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Human impacts on Nitrogen and Phosphorus concentrations in the North Sea

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Human impacts on Nitrogen and Phosphorus concentrations in the North Sea

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Abstract
This study evaluates the temporal and spatial variations in nutrient concentrations within the period of 1990-2011, with the aim to assess the effectiveness of legislative implementation and associated control measures within the period. Annual nutrient emission data were obtained from OSPAR’s Joint Assessment Monitoring Programmes (JAMP’s) to provide values of riverine flow, direct discharge and atmospheric concentration. Temporal assessments of annual nutrient emissions were carried out on a European and national scale, using statistical analysis within periods of legislative change to assess the regional and sub-regional response to legislative implementation. Nutrient data were also obtained from the EEA database “Waterbase” providing winter concentrations from sampling points within the North Sea region. These data were used for spatial analysis of nutrient concentrations within periods of legislative change, which was achieved via the geospatial plotting of values using GIS software.

Changes in inputs of nitrate and phosphate have been determined within periods of assessment, indicating positive response to legislative change. However variations in response have been seen on both European and regional scale, indicating varied effectiveness of control measures associated with legislative implementation. Spatial variations within assessment periods have also reflected varied responses of nitrate and orthophosphate concentrations to legislative change on a sub-regional level, indicative of a non-linear relationship between the coastal response to land-based controls of diffuse and point source emissions. Where hydromorphological and physio-chemical factors have been considered, it has also highlighted the need for greater evaluation of the coastal interactions which influence the residence time of nutrients and occurrence of eutrophication.

This study concludes that while reductions in emissions have been attributable to legislative change, a combined approach to monitoring and management is required to further understand the behaviour of nutrients within complex aquatic systems and to further the control of emissions and associated impacts of nutrients.
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Introduction
With increasing populations and industrial developments along the coastlines of the North Sea, there is enhanced scope for anthropogenic contributions of nitrogen and phosphorus to the surrounding waters. The North Sea is host to a diverse and complex ecological system; with coastlines that are made up of fjords, estuaries, bays and intertidal zones which provide habitats for a range of marine life. The organic enrichment due to increased nutrient concentrations can have deleterious effects on the water quality and prove detrimental to the ecological health of the marine environment. It is therefore imperative that control measures of emissions are effectively managed to reduce these emissions and their associated impacts. In 1988 OSPAR proposed the target of 50% reduction of nitrogen and phosphorus discharges by surrounding countries from 1985-1995. Whilst some reductions in phosphorus have met and exceeded the 50% target [OSPAR, 2010], nitrogen reductions were not met by the majority of reporting countries [OSPAR, 2010]. Thus, there is a need to evaluate the effects of recent legislation concerning nutrient discharge and assess the efficiency of the associated administration in place in order to prioritise further management action.

1. Background
1.1 Area of Study - The North Sea

The North Sea is a semi-enclosed basin on the continental shelf of north-west Europe (figure 1), opening into the Atlantic Ocean to the north and south and into the Baltic Sea to the east via the Kattegat and Skagerrak [Ducrotoy et al., 2000]. The region of study is defined by OSPAR as 48°-62° N and eastwards of 5° W (extending into the Kattegat) with a surface area of approximately 750,000 km² and relatively shallow average depth of 90m, reaching only depths of 700m in open waters [OSPAR, 2010].

The basin is bordered by the densely populated coastlines of France, Denmark, Belgium, Germany, the Netherlands, Norway, the UK and Sweden, with more than 500 people per km² in some regions [OSPAR, 2010]. Many of these coastal settlements are highly industrialized or used intensively for agriculture, leading to an increased potential for input of anthropogenically derived nutrients via point and diffuse sources [Brion et al., 2004].

1.2. Physical properties

Due to the interaction of oceanic and fresh water, continental shelves play a key role in the biogeochemical cycling of nutrients; where concentrations are generally higher in lower salinity waters, leading to the enrichment of receiving coastal regions [Brion et al., 2004]. This interaction is governed by the hydromorphological properties associated with coastal waters such as the tidal influence, depth and river flow [de Jonge et al., 2002]. The North Sea is mostly poorly stratified due to its relatively shallow depth, with a permanent thermocline situated in open waters at approximately 600m [Ducrotoy et al., 2000]. However, during the spring, surface warming leads to enhanced temperature gradients in open waters, while shallower shelf regions (<30m) remain relatively well mixed throughout the year due to tidal and wind influence [OSPAR, 2010].
The North Sea receives Atlantic and Baltic water, influencing the thermo-haline stratification and circulation within the basin which leads to enhanced tidal mixing in some coastal areas. [Ducrotoy and Elliott, 2008]. There is also a high degree of temporal and spatial climate variability in the North Sea associated with the inflow of Atlantic water and prevailing westerly winds, influencing temperature and precipitation in periods of North Atlantic Oscillation [Thomas et al., 2005]. The interaction of the climate, bathymetry, currents and thermohaline properties of water masses aid the transportation and re-suspension of particulate matter [Van Engeland et al., 2010]. This affects the mixing in the water column, light availability and nutrient availability, influencing the distribution of species, structure of habitats and susceptibility to eutrophic conditions [de Jonge et al., 2002; Ducrotoy and Elliot, 2008].

2. Nutrients and Eutrophication

Nutrients are biologically essential and affect the ecological functioning within the marine environment; however organic enrichment due to increased nutrient input can lead to eutrophic conditions, which can have adverse effects on the marine environment [Lenhart et al., 1997; Paerl, 1997; Anderson et al., 2007].

Eutrophication is the process whereby the addition of potentially limiting nutrients causes increased chlorophyll and macro-algal abundance [Bricker et al., 2007]. The secondary responses include decreased levels of dissolved oxygen, and ultimately hypoxic conditions, due to enhanced respiration following the breakdown of organic matter [Bargu et al., 2011; Claussen, 2009]. This process also has associated impact on human activities; where algal blooms can clog fishing nets and the decaying blooms can create unsightly foam on beaches and unpleasant smells that interfere with tourism and recreation [Mcclusky and Elliot, 2008; OSPAR, 2010].

Eutrophication has been described as being amongst the most potentially damaging of all human influences on the oceans in terms of both scale and consequence, and it is widely recognised to be as a result of organic enrichment via anthropogenic input [GESAMP, 2001; Paerl, 1997; Anderson et al., 2007]. With developments in industry, travel and agriculture, there is increased scope for anthropogenic input of nitrogen and phosphorus via run off, atmospheric deposition and discharge of effluent [Matthias et al., 2010] providing the need to implement effective control measures.

2.1. Nitrogen

The nitrogen cycle consists of the natural conversion of gaseous nitrogen (N₂) to ammonium, by nitrifying bacteria , which is assimilated by plants and animals as organic compounds such as amino acids, proteins and DNA [Li et al., 2012]. However, the utilisation of nitrogen in primary productivity is dependent on light availability; where lower concentrations are seen in the summer than the winter [Brockmann et al., 1990]. The effects of seasonality are however less profound in coastal zones, due to freshwater input of nutrients and turbulent mixing [Brockmann et al., 1990].

Anthropogenic sources of N are often derived from the burning of fossil fuels and use of fertilisers in agricultural practice. The combustion processes associated with
industry, agriculture and transport leads to increased atmospheric concentrations of NO$_3^-$ and NH$_4^+$, which are carried by winds and deposited within precipitation [OSPAR, 2010]. Whereas the application of fertilizers containing NaNO$_3$ (sodium nitrate), NH$_4$NO$_3$ (ammonium nitrate), urea and proteins, leads to the transportation of nitrates to the oceans via soil leaching and run off [Galloway, 2014]. It has been found that the leaching of N is dependent on the solubility of N, and thus retention within soil is dependent on composition; where silt or clay soils have high water retention and facilitate increased mobility of nitrogen to lower catchments [Wiederholt and Johnson, 2005; Hagedorn et al., 2001]. Inputs of nitrogen also occur via point source discharge of wastewater and sewerage, containing organic compounds such as proteins and urea, alongside inorganic compounds such as ammonium, and nitrates used in detergents and food treatment [Li et al., 2012]. However it is widely recognised that diffuse sources of N are amongst the most difficult to manage, where the residence times within soils and sediments allow release to the marine environment for decades after reductions at the original sources [OSPAR, 2010; Scott et al., 1999].

2. 2. Phosphorus

Inorganic phosphorus is used as a respiratory substrate in the form of ATP and is naturally derived from apatite in the earth’s crust or via the remineralisation of organic matter (figure 2) [Mitchell and Baldwin, 1988; Holtan et al., 1988]. Similarly to nitrogen, the phosphorus cycle also displays seasonality regarding mid and surface water concentrations [Lenhart et al., 1997]. However where phosphate is only soluble in reducing conditions [Holtan et al., 1988], the remineralisation process is often constrained to sub-surface, anoxic sediments and phosphates are thus found in higher concentrations directly above surface sediments and in coastal zones [Brockmann et al., 1990; Lenhart et al., 1997]. It is thus recognized that phosphates concentrations are heavily influenced by turbulent mixing and are often seen to be elevated in turbid, coastal regions [Harper et al., 2009].

Phosphate is anthropogenically derived, dominantly via the point source discharge of sewage and waste water effluent containing phosphate compounds within; human excrement and industrial/domestic products, such as detergents and pharmaceuticals [Lenhart et al., 1997]. The introduction of phosphate-free detergents and enhanced sewage treatment plants during the 1980’s has resulted in decreased phosphorus emissions [OSPAR, 2010]. However, where even small fluctuations in phosphorus concentrations can have adverse effects on the marine environment, sufficient treatment to remove phosphates from wastewater is required [De Galan et al., 2004]

3. Legislation and Management

In 1992, the new “Convention for the Protection of the Marine Environment in the North Atlantic” was signed, and in 1997 came into force under the OSPAR commission [Ducrotoy and Elliott, 1997]. OSPAR took over from the previous Oslo and Paris Commission and is the mechanism by which the European governments, together with the European Union, cooperate to protect the marine environment of the North-East Atlantic. Within the OSPAR commission the Joint Assessment Monitoring Programme (JAMP) commits contracting parties to implement OSPAR
and EU measures to reduce emissions, discharges and losses of nutrients to the marine environment [OSPAR, 2010]. This implementation involves the collaborative reporting and monitoring of regional nutrient status, with the intent to also explore new and emerging problems associated with water quality. This information is set within the context of the contemporary socio-economic, physical and biological characteristics of the North Sea in order to define relevant and effective monitoring strategies on a regional level. The aim of this reporting is also to promote consideration of marine eutrophication when implementing EU directives, where reports are used to provide a summary of contemporary knowledge and thus improve the methodology and synergy of monitoring systems in place [CEC, 2002].

3.1. The Urban Waste-Water Treatment Directive (UWWTD)

The European Council Directive (91/271/EEC) was addressed to member states on the 21 May 1991; regarding the collection, treatment and discharge of domestic and industrial waste water and other effluent derived from sewerage and rainwater run-off [CEC, 2005]. The aim of the directive is to protect the environment from the adverse effects of waste water discharge [Ferreira et al., 2011]. The directive defines areas as “agglomerations” in which the population or economic activities are of sufficient concentration for the collection, treatment and discharge of wastewater. The assessment uses the biological oxygen demand BOD to evaluate the organic biodegradable load of waste-water; where a BOD of 60g of $O_2$ per day is equivalent 1 p.e (population equivalent) [EEA, 1991]. Under the UWWTD, member states are committed to ensuring discharges to fresh-water and estuaries are <2000 p.e and discharges to coastal waters are <10000 p.e. The implementation action associated with the directive is outlined in figure 3.

<table>
<thead>
<tr>
<th>Date</th>
<th>Implementation Action</th>
</tr>
</thead>
<tbody>
<tr>
<td>By 30th June 1993</td>
<td>Member States committed to enforce the laws, regulations and administrative provisions required to comply with the directive.</td>
</tr>
<tr>
<td>By 31st December 1994</td>
<td>Comparison of Member states carried out by the commission and necessary proposals made</td>
</tr>
<tr>
<td>By 31st December 1998</td>
<td>Disposal of un-treated sludge to surface waters is phased out &amp; urban waste-water collection systems provided for agglomerations of more than 10000 p.e.</td>
</tr>
<tr>
<td>By 31st December 2000</td>
<td>Urban waste-water collection systems provided for agglomerations of more than 15000 p.e.</td>
</tr>
<tr>
<td>By 31st December 2005</td>
<td>Urban waste-water collection systems provided for agglomerations of 2000-15000 p.e.</td>
</tr>
</tbody>
</table>

Member states are also required to assign “appropriate treatment” to meet the relevant quality objectives of the directive; where “Primary treatment” consists of the settlement of suspended solids, and 20% reduction in BOD whereas “Secondary treatment” involves further chemical treatment to meet the requirements outlined in the directive. Member states are required to identify (and review every 4 years) “sensitive areas” as defined under Article 5 of the directive, and publish situation
reports on the disposal of urban waste-water and sludge in their region every 2 years.

3. 2. The Nitrates Directive (ND)

The European Council directive (91/676/EEC) was addressed to member states on the 12th December 1991. The directive concerns the protection of waters against pollution caused by nitrates from agricultural sources, with the aim to protect human health, living resources and aquatic ecosystems. The implementation of the directive involves the identification of Nitrate Vulnerable Zones (NVZ’s), establishment of good agricultural practice (GAP) and national reporting [CEC, 1991]. The Directive focuses primarily on surface and ground waters (especially those) intended for the abstraction of drinking water and identifies polluted waters as those that contain 50mg/l of nitrates (in accordance with Directive 75/440/EEC) [EEA, 1991]. The establishment of codes of GAP includes measures to:

- Limiting the periods of fertilizer application
- Limiting the conditions of fertilizer application (e.g. avoid application on steeply sloping ground or ground close to water courses)
- Limitations regarding the storage capacity of livestock manure
- Practice of crop rotation and use of “catch-crops” to prevent nitrate leaching and run off

The implementation of GAP is on a voluntary basis, unless farms are within NVZ’s, in which case action plans are implemented, consisting of measures to further limit fertilizer use based on crop needs and background nitrogen levels. Further assessment associated with NVZ’s also considers the hydrogeology, soil composition, livestock abundance, crop type and precipitation levels associated within an area, to account for justifications of increased nitrate outputs [CEC, 1991]. National monitoring and reporting by member states every 4 years is also mandatory practice and is to include:

- Nitrate concentrations in ground waters and surface waters
- Eutrophication status
- Assessment of agricultural practice and action plans (where applicable)
- Revision of NVZ’s and action plans
- Estimation of future trends

These reports are used as a basis for a 4-yearly report by the European Commission; set out to provide member states with contemporary scientific and technical knowledge. The implementation action associated with the directive is outlined in figure 4.

Within NVZ’s nitrate concentrations are to be measured for a one year period at monthly intervals (more frequently during flood periods) and the designation of NVZ’s is to be reviewed every four years unless all previous samples are below 25mg/l.
Table 2: Implementation Action Plan of the Nitrates Directive

<table>
<thead>
<tr>
<th>Date</th>
<th>Implementation Action</th>
</tr>
</thead>
<tbody>
<tr>
<td>By 12th December 1993</td>
<td>Member States shall bring into force the laws, regulations and administrative provisions necessary to comply with this Directive</td>
</tr>
<tr>
<td>By 12th December 1997</td>
<td>Review of designations for NVZ’s &amp; Reduction of livestock manure allowance from 210kgN per hectare yr⁻¹ to 170kgN per hectare yr⁻¹</td>
</tr>
<tr>
<td>By 1st January 1998</td>
<td>The Commission shall submit a report to the Council with any appropriate proposals for revision of this directive and associated measures/practice</td>
</tr>
</tbody>
</table>

3.3. The Water Framework Directive (WFD)

The European council directive (2000/60/EC) was brought into force on the 23rd October 2000; with the aim to achieve good ecological and chemical status for all surface and ground waters within 15 years of implementation, in accordance with provisions in annex V of the directive. [EEA, 2000].

The directive establishes a framework for community action with the intent to enhance, protect and restore artificially and heavily modified waters that may be subject to adverse effects regarding ecological and/or chemical status [EEA, 2000]. The framework includes the monitoring and management of water bodies by promoting sustainable water use, reducing discharges/losses and setting emission limit values for hazardous substances, with consideration to areas designated by the Nitrates Directive and UWWTD [CEC, 2002].

As part of the framework, member states are required to produce a ‘single river basin management plan’ in order to provide a holistic approach to management measures [CEC, 2002]. Within each river basin district, member states are committed to ensuring that surface and ground waters, especially those intended for the abstraction of drinking water (>10m³ per day) must meet the requirements of the drinking water directive (98/83/EC). The directive also establishes annual surveillance monitoring to take into account changes in land use within the catchment and account for associated human impacts [EEA, 2000]. Reports on management are to be published 9 years after directive implementation and reviewed at every 6 years thereafter. The implementation action associated with the directive is outlined in figure 5.
Table 3: Implementation Action Plan of the Water Framework Directive

<table>
<thead>
<tr>
<th>Date</th>
<th>Implementation Action</th>
</tr>
</thead>
<tbody>
<tr>
<td>2003</td>
<td>Identification of river basin districts and notification of relevant authorities</td>
</tr>
<tr>
<td>By 23 October 2006</td>
<td>Establishment of national monitoring network</td>
</tr>
<tr>
<td>By 23 October 2009</td>
<td>Full implementation of river basin management plan, including programme of measures and initial report to commission</td>
</tr>
<tr>
<td>By 23 October 2012</td>
<td>Best environmental practice achieved by all member states</td>
</tr>
<tr>
<td>2015</td>
<td>Meeting of good ecological and chemical environmental status</td>
</tr>
</tbody>
</table>

Aims and Objectives

The aim of this report is to evaluate the impacts associated with the implementation of European legislation concerning water quality and nutrient emissions.

1. The first objective is to assess the temporal variations of N&P emissions between years of legislative implementation. In order to achieve this, nutrient emission data has been obtained from OSPAR’s Joint Assessment Monitoring Programme. Statistical analysis has been carried out on this data within periods of legislative change, in order to determine significance of changes occurring within the specified time periods.

2. The second objective was to investigate spatial variations in N&P concentrations to assess the anthropogenic pressures associated with human activity. In order to achieve this, winter nutrient concentration data from sampling points within the North Sea was obtained from the EEA database. This data was geospatially plotted using GIS software, in order to allow spatial analysis of nutrient concentrations and investigate the factors influencing areas of elevated concentrations.

3. The third objective was to assess the effectiveness of management in place and investigate the relationship between emission controls and coastal concentrations. This was achieved by the comparison of temporal and spatial assessments to see where reductions in emissions were reflected in receiving water bodies. This process also involves the review of legislative implementation and findings from previous studies to gain a holistic understanding of the factors influencing the occurrence of N & P.

4. Methodology

Policy relevant and quality assured nutrient data was obtained from OSPAR’s Joint Atmospheric Monitoring Programmes and the EEA database ‘Waterbase’ for selected European countries; UK, NL, FR, BE, DE, DK, SE and NO over the time period of 1990-2011. Data has been processed into a uniform format using a formulated template, to allow comparison between datasets. Statistical analysis of
the data has then been performed within periods of legislative change; 1990-1993, 1993-2000 and 2000-2011, to determine the significance of associated changes.

**4.1. Methodology associated with data concerning river input, direct discharge and atmospheric concentrations.**

The data concerning anthropogenic nutrient inputs into the North Sea were obtained from programmes within OSPAR’s (JAMP). Data from the “Comprehensive Study on Riverine Inputs and Direct Discharges” (RID) was used to acquire annual diffuse and point source emissions of nitrate (NO$_3$) and phosphate (PO$_4$). Data was also obtained from the “Coordinated Atmospheric Monitoring Programme” (CAMP), to acquire the annual concentrations of NO$_3$ and NH$_4$ in precipitation taken from coastal monitoring stations around the North Sea. The methodology associated with the processing of emission data is outlined in figure 6.

1. Calculate cumulative inputs for individual years from 1990-2011
   Justification: to provide a holistic overview of legislative change on a ‘European’ scale by assessing temporal trends in the cumulative input of nutrients by selected countries

2. Calculate 4 year mean from data available from (1990-1993) to be used as a baseline
   Justification: to provide indication of overall impact of legislative change and represent the nutrient status prior to implementation action of the Nitrates Directive and UWWTD.

3. Graphically represent inputs using a moving average of ≤4 yearly values
   Justification: temporally assess general trends with ‘smoothing’ of inter-annual fluctuations (< 4yrs) used where 4yrs of consecutive data not available

   Justification: determine the significance of trends within periods of legislative change. Significance given as a function of degrees of freedom (DF) to account for varied data availability within time periods and differing length of time periods used.

5. Extract data from individual countries for years 1990-2011 and repeat steps 3 & 4
   Justification: determine relative contributions to cumulative nutrient load and assess impact of legislative change on a sub-regional basis

**Figure: 1** Methodology: Processing of OSPAR’s annual emission data
4.2. Methodology associated with geospatial plotting of nutrient concentration data

Nutrient concentration data was obtained from the EEA database “Waterbase” (WaterTCMv10) containing data collected by EU member states through the ‘WISE-SoE’ data collection process, from sampling points in coastal and marine waters. The data was extracted for selected countries (BE, DK, DE, NO, SE, FR, NL, UK) for the period of 1990-2011 to allow for comparison with input data from OSPAR monitoring programmes. N&P values were extracted from within winter months (December, January, and February); in order to minimise the impact of seasonal variation on the results. Where nutrient concentrations were given in µmol l⁻¹, values were converted to mg l⁻¹ (N) and µg l⁻¹ (P) for comparison against legislative values using the following unit conversion equation [ICES, 2014].

\[ \text{gl}^{-1} = \text{mol l}^{-1} \times \text{Molecular Weight (MW)} \]

Where MW: \( \text{NO}_3 = 62.0049, \text{PO}_4 = 94.9715 \) [Wells, 1984]

The aim of using this database was to spatially assess the nutrient status within the North Sea over the period of study.

1. Determine Eastings and Northings of each sampling point using National Station ID (unique identifier for each sampling station)

2. Projection of data points onto ETRS89-LAEA European Grid
   - Northings and easting values interpreted using the UTM 32 North projection

3. Projection of shape files containing European country locations to visually display European coastline

4. Aggregation of data projections into 50km² grid cells
   - To allow comparison of grid cells between years

4. Spatial Join created between data points and sampling grid to calculate means of values within 50km² cells
   - To improve the reliability of temporal comparison between cells

5. Colorimetric scale created for grid cells using discrete, quantified colour categories in which to improve visual assessment of nutrient concentrations

Figure: 2 Methodology: Processing of EEA’s Waterbase nutrient concentration data

In order to achieve this, the values of nitrate and phosphate concentrations were plotted with their corresponding sampling location using geospatial (GIS) software ESRI ArcMap. This process is outlined in figure 7.
This process was repeated nitrate and phosphate values between time periods of legislative change (1990-1993, 1993-2000, 2000-2011) in order to display spatial variations in nutrient concentrations and allow temporal assessment alongside results of nutrient emission data.

5. Results

Temporal variations of riverine input, direct discharge and atmospheric concentration are shown in figures 9-11 as cumulative (a) and individual (b) values for NO$_3$ and PO$_4$ (NH$_4$ atmospheric). Statistical analysis has been achieved using the PEARSON statistical test; measuring the correlation between the two variables (time and concentration), providing an R value of between +1 and -1, indicating maximum positive and negative correlation respectively. Results of statistical analysis are given below relevant figures where significant; with ✔/☒ indicating significant decrease/increase in nutrient emissions. Complete tables of statistical results are included within the appendices. Years of implementation action of the UWWTD, ND and WFD are also shown in figures 9-11 to aid visual assessment of trends within periods of legislative change. An implementation line of the Marine Strategy Framework Directive (MSFD) has also been included, however where no action associated with the directive has occurred within the study period, this line is to provide indication of the baseline level from which the future implications of the directive can be assessed.

Overall there is a general increase in riverine nitrate input within the study period (from 561.76 to 711.20 kt/a), with all post-1993 values above the baseline (figure 9a). No significant periods of decrease in nitrates were observed until the period 2000-2011, where reductions from 795kt/a to 711.2kt/a (R= -0.728, p < 0.02) occurred, indicative of some positive response to legislative change within this period. This general increase is also seen to continue, where increasing nitrate input is observed from 2006 onwards, thus indicating the need for further assessment of management where emission controls have been insufficient.

For phosphates there have been significant decreases in the both periods of 1993-2000 (R=-0.647, p <0.1) and 2000-2011 (R=-0.959, p<0.001) with the greatest decrease occurring in the latter period (from 30.65 kt/a to 17.29kt/a). All values from post-1995 are below the baseline, with additional reductions seen with the further implementation of legislation, indicating a positive response to the implementation of legislation.
Figure 3(a): Cumulative Riverine Input (Europe), showing the moving average (<4) of nitrate and phosphate concentrations. Note: baselines are calculated as a four year average from 1990-1993 and can be expected to be higher than represented where missing data from DK occurs within this period.

Table 4: Results of statistical analysis for cumulative nitrate and phosphate concentrations via riverine input

<table>
<thead>
<tr>
<th>Years</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R Value</th>
<th>DF(n-2)</th>
<th>Significance</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nitrate</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1990-1993</td>
<td>546.357</td>
<td>631.965</td>
<td>585.584</td>
<td>38.883</td>
<td>0.885</td>
<td>2</td>
<td>p &lt; 0.20</td>
</tr>
<tr>
<td>1993-2000</td>
<td>631.965</td>
<td>848.504</td>
<td>752.746</td>
<td>66.537</td>
<td>0.264</td>
<td>6</td>
<td>none</td>
</tr>
<tr>
<td>2000-2011</td>
<td>581.922</td>
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<td>1990-1993</td>
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<td>p &lt; 0.1</td>
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<td>2000-2011</td>
<td>17.289</td>
<td>30.684</td>
<td>25.373</td>
<td>5.066</td>
<td>-0.959</td>
<td>8.000</td>
<td>p &lt; 0.001</td>
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</table>

Riverine inputs of nitrate and phosphate have displayed variation in temporal trends between countries (figure 9b), indicating of differences response to legislative change on a regional level. Countries with the highest emissions for both nitrate and phosphate were consistently NL, FR, DE and the UK. The most significant increases in nitrates were seen during the period of 1993-2000; from the UK (R=0.513, p<0.1) and DK (R=0.885, p<0.01). However, in the period of 2000-2011, reductions were observed from the UK, BE, FR and NL, suggesting positive response to legislative change in the south-west region post-2000. It can also be seen in figure 9(b) that similar temporal trends occur between NL, DE and FR, with synchronized peaks in 1995 and troughs in 2006. While this may be indicative of a regional response to emission, this may also be indicative meteorological, or hydromorphological influence within the southern end of the North Sea basin.
Phosphate inputs by individual countries showed little temporal similarity, however FR, NL and the UK were consistently the main contributors of phosphates; with NL having highest values in periods 1990-1993 and 1993-2000. Significant decreases from FR and DE were seen in the period of 1993-2000, with the most significant decrease in DE (R=-0.873, p<0.01). However, during the same time period, increases occurred in DK, NO and the UK, indicating varied response to legislative measures on a regional level. In the period 2000-2011 significant reductions were however seen by the UK with a decrease to 3.61kt/a; lower than the value prior to 1993, indicating a positive overall response during the study.

*Figure 3(b):* Individual Riverine Inputs (per country), showing the moving average (<4) of nitrate and phosphate concentration in riverine inputs from each country to the North Sea

Cumulative direct discharge was found to decrease for both nitrates and phosphates during the period of study (figure 10a), however nitrate inputs via direct discharge were considerably lower than those via riverine input. Significant reductions in nitrate were seen from 1993-2011, with the most significant decrease in 2000-2011 (R=-0.914, p<0.001) from 13.488-11.141kt/a. Significant reductions in phosphates are also seen, however from 1990-2011, again with greatest reductions occur in the period 2000-2011 (R=-0.973, p<0.001). This is indicative of positive overall response to legislative change, with increasing reductions seen with further legislative implementation where all values post-1993 are below the baseline. It is however important to note that values are expected to be higher than represented where data is not available and thus reductions may be exaggerated where values are missing from one or more countries.
Table 5: Results of statistical analysis for nitrate and phosphate concentrations via riverine inputs from individual countries

<table>
<thead>
<tr>
<th>Year</th>
<th>Country</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R value</th>
<th>DF (n-2)</th>
<th>Significance</th>
<th>G/</th>
<th>Note</th>
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<td></td>
<td></td>
<td></td>
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<td></td>
</tr>
<tr>
<td>1990-1993</td>
<td>Belgium</td>
<td>24.400</td>
<td>35.314</td>
<td>30.676</td>
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<td>0.918</td>
<td>2</td>
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<td></td>
</tr>
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<td>212.00</td>
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<td>36.738</td>
<td>0.848</td>
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</tr>
<tr>
<td></td>
<td>Sweden</td>
<td>1.540</td>
<td>21.010</td>
<td>15.660</td>
<td>9.454</td>
<td>0.823</td>
<td>2</td>
<td>p &lt; 0.2</td>
<td></td>
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</tr>
<tr>
<td>1993-2000</td>
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<td>131.602</td>
<td>89.440</td>
<td>28.333</td>
<td>0.513</td>
<td>6</td>
<td>p &lt; 0.1</td>
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<td></td>
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<tr>
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<td>Denmark</td>
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<td>15.800</td>
<td>9.012</td>
<td>3.950</td>
<td>0.885</td>
<td>6</td>
<td>p &lt; 0.01</td>
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<td></td>
</tr>
<tr>
<td>2000-2011</td>
<td>UK</td>
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<td>153.032</td>
<td>91.304</td>
<td>36.199</td>
<td>-0.434</td>
<td>10</td>
<td>p &lt; 0.2</td>
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<td></td>
<td>Belgium</td>
<td>23.243</td>
<td>46.680</td>
<td>34.900</td>
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<td>-0.501</td>
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<td></td>
<td>France</td>
<td>106.742</td>
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<td>46.796</td>
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<tr>
<td></td>
<td>Netherlands</td>
<td>129.328</td>
<td>307.790</td>
<td>215.340</td>
<td>53.825</td>
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<td>9</td>
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**Phosphate**

<table>
<thead>
<tr>
<th>Year</th>
<th>Country</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R value</th>
<th>DF (n-2)</th>
<th>Significance</th>
<th>G/</th>
<th>Note</th>
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</thead>
<tbody>
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<td>1990-1993</td>
<td>Sweden</td>
<td>0.029</td>
<td>0.219</td>
<td>0.136</td>
<td>0.079</td>
<td>0.963</td>
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<td>p &lt; 0.05</td>
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<td></td>
</tr>
<tr>
<td>1993-2000</td>
<td>UK</td>
<td>7.235</td>
<td>11.451</td>
<td>8.564</td>
<td>1.689</td>
<td>0.850</td>
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<tr>
<td></td>
<td>Denmark</td>
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<td>0.198</td>
<td>0.114</td>
<td>0.052</td>
<td>0.517</td>
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<td>p &lt; 0.2</td>
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<td>3.600</td>
<td>2.704</td>
<td>0.580</td>
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<td></td>
<td>France</td>
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<td>7.591</td>
<td>6.561</td>
<td>0.644</td>
<td>-0.606</td>
<td>6</td>
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<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>Norway</td>
<td>0.059</td>
<td>0.130</td>
<td>0.097</td>
<td>0.028</td>
<td>0.806</td>
<td>6</td>
<td>p &lt; 0.02</td>
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</tr>
<tr>
<td>2000-2011</td>
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<td>2.646</td>
<td>25.072</td>
<td>7.165</td>
<td>6.318</td>
<td>-0.501</td>
<td>10</td>
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<tr>
<td></td>
<td>Belgium</td>
<td>1.401</td>
<td>2.366</td>
<td>1.707</td>
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<td>-0.604</td>
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<td>0.078</td>
<td>0.184</td>
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<td>0.586</td>
<td>6</td>
<td>p &lt; 0.2</td>
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</table>

Figure 4(a): Cumulative Direct Discharge (Europe), showing the moving average (<4) of cumulative direct discharge of nitrate and phosphate. Note: where data is missing, represented values are lower than expected.
Table 6: Results of statistical analysis for cumulative nitrate and phosphate concentrations via direct discharge

<table>
<thead>
<tr>
<th>Years</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R Value</th>
<th>DF(n-2)</th>
<th>Significance</th>
</tr>
</thead>
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<td>15.060</td>
<td>16.092</td>
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<td>0.435</td>
<td>-0.152</td>
<td>2.000</td>
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<tr>
<td>1993-2000</td>
<td>12.940</td>
<td>15.631</td>
<td>14.007</td>
<td>1.011</td>
<td>-0.856</td>
<td>6.000</td>
<td>p &lt; 0.01</td>
</tr>
<tr>
<td>2000 – 2011</td>
<td>11.031</td>
<td>14.067</td>
<td>12.534</td>
<td>1.237</td>
<td>-0.914</td>
<td>10.000</td>
<td>p &lt; 0.001</td>
</tr>
<tr>
<td><strong>Phosphates</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1990-1993</td>
<td>6.804</td>
<td>7.642</td>
<td>7.324</td>
<td>0.385</td>
<td>-0.952</td>
<td>2.000</td>
<td>p &lt; 0.05</td>
</tr>
<tr>
<td>1993-2000</td>
<td>5.158</td>
<td>6.804</td>
<td>5.521</td>
<td>0.547</td>
<td>-0.688</td>
<td>6.000</td>
<td>p &lt; 0.1</td>
</tr>
<tr>
<td>2000 – 2011</td>
<td>2.364</td>
<td>5.158</td>
<td>3.685</td>
<td>0.877</td>
<td>-0.973</td>
<td>10.000</td>
<td>p &lt; 0.001</td>
</tr>
</tbody>
</table>

In the period of 1990-2000 significant decreases of nitrate emissions were seen from countries except for the UK (figure 10b), however in the period 2000-2011, significant reductions were made by the UK (R=-0.558, p<0.1). While this is indicative of response to legislative change, where emissions are only reduced from 10.858kt/a to 8.990kt/a, it indicated a need for further control of emissions. Reductions in phosphate emissions from the UK were more efficient during the study period (from 6.70kt/a to 2.02kt/a).

Significant reductions in phosphate emissions were made by DK, DE, NO and SE, with greatest decrease in latter periods, indicative of more effective management of point source discharges of phosphates, with progressive response to legislative measures. It should however be noted that NL, FR and BE did not submit data within this time period and are therefore not included within figure 10(b).

![Figure 4(b): Individual Direct Discharge (per country), showing the moving average (<4) for individual direct discharge of nitrate and phosphate inputs](image-url)
Table 7: Results of statistical analysis for individual nitrate and phosphate concentrations via direct discharge.

<table>
<thead>
<tr>
<th>Year</th>
<th>Country</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R value</th>
<th>DF (n-2)</th>
<th>Significance</th>
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<td></td>
<td></td>
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</tr>
<tr>
<td>1990-1993</td>
<td>Denmark</td>
<td>0.250</td>
<td>1.000</td>
<td>0.688</td>
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<td>-0.975</td>
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<td>2.120</td>
<td>2.723</td>
<td>2.322</td>
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<td>-0.869</td>
<td>2</td>
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<td>Norway</td>
<td>0.185</td>
<td>0.193</td>
<td>0.190</td>
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<td>-0.961</td>
<td>2</td>
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<td>☺</td>
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<tr>
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<td>Netherlands</td>
<td>0.530</td>
<td>1.063</td>
<td>0.864</td>
<td>0.255</td>
<td>0.943</td>
<td>2</td>
<td>p &lt; 0.1</td>
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<td>Sweden</td>
<td>0.463</td>
<td>0.900</td>
<td>0.691</td>
<td>0.242</td>
<td>-0.912</td>
<td>2</td>
<td>p &lt; 0.1</td>
<td>☺</td>
</tr>
<tr>
<td>1993-2000</td>
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<td>0.260</td>
<td>0.211</td>
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<td>-0.882</td>
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<tr>
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<td>1.750</td>
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<td>0.735</td>
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<td>2000-2011</td>
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<td>8.345</td>
<td>10.888</td>
<td>9.453</td>
<td>0.758</td>
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<tr>
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<td>Netherlands</td>
<td>1.067</td>
<td>1.758</td>
<td>1.302</td>
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<td>0.621</td>
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<td>p &lt; 0.2</td>
<td>☺</td>
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<td>-0.904</td>
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<td>1993-2000</td>
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<td>0.089</td>
<td>0.005</td>
<td>-0.731</td>
<td>6</td>
<td>p &lt; 0.05</td>
<td>☺</td>
</tr>
<tr>
<td>2000-2011</td>
<td>UK</td>
<td>1.549</td>
<td>4.530</td>
<td>2.946</td>
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<td>10</td>
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<td>p &lt; 0.02</td>
<td>☺</td>
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<td>Germany</td>
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<td>0.008</td>
<td>-0.748</td>
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<td>0.217</td>
<td>0.271</td>
<td>0.244</td>
<td>0.019</td>
<td>0.737</td>
<td>10</td>
<td>p &lt; 0.01</td>
<td>☺</td>
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</table>

The concentration of NO₃ and NH₄ in precipitation showed similar temporal trends on a European scale, however with slightly higher values of NH₄ (figure 11a). In the period of 1993-2000 both NO₃ and NH₄ showed significant decrease of (R= -0.611, p<0.1) and (R= -0.811, p < 0.02), indicating positive response to legislative change post-1993. In the period 2000-2011 significant decrease was only seen in NO₃ indicating further response to additional legislative measures. Whilst there are reductions in both forms of nitrogen, values are only reduced slightly from that of the baseline (based on 1993-1996), thus indicating the need for further emission controls.

Figure 5(a): Cumulative Concentration of NO₃ and NH₄ in Precipitation (Europe), showing the moving average (<4) of cumulative NO₃ and NH₄ concentrations. Note: tentative baselines have been based on an average of values from (1993-1996) due to lack of data from as many as 3 countries at any year in 1990-1993
Table 8: Results of statistical analysis for cumulative NO$_3$ and NH$_4$ concentrations in precipitation

<table>
<thead>
<tr>
<th>Years</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R Value</th>
<th>DF(n-2)</th>
<th>Significance</th>
<th>NO$_3$</th>
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<td>1990-1993</td>
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<td>3.510</td>
<td>3.092</td>
<td>0.315</td>
<td>0.912</td>
<td>2.000</td>
<td>p &lt; 0.1</td>
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</tr>
<tr>
<td>1993-2000</td>
<td>3.285</td>
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<td>0.236</td>
<td>-0.611</td>
<td>6.000</td>
<td>p &lt; 0.1</td>
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</tr>
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<td>2000-2011</td>
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<td>3.418</td>
<td>0.151</td>
<td>-0.659</td>
<td>10.000</td>
<td>p &lt; 0.02</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
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<th>Years</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R Value</th>
<th>DF(n-2)</th>
<th>Significance</th>
<th>NH$_4$</th>
</tr>
</thead>
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<tr>
<td>1990-1993</td>
<td>3.113</td>
<td>4.254</td>
<td>3.475</td>
<td>0.524</td>
<td>0.692</td>
<td>2.000</td>
<td>none</td>
<td></td>
</tr>
<tr>
<td>1993-2000</td>
<td>3.670</td>
<td>4.254</td>
<td>3.892</td>
<td>0.224</td>
<td>-0.811</td>
<td>6.000</td>
<td>p &lt; 0.02</td>
<td></td>
</tr>
<tr>
<td>2000-2011</td>
<td>3.670</td>
<td>4.110</td>
<td>3.886</td>
<td>0.139</td>
<td>-0.024</td>
<td>10.000</td>
<td>none</td>
<td></td>
</tr>
</tbody>
</table>

NO$_3$ concentration values show similar temporal trends within each participating country (figure 11b). Significant reductions have been made by NO, DE, DK, SE and SL in periods 1993-2000 and 2000-2011, with significant increase only occurring in FR during the period of 2000-2011 (R=0.505, p<0.1). This is indicative of positive regional response legislative change post-1993, however lack of full data submission by countries from 1990-2000 creates uncertainties in pre-2000 trends. For NH$_4$ concentrations; similarities in temporal variations also exist, however are less apparent than those of NO$_3$. During the period 1990-2000 significant decreases were observed for DK, SE, FR and NO indicating some positive response during this period, however with fewer countries showing significant reductions than that NO$_3$, indicating lesser regional response.

![Figure 5(b): Individual Concentration in Precipitation (per Country), showing the moving average (<4) of NO3 and NH4 concentrations from individual countries.](image-url)
his influence can be seen approximately as cumulative values within orthophosphates within the embayed area. Discharge by DK and SE (figure 10b), suggesting increased persistence of nitrate in the coastal embayment within reflection to control of point source discharge. However, this response was not seen in the coastal embayment within the southern Kattegat, despite reductions of direct discharge by DK and SE (figure 10b), suggesting increased persistence of orthophosphates within the embayed area.

Table 9: Results of statistical analysis of NO3 and NH4 concentrations in precipitation from individual countries

<table>
<thead>
<tr>
<th>Year</th>
<th>Country</th>
<th>Min</th>
<th>Max</th>
<th>Mean</th>
<th>SD</th>
<th>Pearson, R value</th>
<th>DF [n-2]</th>
<th>Significance</th>
<th>O/Ω</th>
</tr>
</thead>
<tbody>
<tr>
<td>NO3</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1993-2000</td>
<td>France</td>
<td>0.284</td>
<td>0.520</td>
<td>0.384187097</td>
<td>0.094</td>
<td>-0.918</td>
<td>2</td>
<td>p &lt; 0.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Norway</td>
<td>0.431</td>
<td>0.550</td>
<td>0.491</td>
<td>0.046</td>
<td>-0.861</td>
<td>6</td>
<td>p &lt; 0.001</td>
<td></td>
</tr>
<tr>
<td></td>
<td>UK</td>
<td>0.292</td>
<td>0.530</td>
<td>0.428</td>
<td>0.059</td>
<td>-0.482</td>
<td>10</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Denmark</td>
<td>0.393</td>
<td>0.647</td>
<td>0.525</td>
<td>0.084</td>
<td>-0.574</td>
<td>10</td>
<td>p &lt; 0.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Germany</td>
<td>0.368</td>
<td>0.653</td>
<td>0.475</td>
<td>0.091</td>
<td>-0.740</td>
<td>10</td>
<td>p &lt; 0.01</td>
<td></td>
</tr>
<tr>
<td></td>
<td>France</td>
<td>0.221</td>
<td>0.557</td>
<td>0.339</td>
<td>0.097</td>
<td>0.5016</td>
<td>9</td>
<td>p &lt; 0.1</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Netherlands</td>
<td>0.338</td>
<td>0.511</td>
<td>0.391</td>
<td>0.047</td>
<td>-0.662</td>
<td>10</td>
<td>p &lt; 0.02</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden</td>
<td>0.354</td>
<td>0.575</td>
<td>0.450</td>
<td>0.061</td>
<td>-0.805</td>
<td>10</td>
<td>p &lt; 0.01</td>
<td></td>
</tr>
<tr>
<td>NH4</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>1990-1993</td>
<td>Denmark</td>
<td>0.65</td>
<td>1.068</td>
<td>0.823</td>
<td>0.181</td>
<td>-0.972</td>
<td>2</td>
<td>p &lt; 0.05</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden</td>
<td>0.56</td>
<td>0.639</td>
<td>0.596</td>
<td>0.038</td>
<td>-0.875</td>
<td>2</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
<tr>
<td>1993-2000</td>
<td>France</td>
<td>0.225</td>
<td>0.573</td>
<td>0.386</td>
<td>0.135</td>
<td>-0.813</td>
<td>1</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Norway</td>
<td>0.341</td>
<td>0.510</td>
<td>0.429</td>
<td>0.064</td>
<td>-0.855</td>
<td>6</td>
<td>p &lt; 0.01</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Belgium</td>
<td>0.28</td>
<td>0.712</td>
<td>0.571</td>
<td>0.109</td>
<td>0.436</td>
<td>10</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Denmark</td>
<td>0.541</td>
<td>0.942</td>
<td>0.670</td>
<td>0.137</td>
<td>-0.480</td>
<td>10</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Sweden</td>
<td>0.372</td>
<td>0.666</td>
<td>0.505</td>
<td>0.088</td>
<td>0.414</td>
<td>10</td>
<td>p &lt; 0.2</td>
<td></td>
</tr>
</tbody>
</table>

The spatial variations in nutrient concentration are shown in figures 12-14 with nitrate (i) and orthophosphate (ii) concentrations represented as averaged (mean) concentrations of values sampled within 50km² grids. This has been done to allow temporal comparison between grid cells, where sampling point locations vary between years. These results are presented as cumulative values within periods of legislative change (a) to provide an overview of the nutrient status within the time period concerned, and are also displayed at the start (b) and end (c) years of each period to indicate changes occurring within the timeframe.

It can be seen in figure 12(i) that the majority of open-water nitrate values are between 0-1mg/l, with coastal values up to 6mg/l, indicative of a freshwater influence on coastal waters. This influence can be seen approximately 100km from the coast in non-enclosed areas, however is seen with greater proximity in embayed areas such as the north-west German coast and west coast of Denmark. Temporal variations of nitrate from 1990(b) to 1993(c) show progressively more dominant costal values; with values off the German coast increasing from 0-4μg/l (in 1990) to 4.01-6μg/ (in 1993). The increase in nitrate concentration in this period is concurrent with those found in river inputs (figure 9a). This is suggestive of riverine influence within this region and can be seen where elevated concentrations are present in the south-east of the UK and north-west coast of Germany, where major rivers such as the Thames and Elbe exist.

Orthophosphates are also seen in higher concentrations in coastal areas (figure 12ii), however with some elevated open water concentrations, suggesting possible off-shore influence. Temporal changes in figures 12ii (b) & (c) show contrasting behaviour to that of nitrate; where values off the German coasts decrease from 100.01-200μg/l to 0-100μg/l during the period of 1990-1993. This is concurrent with reductions in direct discharge made by Germany (figure 10b) suggesting positive reflection to control of point source discharge. However, this response was not seen in the coastal embayment within the southern Kattegat, despite reductions of direct discharge by DK and SE (figure 10b), suggesting increased persistence of orthophosphates within the embayed area.
Figure 6(i): Geospatial Plot: Nitrates (1990-1993), showing the spatial and temporal variation of mean nitrate concentrations within the North Sea region from 1990-1993.

Figure 6(ii): Geospatial Plot: Orthophosphates (1990-1993), showing the spatial and temporal variation of mean orthophosphate concentrations within the North Sea region from 1990-1993.
In the period of 1993-2000, nitrates are still shown at elevated concentrations in coastal areas (figure 13i). Elevated values are still present in the south-east of England, concurrent with increased riverine nitrate inputs by the UK seen in figure 9(b). However, reductions are seen at the German-Netherlands boarder, where values have decreased from 5-6mg/l to 1-2mg/l, which reflect reductions in direct discharge of phosphates seen from both countries (figure 11b).

In the year 2000 (b), no elevation in coastal values can be seen, where values previously ranged from 0-6mg/l in the south east region of the basin. This is indicative of positive response to nitrate emissions controls of both atmospheric and direct discharges observed in figures 10 and 11. It is however important to note that data samples in 2000 were not collected at some sites where elevated concentrations had previously occurred, thus impeding analysis of temporal trends in north-west German and south-east UK values.

![Figure 7(i): Geospatial Plot: Nitrates (1993-2000), showing the spatial variation of nitrate concentrations within the North Sea region from 1993-2000](image-url)
Significant reductions in orthophosphate concentrations can also be seen in the period 1993-2000 (figure 13ii), showing a general decrease since 1990-1993. This can be observed at the north-west German coast, where values have decreased from 100-200μg/l to 0-100μg/l, indicative of positive coastal response to reductions in direct discharge made by Germany within this period (figure 10b). Temporal variations of orthophosphates within embayed areas from 1993-2000 are similar to those of nitrate, with reductions seen at the north-east coast of Germany from 500-600μg/l (a) to 100-200μg/l (b). However unlike nitrates, orthophosphate are still seen at elevated concentrations in the southern Kattegat, giving further evidence of phosphate residence within this enclosed area.

![Figure 7(ii): Geospatial Plot: Orthophosphates (1993-2000), showing the spatial variation of orthophosphate concentrations within the North Sea region from 1993-2000](image)

Increases in nitrate concentrations are seen in figure (Figure 14i); with higher coastal values present at south-east coast of England and the north coast of Germany in 2011 despite reductions in direct discharge made by the UK (figure 9b) and atmospheric reductions in Germany (figure 11b). It can also be seen in 2011 that elevated concentrations exist in the channel between the UK and Netherlands, again despite reductions in direct discharge from the UK (figure 10b) and river input
reductions from the Netherlands, Belgium, France and UK (figure 9b). This is suggestive of little coastal response to control measures on direct discharge and riverine input, however where these elevated concentrations occur with increased distance from the coast, it is suggestive of off-shore influence. This could be as a result of hydromorphological influence or increased atmospheric deposition, where increasing atmospheric NO$_3$ concentrations from France (figure 11b) have been seen within this period. While increases in nitrate concentrations have been observed from 2000(a) to 2011(b), values are generally lower than those in previous periods (Figure 14i), indicative of a positive overall coastal response to legislative measures.

**Figure 8(i):** Geospatial Plot: Nitrates (2000-2011), showing the spatial variation of mean nitrate concentrations within the North Sea region from 2000-2011

Orthophosphate concentrations are seen to increase within the period of 2000-2011, also showing elevated values seen between the coasts of the Netherlands and the UK in 2011. However, unlike nitrates, coastal response to emission controls of river input can be seen, where significant increase in UK river input of phosphates are reflected in coastal values during 2000-2011 (figure 9b). This influence can also be seen where increased concentrations occur in the coastal waters of Denmark,
reflective of increases in direct discharges of phosphates from Denmark within the same period (figure 10b).

While there have been coastal increases in both nitrate and orthophosphate concentrations within the period 2000-2011, values have shown an overall reduction throughout the study period, indicating positive response by participating countries during periods of legislative change. It should however be noted that the UK has continued to show elevated coastal values of nitrate and orthophosphate, reflective of the high values of direct discharge seen by the UK throughout the study (figure 10b).

**Figure 8(ii):** Geospatial Plot: Orthophosphates (2000-2011), showing the spatial variation of orthophosphate concentrations within the North Sea region from 2000-2011.
6. Discussion

6.1 Temporal assessment of nutrient emissions and associated control measures

The periodic assessment of cumulative nutrient emissions has provided a holistic overview of the response to legislative change on a European scale and has thus provided a means to evaluate the effectiveness of associated emissions controls. Within this assessment, responses to legislative change have varied between riverine input, direct discharge and atmospheric concentrations, with behavioural differences also seen between nitrate and phosphate.

6.1.1 Riverine and Direct discharge

In the period of 1993-2000, significant reductions were seen for the first time in the direct discharge of nitrates, reflecting a positive response to measures taken to reduce point source discharges by the UWWTD [MEMG, 2004; Maier et al., 2008]. However no significant reductions of nitrates were seen via riverine input, indicating little positive response to the regulation of agricultural practices in NVZ’s within this period. This differing response between sources is thought to be attributable to the pathways of nutrients; where point source discharge can be controlled at the location of discharge, whereas diffuse sources are subject to many external influences prior to reaching the aquatic environment [Scott et al., 1999]. For instance, where most agglomerations are now provided with waste water treatment facilities, the response to the UWWTD can be mostly attributed to removal of nutrients via biological and chemical treatment prior to discharge [OSPAR, 2010; Smith et al., 1999]. However, river inputs may contain nutrients from sources such as land-run off, soil leaching and atmospheric deposition, and are therefore subject to factors which influence the amount of nutrients they receive. These factors include; precipitation, land use, residence time of nutrients in soils/ sediments and geological characteristics of the surrounding area. These interactions can provide a lagged response time management change, which can differ greatly between marine regions. It is therefore important that sub-regional assessment of these interactions is carried out to gain a better understanding of the delayed response associated with emission controls; where nutrients may be released into waters after the original source has been reduced [Scott et al., 1999; Maier et al., 2008].

In the later period of 2000-2011, significant reductions of nutrient emissions were observed by both riverine inputs and direct discharges. While this indicates a positive response to the implementation of the WFD, it is also thought that this response is attributable to the combination of legislation in place; whereby the WFD classification of ‘artificial’ or ‘heavily modified’ water bodies also considers areas defined as ‘sensitive’ or ‘vulnerable’ in the Nitrates Directive and UWWTD, thus increasing the areas which legislative measures under this directive will be applied to [CIS, 2005]. This response is also thought to be enhanced by the further implementation of existing directives; where the classification of ‘NVZ’s was broadened in the UK to cover waters which “either already are or are likely to become eutrophic” leading to 55% of land being designated and thus subject to emission controls [CEC, 2002; Maier et al., 2008]. Continued reductions in point source discharge are also thought to be attributable to further implementation of the UWWTD in 1995; where ‘sensitive areas’ (subject to secondary treatment) were increased from 38 to 347 designations.
[Maier et al., 2008]. The implementation of a single basin management plan and subsequent regional frameworks under the WFD are also thought to improve the synergy of legislation in place, by encouraging international cooperation and setting ecological and chemical status parameters to be adhered to [OSPAR, 2010].

6.1.2 Atmospheric Concentrations

Behavioural differences were also seen between NO$_3$ and NH$_4$ concentrations in precipitation within the study. Where significant reductions in NH$_4$ concentrations were seen only in 1993-2010, progressive reductions in NO$_3$ were from 1993-2011. Where oxidized nitrogen is largely attributable to combustion emissions, this response is thought to be as a result of pollution control in industry and stricter emissions standards set for motor vehicles by the EU National Emissions Ceiling Directive of 2001 [OSPAR, 2010; Maier et al., 2008]. However, the lack of long term response seen in NH$_4$ has led to difficulties in assessing the response to land based management where expected response to increased areas of NVZ’s (and associated limitations of agricultural ammonia use) were not seen. This behavioural difference is thought to be due to the shorter atmospheric residence time of NH$_4$ (5hrs) compared to that of NO$_3$ (30hrs) and must be considered when assessing the response to management associated with atmospheric concentrations [NEGTAP, 2001; Matthias et al., 2010].

6.1.3 Limitations of Temporal Assessments

While cumulative assessments have been useful in determining the response to management on a European scale, individual country assessments of nutrient emissions have highlighted where European assessments can misrepresent the management on a regional level. This has been found where cumulative assessments have been representative of trends only in countries with highest emissions, however providing misleading representation of countries with lesser contribution. This was seen where the cumulative assessment of direct discharge was reflective of reductions in the UK, however did not represent significant increases that had occurred in Norway and Denmark. Future implications of such assessment should include the use of contribution-based assessment; where inputs are given proportional to the countries attributes, such as population density (e.g. kt/a per 1000people) or based on known emissions as a function of area (e.g. kt/a per km$^2$). This would provide a more accurate representation of differing regional emissions and give a more accurate indication to management success on a European level.

The individual assessment of nutrient emissions has thus proven a useful tool in evaluating responses to legislative change on a sub-regional level; indicating countries with the highest contributions of emissions and assessing the response to management on a sub-regional level. Assessing the similarities of temporal trends also allowed the evaluation of synergy between countries; where similar temporal trends are indicative of an effective response to European frameworks on a sub-regional level. This assessment also gives indication to other influencing factors on nutrient input, for instance where synchronised highs or lows of values occur in nutrient emissions, these changes may be attributable to non-anthropogenic factors, such as meteorological conditions that affect the entire region. This was seen for riverine nitrate input in 1995, where synchronized peaks were not representative of expected time-lags associated with diffuse sources, and were thus thought to be attributable to the above-normal precipitation associated with the negative phase of
the North Atlantic Oscillation during the same year [Halpert & Bell, 1996; Thomas et al., 2005].

6.2 Assessment of spatial trends in nutrient concentration

The use of geospatial plotting allowed the spatial analysis of nutrient occurrence within the North Sea, identifying areas of elevated concentrations and assessing the coastal influence of anthropogenic inputs. The use of periodic assessment was also a useful tool in assessing the temporal variations in nutrient concentrations and evaluating the coastal response associated with land based management between periods of legislative change.

Results from the geospatial plotting of winter nutrient values found elevated concentrations in coastal areas, indicative of land based input of nutrients and increased nutrient remineralisation associated with turbulent mixing of sediments [Lenhart et al., 1997; Van Engeland., 2010]. Spatial analysis also provided indication to specific areas with consistently elevated concentrations. Elevations in nitrate and orthophosphate concentrations were seen at the German Bight throughout the study, and were also shown to reflect reductions in riverine inputs and direct discharges from Germany; indicative of anthropogenic influence at the coast. It is however also thought that the Bight receives nutrients via the input of Atlantic water, which is carried by coastal currents and becomes progressively enriched by nutrients as it moves through the Channel [OSPAR, 2010].

Elevated nutrient concentrations were also be seen in the southern embayment of the Kattegat, however with further proximity from the coast than seen in open areas such as the German Bight (200km compared to 100km). However within this area, responses to significant reductions in direct discharge by DK and SE were not seen. This lack of response to anthropogenic emission controls is thought to be as a result of the physical properties associated within the embayment, whereby restricted tidal flow leads to a lack of dispersion within the embayment, and thus increased nutrient residence time [Ducrotoy and Elliot, 2008; Scott et al., 1999]. These elevated concentrations are also thought to be as a result of nutrient-rich water from the German Bight being transported along the west coast of the Jutland to the Kattegat, together with outflow from the Baltic Sea and local anthropogenic loading [OSPAR, 2010]. This effect of nutrient transport highlights the need for international cooperation and effective assessment of transitional waters, where emissions from one country may affect the nutrient status within another area.

Open areas have also shown varying degrees of response to legislative change; for instance in the south west of England, elevated concentrations have been observed throughout the study. This is thought to be a result of the initial designation of the river Thames as a ‘less sensitive area’ having ‘high natural dispersion’ and thus measures regarding the UWWTD were not taken within the initial instatement of the directive [DEFRA, 2003; OSPAR, 2005]. The Thames location also represents the need for sub-regional assessment within countries, where overall reductions of direct discharge and riverine inputs of nutrients within the UK are not necessarily representative of those in river and estuaries [Maier et al., 2008].

Further evidence of the physical influence on nutrient concentrations was observed in the period of 2000-2011; where elevated concentrations were seen off the Belgian
coast & in the channel between the UK and Netherlands. These concentrations were not reflective of the reductions in direct discharge and river inputs within these areas and are therefore thought to not be as a result of coastal anthropogenic influence. It is however though that this phenomenon is due to the interaction of the bathymetry and tidal currents within the basin [Van Engeland et al., 2010]. This interaction involves the anticlockwise circulation of water within the North Sea basin causing deflection of water entering from the English channel towards the Belgian and Dutch coasts [Ducrotoy & Elliot, 2008] This interaction leads to the transport of nutrients from the rivers Scheldt, Meuse and Rhine and thus causing elevated concentrations in the south west region of the basin [Brion et al., 2004; Ducrotoy & Elliot, 2008].

6.3 Classification of areas sensitive to eutrophic conditions

The combined use of temporal and spatial assessment has provided insight into the regional and sub-regional responses to legislative change. However it has also demonstrated that non-linearity occurs in the relationship between inland and coastal waters when assessing the effects of nutrient enrichment. This is due to the complex biogeochemical and physical interactions that occur within transitional and coastal waters, including; current-transport, residence time of nutrients in soils/sediments, meteorological influence, seasonality and flushing [Scott et al., 1999; Maier et al., 2008; Ducrotoy and Elliot, 2008]. This complexity is furthered when assessing the eutrophication status of an area; where the primary production and ecological function is dependent on other physio-chemical factors, such as oxygen levels, temperature and salinity. It is thus important to the designation of ‘sensitive areas’ that these factors are considered, alongside an understanding of the difference between ‘hypernutrified’ and eutrophic conditions, whereby hypernutrified waters may not present the ecological symptoms associated with eutrophication [Maier et al., 2008]. This provides further need for the consideration of physical factors, such as current-transport, whereby the transport of water from a hypernutrified however, may lead to organic enrichment and eutrophic conditions in another region with different physio-chemical characteristics [Maier et al., 2008]. Thus, when assessing nutrient control measures, especially those associated with diffuse inputs, it is imperative that that a multi-dimensional approach is taken, with collaborative frameworks in place that are based on contemporary scientific knowledge of the hydromorphological and physio-chemical properties associated within an area [Nimmo-Smith et al., 2007].

6.4 Implementation of legislative measures

The designation of NVZ’s and implementation of codes of agricultural practice has been found to show significant reductions in riverine nutrient emissions, while significant reductions in point source discharge have been seen as a result of the designation of areas sensitive to eutrophication and the subsequent removal of nutrients within treatment plants [Nimmo-Smith et al., 2007]. However there are still areas of concern within the North Sea region where more rigorous implementation of legislative measures are needed to meet required reductions in nutrient emissions; for example areas of high industrial or agricultural output, or areas where there is enhanced residence time of nutrients due to soil/sediment composition or restricted flow.
It is recognized that successful implementation of individual directives may require stricter guidelines; where it is argued that nitrogen emission limits are set to high (based on maximum economic potential) within certain areas and derogation clauses associated within the UWWTD allow for limited measures in areas of “sufficient carrying capacity to degrade, disperse and assimilate the Materials” [Nimmo-Smith et al., 2007; Ducrotoy et al., 2000]. Where DEFRA recognise the difficulties in monitoring diffuse nutrient sources, it has also been proposed that further implementation includes reduced stocking densities or ceasing of production all together within areas designated as sensitive to eutrophication [DEFRA, 2007; Maier et al., 2008].

6.4.1 Future implications

In order to achieve the 15 year goal of the WFD of ‘good ecological status of all ground and surface waters”, alongside the future aims of the Marine Strategy Framework Directive to “achieve good environmental status of EU waters by 2020”; it is important that synergy is achieved between legislative frameworks and that measures are applied by all member states. There are however socio-economic issues surrounding such legislative implementation, where it is argued that the success of legislative frameworks is dependent on the enforcement by local organisations to act upon recommendations set out by commissioning bodies such as OSPAR [Ducrotoy et al., 2000]. It is also argued that regional management difficulties arise in national scale frameworks where international rivers such as the Rhine and Elbe have catchments spanning into neighbouring countries which may not be included within European legislation [Salomons et al., 1989]. It is therefore important that the connections between policy drivers and the associated environmental impacts are well understood by local organisations and are set out in the context of socio-economic impact upon individual regions [Maier et al., 2008].

Nitrate concentrations within the North Sea basin were seen below the threshold of 50mg/l (in accordance with Directive 75/440/EEC) for the entire period of the study, however elevated concentrations of up to 6mg/l were consistently seen in coastal waters [EEA, 1991]. Orthophosphate values, based on the chemical classification of the WFD, were seen to generally range from ‘high’ to ‘good’ status in open waters (0-120μg/l) and ‘moderate’ to ‘poor’ status in coastal regions (150-600μg/l) [UK TAG, 2008]. While reductions in nitrate and phosphate concentrations have been observed (with many values below legislative thresholds), it is important to note that the values in coastal regions are representative of ‘dispersed’ values. Thus, where elevated concentrations occur, they are indicative of the influence from inland surface and ground waters which may still not meet ecological and chemical standards. This highlights the need for assessment of transitional and inland water bodies, in order to bridge the gap of knowledge between nutrient emissions and coastal concentrations and provide a better understanding of the interactions between the two.

It is therefore imperative that with developments of legislation and the addition of future directives, further monitoring of coastal and transitional waters will be implemented to gain a between understanding of these interactions on a sub-regional level. This is hoped to be seen with the further assessment of hydromorphological quality elements in estuaries and coastal waters within the WFD [WFD UK, 2012], alongside objectives of the Marine Strategy Framework Directive; to consider the socio-economic and physiochemical factors associated within marine regions.
7. Conclusion

The use of spatial and temporal analysis of nutrient occurrence has been a useful method in assessing the human impacts on nutrient emissions and evaluating the response to legislative change. Temporal analysis of cumulative emissions has revealed that significant reductions in nitrate and phosphate inputs have been made on a European scale. However, individual country assessment has shown that response to European legislation has been varied amongst member states, and has thus indicated the differences in effectiveness of management on a regional scale. Response to legislation has also varied between emission sources; where reductions in direct discharge have been found more significant than those of riverine input and atmospheric concentration. This has been attributed to the differences in management effectiveness associated with diffuse and point sources, highlighting the complexity of interactions associated with diffuse sources. The use of statistical analysis between implementation years has also allowed the quantified assessment of response to emission controls within periods of legislative change. The most significant emission reductions have been seen in later periods of assessment, indicating the greatest response to combined legislative implementation and the further development of directives. This has been largely attributed to the increase in areas that legislative measures are applied, due to broadening of classification by directives. It has thus been concluded that to further reduce nutrient emissions, it is imperative that synergy is achieved between legislative frameworks, and that the measures associated with individual directives are applied rigorously across all member states.

Periodic assessment of geospatial data has proved a useful tool in detecting areas of elevated nutrient concentrations and assessing the coastal response to land based emission controls within periods of legislative change. This assessment also provided increased spatial resolution regarding the emissions of individual countries; where elevated coastal concentrations have given indication to the emissions within sub-regions of countries. Coastal response to direct discharge, riverine input and atmospheric concentration has also been found to vary between different regions of the North Sea. This has shown the non-linear relationship between coastal waters and land-based emission control, and has highlighted the complexity of interactions associated with the coastal zones.

It has thus been concluded that in order to effectively further assess the human impacts on nutrient concentrations in the North Sea; it is necessary to evaluate the nutrient concentrations in transitional and coastal waters on a sub-regional scale. This is required to gain a better understanding of the hydromorphological and physio-chemical characteristics associated within regions and further evaluate the response to European legislation on a sub-regional scale.

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Appendices for this work can be retrieved within the Supplementary Files folder which is located in the Reading Tools menu adjacent to this PDF window.