean irrigating in periods when soil did not need extra moisture. Besides this, some water was used twice. However, applying nutrients is reported to have been difficult and not of big significance, as the river water is reported to have been generally poor in nutrient content.

As an example: if irrigation water had an average N concentration of 1 mg/l and the above estimated water amounts are applied, the nitrogen application would be app. 150 kg/ha, which is a very significant amount of N input. The irrigated wet meadows with a permanent grass cover were ideal sites for uptake and retention of nitrogen through denitrification alike the modern recreated floodplain meadows receiving tile drainage and/or river water. Assuming a 50% nitrogen trapping efficiency will yield a nitrogen retention of 750 ton/ yr. A trapping efficiency of 75% yields a nitrogen retention of 1125 ton/ yr. The trapping efficiency depends on (among other things), the fraction of irrigation water returned to the river, the irrigation water nitrogen concentration, the crop nitrogen uptake. Assuming 25% of all meadow irrigation area was located upstream the Allergårde hydrometric station on the Skjern river (estimated from fig 6.4), this would amount to 188 ton/yr nitrogen retention in this area (50% trapping efficiency). Relating this amount to the total surface water nitrogen retention for the year 1900 scenario (Table 6.1) of 1530 ton/yr it would been increased to 1718 ton/yr and make up 12% of the total surface water nitrogen retention. Meadow irrigation could therefore have produced significant amounts of nitrogen retention in the areas where it was installed. However, meadow irrigation as around the year 1900 is essentially not in use today. Therefore, the nitrogen retention associated with it has also disappeared.

Figure 6.4. Meadow irrigation canals existing abt. 1910. The map only includes the regions relevant to the problem discussed. It is principally attached to streams in West- and South-West Jutland and mainly to outwash plains along streams of the old moraine irrigation of meadows was found to a lesser extent. Characteristic of the irrigation canals along the streams on outwash plains (Karup Å, Skjern Å etc.) is that they are mainly found along the upper and middle reaches of the streams, which is reasonable because of the need for a sufficient gradient. However, canals are also seen at the mouths of some streams (South-West Jutland) in most cases it was necessary to establish an artificial link between streams and canals by way of pumps.

6.11 Conclusions

The national nitrogen model has shown that landscape N retention around the year 1900 most likely varied depending on the region. The N retention in wetlands more than doubles in the River Skjern catchment (Ahlergaarde monitoring station). The three sites yield increases in wetland N-retention of 93%, 94% and 140%. Thus, the total N retention in surface waters, regarding only changes in wetland areas around the year 1900, was 37% in the River Odense compared to 28% in the present period, 58% in the River Skjern catchment compared to 41% in the present period, and 65% in River Suseå as compared to 60% in the present day (baseline) situation. The increase is solely due to the existence of larger wet riparian areas, such as wet meadows and wetlands surrounding the watercourses in the River Odense, River Skjern and River Suseå catchments around the year 1900. The calculated N retention around the year 1900 is expected to be a maximum ("best case") estimate. Moreover, around 30% less tile drainage of fields around the year 1900, as compared to today, has been shown in the SWAT model applied on the River Odense catchment to decrease riverine N loadings by approx. 13% as compared to present day N loadings. Lastly, a SWAT scenario comparing present day climate with the climate around the year 1900 showed a lower runoff (-26%) and nitrate-N transport (-12%) around the year 1900 than at present. The two latter findings would give rise to a higher N retention in groundwater around the year 1900 than the current national average of 62%. The average N retention in groundwater in Denmark was higher around the year 1900 than at present conditions due to less tile drainage and a dryer climate around year 1900 than today.

The differences found in N retention around the year 1900 are heavily dependent on local landscape changes from around the year 1900 until the landscape of today and current draining characteristics. A better spatial knowledge of this would require a modeling of more catchments in different regions of Denmark. Moreover, the expected increase in N retention in groundwater due to higher percolation to groundwater aquifers without tile drainage around the year 1900 would require use of 3D groundwater models together with best estimates of the depth to the redox zone in regions where drainage conditions has changed between the year 1900 and present days.

7 Synthesis

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7.1 Background and aim

The overall aim of this report is to investigate the factors, that could have influenced the annual mean nitrogen (N) concentrations from source to sea around the year 1900 in order to estimate the N concentration level in stream runoff to sea at that time in Denmark. Fulfilling such an aim requires, that a range of parameters, that are known to influence the N cycle such as climate, hydrology, land use, agricultural practices, drainage, landscape, etc. , are best possible described for the period around the year 1900. Very few historical measurements of N-concentrations in streams and rivers are available from that time in Denmark as well as in other countries. Therefore, other methods are required to obtain estimates of the N-concentrations.

A number of different methods, that could assist in estimating and / or finding indications of the N concentrations around the year 1900 in soil water, groundwater and surface waters have been discussed in the previous chapters – like agricultural statistics, trends in climate and hydrology, review of historical measurements found in the international and Danish literature or use of model estimates. This chapter synthesizes these indicators (or the majority of them) to provide an estimate of what could have been the range of N-concentration in Danish soil water, groundwater and surface waters around the year 1900.

Aggregated results from the chapters in this report together show, that agricultural activities were the main anthropogenic source of nitrogen load to the coastal areas around the year 1900. The synthesis, therefore, focuses on this main source.

7.2 Danish agriculture and landscape around 1900

Danish agriculture around the year 1900 was very different from today, much more extensive, but it occupied a higher percentage of the total land area compared to the present situation. No use of chemical fertilizers, low yield, no modern agricultural machines, mostly small scale farms, less drainage with tiles (only 22% of the agricultural area), and different composition of the livestock with more mixed farming across the country are the main differences from the current situation. Some of the agricultural practices around the year 1900 have on the other hand led to relatively high N loss rates – like application of manure in the autumn or the use of bare fallow. Moreover, the increase of land under cultivation and the increase in tile drained area, which occurred in the decades preceding 1900, initiated a decline in the soil organic N pool. This is considered to have contributed to an additional risk of N leaching loss from the root zone around the year 1900.

The extensive farming is also the reason, why the past landscape looked very different from today's, with a much higher percentage of grasslands, non-drained land and wetlands and watercourses in a much more natural form meandering in the river valleys. Another important difference is the climate –

with lower temperatures, lower precipitation and lower stream runoff around the year 1900 as compared with the present situation.

7.3 Nitrogen pathway from root zone to coastal areas

The N concentration in water undergoes a substantial reduction from the root zone to the coast through denitrification in groundwater and surface waters (wetlands and lakes / rivers). This is illustrated in Figure 7.1, where the N concentration in the root zone is routed to the coast.

7.3.1 Root zone concentration

Concentrations of N in leachates from agricultural soils around the year 1900 is estimated to typically have ranged between 5 and 15 mg N / l depending on crop and management. Concentrations have been higher in leachates from land subject to prolonged grazing periods, land with manure application in the autumn and land left bare after ploughing. From long-term experiments with current organic farming systems that have similar inputs as farming systems around the year 1900 it is estimated, that the concentration of N in root zone leachates averaged 12 mg N / l from agricultural land with grassland in rotation with arable crops. This also spans lower N leaching from permanent grassland areas and substantially higher leaching from land in bare fallow, which constituted 8% of the total land area around the year 1900.

The weighted average root zone N-concentration for all of Denmark is found by using a root zone concentration of 12 mg N / l for agricultural areas. For nature areas, a concentration of 1 mg N / l is used.

It is anticipated, that 75% of Danish land area was used for agriculture and 25% was mature nature areas. In this respect it is assumed, that infrastructure and built-up areas only occupied a very small part of the total area. For urban areas – see below.

The combination of these estimates results in a weighted average root zone concentration of 9 mg N / l for the entire Danish area around the year 1900.

7.3.2 Denitrification

The described landscape around the year 1900 is very important for the fate of the nitrogen leaving the root zone compared with the present situation. Nitrogen is removed through the process of denitrification (also called N retention) in groundwater and, as the drained part of the agricultural area was significantly lower around the year1900 than today, a higher proportion of the percolating water from fields drained to the groundwater aquifers and not directly to surface waters. Moreover, a colder and drier climate around 1900 increased the retention, particularly in groundwater. This means that the overall denitrification in groundwater was higher than the present estimate of 62% (Højbjerg et al., 2015a+b), but is has not been possible to determine a figure or a range for the denitrification in groundwater around the year 1900. Therefore the present estimate of 62% has been used for the further estimates around the year 1900 bearing in mind, that this definitely is a minimum estimate.

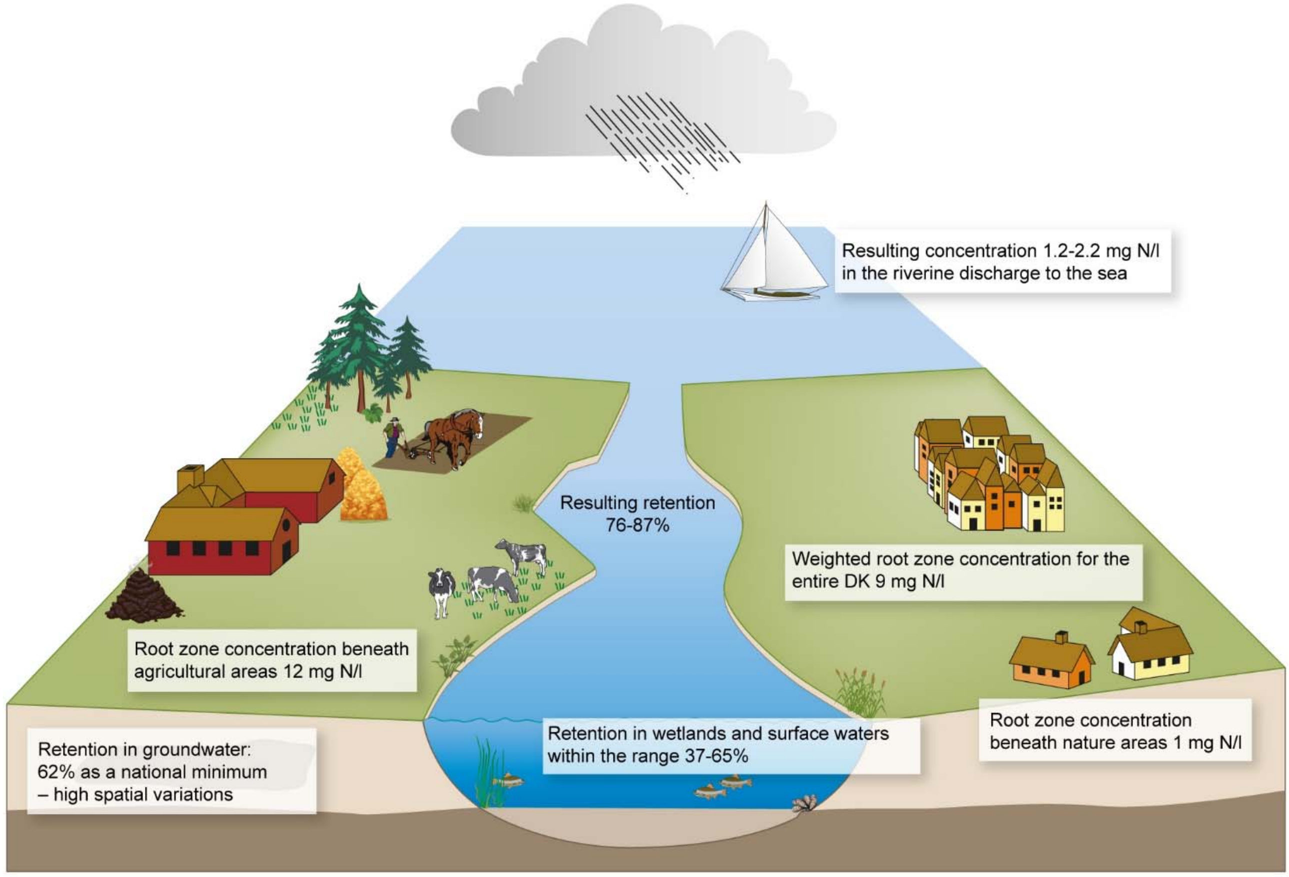

Figure 7.1. Diffuse N concentration from root zone to coastal areas on a national level.

The wetland areas (wet meadows, wetlands, etc.) surrounding streams and lakes were significantly larger around the year 1900 than today, as evidenced from old maps, implying that N retention in riparian and surface areas was higher than at present. The retention in surface water areas around the year 1900 was calculated for three different catchments and was found to range from 37 to 65%. As the three catchments represent different parts of the Danish territory in the year 1900, this range is assumed to be a fair estimate of the surface water retention for most of the Danish land area. The resulting retention around the year 1900 (groundwater + surface water areas) can be estimated to between 76% and 87% compared to the present 65-70% according to the findings in chapter 6.

7.3.3 Point sources and atmospheric deposition

It is assumed, that the atmospheric deposition on land (estimated in general to 4 kg N / ha) around the year 1900 is incorporated in the estimated root zone concentration both on agricultural and nature land.

Another more direct input of N was point sources – and locally they may have had a significant impact. It is however assumed, that on the national scale, point sources around the year 1900 was negligible as described in chapter 4.

7.4 Resulting N concentration to coastal areas around 1900

Using the approaches and assumptions described above, it can be estimated, that the resulting N concentration in the water discharged into coastal areas around the year 1900 was in the range of 1-2 mg N / l (the calculated maximum result from Fig. 7.1 of 1.2-2.2 mg N / l is rounded to the nearest integer – both for taking into account the uncertainty and that the resulting retention is an absolute minimum). It should be noted, that this is a national estimate, and there may have been substantial regional and local variation.

7.5 Supplementing estimates of N concentration in the surface water around 1900

As previously described, a range of other methods / data has been included in the analysis. The main results from these analyses are summarized in Table 7.1 and all refer to surface water concentrations. It is very important to note, that the concentrations given in Table 7.1 are not directly comparable as they represent different forms of nitrogen – total-N, NO_3-N, organic N – and are measured / modelled at different stages from the root zone to the coastal areas; thus they do not all represent the N concentration in the discharge to the coastal areas.

Table 7.1. Estimates and measurements of nitrogen concentrations around 1900.

	N form	Concentration mg N/l	Comments
	Measurements around r 1900		
Six Danish rivers Chapter 5	NO_3-N + Org. N	0.7 – 2.7	Dominated by organic N. Probably not accounting for “full” surface water retention to the seashore.
Larger European rivers Chapter 5	NO_3-N	0.1-0.5	Only catchments with more than 25% cropland included. Probably not accounting for “full” surface water retention to the seashore.
The Thames Chapter 5	NO_3-N	2	Probably low groundwater retention and not accounting for “full” surface water retention to the seashore.
	Modelling		
Based on oxic groundwater Chapter 5	NO_3-N	2	Model estimate using current climatic conditions and retention – a maximum estimate.
Modelling by MONERIS Chapter 5	Total N	0.7	Mean concentration in the German part of the Baltic Sea catchment around 1880.
	Estimates based on present data		
Extrapolation of data 1990-2014 Chapter 5	Total N	2	Model estimate using current climatic conditions and retention – a maximum estimate.
Bøgestrand et al. (2014)	Total N	0.6-1.5	Based on data from 1990-2012. Not accounting for “full” surface water retention to the seashore.
Nature areas Chapter 5	Total N	0.1-0.2	Estimated to 1 mg N/l in the root zone.

The few measurements available from rivers around the year 1900 indicate that nearly all nitrate was denitrified in groundwater and surface waters, so the load of nitrogen to the sea consisted mainly of organic N.

The estimations from the sandy catchment support the findings from the few historical measurements conducted in Danish streams, suggesting that past nitrate concentrations were very low (traces), especially in the sandy regions of Denmark.

Utilization of the two empirically established models indicates an N concentration in surface waters of maximum 2 mg N/l (relative to current climate conditions and retention) – the same for the two estimates based on present data.

It is also important to note, that for some of the existing data from around 1900, the nitrate concentrations measured in, for instance, rivers are not routed all the way to the seashore, meaning that the results do not express the full retention in surface water.

The results summarized in Table 7.1 are, in general, lower than or in the lower end of the range 1-2 mg N/l estimated from the root zone concentration and retention. However, the fact that many of the low concentrations are nitrate and do not include, for example, organic nitrogen should be taken into account.

7.6 Geographical distribution

The resulting N concentration in the water running from land to coastal areas around the year 1900 have probably been geographically differentiated due to factors such as type of farming, soil type (i.e. groundwater retention), differences in drainage, differences in surface water retention and/or differences in precipitation. These factors probably also contribute to the often wide range in concentrations shown in Table 7.1. In Bøgestrand (2014b), the background concentration of nitrogen was geographically differentiated, with the lowest concentration (< 1 mg N/l) occurring in the western sandy regions and the highest (1-1.5 mg N/l) in the eastern part of Denmark.

7.7 Uncertainties

In our attempt to estimate the N concentration in water running to the sea around the year 1900, multiple expert judgements and assumptions have been made for nearly all elements included in the analysis. This means that the result of the individual element is probably associated with relatively high uncertainty, but nearly all elements point in the same direction (same range).

7.8 Conclusion

The climate around the year 1900 differed from today's climate and was characterized by less precipitation and lower temperatures app. 1.5 °C.

Around the year 1900 67% of the land was cropped, 8% was bare fallow and 25% was mature nature areas. For agricultural land in total it may be assumed, that the nitrate concentration leaving the root zone was in average 12 mg N/l, corresponding to the level found in today's organic arable crop farming.

Approximately 22% of the area used for agriculture was drained around 1900 (16% of the total area) compared to app. 50% of the agricultural area today.

The model-calculated total N-retention was considerably higher than the present retention, lying within the range 76–87% around the year 1900 regarded as an absolute minimum.

The conceptual "model" routing the nitrogen concentration from the root zone to the coast showed a concentration of 1-2 mg N/l in the water running into coastal areas around the year 1900.

Empirical models (surface water and groundwater related) all demonstrate concentrations within the range 1-2 mg N/l without accounting for the full retention in surface water.

None of the other approaches yields results, that are significantly outside the range 1-2 mg N/l.

From the indications etc. described in the chapters and synthesized above, the best estimate of the nitrogen concentration in the water running into the Danish coastal areas around the year 1900 therefore lies within the range 1-2 mg N/l.

7.9 Perspectives

We have provided some indications for the importance of the two main factors accounting for the lower nitrogen concentration around the year 1900 – climate and landscape - but only through three selected catchment case studies in Denmark. To fully understand how these factors have interacted to make the landscape more resilient to N loadings around the year 1900 in the different regions of Denmark, a further integrated model analysis is needed in selected representative catchments for the Danish regions. A fully integrated regional model analysis may not only provide a historical view to human effects on N pollution of groundwater and surface waters assisting in setting geographically differentiated reference targets for these ecosystems. It would also provide guidance on future management of the landscape, not least in the light of the needs to adapt to climate change.

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Appendix 1

Table for trends in yearly runoff. Year 1900 trend estimated runoff value and 95%-confidensinterval for the trend in brackets.

Stream	Trendanalsis (Z)	Trendanalyis (P)	Slope (*1000 mm yr^{-1})	Intercept (Start year-1) (*1000 mm yr^{-1})
Uggerby Å	2,95	0,0032	121,1	43808 (41870;[40674;43126])
Tude Å	2,07	0,039	96,70	23818 (20724;[17575;23675])
Harrested Å	2,76	0,0059	12,86	2423 (2153;[1967;2347])
Aamose Å	2,82	0,0047	239,6	48203 (43411;[40405;46759])
Brende Å	1,95	0,052	44,41	17135 (16336;[15648;17159])
Odense Å	1,90	0,058	169,7	84478 (81593;[78781;84612])
Brede Å	4,73	< 0,0001	597,2	86447 (73906;[69601;78520])
Ribe Å	3,98	< 0,0001	1357	228788 (185364;[164263;205265])
Aarhus Å	0,43	0,67	15,09	31099 (30812;[29585;32226])
Alergaard	4,57	< 0,0001	1393	397141 (369281;[357925;396339])
Aastedbro	2,80	0,0051	185,6	62262 (59292;[57288;61346])
Tvilum	2,26	0,024	626,4	484099 (474077;[466400;482973])
Lindenborg Å	1,30	0,19	50,26	73370 (72164;[70303;74008])
Aarup Å	3,23	0,0012	144,8	36667 (31599;[30690;35095])
Lindholm Å	2,27	0,024	69,85	26361 (25174;[24143;26189])
Saltø Å	1,97	0,049	41,28	10950 (10207;[9542;10941])
Suså	1,74	0,082	437,8	155748 (140863;[121870;157968])
Tryggevælde Å	1,36	0,17	45,60	24909 (24134;[22937;25259])

Table for trend analyses in yearly max runoff.

Vandløb	Trendanalyse (Z)	Trendanalyse (P)	Hældning (*1000 mm yr^{-1})	Afskæring (Startår-1) (*1000 mm yr^{-1})
Uggerby Å	-0,233	0,82	-0,179	670,9
Tude Å	-0,0298	0,77	-0,263	475,9
Harrested Å	1,97	0,048	0,292	53,01
Aamose Å	0,793	0,43	0,810	693,0
Brende Å	-4,03	<0,0001	-0,976	360,9
Odense Å	0,439	0,66	0,679	1245
Brede Å	3,23	0,0013	4,81	804,5
Ribe Å	2,72	0,0066	9,43	1642
Aarhus Å	-0,836	0,40	-0,349	417,4
Alergaard	1,97	0,048	8,20	2722
Aastedbro	1,63	0,10	1,99	850,4
Tvilum	0,487	0,63	1,00	2793
Lindenborg Å	0,041	0,97	0,014	517,0
Aarup Å	2,51	0,012	1,22	308,0
Lindholm Å	-3,99	<0,0001	-1,63	593,9
Saltø Å	0,082	0,93	0,029	309,0
Suså	-1,13	0,26	-3,03	2203
Tryggevælde Å	-1,13	0,26	-0,858	771,4

Table for trend analyses in yearly minimum runoff.

Stream	Trendanalysis (Z)	Trendanalysis (P)	Slope(*1000 mm yr^{-1})	Intercept (Start year-1) (*1000 mm yr^{-1})
Uggerby Å	4,05	<0,0001	0,190	16,90
Tude Å	-1,10	0,27	-0,014	7,874
Harrested Å	-0,057	0,95	0	0,505
Aamose Å	5,87	<0,0001	0,210	-2,043
Brende Å	2,98	0,0029	0,019	0,753
Odense Å	2,55	0,011	0,135	29,74
Brede Å	5,14	<0,0001	0,650	16,44
Ribe Å	5,38	<0,0001	1,98	80,98
Aarhus Å	2,71	0,0067	0,074	10,93
Alergaard	3,54	0,0004	1,45	553,1
Aastedbro	0,927	0,35	0,048	69,45
Tvilum	3,85	0,0001	1,57	532,7
Lindenborg Å	2,13	0,034	0,159	122,8
Aarup Å	0,083	0,93	0,007	45,33
Lindholm Å	3,29	0,0010	0,077	1,938
Saltø Å	0,859	0,39	0,0014	0,556
Suså	4,61	<0,0001	0,640	-15,21
Tryggevælde Å	-0,703	0,48	-0,006	4,245

All discharge statins show homogenous monthly trend analyses. All tests for homogeneity are not significant.

Tabel for trend analyses in monthly runoff.

Stream	Trendanalysis (Z)	Trendanalysis (P)	Slope (*1000 mm yr^{-1})	Intercept (Start year-1) (*1000 mm yr^{-1})
Uggerby Å	J: 1,20	J: 0,23	J: 12,57	J: 5521
	F: 0,902	F: 0,37	F: 8,285	F: 4314
	M: 1,30	M: 0,19	M: 10,71	M: 4477
	A: 0,411	A: 0,68	A: 2,121	A: 3554
	M: 3,81	M: 0,00014	M: 10,69	M: 1510
	J: 4,13	J: < 0,0001	J: 8,917	J: 982,1
	J:3,91	J: < 0,0001	J: 9,431	J: 755,0
	A: 2,76	A: 0,0057	A: 7,642	A: 1038
	S: 1,90	S: 0,058	S: 7,030	S: 1768
	O: 2,16	O: 0,030	O: 13,57	O: 2517
	N: 1,70	N: 0,089	N: 13,56	N: 3913
	D: 2,27	D: 0,023	D: 17,18	D: 4257
Tude Å	J: 2,42	J: 0,016	J: 30,08	J: 1689
	F: 0,502	F: 0,62	F: 5,601	F: 3647
	M: 0,281	M: 0,78	M: 2,477	M: 3565
	A: -1,20	A: 0,23	A: -8,427	A: 3288
	M: 0,436	M: 0,66	M: 1,187	M: 1016
	J: 1,44	J: 0,15	J: 1,727	J: 384,3
	J: -0,869	J: 0,39	J: -0,896	J: 488,7
	A: -0,314	A: 0,75	A: -0,432	A: 419,0
	S: 0,518	S: 0,60	S: 0,747	S: 410,5
	O: 1,50	O: 0,13	O: 4,35	O: 288,5
	N: 1.91	N: 0,056	N: 12,46	N: 421,4
	D: 2,21	D: 0,027	D: 24,12	D: 1315

Harrested Å	J: 2,13	J: 0,033	J: 3,074	J: 235,7
	F: 1,83	F: 0,067	F: 1,909	F: 250,5
	M: 0,118	M: 0,91	M: 0,133	M: 424,8
	A: -0,617	A: 0,54	A: -0,300	A: 244,9
	M: 0,604	M: 0,55	M: 0,138	M: 86,84
	J: 3,06	J: 0,0022	J: 0,368	J: 15,72
	J: 2,14	J: 0,032	J: 0,152	J: 19,20
	A: 2,69	A: 0,0070	A: 0,191	A: 12,84
	S: 1,64	S: 0,10	S: 0,171	S: 20,82
	O: 1,68	O: 0,092	O: 0,408	O: 28,23
	N: 1,71	N: 0,087	N: 0,893	N: 100,4
	D: 2,29	D: 0,022	D: 2,805	D: 133,8
Aamose Å	J: 1,97	J: 0,048	J: 44,83	J: 5235
	F: 1,57	F: 0,11	F: 27,96	F: 5695
	M: 1,21	M: 0,23	M: 19,70	M: 5638
	A: 0,103	A: 0,92	A: 1,310	A: 5570
	M: 2,65	M: 0,0082	M: 13,51	M: 1308
	J: 3,57	J: 0,0004	J: 10,62	J: 443,7
	J: 3,67	J: 0,0002	J: 7,83	J: 326,9
	A: 3,69	A: 0,0002	A: 6,96	A: 223,0
	S: 2,78	S: 0,0054	S: 7,90	S: 360,8
	O: 2,09	O: 0,037	O: 11,78	O: 986,6
	N: 1,34	N: 0,18	N: 15,66	N: 2759
	D: 2,27	D: 0,023	D: 41,54	D: 3787
Brende Å	J: 0,674	J: 0,50	J: 4,959	J: 2899
	F: 1,02	F: 0,31	F: 5,576	F: 2111
	M: 0,256	M: 0,80	M: 1,846	M: 2332
	A: -0,889	A: 0,37	A: -2,307	A: 1857
	M: -0,142	M: 0,89	M: -0,285	M: 838,1
	J: 1,72	J: 0,086	J: 1,485	J: 226,5
	J: 2,17	J: 0,030	J: 1,373	J: 103,3
	A: 1,59	A: 0,11	A: 1,166	A: 101,5
	S: 1,47	S: 0,14	S: 1,53	S: 157,4
	O: 1,03	O: 0,30	O: 2,47	O: 427,5
	N: 1,21	N: 0,23	N: 5,29	N: 1251
	D: 2,05	D: 0,041	D: 11,018	D: 1416
Odense Å	J: 0,939	J: 0,35	J: 23,55	J: 12006
	F: 0,583	F: 0,56	F: 13,61	F: 10835
	M: 0,83	M: 0,41	M: 16,02	M: 9608
	A: -0,614	A: 0,54	A: -7,24	A: 8479
	M: 0,258	M: 0,80	M: 1,46	M: 4377
	J: 1,26	J: 0,21	J: 4,94	J: 2275
	J: 0,467	J: 0,64	J: 1,35	J: 2159
	A: 1,02	A: 0,31	A: 3,29	A: 2028
	S: 0,209	S: 0,83	S: 0,686	S: 2439
	O: 1,23	O: 0,22	O: 9,88	O: 2845
	N: 1,31	N: 0,19	N: 20,57	N: 5052
	D: 2,11	D:0,035	D: 43,73	D: 7255

Brede Å	J: 2,50	J: 0,013	J: 66,10	J: 9459
	F: 2,67	F: 0,0075	F: 53,16	F: 7143
	M: 3,06	M: 0,0022	M: 50,34	M: 6987
	A: 3,02	A: 0,0025	A: 32,43	A: 5244
	M: 3,19	M: 0,0014	M: 23,48	M: 3735
	J: 4,50	J: <0,0001	J: 23,16	J: 2046
	J: 4,07	J: <0,0001	J: 22,80	J: 1246
	A: 2,60	A: 0,0093	A: 24,70	A: 1756
	S: 2,74	S: 0,0061	S: 36,83	S: 2308
	O: 2,53	O: 0,011	O: 53,35	O: 3780
	N: 2,97	N: 0,0030	N: 69,72	N: 5644
	D: 3,40	D: 0,0007	D: 79,70	D: 6458
Ribe Å	J: 2,65	J: 0,0081	J: 189,0	J: 15214
	F: 1,74	F: 0,082	F: 85,53	F: 21581
	M: 3,26	M: 0,0011	M: 160,7	M: 11601
	A: 2,06	A: 0,040	A: 69,12	A: 14815
	M: 3,57	M: 0,0004	M: 90,29	M: 6715
	J: 4,83	J: <0,0001	J: 75,61	J: 4789
	J: 3,21	J: 0,0013	J: 42,30	J: 7102
	A: 2,88	A: 6613	A: 2,88	A: 6613
	S: 3,10	S: 0,0019	S: 84,28	S: 6167
	O: 2,93	O: 0,0033	O: 129,9	O: 6846
	N: 3,29	N: 0,0010	N: 194,5	N: 8307
	D: 3,30	D: 0,0010	D: 202,3	D: 10363
Aarhus Å	J: 1,66	J: 0,097	J: 13,67	J: 2988
	F: 0,332	F: 0,74	F: 2,944	F: 4155
	M: 0,225	M: 0,82	M: 1,390	M: 3647
	A: -1,92	A: 0,055	A: -8,645	A: 3702
	M: -1,53	M: 0,13	M: -3,855	M: 2234
	J: -0,927	J: 0,35	J: -1,366	J: 1287
	J: -0,845	J: 0,40	J: -1,385	J: 1103
	A: -1,07	A:0,28	A: -1,858	A: 1175
	S:-0,573	S:0,57	S: -1,121	S: 1158
	O: 0,022	O:0,98	O: 0,074	O: 1397
	N:-0,605	N:0,55	N: -3,063	N: 2583
	D: 1,53	D:0,13	D: 9,796	D: 2363
Alergaard	J: 2,84	J: 0,0045	J: 177,3	J: 36244
	F: 2,39	F: 0,017	F: 123,1	F: 35484
	M: 3,72	M: 0,0002	M: 158,0	M: 31186
	A: 4,67	A: <0,0001	A: 157,0	A: 23298
	M: 5,51	M: <0,0001	M: 134,1	M: 18911
	J: 4,49	J: <0,0001	J: 87,58	J: 18282
	J: 1,98	J: 0,048	J: 38,85	J: 22299
	A: 1,88	A: 0,060	A: 43,32	A: 23867
	S: 2,27	S: 0,023	S: 67,04	S: 23282
	O: 2,03	O: 0,042	O: 79,73	O: 28748
	N: 2,16	N: 0,031	N: 109,8	N: 34627
	D: 3,43	D: 0,0006	D: 161,1	D: 33822

Aastedbro	J: 1,45	J: 0,15	J: 26,93	J: 8121
	F: 1,48	F: 0,14	F: 19,32	F: 6986
	M: 1,59	M: 0,11	M: 18,47	M: 6642
	A: 0,319	A: 0,75	A: 1,980	A: 5748
	M: 1,01	M: 0,31	M: 3,705	M: 3859
	J: 1,44	J: 0,15	J: 3,485	J: 3007
	J: -0,816	J: 0,41	J: -1,680	J: 3141
	A: -0,694	A: 0,488	A: -1,856	A: 3261
	S: -0,215	S: 0,83	S: -1,028	S: 3773
	O: 0,632	O: 0,53	O: 5,344	O: 4957
	N: 1,01	N: 0,314	N: 13,23	N: 6685
	D: 2,60	D: 0,0092	D: 32,79	D: 6294
Tvilum	J: 1,04	J: 0,30	J: 65,31	J: 52535
	F: 1,72	F: 0,085	F: 89,72	F: 46294
	M: 1,20	M: 0,23	M: 63,07	M: 46932
	A: 0,636	A: 0,53	A: 22,86	A: 43324
	M: 1,05	M: 0,30	M: 25,16	M: 32796
	J: 2,55	J: 0,011	J: 54,63	J: 23646
	J: 1,49	J: 0,14	J: 26,57	J: 25767
	A: 0,780	A: 0,44	A: 15,87	A: 27246
	S: 1,65	S: 0,099	S: 37,60	S: 26754
	O: 1,62	O: 0,11	O: 49,61	O: 30418
	N: 1,09	N: 0,28	N: 44,92	N: 37589
	D: 1,84	D: 0,065	D: 91,70	D: 43189
Lindenborg Å	J: 1,73	J: 0,084	J: 11,28	J: 6429
	F: 0,878	F: 0,38	F: 5,483	F: 6114
	M: 1,26	M: 0,21	M: 7,005	M: 6289
	A: -0,620	A: 0,54	A: -2,415	A: 6509
	M: 0,630	M: 0,53	M: 1,745	M: 5630
	J: 0,994	J: 0,32	J: 3,499	J: 4794
	J: 0,466	J: 0,64	J: 1,831	J: 5012
	A: 1,28	A: 0,20	A: 4,712	A: 4656
	S: 1,40	S: 0,16	S: 5,036	S: 4845
	O: 2,12	O: 0,034	O: 9,421	O: 5134
	N: 0,699	N: 0,48	N: 3,274	N: 5989
	D: 1,82	D: 0,068	D: 8,142	D: 6215
Aarup Å	J: 2,96	J: 0,0030	J: 33,78	J: 1907
	F: 1,81	F: 0,070	F: 19,25	F: 2684
	M: 1,84	M: 0,066	M: 17,87	M: 3023
	A: 0,356	A: 0,72	A: 2,141	A: 3307
	M: 0,120	M: 0,90	M: 0,3303	M: 2514
	J: -0,411	J: 0,68	J: -1,075	J: 2148
	J: -0,278	J: 0,78	J: -0,886	J: 1917
	A: 0,0291	A: 0,98	A: 0,0701	A: 1769
	S: 1,13	S: 0,26	S: 4,951	S: 1535
	O: 1,99	O: 0,046	O: 15,04	O: 1466
	N: 2,10	N: 0,036	N: 21,17	N: 2209
	D: 2,12	D: 0,034	D: 18,40	D: 3114

Lindholm Å	J: 0,866	J: 0,39	J: 7,037	J: 4187
	F: 0,325	F: 0,75	F: 2,602	F: 3493
	M: 1,12	M: 0,26	M: 7,628	M: 2879
	A: 0,356	A: 0,72	A: 1,225	A: 2028
	M: 3,07	M: 0,0021	M: 5,550	M: 695,7
	J: 4,26	J: <0,0001	J: 5,742	J: 121,5
	J: 2,55	J: 0,011	J: 2,992	J: 260,8
	A: 1,52	A: 0,13	A: 2,578	A: 377,4
	S: 1,96	S: 0,050	S: 4,640	S: 481,9
	O: 2,76	O: 0,0057	O: 11,17	O: 616,0
	N: 1,66	N: 0,097	N: 10,73	N: 2013
	D: 2,14	D: 0,032	D: 14,38	D: 2690
Saltø Å	J: 0,820	J: 0,41	J: 4,891	J: 2043
	F: 0,826	F: 0,41	F: 4,067	F: 1761
	M: 0,484	M: 0,63	M: 1,889	M: 1746
	A: -0,548	A: 0,58	A: -1,187	A: 939,8
	M: -0,079	M: 0,94	M: -0,0497	M: 336,1
	J: 0,351	J: 0,73	J: 0,1259	J: 131,9
	J: 1,28	J: 0,20	J: 0,2355	J: 48,32
	A: 1,84	A: 0,066	A: 0,2858	A: 30,31
	S: 1,21	S: 0,23	S: 0,3464	S: 51,67
	O: 1,29	O: 0,20	O: 0,9634	O: 155,3
	N: 1,28	N: 0,20	N: 3,606	N: 457,1
	D: 2,26	D: 0,024	D: 11,83	D: 703,3
Suså	J: 1,74	J: 0,082	J: 132,0	J: 16971
	F: 0,428	F: 0,67	F: 21,66	F: 24524
	M: -0,045	M: 0,96	M: -7,464	M: 24335
	A: -1,49	A: 0,14	A: -61,95	A: 22755
	M: 0	M: 1	M: 0,0673	M: 7406
	J: 0,143	J: 0,89	J: 1,185	J: 4576
	J: 1,11	J: 0,27	J: 8,997	J: 2700
	A: 1,01	A: 0,31	A: 5,859	A: 2636
	S: 1,20	S: 0,23	S: 9,522	S: 2154
	O: 2,08	O: 0,037	O: 37,30	O: 982,8
	N: 2,36	N: 0,018	N: 85,21	N: 1435
	D: 2,31	D: 0,021	D: 145,7	D: 7337
Tryggevælde Å	J: 0,595	J: 0,55	J: 6,778	J: 4307
	F: 0,381	F: 0,70	F: 3,353	F: 3533
	M: 0,804	M: 0,42	M: 6,256	M: 3062
	A: -0,510	A: 0,61	A: -2,445	A: 2332
	M: 1,23	M: 0,22	M: 2,530	M: 811,8
	J: 1,72	J: 0,086	J: 1,841	J: 250,4
	J: 0,761	J: 0,45	J: 0,4886	J: 219,9
	A: 1,59	A: 0,11	A: 1,072	A: 142,2
	S: 0,368	S: 0,71	S: 0,4901	S: 316,8
	O: 0,706	O: 0,48	O: 1,502	O: 694,7
	N: 1,15	N: 0,25	N: 6,273	N: 1199
	D: 2,11	D: 0,035	D: 17,61	D: 2256

Table for trends in yearly precipitation.

Climate station	Trendanalysis (Z)	Trendanalysis (P)	Slope (mm yr^{-1})	Intercept (Start year-1) (mm yr^{-1})
Grønbæk	4,14	< 0,0001	2,216	562,9
Nordby	3,94	< 0,0001	1,642	641,1
Vestervig	4,18	< 0,0001	2,118	637,0
Broderup	3,11	0,0019	3,509	698,0
København	3,66	0,00025	1,23	522,8

The development in monthly precipitation is non homogenous for the five climate stations.

Table for trends in monthly precipitation.

Klimastation	Trendanalyse (Z)	Trendanalyse (P)	Hældning (mm yr^{-1})	Afskæring (Startår-1) (mm yr^{-1})
Grønbæk	J: 5,04	J: <0,0001	J: 0,304	J: 29,48
	F: 3,14	F: 0,0017	F: 0,157	F: 24,61
	M: 2,83	M: 0,0046	M: 0,124	M: 28,31
	A: 0,077	A: 0,94	A: 0,0034	A: 36,15
	M: 1,18	M: 0,24	M: 0,060	M: 32,75
	J: 2,37	J: 0,018	J: 0,151	J: 35,71
	J:-0,180	J: 0,86	J: -0,011	J: 64,74
	A: -0,814	A: 0,42	A: -0,068	A: 81,46
	S: 1,12	S: 0,26	S: 0,073	S: 53,66
	O: 1,47	O: 0,14	O: 0,117	O: 58,08
	N: 3,39	N: 0,00070	N: 0,209	N: 47,06
	D: 3,84	D: 0,00013	D: 0,243	D: 37,22
Nordby	J: 3,70	J: 0,00022	J: 0,219	J: 38,11
	F: 2,29	F: 0,022	F: 0,122	F: 31,08
	M: 1,51	M: 0,13	M: 0,076	M: 34,20
	A: 0,046	A: 0,96	A: 0,0019	A: 34,82
	M: 1,37	M: 0,17	M: 0,059	M: 30,89
	J: 1,44	J: 0,15	J: 0,081	J: 40,72
	J: 0,517	J: 0,61	J: 0,036	J: 55,76
	A: -1,13	A: 0,26	A: -0,095	A: 85,87
	S: 1,68	S: 0,093	S: 0,143	S: 62,70
	O: 0,995	O: 0,32	O: 0,086	O: 82,37
	N: 3,78	N: 0,00016	N: 0,269	N: 53,36
	D: 3,16	D: 0,0016	D: 0,213	D: 50,89
Vestervig	J: 3,87	J: 0,00011	J: 0,243	J: 42,56
	F: 1,87	F: 0,061	F: 0,097	F:32,30
	M: 0,993	M: 0,32	M: 0,060	M: 40,66
	A: 0,446	A: 0,66	A: 0,017	A: 35,48
	M: 0,603	M: 0,55	M: 0,026	M: 37,71
	J: 1,59	J: 0,11	J: 0,087	J: 34,90
	J: -0,582	J: 0,56	J: -0,037	J: 59,29
	A: -0,131	A: 0,90	A: -0,014	A: 77,92
	S: 1,99	S: 0,047	S: 0,149	S: 66,38
	O: 0,931	O: 0,35	O: 0,098	O: 82,23
	N: 3,59	N: 0,00033	N: 0,251	N: 61,67
	D: 2,47	D: 0,014	D: 0,177	D: 56,88

Broderup	J: 1,93	J: 0,054	J: 0,283	J: 39,31
	F: 1,96	F: 0,050	F: 0,196	F: 20,60
	M: 2,82	M: 0,0048	M: 0,303	M: 12,17
	A: -0,733	A: 0,46	A: -0,056	A: 45,38
	M: 1,02	M: 0,31	M: 0,107	M: 41,67
	J: 3,21	J: 0,0013	J: 0,367	J: 28,28
	J: 0,395	J: 0,69	J: 0,056	J: 70,41
	A: -0,094	A: 0,92	A: -0,017	A: 98,29
	S: 1,18	S: 0,24	S: 0,180	S: 58,16
	O: 1,57	O: 0,12	O: 0,249	O: 62,91
	N: 1,37	N: 0,17	N: 0,230	N: 55,64
	D: 3,407	D: 0,00066	D: 0,497	D:24,45
København	J: 3,84	J: 0,00012	J: 0,196	J: 28,78
	F: 2,03	F: 0,042	F: 0,083	F: 27,49
	M:0,586	M: 0,56	M: 0,022	M: 29,53
	A: 0,305	A: 0,76	A: 0,013	A: 33,22
	M: 1,52	M: 0,13	M: 0,069	M: 34,20
	J: 1,95	J: 0,051	J: 0,099	J: 40,24
	J:-0,743	J: 0,46	J: -0,043	J: 62,38
	A: -0,658	A: 0,51	A: -0,053	A: 66,81
	S: 0,686	S: 0,49	S: 0,043	S: 49,19
	O: 0,074	O: 0,94	O: 0,0070	O: 58,35
	N: 2,10	N:0,036	N: 0,111	N: 42,14
	D: 3,04	D: 0,0024	D:0,149	D: 39,87

Table with results of the NAM simulated parameters for Nordby climate station and Ribe stream – yearly values. Estimated and simulated values for year 1900 respectively in brackets.

Variabel	**Trendanalyse (Z)**	**Trendanalyse (P)**	**Hældning**	**Afskæring (Startår-1)**
Epot	4,04	< 0,0001	0,188	285,2
EA	5,02	< 0,0001	0,0109	13,08
Q	4,95	< 0,0001	7,11	2161
				(2360;[2285;2432])(1998,78)

Q shows a non homogeneous developmet for months.

Table with results of the NAM simulated parameters for Vestervig climate station and Aarup stream – yearly values. Estimate and simulated values for year 1900 respectively in brackets.

Vandløb	**Trendanalyse (Z)**	**Trendanalyse (P)**	**Hældning**	**Afskæring (Startår-1)**
Epot	5,96	< 0,0001	0,265	357,4
EA	5,64	< 0,0001	0,0097	12,80
Q	4,84	< 0,0001	1,18	376,0
				(409;[397;421])(418,11)

Q shows a non homogeneous developmet for months.

Table with results of the NAM simulated parameters for københavn climate station and Tryggevælde stream – yearly values. Estimate and simulated values for year 1900 respectively in brackets.

Vandløb	Trendanalyse (Z)	Trendanalyse (P)	Hældning	Afskæring (Startår-1)
Epot	-4,62	< 0,0001	-0,275	442,8
EA	-3,50	0,0005	-0,137	376,1
Q	5,43	< 0,0001	1,16	200,8
				(233;[222;244])(303,39)

Epot and Ea Q shows a non homogeneous developmet for months.

Appendix 2

Figure AP2.1. Yearly runoff from Uggerby stream station, based on measured data and statistical trend model (red line).

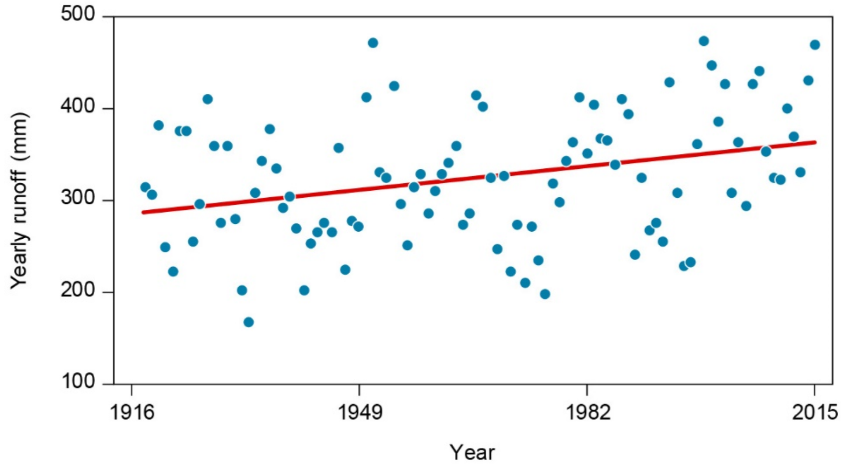


Figure AP2.2. Yearly runoff from Lindholm stream station, based on measured data and statistical trend model (red line).

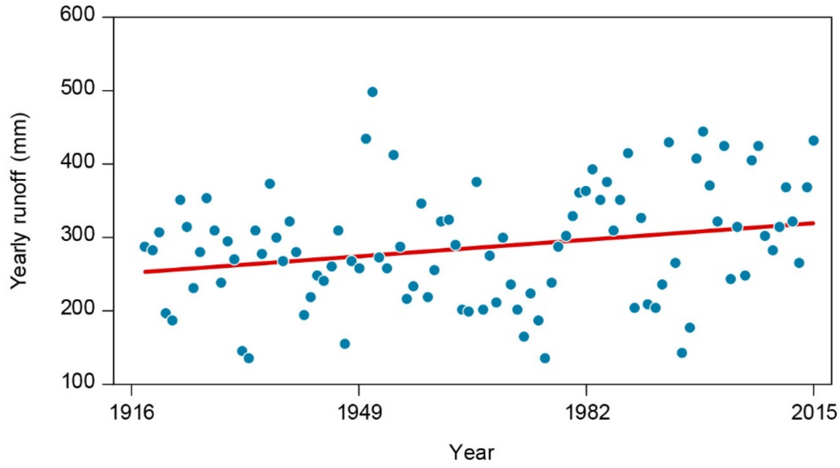
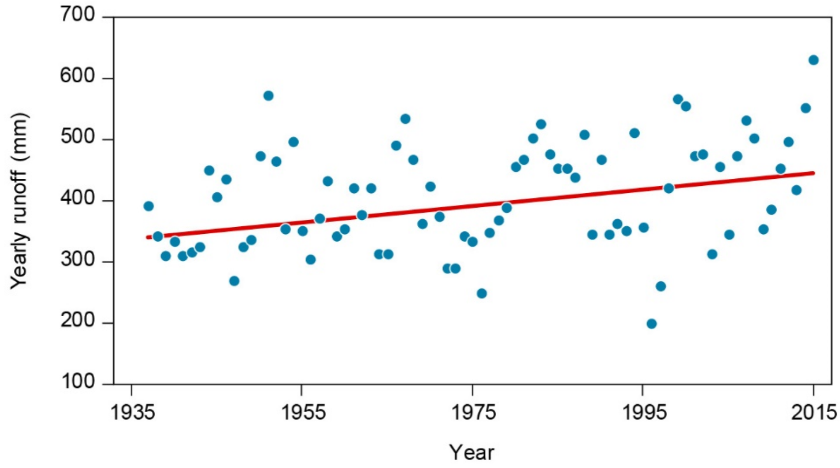


Figure AP2.3. Yearly runoff from Aarup stream station, based on measured data and statistical trend model (red line).



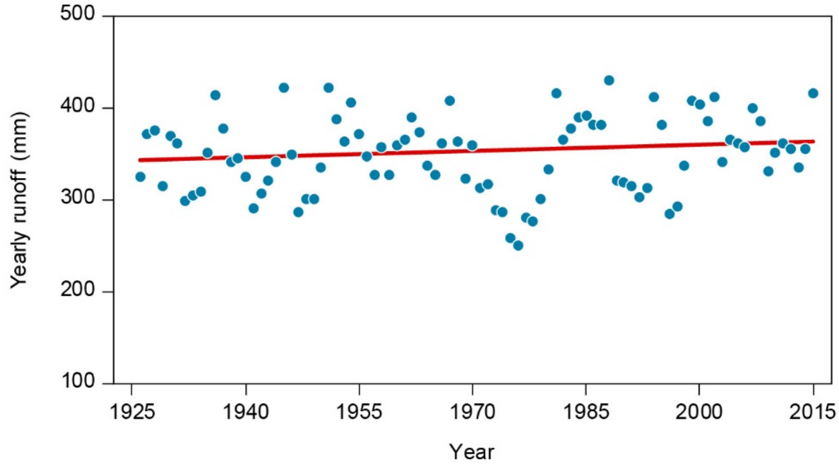


Figure AP2.4. Yearly runoff from Lindenborg stream station, based on measured data and statistical trend model (red line).

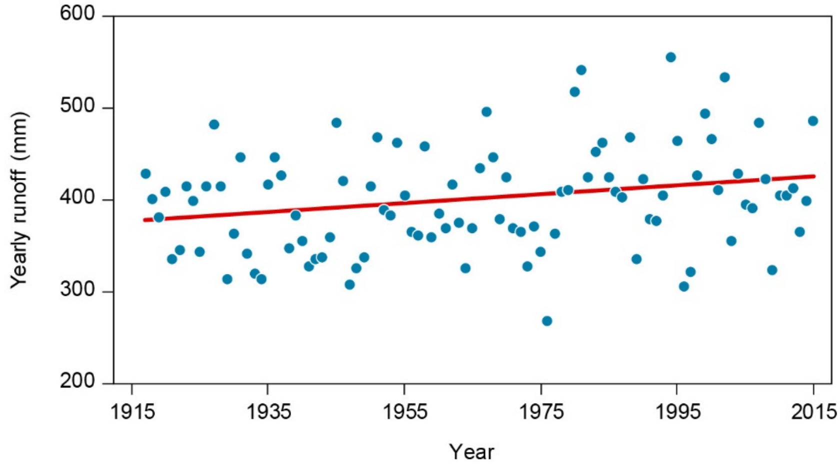


Figure AP2.5. Yearly runoff from Tvilum stream station, based on measured data and statistical trend model (red line).

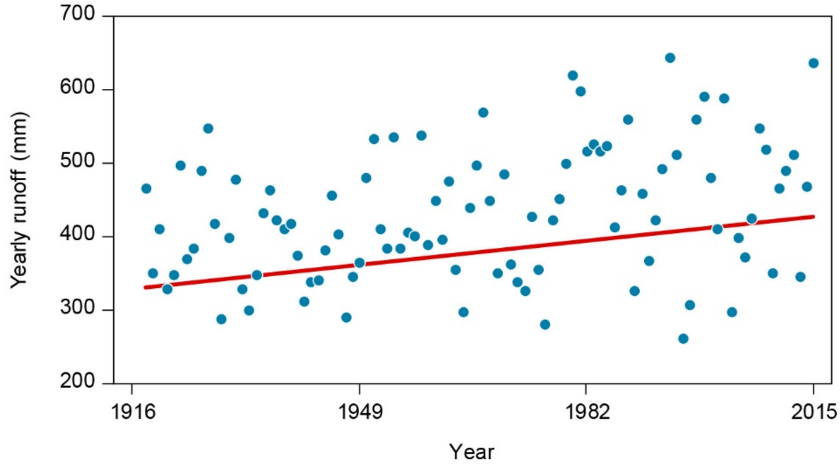


Figure AP2.6. Yearly runoff from Aasted stream station, based on measured data and statistical trend model (red line).

Figure AP2.7. Yearly runoff from Aasted stream station, based on measured data and statistical trend model (black line) and 95% confidence interval for the trend (red lines).

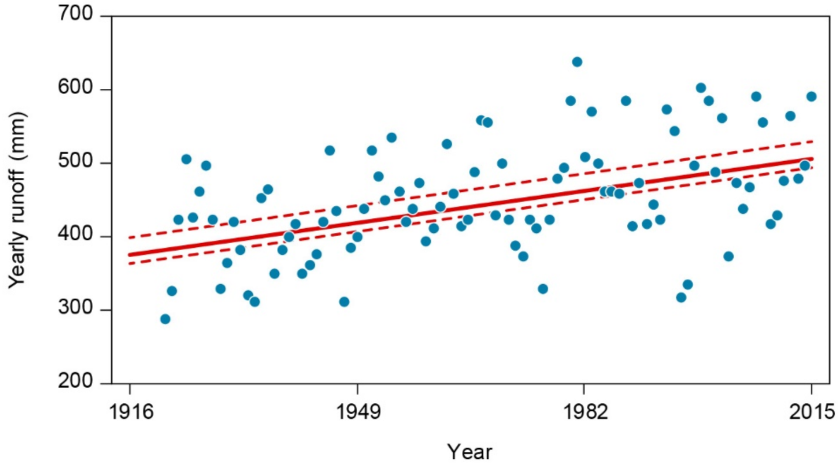


Figure AP2.8. Yearly runoff from Aarhus stream station, based on measured data and statistical trend model (red line).

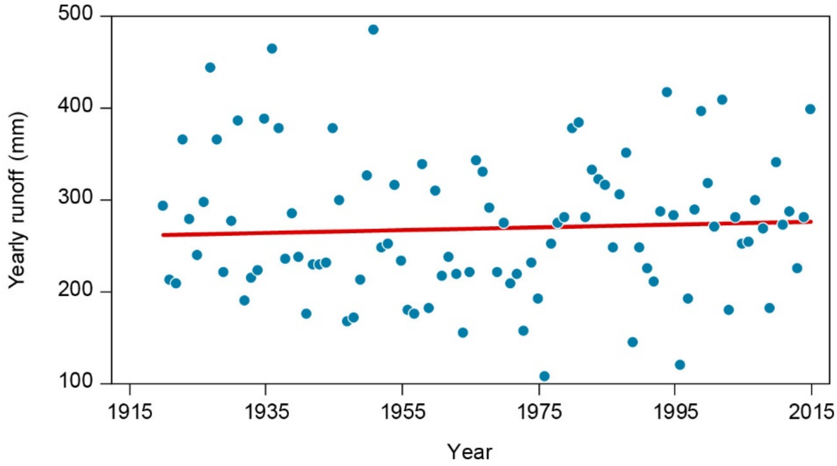


Figure AP2.9. Yearly runoff from Ribe stream station, based on measured data and statistical trend model (red line).

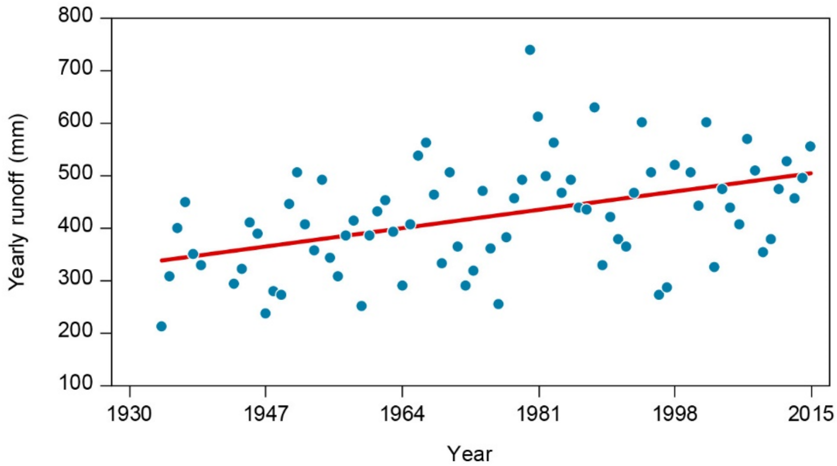


Figure AP2.10. Yearly runoff from Brede stream station, based on measured data and statistical trend model (red line).

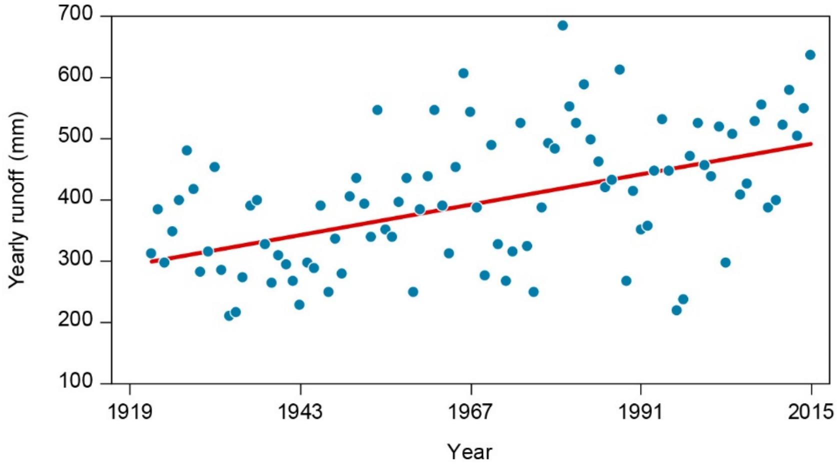


Figure AP2.11. Yearly runoff from Odense stream station, based on measured data and statistical trend model (red line).

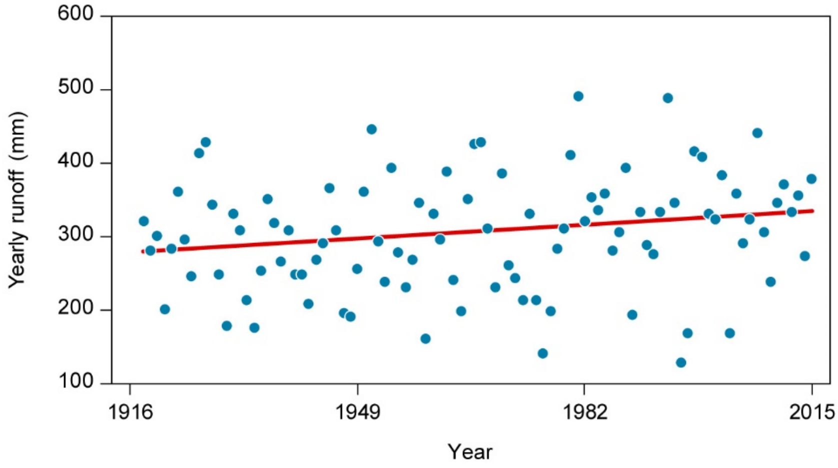


Figure AP2.12. Yearly runoff from Brende stream station, based on measured data and statistical trend model (red line).

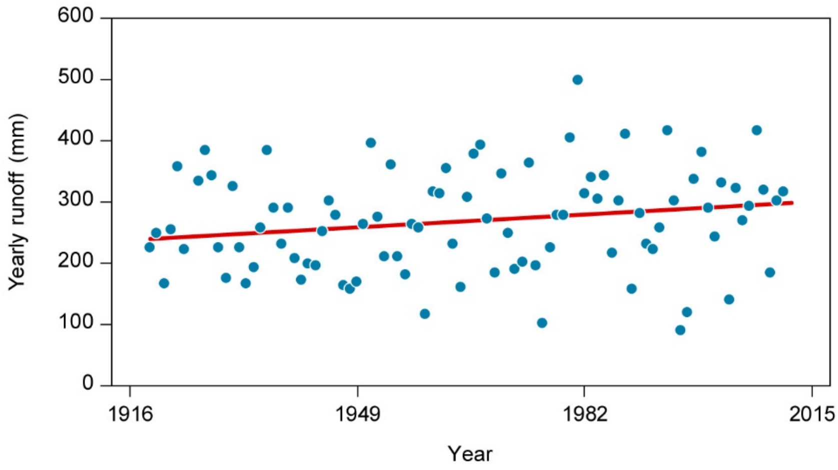


Figure AP2.13. Yearly runoff from Åmose stream station, based on measured data and statistical trend model (red line).

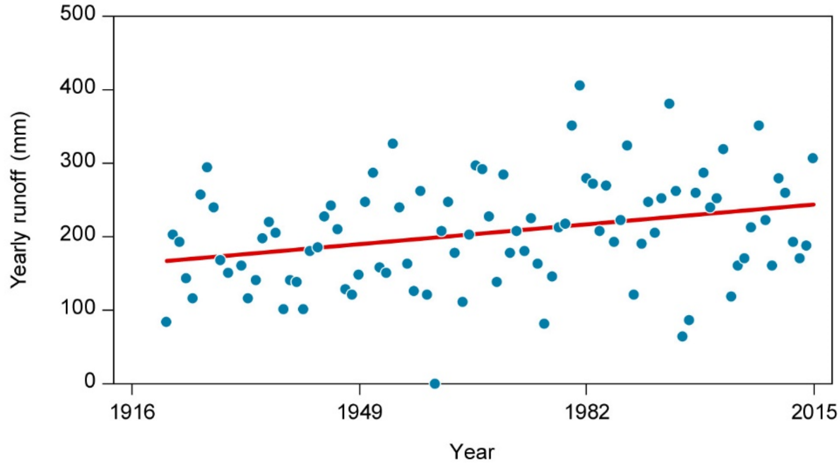


Figure AP2.14. Yearly runoff from Harrested stream station, based on measured data and statistical trend model (red line).

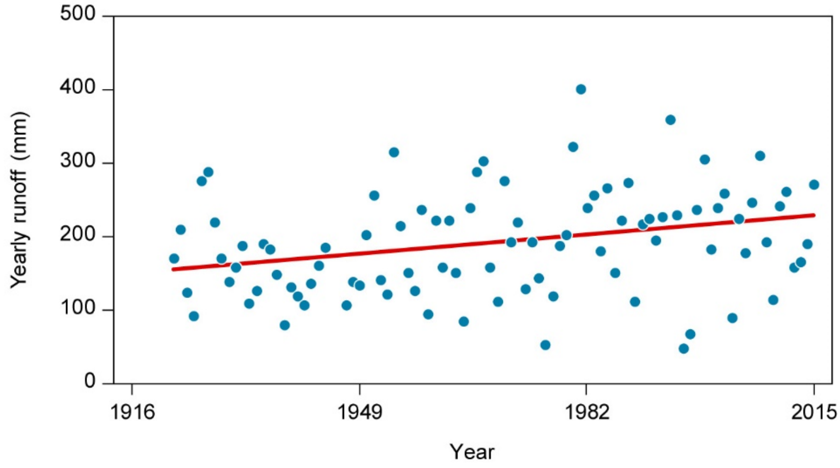


Figure AP2.15. Yearly runoff from Tude stream station, based on measured data and statistical trend model (red line).

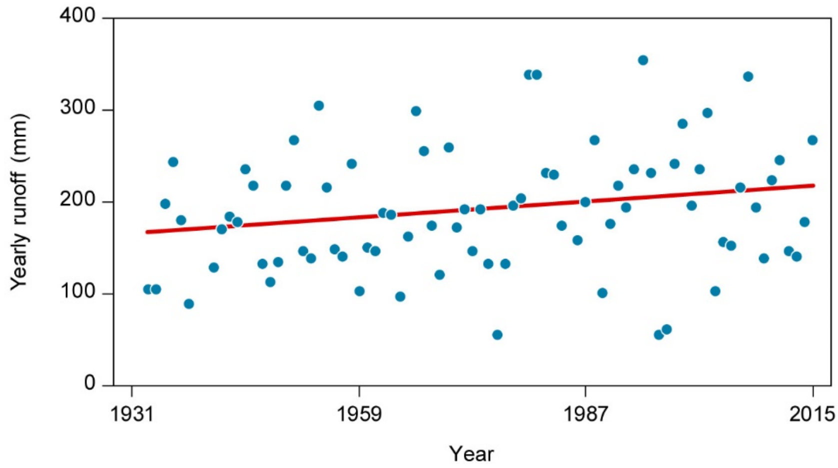


Figure AP2.16. Yearly runoff from Saltø stream station, based on measured data and statistical trend model (red line).

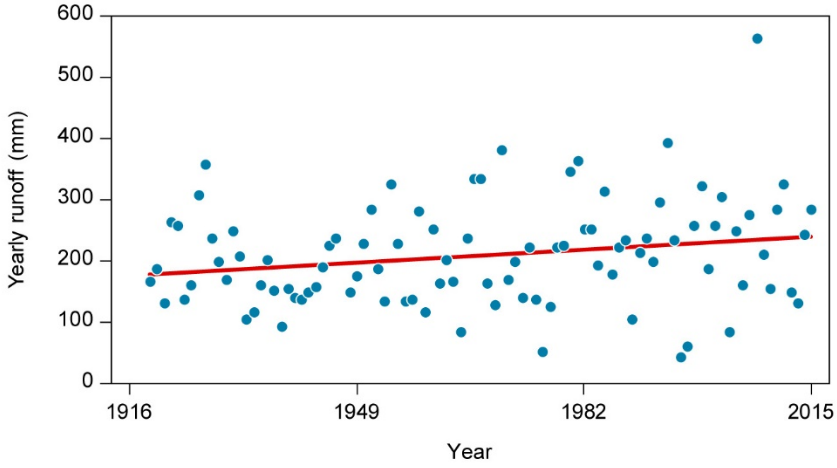


Figure AP2.17. Yearly runoff from Suså stream station, based on measured data and statistical trend model (red line).

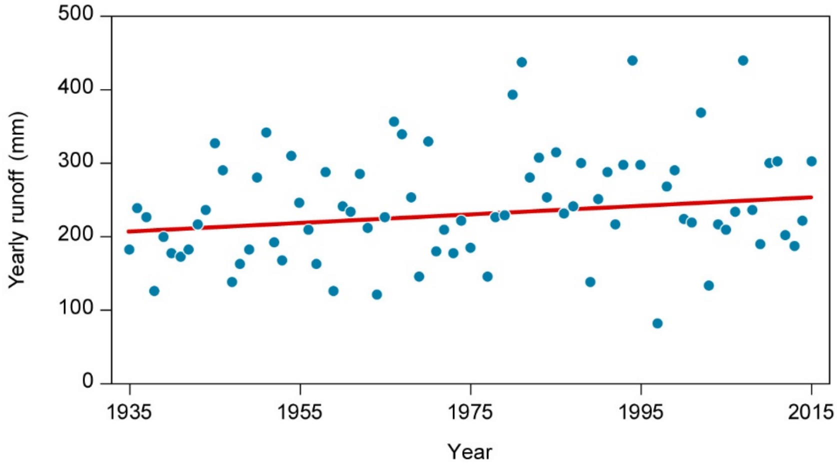
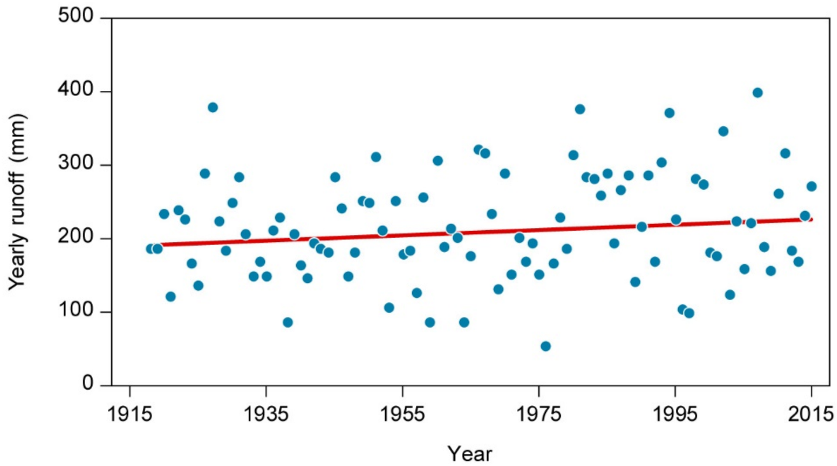


Figure AP2.18. Yearly runoff from Tryggevælde stream station, based on measured data and statistical trend model (red line).



Appendix 3

International Workshop on Estimation of nitrogen loads to the marine environment around the time of the year 1900

Date:
14-15 November 2016

Location:
The Sandbjerg Estate – Aarhus University's Conference Centre (http://www.sandbjerg.dk/en/)
Sandbjergvej 102, DK-6400 Sønderborg, Denmark

Programme

14 November:

Travel and arrival at Sandbjerg (check in at Sandbjerg Estate: from 17.00)

18.30:	Dinner and get together

15 November:

7.30-8.30:	Breakfast
8.30 – 8.40	Welcome by Director of DCE - Da Hanne Bach
8.40 - 9.30	Introduction • **A short introduction to reference conditions** by Chief Consultant Poul Nordemann Jensen (15 min) • **Reconstruction of nutrient inputs and Baltic Sea environment during 20th Century** by Director of BNI Sweden Bo Gustafsson, Stockholm University, Sweden (25+5 min)
9.30 – 10.15	Session 1 (Chair: Professor Jørgen Eivind Olesen): **Land use, agriculture, landscape and drainage around the time of the year 1900 in Denmark and North West European countries.** • **A proxy for nitrogen concentrations in Danish streams around year 1900** by Professor Brian Kronvang, Aarhus University, Denmark (12+3 min) • **Agricultural land use and management: Implications for nitrogen leaching losses** by Professor Bent Tolstrup Christensen, Aarhus University, Denmark (12+3 min) • **Assessment of changes in nitrogen retention in the landscape from year 1900 until present days** by Senior Advisor Hans Thodsen, Aarhus University, Denmark (12+3 min)

10.15 – 10.45	Coffee break
10.45 – 12.15	Session 2 (Chair: Professor Brian Kronvang): **Utilizing historical data to detect changes in hydrology and nitrogen concentrations and loadings around the time of the year 1900** • **Analyses of background nitrogen concentrations in Sweden: evolution of landscape processes and hydrology** by Senior Researcher Alena Bartosova, SMHI, Sweden (25+5 min) • **On the precipitation and temperature climate in Denmark around the time of the year 1900** by Senior Advisor Flemming Vejen, DMI, Denmark (12+3 min) • **Trends in discharge and climate during the last ca. 100 years** by Post Doc. Jane Rosenstand Poulsen, Aarhus University, Denmark (12+3 min) • **Atmospheric deposition of nitrogen around year 1900** by Senior Researcher Jesper Heile Christensen, Aarhus University, Denmark (12+3 min)
12.15 – 13.00	Lunch
13.00 – 14.45	Session 3 (Chair: Professor Jørgen Eriksen): **Methods applied to estimate hydrology and nitrogen concentrations and loadings around the time of the year 1900** • **Modelling nutrient input to Central European surface waters in the 1880s to estimate reference conditions using the model MONERIS** by Scientist Markus Venohr, IGB, Germany (25+5 min) • **Historical records of nitrate in oxidized groundwater in Denmark** by Senior Scientist Birgitte Hansen, GEUS, Denmark (25+5 min) • **Can marine observations and models assist in setting nitrogen loadings around year 1900?** by Professor Stig Markager, Aarhus University, Denmark (12+3 min) • **The Dutch approach for apportionment of diffuse sources of nutrients in surface waters** by Scientist Erwin van Boekel, Wageningen University, Holland (25+5 min)
14.45 – 15.45	Discussion and future perspectives (Chair: Chief Consultant Poul Nordemann Jensen)
15.45 – 16.00	Concluding and closing session

List of participants

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Research Assistant Mette Vodder Carstensen	Aarhus University, Denmark	mvc@bios.au.dk

Appendix 4

Nationalt seminar om Estimering af kvælstofkoncentration fra jord til fjord omkring år 1900

Dato:
6. april 2017 kl. 9.30 – 14.00

Sted:
Moesgaard Museum (http://www.moesgaardmuseum.dk/)
Moesgård Allé 15, 8270 Højbjerg
Lokale 4240-301 (Moesgaard Konferencelokale)

Program

9.30 – 10.00:	Ankomst og morgenkaffe
10.00 – 10.10	Velkomst v. Hanne Bach, Direktør for DCE
10.10 - 10.20	Introduktion v. Poul Nordemann Jensen
10.20 – 11.30	Centrale emner fra rapporten ”Estimation of Nitrogen Concentrations from root zone to marine areas around the year 1900” • Klima og hydrologi v. Jane Rosenstrand Poulsen • Landbrug og N-tab fra rodzonen v. Bent T. Christensen • Historiske data m.m. v. Jørgen Windolf • Retention v. Hans Thodsen
11.30 – 11.50	Kaffepause
11.50 – 12.10	Syntese v. Poul Nordemann Jensen
12.10 – 12.50	Diskussion (ordstyrer: Brian Kronvang)
12.50 – 13.00	Orientering om videre forløb og afslutning af workshop v. Hanne Bach
13.00 – 14.00	Frokost (i museets café)

Baggrund
I 2014 bad Miljøstyrelsen DCE – Nationalt Center for Miljø og Energi om at vurdere størrelsen af udledningen af kvælstof omkring år 1900. DCE udarbejdede efterfølgende notatet ”Næringsstof-belastningen til vandområder omkring år 1900”, hvori det blev vurderet, at koncentrationsniveauet af kvælstof i det vand, der løb til havet omkring år 1900 var på omkring 1 mg N pr. liter.

I et udredningsprojekt er AU-forskere fra DCE – Nationalt Center for Miljø og Energi og DCA – Nationalt Center for Fødevarer og Jordbrug nu gået endnu dybere ned i arbejdet med at undersøge, hvordan kvælstofkoncentrationen så ud omkring år 1900.

På det tidspunkt var der eksempelvis ikke mulighed for at anvende sprøjtemidler til ukrudtsregulering. I stedet kunne ukrudtsproblemer forebygges ved at holde jorden sort (brak) gennem længere perioder;

hvilket gav en øget udvaskning af kvælstof. Tilsvarende var udnyttelsen af kvælstof i husdyrgødning også markant lavere end i dag, bl.a. fordi en del af husdyrgødningen blev bragt ud om efteråret.

Til gengæld var der dengang langt flere vådområder og slyngede vandløb m.m., som medvirker til at kvælstof omdannes til uskadeligt, luftformigt kvælstof. Desuden var der færre drænrør i markerne end nu og dermed strømmede der mindre kvælstof direkte ud i vandmiljøet. Omkring år 1900 var såvel udnyttelsen som mængden af kvælstof i planteproduktionen betydeligt mindre end i dag, og anvendelsen af kvælstof i handelsgødning var ubetydelig. Der er således mange, og ofte modsatrettede faktorer, der påvirker udledningen.

Aarhus Universitet besluttede at gennemføre en tilbundsgående analyse af landbrugsdrift og andre faktorer, som påvirkede koncentrationen på det pågældende tidspunkt og som dermed kan have påvirket koncentrationsniveauet i det vand, der løb til havet omkring år 1900.

Undersøgelsen er blevet gennemført ved analyse af arealanvendelse og landbrugspraksis omkring år 1900 sammenholdt med de forhold i landskabet, der påvirker transport og omsætning af kvælstof fra jord til fjord. Der er fx blevet kigget på forhold, der kan have påvirket kvælstofudvaskningen fra landbruget, herunder klima, landbrugspraksis, dræningsgraden, kvælstoffikserende afgrøder, kvælstofdeposition og opdyrkningshistorie.

Desuden har forskerne udbygget det empiriske grundlag for fastsættelse af kvælstofkoncentrationer i vandløb med yderligere historiske målinger fra vandløb og grundvand. Disse forhold er sammenstillet som grundlag for kvalificering af niveau og usikkerheder omkring koncentration og mængde af kvælstof udledt til det marine miljø omkring år 1900. Endelig har man analyseret, hvordan den forekomst af ålegræs, man fandt omkring år 1900, svarer til niveauet for kvælstofudledning.

I forbindelse med projektet afholdtes en international workshop med inddragelse af nationale og internationale eksperter og interessenter på området, som bidrog med erfaringer og drøftelser af metodevalg.

Resultaterne af projektet er beskrevet i rapporten ”Estimation of Nitrogen Concentrations from root zone to marine areas around the year 1900” (under udarbejdelse).

Appendix 5

Lars Bergström, SLU, Sverige:

Comments on the technical report from DCE: Estimation of nitrogen concentrations from root zone to marine areas around the year 1900

General impression:

The objective of this report is to evaluate how different factors such as climate, land use, agricultural activities etc. influence nitrogen (N) concentrations reaching marine environments surrounding Denmark in the year 1900. This should form a basis for comparison with the situation today and if deviations occur, will they comply with the EU Water Framework Directive in terms of 'good ecological status' in coastal waters. In order to compensate for the lack of data on N concentrations in streams from 1900, agricultural statistics and model estimates were used in the evaluations. This certainly introduces a great deal of uncertainty in the estimates, which the authors are aware of and point out in several places in the report. Still, the final results obtained are considered to be rather 'robust'.

Overall, it is a very comprehensive report covering climate and hydrology, agricultural non-point source pollution, N deposition, point sources and N retention in streams, all which have a major impact on N levels in soil and waters.

Appendix 6

General remarks from Marcus Vernohr, IGB-Berlin, Tyskland

This is very interesting and in total one of the most comprehensive study on nutrient flux analysis around 1900. A strength of the report is to address most important drivers impacting nutrient concentrations in surface water and loads to the sea, including climate and run-off, which was neglected in other approaches, as e.g. such in Germany. However the report also has some short comings, as briefly concluded in the following:

- As described in the preface of the report, the EU-WWFD requires a good ecological status for all limnic and marine ecosystems systems and for groundwater. Based on an eelgrass modelling, reference conditions and a good to moderate threshold were derived. The findings from this report shall be used to model reference conditions for other quality elements like Chl-a (i.e. Algae). For this purpose other parameters like phosphorus concentrations, shading and water temperature would be needed, which are not address by this report.

- Chapter 7, which was not available to me, should provide a more detailed justification than such given in the introduction. This would also suggest locating chapter 7 at the beginning of the document as it is the basis for all following evaluations. Without knowing the content of chapter 7 the justification of assuming the good-moderate threshold as N concentrations at 1900 + 26 % is quiet vague. I think at least different approaches for Baltic Sea and North Sea would be needed. Compared to current conditions the spatial pattern of emissions and loadings can be assumed different from historical conditions. I recommend to include such an assessment in the report. Here, also a comparison to the targets derived from other groups (e.g. BLANO, HELCOM, ..) – if not given in chapter 7 – is missing. In approaches developed in Germany the conditions around 1900 are used as the good-moderate threshold, as already some impact of human activities could be shown. His difference should also be addressed in the report.

- The different chapter are written by different author groups. This results in different quality but also different depth of the analyses. Further, not all results seem to be fully harmonized between the chapters. Where obvious I addressed apparent inconsistencies in the pdf and in the following detailed remarks. Beyond that, a general revision of language-style and grammar (in particular for chapter 4) is recommended to make the report a more homogeneous piece of work.

- The report provides a full range of information on conditions around 1900, unfortunately I have the impression that the considered elements and derived nutrient balance (9 mg/l in root zone * 80-90% retention = 0.9-1.8 mg /l to the sea) seems to be over simplified. What about surface runoff and tile drainages (for which no retention gw can be assumed). I also doubt that zero emissions from 2.5 mill. inhabitants can be assumed. In chapter 6 all tools are available to model N fluxes at 1900 including all described driving parameters together. Unfortunately, such results have not been derived or are not shown. Without this, the main goal of estimating N concentrations to the sea around 1900 seems to be incomplete. I did not follow the numbers and calculations in that depth, but I am not sure if Nitrate and TN is always clearly distinguished between the chapters and for the conclusions.

Concluding, I think the report is a decent piece of work and delivers valuable information for the process to derive reductions goals for marine systems. When reading the report I tried to be critical and addressed all open issues apparent to me. However, some of the comments and mentioned short comings might only be relevant for a second phase, for which I recommend to directly include other parameters such a phosphorus, light or water temperature.

ESTIMATION OF NITROGEN CONCENTRATIONS FROM ROOT ZONE TO MARINE AREAS AROUND THE YEAR 1900

Factors affecting the nitrogen concentration from root zone to the coast around the year 1900 have been investigated. The aim of this investigation is to establish a nitrogen concentration in the water discharged into marine areas around the year 1900. Besides this, the national and international literature have been consulted to find references to the nitrogen concentration around the year 1900. The best estimate of the nitrogen concentration back in 1900 is within the range of 1-2 mg N/l.

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