Estimations of background N loads to rivers and coastal waters in Denmark for defining reference conditions – a review

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Executive Summary

This report provides a review of the estimations of nitrogen loads from Danish territory to Danish coastal waters around 1900. These estimations aim at supporting the setting of reference conditions for Biological Quality Elements (BQE) as demanded by the Water Framework Directive (2000/60/EC).

Based on long records of the depth-distribution of eelgrass in the Danish coastal waters and on the assumption that human impacts on the coastal waters were minor, it has been suggested that a reference condition existed for the BQE eelgrass in the Danish coastal areas around 1900. Next, it was inferred that the nitrogen input from the terrestrial area in Denmark to the coastal waters around the year 1900 would provide the reference nitrogen load to the coastal waters. Nitrogen is essential for all life on earth, but excess nitrogen in coastal waters stimulates phytoplankton growth, which decreases the transparency of the water and thereby suppresses eelgrass. Nitrogen inputs to Danish coastal waters mainly originate from agriculture through nitrate leaching, but a part originates from households and industry through discharges, and from atmospheric deposition. In addition, there is influx of nitrogen to the Baltic Sea from surrounding countries.

Danish researchers have explored three approaches to estimate reference nitrogen loads from Danish territory to Danish coastal waters (i) measurements of current nitrogen loads in small watercourses, which drain catchment areas with a very small level of cultivation, (ii) estimations of nitrogen loads by rivers to coastal waters around 1900 through a synthetic integrated modelling analysis using historic data, and (iii) based on an extrapolation of the relationship between calculated nitrogen surpluses and measured nitrogen loads in rivers during the last few decades. The review has identified strength and weaknesses and pros and cons of the three approaches. All three approaches have advantages and disadvantages, but these seem insufficiently articulated and examined thus far. All three approaches may be improved, which may increase the consistency in the approaches and the confidence in the estimations ultimately.

The reference nitrogen loads estimated via the three approaches differ by a factor of 2 to 3. This is a relatively small range when considering the large differences in approaches and the large uncertainties involved. No find clear and convincing scientific arguments have been found in the Danish studies to prefer one of the approaches above the other.

1. Introduction

The EU Water Framework Directive (WFD) stipulates that EU Member States define types of water bodies (fresh waters, marine coastal waters) and baseline conditions (or Reference Conditions; RCs) for the ecological, physical-chemical and hydrological status of these types of water bodies. Further, the WFD demands that all surface waters (e.g. lakes, rivers, coastal waters) should achieve at least good ecological status (GES) and good chemical status (GCS).

The ecological status of a water body expresses the quality, structure and functioning of an aquatic ecosystem; for coastal waters this encompasses the composition, abundance and biomass of phytoplankton, the composition and abundance of other flora (macroalgae and angiosperms) and the composition and abundance of benthic invertebrate fauna. The ecological status is classified into five classes (High, Good, Moderate, Poor, and Bad), which are defined relative to RCs, using Ecological Quality Ratio (EQR) values. The WFD demands a Good Ecological Status (GES) and a Good Chemical Status (GCS); measures have to be taken when surface waters have a status less than GES and GCS.

Many water managers and scientists have been struggling to transform the normative definitions of the WFD into quantitative goals and operational measures, from the time of adoption of the WFD by EU Member States in 2000 (Carvalho et al., 2018; Skarbøvik et al., 2020). One of the main challenges is to establish solid RCs, also because RCs serve different purposes and may have large impacts. Too high RCs and boundaries between Good and Moderate quality for, for example, nutrients may result in enhanced eutrophication and risks of harmful algal blooms, whereas too strict RCs and boundaries may cause implementation of impossible/unnecessary nutrient reduction measures with significant economic consequences.

Denmark is in the process of defining/revising RCs and reference nutrient loads for coastal waters. Two main approaches have been suggested for deriving reference nutrient inputs to coastal waters for the Water Plans (Timmermans 2020), i.e., (i) based on nutrient concentrations in smaller watercourses, which drain catchment areas with a very small level of cultivation multiplied by a present-day waterflow, and (ii) based on the nutrient loading to rivers and coastal waters around 1900 when human influences on environmental conditions were thought to be minor, as reflected in the depth distribution of eelgrass in Danish coastal waters (Krause-Jensen et al., 2005).

The objectives of this study[1] are to (i) Analyse the approaches used by Denmark to define reference conditions; (ii) Evaluate the indicators chosen for assessment of the status of the biological quality elements (BQE); and (iii) Examine the adequacy[2] and consistency[3] of the measures proposed to enable the coastal systems to meet the reference condition for eelgrass, in particular related to reference nitrogen loads. This report provides a review of the estimations of nitrogen loads to coastal waters around 1900. It accompanies reports by Ferreira (2021), who addresses the 2nd objective and Van Calster and Garnett (2021) who address the 1st and 3rd objectives.

[1] At the request of SEGES, a private non-profit advisory, test and research association (https://www.seges.dk/).
[2] Adequacy of measures means they will succeed in meeting the stated objectives for a desired ecological status.
[3] Consistency of measures means that robust criteria are used in their definition, in the context of the objectives the measures are designed to fulfil.

2. Establishing reference conditions according to the Water Framework Directive

Guidance document 5 of the Common Implementation Strategy for the WFD[4] explains the concept of biological reference conditions, and the hydro-morphological and physio-chemical elements supporting the biological elements, and it explains how to use these concepts in practice: *"The reference condition is a description of the biological quality elements that exist, or would exist, at high status. That is, with no, or very minor disturbance from human activities. The objective of setting reference condition standards is to enable the assessment of ecological quality against these standards. In defining biological reference conditions, criteria for the physical-chemical and hydro-morphological quality elements at high status must also be established." (Section 4; page 36)*.

Hence, the reference condition is a description of the biological quality elements only, while physical-chemical and hydro-morphological quality elements are supporting the biological reference conditions. Reference conditions have to be determined for each type of water body. The definition of RCs leaves some space for interpretation, especially in terms of 'biological quality elements that exist or would exist', and 'conditions with no or very minor disturbances from human activities'.

The WFD identifies four options for deriving reference conditions. Guidance document 5[4] suggests to use "*a hierarchical approach in the following order* (Section 4.5; page 42):
1. An existing undisturbed site or a site with only very minor disturbance; or
2. Historical data and information; or
3. Models; or
4. Expert judgement.
Hence, different methods may be used for defining RCs, but with validation checks: "*Where a site with 'very minor disturbance' is used to derive reference conditions it should be validated to ensure that it meets the definitions of high status".* And *'It may be possible to use historical information to derive reference conditions if the historical data are of assured quality. If reference conditions are derived from historical conditions, these should be based upon the condition of water bodies at times of no or very minor anthropogenic influence* (Section 4.5; page 42).

Variability in RCs have to be considered as well: '*Reference conditions must summarise the range of possibilities and values for the biological quality elements over periods of time and across the geographical extent of the type. The reference conditions represent part of nature's continuum and must reflect natural variability* (section 4.2; Page 37). Further, "*It is likely that the natural variability of a quality element within a type may be as large as the natural variability between types".* Further, '*Reference conditions are not permanent. Climate, land cover and marine ecosystems vary naturally over many periods relevant to the WFD. Every six years from 2013, Member States must review the characterisation of water bodies, including reference conditions. It is accepted that many of these variables are not fully understood in the marine environment' (section 4.9; page 46).*

[4] EC, 2003 Common Implementation Strategy for the Water Framework Directive (2000/60/EC) Guidance Document No 5. Transitional and Coastal Waters – Typology, Reference Conditions and Classification Systems.

3. Deriving reference N inputs from undisturbed sites and sites with minor disturbance

Timmermann (2020) recently argued that for Water plan 3 '*the reference nitrogen input to coastal waters in Denmark has to be based on concentrations in smaller watercourses, which drain catchment areas with a very small level of cultivation multiplied by a present-day waterflow'.*

This approach has been used also for Water Plan 2 and has been published in English language by Kronvang et al (2015)[5]. In this approach the flow-weighted nitrate (NO3), ammonium (NH4) and total organic nitrogen (TON) concentrations have been determined in 19 streams in catchments with less than 10% of the area in agricultural land in Denmark. These streams were sampled once per season (hence four times per year) in one year and combined with measurements of the 'instantaneous discharge' during 2004-2005 (snapshot sampling). The results of the measured 19 streams were linked to the six main landscape types in Denmark, and mean annual total N concentrations in the runoff were calculated for these six landscape types.

Authors found that the mean flow-weighted concentrations of NH4 (0.05±0.01 mg N/l) and TON (0.53±0.07 mgN /l) were similar in all landscape types, but that mean NO3 concentrations differed between the landscape types, mainly due to differences in soil types (range: 0.06 to 0.83 mg N/l)[6]. The mean total N concentrations (NO3+NH4+TON) per landscape type (range 0.6-1.4 mg N/l) were upscaled to the whole territory of Denmark using a 5 × 5 km grid; for each grid cell a flow-weighted mean total N concentrations was assigned according to the dominant (>50%) landscape type. The 5 × 5 km grid map of flow-weighted mean annual total N concentrations in surface water bodies was used to calculate the 'background' total N loading to coastal waters in Denmark, using a national-scale hydrological-biogeochemical model (Windolf et al. 2011). The average annual background N load to streams amounted to 16,000 tons N per year during the period 2004–2005. However, just around 13,000 tons N per year ended up in the coastal waters, because of N retention in surface freshwaters (streams, lakes and wetlands).

Kronvang et al (2015) report that the aforementioned 19 streams included five streams with long-term monitoring data (1990-2011). Concentrations of total N in these streams decreased significantly over time in four out of the five streams; this decrease (0.029–0.061 mg N per liter per year) was ascribed to the decrease in atmospheric N deposition (31%) during the monitoring period. The decrease in mean total N concentrations for all five streams was 24 ± 11% during the period 1990–2011.

Authors indicate that the mean total N concentrations in streams draining natural catchments in Denmark agree reasonably well with reported total N concentrations in stream draining natural catchments elsewhere in the world.

[5] Based on 'Bøgestrand et al. (2014) Baggrundsbelastning med total N og nitrat-N, Notat fra DCE-Nationalt Center for Miljø og Energi'
[6] Are the relatively high mean N concentrations in catchments with loam/clay soils (compared to the N concentrations in catchments with sandy soils) related to differences in N inputs? Or are they related to shorter traveling times, less N retention, and less downward leaching?

Kronvang et al (2015) list two possible shortcomings in their study for using mean N concentrations in streams draining small and little disturbed catchments for estimating 'background' N loads from Denmark to coastal waters. First, the measured N concentrations may incorrectly reflect 'background' N concentrations due to the possible impact of farming in the catchments (i.e., some catchments had up to 10% of the area in agriculture). Second, the possibly incorrect assumption that N concentrations in small streams are similar to those of large rivers. Authors argue that catchment with large rivers have more lakes and wetland and hence have greater retention of NO3 (through denitrification) and therefore possibly lower mean N concentrations than small catchments with small streams.

A few questions pop-up, when using the results of the study of Kronvang et al (2015) for estimating the reference nitrogen input to coastal waters in Denmark.

First, the WFD demands that water type-specific biological reference conditions have to be established for biological quality elements and, subsequently, that water type-specific hydro-morphological and physico-chemical reference conditions have to be established for the hydro-morphological and physico chemical elements (Chapter 2 of this report). The hydromorphological and physico-chemical elements support the biological quality elements. The question here is 'to which biological quality elements are the estimated mean total N concentrations in streams and total N loading to coastal waters related'? Are the mean total N concentrations in streams and total N loading to coastal waters determined in 2004-2005 meant to be 'supportive' to the eelgrass vegetation cover in 1900 and associated phytoplankton concentrations? The link between the biological quality elements and the physico-chemical and hydro-morphological quality elements is unclear in Kronvang et al (2015).

Second, the WFD indicates that RCs have to be determined for each type of water body. To which water body types do the background N loading relate? Are the five landscape types distinguished by Kronvang et al linked to different water body types?

Third, great care have been taken to sample streams in undisturbed catchments or in catchments with only very minor anthropogenic disturbances. This has been interpreted as catchments with less than 10% of the area in agriculture (the inclusion of up to 10% agricultural land was identified by the authors as a first possible shortcoming of the study). The question remains whether the examined catchments are representative for the impact that an 'undisturbed' Danish terrestrial biosphere has on the chosen (targeted) biological reference conditions of a specific water type? Do the 19 catchments represent 'the mean undisturbed or minimally disturbed' Danish terrestrial biosphere? This question was only indirectly and implicitly addressed during the upscaling procedure – each of the 19 catchments were linked to the 5 distinguished geomorphological landscape types. But are the 3 to 5 streams/catchments representative for the 5 landscape types? Indeed, Kronvang et al (2015) argued that the chosen catchments (mean area 3.9 km^2; range 0.05 to 15.7 km^2) may not be representative, because they are much smaller than most catchments, and have less lakes and wetlands. Further, it may not be excluded that these small natural catchments are 'leftovers' in the landscape after people occupied the Danish territory. Early settlers likely used the better land and left the poor land aside (left to nature) following the occupation of the land. It is well-documented that almost all early settlements started in river deltas, and on fertile land. Thus, the question is whether the smaller catchments have low natural fertility and/or low potential for agricultural production than the land that has been utilized for agriculture and infrastructure? Are the 19 catchments representative for the territory of Denmark as regards the background N loading to the different water types of the coastal waters?

Fourth, what is the scientific underpinning for the 10% cut-off value for agricultural land? Is there empirical evidence that catchments with 10% agricultural land do not differ statistically significant from catchments with 0 or 5% of agricultural land. And do these catchments differ statistically significant from catchments with 15% or 20% of agricultural land?

Fifth, the 19 catchments were sampled only four times in one year (5 catchments were monitored intensively for decades). Do the averages of the four samples provide a robust estimate of the mean N concentration in a year and is this mean representative for the long-term average? Authors do not address this question. The long-term monitoring results of the 5 catchments show indeed large interannual variations (seasonal variations were not shown).

Sixth, where does the N in streams of natural catchments come from? This question is also not addressed, although authors relate the decrease in mean total N concentrations in the long-term monitoring catchments to decreases in atmospheric N deposition, following governmental measures aimed at decreasing ammonia (NH_3) emissions from agriculture and nitrogen oxide (NO_X) emissions from combustion sources. Total N concentrations decreased by on average 24% during 1990-2003, while atmospheric deposition decreased by 31% (according Kronvang et al., 2015), and by 23% according to Andersen et al (2006) during the period 1990-2004. Mean atmospheric deposition was 16 kg N per ha per yr in 2004, which suggest that the mean N deposition was (16 x 1.23 =) 19.7 kg per ha per yr in 1990. Hence a decrease in N input of approximately 4 kg per ha during the period 1990-2004. Using a similar 'back-of-the-envelop' calculation for the decrease in mean N concentration in natural catchments suggests that the overall mean N concentration has decreased by ~0.2 mg N per liter during the period 1990-2004 (estimated from a decrease of 24% and an overall mean N concentration of ~1 mg/l). Hence, a decrease in mean N input of 4 kg would lead to a decrease in N concentration in the runoff (stream flow) of ~0.2 mg per l[7]. Is that feasible/reasonable?

What are other possible N inputs? Biological N_2 fixation (BNF) is commonly a dominant N input in natural systems. Total N inputs via BNF are unknown for the 19 catchments, but possibly range between 1 and 50 kg per ha, depending also on the fertility of the sites and the natural vegetation (Vitousek et al., 2003). Another possibly (temporary) source of inorganic N is the soil itself, through net mineralization. This is also a highly uncertain source, but it cannot be excluded that some of the annual variations in total N content in leachates is related to temporal variations in net

[7] A mean decrease in N concentration of ~0.2 mg per liter translates to a mean decrease in N leaching loss of ~0.66 kg per ha (assuming a mean runoff of 328 mm per year according Anderson et al., 2006), which is ~16% of the decrease in atmospheric N deposition. Evidently, this holds only under the assumption of linearity and steady state conditions. In 2004-2005, total N deposition was on average 16 kg per ha (Anderson et al., 2006); if this deposition decreases further, and the assumed linear relationships between atmospheric N deposition and mean N concentration of stream waters hold, mean N concentration of the stream waters will be ~0 mg/l when atmospheric N deposition in a hypothetic case would decrease to 0 kg per ha.

mineralization of soil organic N (in the range of +50 to -50 kg per ha per year). A bit more quantitative insight in the possible sources of the N in the streams of natural catchments would provide more confidence in the appropriateness of using these streams for estimating background N loads.

In conclusion, there are a number of uncertainties related to the derivation of background N loads to coastal waters in the study of Kronvang et al (2015). These uncertainties question the confidence in the estimated background N concentrations and background N loading. Further, it is insufficiently clear that the estimated RCs for the N loading and those for the related biological quality elements have been derived in a consistent manner.

4. Deriving reference N inputs from historic data around the year 1900

Based on long records of the depth-distribution of eelgrass in the Danish coastal waters[8] and on the assumption that human impacts on the coastal waters were minor around 1900, it has been suggested that a reference condition existed for the biological quality element eelgrass in the Danish coastal areas around 1900. Based on this water quality status, it was inferred that the nitrogen (N) input from the terrestrial area in Denmark to the coastal waters around the year 1900 would provide the reference N load to the coastal waters. These thoughts have been the basis of a collection of studies published in Jensen (2017)[9] about the estimation of the net N load from the terrestrial areas to the coastal waters around 1900.

Agriculture, atmospheric deposition, and urban sites (households) were considered to be N sources for N leaching to the coastal waters in the studies (Jensen, 2017). Subsoils, wetlands and surface waters were considered to act as sinks of N (through denitrification processes), and climate (excess rainfall; runoff) the driving force for moving N from the sources via the sinks to the coastal waters.

In the synthesis chapter (Chapter 7), authors conclude that the runoff from the terrestrial areas to the coastal areas in the year 1900 contained on average 1-2 mg of N per liter. The range reflect the uncertainty in the estimate; the lower end of the range is representative for sandy areas (Jutland) and the higher end of the range is more representative for the clayey soil areas (Zealand). The estimated range concords with various other estimates presented in Chapter 7. The range is an estimate for the whole territory of Denmark that drains into the coastal areas. Authors indicate that further studies are needed to differentiate between geographical areas (catchments) in N concentrations in the runoff from land to coastal areas around the year 1900. Geographic differences in N concentration in the runoff likely result from differences in soils, landscapes, farming systems and drainage.

[8] Krause-Jensen et al. (2005) Eelgrass as a Bioindicator Under the European Water Framework Directive. Water Resources Management, 19:63-75.
[9] Jensen, P.N. (Ed.) 2017. Estimation of Nitrogen Concentrations from root zone to marine areas around the year 1900. Aarhus University, DCE – Danish Centre for Environment and Energy, 126 pp. Scientific Report from DCE – Danish Centre for Environment and Energy No. 241. http://dce2.au.dk/pub/SR241.pdf.

How to assess the methods and approaches used to obtain the overall mean estimate of 1-2 mg N per liter in the runoff from the terrestrial area to the coastal waters in 1900? And how to assess the appropriateness of this estimate as a RC? Below, I tried to answer these questions, first by reviewing the report and second by reflecting on the criteria mentioned in the report, WFD and the WFD CIS Guidance Document No. 5 (see footnote 4).

Chapter 1 presents an analysis of the climate and hydrology in Denmark around 1900, with a focus on runoff. Based on data from 18 stream hydrometric stations with at least 80 years' of runoff data and five climate stations, authors conclude that the overall mean annual runoff in 1900 was 24% lower than to-day. They estimate that the average total runoff from Denmark in year 1900 was 256 mm. Annual variations in runoff were very large (range ~100 to 400 mm). Further, winter temperature was lower in 1900 than it is to-day. These results have implications for the estimation of N concentrations in the runoff to the coastal waters in 1900 but also for the hydro-morphological conditions in the coastal waters, especially when the overall mean annual runoff in 1900 was also lower in other countries surrounding the Baltic Sea.

Chapter 2 reports on the estimation of the N concentration in the runoff from agriculture around 1900. The N concentration in the leachate below the root zone of agricultural land was estimated at on average 12 mg/l (range 5 to 15 mg/l). This estimate is based on measured N concentrations in the leachate from below the root zone in current (organic) farming systems with a management that was supposed to be relevant for the situation around 1900. Authors argue that farm-gate N balances and field N balances of agriculture in and around 1900 cannot be used to estimate N leaching losses from the root zone, because the relationship between measured N leaching losses and farm-gate N balances established during the last few decades[10] do not hold for the situation around 1900. However, authors use results from field experiments carried out during last few decades (but with assumed farm management relevant for the period around 1900) to derive N leaching losses from agriculture around 1900. This way of thinking seems not consistent. Are farm management and crop types and rotations and N inputs in current organic farming field experiments similar to those around 1900? And are current soils and climate also similar to the situation around 1900?

The average N leaching loss from agricultural land in 1900 would have been about 30 kg per ha per year, if the average N concentration in the leachate below the root zone was 12 mg per liter, and the average runoff was 250 mm per year (Chapter 1). This leaching loss seems at the upper end of what would be possible, because the estimated average farm-gate N surplus ranged between 20 and 40 kg per ha per year in Denmark during the period 1900-1920 (Figure 4; Kyllingsbæk, 2008). This would suggest that the whole farm-gate N surplus was lost via nitrate leaching, and that there were no denitrification losses in the root zone (or that denitrification losses were equal to net soil N mineralization). This seems unlikely. Further, reported N balances of organic mixed farms during the last few decades are a factor of 2 to 4 higher than the reported mean N balance for Denmark around 1900 (Hallberg et al., 1995; 2010; Knudsen et al.,2006; Kristensen and Hermansen, 2008), suggesting that the current organic farms are different from the farming systems in Denmark around 1900.

[10] This approach was used by Conley et al (2007).

Chapter 3 presents an analysis of the atmospheric N deposition around 1900. Authors argue that the early records of wet atmospheric N deposition are likely biased (due to sampling at few sites only and due to possible contamination of samples) and therefore they use estimates from model calculations and compare these with other modelling studies. They arrive at mean N inputs of 3 kg per ha per year for the coastal areas and of 4 kg per ha per year for the terrestrial system around 1900, with estimated uncertainty of 'at least a factor 2 of the average estimate'. Both, the used approaches and the estimates seem 'reasonable' for the purpose of the study.

Chapter 4 presents a brief qualitative review of point sources around year 1900. A point source is defined as a single identifiable source of water, including wastewater treatment plants (mostly treating urban residential wastewater), industrial wastewater, fish farms and manure heaps and liquid manure containers. Authors conclude that there is (i) very little quantitative information about N losses from point sources, and (ii) that the total N losses from point sources were likely small, because Denmark had a 'circular economy' at that time (as regards recycling urban waste). Hence, no attempt was made to estimate N losses from point sources quantitatively. This is somewhat disappointing[11]. Locally and regionally, N losses from point sources may have been significant, especially around settlements and farms with livestock. For example, Kristensen et al (2015)[12] estimated a mean runoff loss from dairy manure storage of 6 kg N per ha per year in 1920. It cannot be excluded that runoff losses from pig manure in barns and storages were also significant at that time. In conclusion, runoff N losses to rivers and coastal waters around 1900 losses may have been significant, at least regionally.

Chapter 5 presents an historic overview of nitrogen concentrations in streams and rivers, using data bases and simulation models. The earliest empirical records are from snapshot sampling in six major Danish rivers, upstream major towns, in 1889 and 1892. Authors conclude that these results are not very reliable because of possible sampling and measurement artefacts. Solid time-series of N concentration measurements in streams and rivers started in the 1960s. Trends in N concentrations in streams between 1960 and 2015 followed more or less the same trend as the total N surplus of Danish agriculture, but with large delays for some streams. Authors then used empirical (statistical) models for the relationships between flow-weighted N concentrations and N surpluses in agriculture for two periods to estimate N concentrations in streams around the year 1900. They arrive at a mean estimate of about 2 mg N/L. Authors argue that these estimates are uncertain, but these estimates fall in the range of reported N concentrations in streams around 1900.

Chapter 6 presents an analysis of nitrogen retention in the landscape around year 1900, using historical data and information and simulation models. Authors do not explicitly define what they mean by 'N retention', but this reviewer understands that N retention is here equivalent to N losses via gaseous N emissions (NO, N_2O, N_2, NH_3) following (de)nitrification and NH_3 volatilization, although N retention through immobilization, fixation and sedimentation may possibly not be excluded. Further, authors do not define the system boundaries for the retention; is the bottom end

[11] Surprisingly, quantitative N loads from point sources were included in the modelling study in Chapter 6 of Jensen (2017)
[12] Troels Kristensen, OleAaes, Martin Riis Weisbjerg (2015) Production and environmental impact of dairy cattle production in Denmark 1900–2010. LivestockScience 178, 306–312. http://dx.doi.org/10.1016/j.livsci.2015.06.012.

of the root zone, as defined in Chapter 2 of Jensen (2017) a boundary, or is the N retention in the root zone included in the estimations? And is the coastline another boundary; is the N retention in streams and surface waters included up to the coastline with the sea? Hence, there are a number of uncertainties related to the estimation of N retention.

Estimating N retention in subsoils, wetlands and streams is probably the most complicated effort and yields probably the most uncertain outcome of all studies compiled in this Jensen (2017) report. Authors include the effects of a wide range of land and land use changes in the model simulation, including meadow irrigation. This reviewer was amazed by the huge amount of irrigation water used (150000 m^3/ha/year, which would translate to a water height of 15 m per year......). Authors conclude that ”The N retention in surface waters[13] 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”. Further, authors conclude that the overall mean N retention in groundwater around the year 1900 was higher than at present (current estimation is 62%), but authors do not explicitly indicate by how much. For readers, it is difficult to find out how the borders are defined between surface waters and groundwaters. Authors state that ‘the calculated N retention around 1900 is expected to be a maximum (“best case”) estimate’, but it remains unclear what this estimate is and why this is a ‘best case estimate’.

<u>Chapter 7</u> presents an overall synthesis. Authors state at the end of section 7.1 that “*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*”. This emphasis is much narrower than stated in the foreword of the report, and conflicts somewhat with the notion of RCs, as defined in the WFD, and hence with the scope of the study.

For agricultural land covering 75% of the total land area, N concentration in the leachate from the root zone was estimated at 12 mg/l (based on the analysis presented in chapter 2), and for natural areas covering 25% of the total land area, the N concentration in the leachate from the root zone was set at 1 mg/l (I did not find an underpinning for this estimate). For N retention, a distinction was made between retention in groundwater bodies (62%) and retention in wetlands and surface waters (37-65%). Overall mean N retention was then estimated to range between 76 to 87% (based on chapter 6, where I did not find these numbers) and was reported to be a minimum estimate.

The effect of atmospheric N deposition was assumed to be included in the estimated N concentration of the leachate, and the influence of point sources was assumed to be negligible (which may not be true as argued before). The overall mean N concentration in streams discharging to the sea (1-2 mg/l), as derived in Chapter 7, was compared with various other (literature) estimates, and was found to be in the similar range or at the higher end of the reported values (in Table 7.1). Finally, authors argue that ‘a fully integrated regional model analysis may provide a better historical view on the effects of humans on N pollution of groundwater and surface waters, and may assist in setting geographically differentiated reference targets’.

[13] Not sure this includes wetlands, and the soil layer between the bottom of the root zone and the surface of the groundwater bodies (acquifers).

Chapter 7 does not present a number for the total N load to the coastal waters around 1900. However, this load may be estimated on the basis of the data presented in the report and a 'back-of-the-envelop' calculation. The terrestrial area of Denmark is 42,000 km^2 (rounded off value). The mean runoff was 256 mm rainwater in 1900 (Chapter 1). The overall mean N concentration in streams discharging to the sea was 1 to 2 mg/l (Chapter 7). Hence, total N load to the coastal waters ranged between 10,8 to 21,6 kton per year around 1900.

Two presentations related to the estimations of the reference N loading to coastal waters in 1900 were given at the recent Plantekongres in Herning (14-15 January 2020), by Anker Lajer Højberg (from GEUS) and by Jørgen Eriksen et al (from Aarhus University). The central message of these powerpoint presentations was similar as the central message of the aforementioned report (Jensen, 2017). Large areas of Denmark were used for agriculture and human influence on N cycling and N loadings to streams, rivers and coastal areas were likely noticeable in runoff of N. Højberg concludes that the recently updated N losses were larger than earlier estimates; they arrived at an overall mean N concentration in streams of 2.5 mg/l, likely because point source losses were now included. This would mean that the total N load to the coastal waters was 27 kton per year around 1900. Unfortunately, the report and the underlying data and analyses are as yet not available – the report is under external review (till about May 2021).

Two additional papers became available recently[14]; these provide additional data and views. Brudler et al (2020) made a detailed assessment of point sources (humans, animals in towns, and industry) around 1900. They estimated that the N loading to surface waters at 2.5 kton and the P loading at 0.46 kton per year around 1900. They indicated that large parts of the population migrated to towns in the second half of the 19th century. By 1901, 39% of the Danish population lived in towns. Emissions from industries were largely unregulated, and wastewater treatment did not exist. They conclude that nutrient emissions from human activities (point sources) were already significant in 1900 and that many surface waters were 'brown and foul-smelling of diseases'.

Christensen et al (manuscript accepted for publication) refined/updated the estimation of the N leaching from the rootzone around 1900 (in chapter 2 of Jensen (2017), using historical data related to land use and N leaching estimates from current land use, but 'relevant for the situation around 1900'. They estimated a mean N concentration in the leachate from the rootzone of agricultural land of 12 mg per liter. This estimate is similar to the estimate reported in chapter 2 of Jensen (2017). The mean N concentration of the leachate from the whole Danish territory was estimated at 9.6 mg per liter. Authors do not estimate the N retention in the landscape and do not provide an estimate of the total N loading to the coastal waters. Authors conclude that agricultural activities were significant around 1900 with a substantial potential for N losses to the environment. Authors argue that N balances are unsuitable for providing insight in the potential N leaching loss, because these balances do not consider changes in net mineralisation of soil organic N in the crop rotations prevailing around 1900. They suggest a significant net mineralisation of soil organic N (as a source of N leaching) during the second half of the 19th century due to land use change, frequent ploughing and harrowing for weed control, and ploughing down grassland in rotation.

[14] Kindly provided by prof Jørgen Eriksen from Aarhus University upon request via email on 05-02-2021

Summarizing, there are no empirical data for estimating the background N load from Denmark to coastal waters around 1900 directly. Hence, the background N load around 1900 has to be inferred indirectly, using model calculations and assumptions. The approaches presented in Jensen (2017) and Christensen (accepted) seem logical from a scientific point of view, but there are a number of uncertainties in the estimations and assumptions, which question the confidence in the estimated background N loading. Most uncertainties relate to the assumptions that (i) N leaching from current organic farming systems (as examined in field trials) is representative for the mean N leaching from farming in Denmark in 1900, and (ii) N retention in the landscape in 1900 can be estimated from a reconstruction of the 1900 landscape and model calculations. The estimated overall mean N concentration in streams discharging to the coastal waters in 1900 (1-2 mg/l) seems reasonable. This estimate was recently adjusted by Højberg and Eriksen (2020) in upward direction, likely because of the updated N leaching from agricultural land by Christensen et al (accepted) and the inclusion of point sources in the calculations, as reported by Brudler et al (2020).

5. Discussion

5.1. Estimated N loading to Danish coastal waters in 1900

The estimated mean N loadings from the territory of Denmark to the coastal waters around 1900 are remarkably similar (Table 1), when considering the large differences in approaches and the large uncertainties in the assumptions. Differences between estimates are within a factor of 2 to 3.

Table 1. Summary overview of estimated mean annual total N (Nt) concentrations in rivers draining into coastal waters, and the annual background N loadings from the main territory of Denmark to coastal waters around 1900.

Source	Mean Nt concentration, mg/l*)	Mean annual N load, kton (million kg)
Kronvang et al (2015)	0.6 - 1.4	13**)
Jensen (2017)	1 - 2	10.8 to 21.6
Højberg and Eriksen (2020)	2.5	27
Conley et al (2007)	2.3 - 2.8	25-30
Erichsen & Timmermann (2020)	1.5	16***)

*) Kronvang et al (2015) included nitrate (NO3), ammonium (NH4) and total organic nitrogen (TON) in Nt, while Jensen (2017), Højberg and Eriksen (2020) and Conley et al (2007) included only nitrate (NO3) in Nt.
**) Kronvang et al (2015) estimated the total N load to streams at 16 kton in 2004‑2005. Due to N retention in fresh surface waters, the total N load to coastal waters was corrected to 13 kton.
***) Erichsen & Timmermann (2020) used the uncorrected (for N retention in surface waters) estimate of Kronvang et al (2015).

Kronvang et al (2015) and Erichsen and Timmermann (2020) estimated background N loadings from natural catchments in 2004-2005. It is not directly clear which biological quality elements these background N loads support.

Conley et al (2007), Jensen (2017), and Højberg and Eriksen (2020) all estimated the N loading to the coastal waters around 1900, with the aim to establish the physico-chemical reference conditions that support the biological quality elements (eelgrass) for the conditions around 1900. Hence, the N loads are linked to the eelgrass distribution around 1900.

Conley et al (2007) estimated total N inputs to the Danish Straits around 1900 at about 25-30 kton (see Fig. 6 in Conley et al 2007), based on N input-output balance (N surplus) calculations and the relationship between calculated N surpluses and measured/calculated N concentrations in runoff during the period 1990-2003. More than half of the total input from the Danish territory to coastal waters originated from point sources and less than half from diffuse sources (agricultural land). The large proportion of point sources contrasts with the conclusion that point sources were negligible around 1990 according to Jensen (2017).

A total N input of 10 to 30 kton per year (maximum range) translates to a net N leaching loss of 3.7 to 11.2 kg per ha per year, when the total N loading would have to come from agricultural land, or translates to a net N leaching loss of 2.4 to 7.1 kg per ha per year when the whole Danish territory is considered. Taking into account N retention in the landscape (76-87%; Jensen, 2017), the actual leaching loss from the root zone would be about 5 times higher. Hence, the mean leaching loss from the rootzone of agricultural land only would range from 18 to 56 kg per ha per year, and the mean leaching loss from the rootzone of the whole territory would range from 12 to 36 kg per ha per year.

How do these numbers compare with the farm-gate N balance around 1900? Total N surplus in agriculture was about 75 kton and the farm-gate N surplus was 25.8 kg per ha in 1900 (Table 2). The fate of this N surplus is unclear, but some will be lost to the atmosphere via NH3 volatilization, another part may be lost via leaching and some may be lost via denitrification (Kyllingsbaek, 2008). In addition, some may be accumulated in the soil (temporarily) as mineral N or more likely as organically bound N. Conversely, there may have been also net mineralization of organically bound N to mineral N (Christensen et al (accepted). Kyllingsbaek (2008) assumed that the amount of soil N remained constant, i.e., the soil was not a sink or source of mineral N.

Table 2. Farm-gate N input-output balance for Denmark around 1900 (Kyllingsbaek, 2008)

Inputs, kg/ha/yr		Outputs, kg/ha/yr	
Synthetic fertilizers	0.3	Harvested crop produce	2.9
Waste	0.8	Harvested animal produce	9.5
Atmospheric deposition	3.8		
Biological N fixation	21.1		
Feed import	12.2	Surplus	25.8
Total	38.2	Total	38.2

The N balance indicates that Denmark had a low N input agriculture in 1900 (Table 2). Most of the N input came from biological N_2 fixation. Feed import provided also a significant N input. Figure 4 in Kyllingsbaek (2008) indicate that the N surplus fluctuated between 20 and 40 kton during the first 20 years of the 20th century, indicating that the net N input did not increase much during these years.

5.2. Which approach is most appropriate for estimating the N load to coastal waters?

Thus far, no convincing scientific arguments have been presented in the reports to be able to choose one approach above the other, for estimating background N loads. A main point here is also that the background N loads have to support the established biological reference conditions, but it is not always clear which biological quality elements, and linked to the elements, which biological reference conditions have been chosen.

All three approaches have advantages and disadvantages. The basis of the approach used by Kronvang et al (2015) is the 'minimal human activity' in the catchments, so as to comply with one of the criteria for deriving RCs according to the WFD. The main disadvantage is the uncertainty about the representativeness of the chosen catchments for the territory of Denmark, and the unclarity (inconsistency) of the relationship between the chosen biological reference conditions around 1900 and background N load estimations in selected catchments in the 2000s. The basis of the Jensen (2017) approach and the Højberg and Eriksen (2020) approach is the consistency in the derivation of RCs, i.e., they try to estimate the reference N loading for the biological reference conditions for eelgrass in the coastal waters around 1900. The main disadvantages of their approach is the assumption that leaching data from current organic farming field trials can be used for estimating N leaching from the root zone in 1900, and the need for sophisticated models that yield uncertain estimates of the N retention in the landscape. The basis of the approach used by Conley et al (2007) is also the reconstruction of the situation in 1900 and the use of a transparent and simple derivation of leaching loss. The main disadvantage of their approach is the uncertainty related to the extrapolation of the relationship between changes in the N surplus of Denmark and changes in the N concentration in the runoff of streams and rivers to coastal waters.

Jensen (2017) does not address explicitly the question whether the estimated runoff N loss to the coastal waters is appropriate for defining the reference N load, and whether their approach complies with the concept of reference conditions as outlined in the WFD and in Guidance document 5 of the Common Implementation Strategy for the WFD. Timmermann (2020) explicitly answers this question by 'no', because '*the nutrient input to the sea cannot reasonably be assumed to have been unaffected or only slightly affected by human activity around 1900*'. Instead, she argues that '*the reference input has to be based on concentrations in smaller watercourses, which drain catchment areas with a very small level of cultivation multiplied by a present-day waterflow. This calculation method is the closest that one can get in Denmark to an unaffected or almost unaffected situation. There is an anthropogenic signal from the atmospheric N deposition, but it is estimated that the effect from here is within the permitted range in the reference condition definition'.* Hence, Timmermans (2020) and Erichsen and Timmermann (2020) propose to use the approach described by Kronvang et al (2015) for estimating the RCs in Water Plan 2. However, they do not discuss the uncertainties associated with this approach (see Chapter 3) and the appropriateness (and consistency) of the approach for establishing reference conditions, according Guidance document 5 of the Common Implementation Strategy for the WFD.

Brudler et al (2020) indicate that the historical observations challenge the choice of the year 1900 as a reference to describe undisturbed conditions. They report that nutrient emissions from human activities to surface waters were already significant in 1900. Christensen et al (accepted) conclude 'that the ecological state of coastal areas around year 1900 is unlikely to serve as WFD reference conditions'. However, they do not provide information about the ecological status of coastal waters, and this conclusion seems to have no underpinning in this paper. It seems that these conclusions relate to just one specific criterion mentioned in the WFD, and that other criteria for setting RCs in a consistent manner have been neglected.

A more fundamental question arises: 'Do scientists have to define / choose the reference conditions?' Given the description and definition of reference conditions (RCs) and the loose formulations in the WFD[15] and in Guidance document 5 of the Common Implementation Strategy for

[15] "The reference condition is a description of the biological quality elements that exist, or would exist, at high status. That is, with no, or very minor disturbance from human activities"

the WFD, it is difficult to decide on scientific grounds only. A better way is probably that scientists gather the data, do the analyses, and provide the pros and cons of the possible options for defining RCs, and that policy makers and politicians then make the decisions.

However, it appears that all pros and cons of the various approaches have not been articulated and analysed systematically yet. Such articulation and analyses could provide a basis for discussion among policy makers and also a basis for decision making.

5.3. Where does the nitrogen in agriculture come from in 1900?

One may conclude indeed that agricultural activities in Denmark around 1900 were widespread and affecting runoff N losses to the coastal waters, although there are large uncertainties about the actual magnitude of these losses (Jensen, 2017). It is also clear that the N input-output balance of Danish agriculture around 1900 (Table 2) indicates that N inputs (and N outputs via harvested produce) were relatively low; agricultural production in Denmark was nitrogen limited around 1900. Overall N use efficiency in agriculture (crop and animal production combined) was about 33%.

Nitrogen is essential for all life on earth and thus also for food production. To be able to produce food and to consume the produced food, there needs to be input of nitrogen. Most of the N input around 1900 originated from biological N_2 fixation (Table 2). Likely, this was also the situation before 1900, although there was less knowledge about N_2 fixing crops in rotations before the 20th and especially before the 19th century. Christensen et al (accepted) argued that significant amounts of mineral N were liberated from the soil organic matter through soil cultivation in the 2nd half of the 19th century, but they do not provide quantitative estimates.

Interestingly, N_2 fixing crops only fix N_2 when there is a shortage of N in the soil, indicating that there is relatively little spillage of N in crop rotations with N_2 fixing crops. This suggest that the cropland in Denmark around 1900 were relatively leak tight. This situation may change when (i) the (grass)land is ploughed down in autumn and kept bare during winter, and (ii) crops are fed to animals and the resulting animal manure is applied to cropland in autumn. Christensen et al (accepted) suggest that this was indeed the situation. This situation will lead to a decrease in soil organic matter, to soil depletion and ultimately to soil degradation. Definitely, this situation is not sustainable for a long time – it will lead to decreases in crop yield.

Nevertheless, crop production was likely N limited and the efficiency of N utilization was modest (Table 2). Given this background, it is difficult to understand from a scientific point of view that the estimated mean leaching losses from Danish agriculture around 1900 ranged from 18 to 56 kg per ha per year. The estimated leaching losses seem at the higher end of what is possible from a N balance point of view; the upper end of the range (56 kg per ha per year) is even much higher than the mean total N input in agriculture (38 kg per ha per year) around 1900.

This raises the question about the background of the N inputs in agriculture and runoff N losses from agriculture around 1900. This questions is also relevant for undisturbed catchments and catchments with little anthropogenic activities. In the documents reviewed, there has been little discussion about the origin and the amounts of N entering natural and agricultural systems in Denmark around 1900. The origin and amounts entering the natural terrestrial systems largely control also the N outputs of these systems, including the output via leaching losses, and hence seem relevant for answering the question how much N from the Danish territory was lost to the Danish coastal waters around 1900.

The Hubbard Brook Ecosystem Study, founded in 1963 in the northern hardwood forest within the White Mountain National Forest in New Hampshire (US), is probably one of the longest running and most comprehensive ecosystem studies in the world[16]. Chapter 7 of the recently published online book 'A Synthesis of Scientific Research at Hubbard Brook' deals with Nitrogen Cycling. In the Introduction of this chapter, author Peter Groffman states "*The N cycle is exceptionally complex, because .... At Hubbard Brook, scientists are wrestling with an as yet unexplained behavior of the forest N budget: despite continuing high atmospheric inputs to the ecosystem, N is strongly retained with only trace levels of N lost in streamflow*"[17]. Missing sources (in the beginning) and missing sinks are as large as 50% (or more) of the total input or total output on the N mass balance over a 50 yrs period, respectively. Such notions of complexity, variabilities over time and uncertainties are important also for estimating runoff losses from small catchments 'unaffected or only slightly affected by human activity'. These notions of complexity and uncertainty are not always noticeable in the reports related to the estimation of the N loadings to Danish coastal waters around 1900. Further, it emphasizes the need for long-term monitoring studies.

5.4. How to interpret 'unaffected or only slightly affected by human activity'?

Humans have greatly influenced global land use and the global C, N and P cycles. The alterations in global nutrient cycles through land use change, agriculture, food processing and transport have increased especially from the 20th century onwards (Vitousek et al 1997; Smil 2001). But human influences started much earlier already.

The first humans arrived in Denmark some 10,000 years ago, but population density and the effect of human activities on the environment were minor initially. Following the first agricultural revolution, which led to sedentary agriculture or settled agriculture, and formed the start of settlements[18], human activities on the environment increased, but originally this occurred outside Europe. Agricultural activities in Denmark started some 4000 yrs ago[19]. During the first millennia, agricultural productivity was probably low, and related to the human population and population growth. This situation changed around the second agricultural revolution in the 18th century; the introduction of improved agricultural methods increased crop and animal production significantly, and as a result, human population increased as well. The second agricultural revolution was accompanied by / associated with the industrial revolution, which induced also significant influences on the environment. The industrial revolution was made possible in part through the increased labor productivity in agriculture; the surplus laborers found employment in industries and urban areas.

The third agricultural revolution (or green revolution) started in the 1950s and led to a strong intensification of agricultural production. The fourth agricultural revolution perhaps started in the 1980s, when awareness of environmental concerns increased and regulations were implemented. The last three revolutions are clearly seen in the N surplus envelope presented by Kyllingsbæk

[16] https://hubbardbrook.org/
[17] https://hubbardbrook.org/online-book/nitrogen-cycling

[18] Mazoyer M and L Roudart (2005) A History of World Agriculture: From the Neolithic Age to the Current Crisis. Montly Review Press.
[19] Rowley-Conwy, P (1985) The Origin of Agriculture in Denmark: A Review of some Theories. Journal of Danish Archaeology 188-195 | Published online: 05 Nov 2012. https://doi.org/10.1080/0108464X.1985.10589950.

(2008) and the envelope of total nitrogen inputs to the Danish straits presented by Conley et al (2007). Following the first three agricultural revolutions, population density also strongly increased.

Evidently, choosing a baseline year, where the environment was ’unaffected or only slightly affected by human activity’ largely depends on the definition of ’unaffected or only slightly affected by human activity’. Depending on the definition, one may argue that the baseline or RCs should be defined before the first agricultural revolution, i.e., for Denmark some 4000 yrs ago, or before the 2nd agricultural revolution, i.e. before the 18th century, or before the 3rd agricultural revolution and especially before the advent of synthetic N fertilisers, i.e. around 1900.

Understanding agricultural history is also important when choosing RCs on the basis of catchments with currently little or no agricultural activities. Here, a relevant question is ‘why is there no or only little agricultural activities in the catchment’? Has the catchment been abandoned at some point in time, or has the catchment never been subjected to agricultural activities? If so, why did no settler ever utilize the land? The answer to these questions are relevant for understanding the representativeness of the catchment for the Danish territory.

The history of the land is important. The Hubbard Brook Ecosystem Study was set-up in virgin, natural forests. Western Europe has a much longer history of human occupation and alteration of the local environment. The early settlers selected preferably the most fertile grounds for agriculture, leaving the poor grounds to nature. Many of the poor grounds have been further impoverished by biomass harvesting and grazing on the ‘communal lands’. This raises the question whether the current catchment areas with a very small level of cultivation are representative as RCs for the nearby rivers and coastal waters. There is need for having a look into the past landuse.

6. Conclusions

The discussion about appropriate RCs for rivers and coastal waters in Denmark is important and contextual. It is important because eutrophication of coastal waters needs to be prevented and/or minimized, while mitigation measures have to feasible. It is contextual because the available WFD guidelines do not provide clear-cut guidance about the best scientific method/approach for deriving RCs (Van Calster and Garnett, 2021; Ferreira, 2021).

The depth-distribution of eelgrass in Danish coastal waters around 1900 has been chosen as the biological quality element of the RCs. Thus far three approaches have been examined to estimate the N loading from the Danish territory to the coastal waters around 1900, while assuming that the N loading into the coastal waters controls the chemical and biological status (eelgrass distribution) in the coastal waters. All three approaches have advantages and disadvantages, but these seem insufficiently articulated and examined. All three approaches may be improved, which may increase the consistency in the approaches and the confidence in the estimations ultimately.

The estimated N loads from the Danish territory to the coastal waters around 1900 differ by a factor of 2 to 3; this is a relatively small range when considering the large differences in approaches and the large uncertainties involved. No clear and convincing scientific arguments have been found in the Danish studies to prefer one of the approaches above the other.

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