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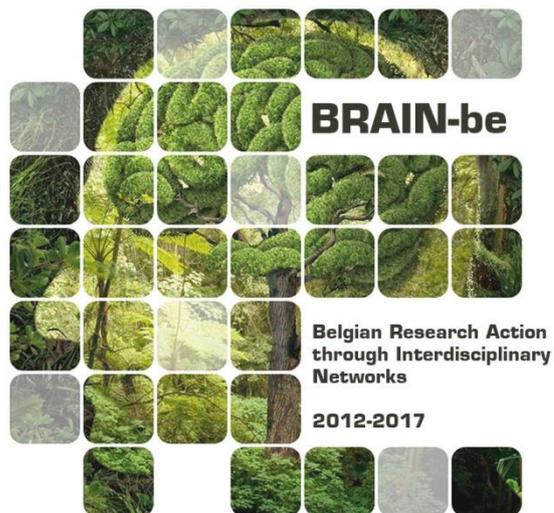
4DEMON

4 Decades of Belgian Marine Monitoring: Uplifting historical data to today's needs

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Axis 3: Cultural, historical and scientific heritage





NETWORK PROJECT

4DEMON

4 Decades of Belgian Marine Monitoring: uplifting historical data to today's needs

Contract - BR/121/A3/4DEMON

FINAL REPORT

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ABSTRACT

Context

Long-term quality checked and integrated datasets for the Belgian Part of the North Sea (BPNS) are essential to detect changes in this complex ecosystem and support policy related decisions.

Objectives

- Compile and safeguard quality checked, intercalibrated and integrated datasets and make them publicly accessible for further research and policy purposes.
- Improve or develop methods and protocols to assess and interpret environmental change in the BPNS and compare trends with neighbouring areas.
- Provide support and advice for policy related decisions and legal measures, like MSFD and OSPAR.

Conclusions

A scheme with data management tools has been worked out for efficient data flow throughout the project. The scheme, including the mandatory metadata fields and standardization, can be used as a guideline for future projects. Inventories of datasources, projects and data-origimators were compiled and the final datasets are available via the central datportal.

Even after compiling and quality checking the long-term datasets, the scientists had to consider some limitations, like changing methodologies and low data resolution, and incorporate these into their trend analyses procedures. Some remarkable environmental changes over time were observed.

Model results of the contaminants showed decreasing trends for heavy metals and PCBs. Zn concentrations, however, were found increasing in marine sediments while As concentrations were found increasing in groyne mussels. Nearby the port of Zeebrugge and the mouth of the Scheldt, PCB concentrations were found slightly increasing again over the last decade.

The study on eutrophication showed that nutrient (N, P) riverine concentrations and loads have decreased continuously from the end of the 1980's to now. However, this did not result in a comparable decrease of marine nutrient concentrations. No clear long-term trend effect was observed on in situ chlorophyll *a* concentrations following the nutrient decrease. However, a clear change in chlorophyll *a* phenology followed the increase in sea surface temperature in the period. Furthermore, biomass, seasonality and structure of diatom and dinoflagellate communities were compared between the 1970s and 2000s for the Belgian Part of the North Sea (BPNS), derived from the newly established Belgian Phytoplankton Database. Distinct changes were observed: changes in diatom and

dinoflagellate biomass and shift of fulcrum; changes in community structure, with a trend towards seasonal homogenization in the diatom community; increased occurrence of harmful diatom (*Pseudo-nitzschia*) and dinoflagellate (e.g. *Prorocentrum*) genera. The observed changes correlate well with overall increases in temperature and changes in nutrient loads and ratios.

And finally, for acidification, pH data reveal an increasing trend from the mid-70's to the mid-80's and a decrease of pH from the mid-80's onwards that seems consistent with changes in primary production patterns. The comparison of CH₄ concentrations obtained in 1990 and 2016, showed a decreasing trend consistent with alleviation of eutrophication in the area.

Based on the outcome of this successful project, the partners defined various recommendations regarding future monitoring strategies for policy makers.

Keywords (max 5)

Contamination, eutrophication, ocean acidification, Southern North Sea, long-term trends

ACCRONYMS and ABBREVIATIONS USED

AA	Auto Analyzes
BPNS	Belgian Part of the North Sea
CDI	Common Data Index
CSR	Cruise Summary Report
EDMERP	European Directory of Marine Environmental Research Projects
EDMO	European Directory of Marine Organisations
EO	Earth Observation
GES	Good Environmental Status
GHG	Greenhouse gas
ICES	International Council for the Exploration of the Sea
IDOD	Integrated and Dynamical Oceanographic Data management
IS	In Situ
MSFD	Marine Strategy Framework Directive
NERC	Natural Environment Research Council
NODC	National Oceanographic Datacentre
OSPAR	Oslo and Paris Convention
PMPZ	Projet Mer – Projekt Zee
PMPZ-DBII	PMPZ - Resuscitation of the data collected during the first years of modern oceanography in Belgium (1970-1982)
PSI	Public Sector Information Directive
RWS	Rijkswaterstaat Nederland
SDC	SeaDataCloud
SST	Sea Surface Temperature
WFD	Water Framework Directive
WoRMS	World Register of Marine Species
WP	Work Package
WWT	Urban Waste Water Treatment Directive

Remark: for codes of parameters, see Annex 2.

1. INTRODUCTION

Since the sixties, general awareness grew of the human induced negative impact on the health of the environment. The increasing production of hazardous and toxic chemicals, such as heavy metals and PCBs resulted in an accumulation of concentrations in marine sediments and biota (Roose, 2005). At the same time, it was found that high nutrient contents in coastal waters promotes eutrophication and by this can lead to biodiversity loss and fish kills (Cloern (2001), Worm et al. (2006)). Seasonal phytoplankton cycles in coastal waters can be modified by changing surrounding conditions which may affect the onset of the bloom, the bloom timing, the length of the bloom and its magnitude (Cloern and Jassby (2008), Kromkamp and Van Engeland (2009), Lewandowska and Sommer (2010), Winder and Cloern (2010)). Furthermore, temperature changes and light regimes are influencing the phytoplankton biomass and community composition Boyce and Worm (2015), Capuzzo et al. (2015), Paerl et al. (2011; 2014), Rühland et al. (2008), Suikkanen et al. (2013)). On top of that, ocean acidification can alter the rates and fates of primary production and calcification of numerous marine organisms and communities (Kleypas et al. 2006, Doney et al. 2009), and thus alter the ocean's carbon sequestration capacity, marine biodiversity and marine ecosystem services and goods. Indeed, some of these calcifying organisms such as bivalves are important economic resources (fishery) and constitute important resources for marine birds (Gutiérrez et al. 2003).

Over the years many policy measures have been taken at international and regional level to bring the human impact on the ecosystem to a minimum. For example, within the Marine Strategy Framework Directive (MSFD; Directive 2008/56/EC), the objective is to achieve a Good Environmental Status (GES) by 2020 (Belgische Staat, 2018). However, there are only few datasets that enable us to perform long-term trend analyses in the BPNS going back to the 1970s. As eutrophication and contamination are both qualitative descriptors (Descriptor 5 and 8) to evaluate the GES, and ocean acidification has a high impact on the health of our food web (Descriptor 4), we found the high need to compile robust centralized and accessible datasets on these themes and make them available for further research and policy makers.

Between 1971 and 1976 the large scaled *Projet Mer/Projekt Zee* (PMPZ), where over 200 scientists integrally studied the Southern North Sea, set the starting point of modern oceanography. Since 1978 the Paris convention (becoming the OSPAR convention in 1992) ensured that the member states set up systematic monitoring programs in their area within the North-Atlantic Ocean. Many other research projects followed. However, data management infrastructures, where standardization is key, were not yet established as today, and a lot of data were inaccessible and scattered around the country. Many data from 1971 until 1982 were already rescued in the project PMPZ-DBII, where it was concluded that intensive quality controls and intercalibration exercises were required to make the historic data comparable (Lagring et al., 2012 a). The seventies were therefore defined as a starting point for 4DEMON.

The compiled intercalibrated and quality controlled long-term datasets on contamination, eutrophication and acidification are of high value for the scientific community and policy makers, as they are made centrally accessible via one data portal and further disseminated at international level. Furthermore, the results of the trend analyses to detect changes in the BPNS provide best practices to analyse the datasets where historic data are integrated with newer data. Thus, uplifting historic values to today's needs, which are mainly to define the human impact on the environment on the one hand and to evaluate whether taken policy measures have been effective on the other hand.

2. STATE OF THE ART AND OBJECTIVES

To reduce the negative impact of human induced changes in the vulnerable marine ecosystems, many measures have been taken at regional and international level. In 1978, the 'Paris Convention for the Prevention of Marine Pollution from Land-Based Sources', meant the start of long-term strategic monitoring in the BPNS. In 1992, this became the OSPAR Convention for the Protection of the Marine Environment of the North-East Atlantic. At European level, many directives were adopted since the 1990s to further control the reduction of human induced environmental damage. For example, the Urban Waste Water Treatment (WWT) Directive (91/271/EEC); the Nitrates Directive (91/676/EEC); the Directive on the regulation on packaging and packaging waste (*i.a.* Heavy metals) (94/62/EC) in 1994; the Directive on the disposal of polychlorinated biphenyls and polychlorinated terphenyls (PCB/PCT) (96/59/EC) in 1996; the Directive on the quality of water intended for human consumption (98/83/EC) in 1998 and the Detergents Regulation (*i.a.* Phosphates) (648/2004) in 2004. Two framework directives, the Water Framework Directive or WFD (2000/60) and the Marine Strategy Framework Directive or MSFD (2008/56/EC), integrated several existing European regulations for water quality of lakes, rivers, marine, transitional and coastal waters (waste water, fishing water, drinking water, bathing water *etc.*). The target of both directives is to bring our waters to a desired good ecological status (GES), which is within the MSFD to be achieved by 2020 (Van Hoey et al., 2010; McQuatters-Gollop A, 2012). This is being evaluated based on ten descriptors, *i.a.* eutrophication (Descriptor 5) and contamination (Descriptor 8) (Belgian State, 2018). For these descriptors, each member state must define common indicators for its regional sea. Therefore, profound knowledge of its ecosystem is indispensable.

The Belgian federal government is responsible for the correct implementation of these directives in the Belgian law. Therefore, in the BPNS, the state of the ecosystem and the environmental effect of installations like windmills, dumping and dredging activities, aquaculture *etc.* are carefully monitored by means of systematic measuring campaigns for specific indicators in water, sediment, biota and species abundances, resulting in a vast amount of data since the 1970s.

Today, public access to data is indispensable thanks to European legislation. The Directive on Public Sector Information (PSI) (2003/98/EC) ensures that public information is readily and widely accessible. Marine data collection, often funded by the government, involves high costs for the vessel, crew, material, laboratories, scientists, data handling, *etc.* It is therefore our duty to make research and monitoring programs as cost-efficient as possible. Unfortunately, a lot of valuable data are still inaccessible, hidden on personal computers or even in logbooks (the so called 'dark data'). They can be non-public, originate from private research projects or industry, or date back from before the open data policy. Especially the historic data, dating back from before modern data management infrastructures, are at risk of getting lost. Therefore, several initiatives (e.g. EMODnet Data Ingestion) are being set up to increase data accessibility of so far locked or hidden data. Other factors that hamper long-term trend detection, next to inaccessibility of data, are changes in sampling strategies and

methodologies (making the data incompatible), lacking meta information (making the data unreliable) and gaps in time and space of measured parameters and co-factors.

To study spatio-temporal changes in the BPNS, to evaluate the situation of this complex coastal ecosystem and to monitor the effectiveness of all the above-mentioned measures, as much information as possible is needed. Indeed, the further one can go back in time, the more one can distinguish and understand the human induced harm versus the natural dynamisms within an ecosystem. Therefore, long-term integrated and accessible datasets covering the relevant indicators are indispensable, however very scarce. On the one hand, there is thus need for tracking back as much data and relevant information as possible to build robust and integrated datasets. On the other hand, protocols are required to perform intensive quality checks and procedures to overcome changes, like intercalibration or normalization scripts, rendering historic data comparable with more recent results. The quality-checked and intercalibrated datasets can then form the basis of integrated statistical analyses to assess and interpret environmental changes in the BPNS. This opens up extended possibilities for research on long-term marine environmental change and provides information to Belgian policy makers for developing effective coastal management and sustainable development strategies in line with societal, economic and ecological needs and international obligations.

The general objective of the project was therefore to build quality checked, intercalibrated and integrated long-term datasets on contamination, eutrophication and ocean acidification, which are important indicators to study the health of the complex marine ecosystem, and use these data for assessing environmental changes in the BPNS, starting with the monitoring data from the 1970s.

To evaluate whether the above objective has been met, three pillars or sub-objectives were defined.

- Pillar 1: Identify, safeguard (historic) monitoring and research data, and increase public availability of and accessibility to quality controlled long-term datasets.
- Pillar 2: Adapt or improve methodologies and protocols to study long-term environmental changes in the BPNS.
- Pillar 3: Provide policy support and justify relevance of selected themes.

Based on these three pillars specific recommendations were defined towards monitoring and policy makers (see Chapter 4).

3. METHODOLOGY AND RESULTS

3.1. Project overview

3.1.1. Project structure and Workflow

The project structure (Fig. 1) ensured efficient cooperation between the data managers WP 2 (Work Package) and the researchers of the three thematic topics (WP 3-5). The separate WP on data analyses enabled the researchers to have a framework to work interdisciplinary. The overall coordination, including follow-up of the planning and organizing annual meetings, was trusted to BMDC with input of all partners.

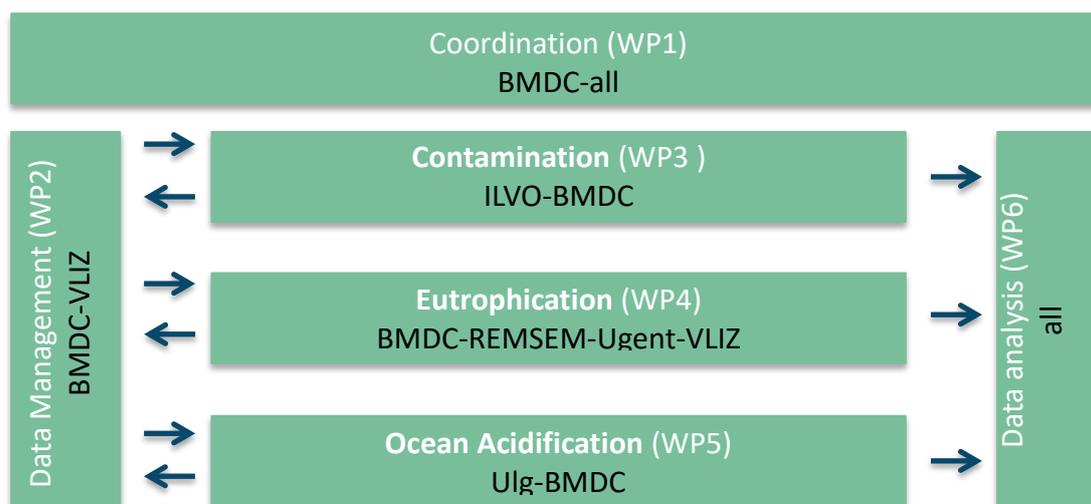


Figure 1: Project structure

Definition of tasks and responsibilities

WP 3-5, which are devoted to the actual development of the integrated and intercalibrated data bases for each specific theme, were in close interaction with WP 2 which streamlines data management from the start of the project by offering common and central data management tools to the partners, from data inventory to database and data dissemination. Within WP6, all information was used in integrated statistical analyses, analyzing and comparing the established time series and comparing with trends in adjoining coastal areas.

Overall timing

The time table (Table 1) of the project, provided in the proposal, was used to carefully follow-up the annual progress of each assigned task. During the first year, identification and gathering of data sources and missing information took place. Based on the missing information, additional intercalibration or sampling exercises were planned, and additional literature was consulted. In the second year, additional sediment samples were taken within WP3. Meanwhile data digitization and import in the database continues and preliminary quality checks were performed. At the same time, data analysis started. In the third year, new analyses were finalized. Data were thoroughly checked and used for intercalibration, normalization and data binning where necessary. During the last year, the evaluation of

environmental change took place. Throughout the project, results, when ready, were disseminated on the website, publications and conferences. As is shown from the table, all foreseen tasks have been dealt with, within the timeframe of the project. Dissemination, of course, is a continuous process and is therefore marked as 'ongoing', together with some import activities.

Table 1: Overview tasks and planning of the project

WP	Task	Year 1	Year 2	Year 3	Year 4	+ 6 mnd	Status
		End 2013	End 2014	End 2015	End 2016	Mid 2018	
WP1	WP1. Coordination						
Task 1.1	Reporting Activities	x	x	x	-->	x	ok
Task 1.2	Annual Meetings	x	x	x			ok
Task 1.3	Technical meetings and workshop						ok
Task 1.4	Dissemination	x	x	x	-->	x	ok
	- <i>Website</i>		x	x	-->	x	ok
	- <i>Publications and presentations</i>		x	x	-->	x	ok
	- <i>Outreach</i>		x	x	-->	x	ok
WP2	WP2. Data Management						
Task 2.1	Archive & Invento	x	x	x			ok
	- <i>Inventory</i>						
	- <i>Archive</i>	x	x	x			ok
Task 2.2	Data formats & pr	x					ok
	- <i>In situ</i>						
	- <i>Diversity</i>	x					ok
	- <i>Along Track(ODAS, MIDAS)</i>			x			ok
Task 2.3	Data Integration			x	-->	x	ongoing
	- <i>IDOD</i>			x	-->	x	ok
	- <i>IMERS and Aphia</i>			x	-->	x	ok
	- <i>GIS</i>						ok
Task 2.4	Dissemination an				-->	x	ongoing
	- <i>Dissemination (data portal)</i>				-->	x	ongoing
	- <i>International</i>				-->	x	ongoing
	- <i>Other</i>				-->	x	ongoing
WP3	WP3. Contamination						
Task 3.1	Inventory	x	x	x			ok
Task 3.2	QC & Intercalibrat	x	x				ok
	- <i>Missing metadata</i>						
	- <i>Conversion due to analytical differences</i>						ok
	- <i>Normalisation</i>			x	-->	x	ok
	- <i>Spatial and temporal calibration and databinning</i>			x	-->	x	ok
WP4	WP4. Eutrophication						
Task 4.1	Inventory	x	x	x			ok
Task 4.2	QC & Intercalibrat	x	x	x	-->	x	ok
	- <i>Taxonomy phyto- and zooplankt</i>						
	- <i>Biomass phyto- and zooplankton</i>	x	x	x	-->	x	ok
	- <i>Nutrients and turbidity</i>		x	x	-->	x	ok
WP5	WP5. Ocean Acidification						
Task 5.1	Inventory	x	x	x			ok
Task 5.2	QC & Intercalibration	x	x	x			ok
WP6	WP6. Data analysis and integration						
Task 6.1	Trend analyses			x	-->	x	ok
Task 6.2	Trend comparison: integration			x	-->	x	ok
Task 6.3	Comparison North Sea areas			x	-->	x	ok

3.1.2. Project website

In the first year, the project website (www.4demon.be) was set up. It contains information on the project, the involved partners and the research topics. A news page gives an overview of important events related to the project. Furthermore, it provides access to the metadata catalogue, the product gallery and the data portal (see Chapter 3.2.1.4).

3.1.3. Dissemination activities

During the four year the project was running, the partners were very active in disseminating the progress or their work and results during multiple events (like attending workshops and conferences) and publications (in scientific and informative research journals). Below, an overview is given of these activities. More detail on these (past) activities is provided under Chapter 5 (Dissemination and valorization). Of course, as dissemination is a continuous process, more activities will occur in the future where the outcomes of the project will be promoted. For example, a general publication is planned in the Science Connection magazine of Belspo, there will be a presentation at the European Marine Biology Symposium (EMBS) in Ostend (September 2018) and publications for Chemosphere are in preparation.

Participation in Conferences:

European Marine Biology Symposium (EBMS), 17-21 September 2018, Oostende, Belgium

4th International Symposium on Research and Management of Eutrophication in Coastal Ecosystems (EUTRO 2018), 18-20 June 2018, Nyborg, Denmark

50th Liège colloquium on Ocean Dynamics, 28 May – 1 June 2018, Liège, Belgium

VLIZ Marine Science Day 2018, 21 March 2018, Bredene, Belgium

Ocean Sciences Meeting 2018, 11-16 February 2018, Portland, USA

EGU General Assembly, 22-28 April 2017, Vienna, Austria

North Sea Open Science Conference, 7-10 November 2016, Ostend, Belgium.

IMDIS 2016, 11-13 October 2016, Gdansk, Poland.

VLIZ Marine Scientist Day, 12 February 2016, Brugge, Belgium

IODE Scientific Conference, 16-20 March 2015, Brugge, Belgium

47th International Liège colloquium, 4-8 May 2015, Liège, Belgium

VLIZ Young Scientist Day, 20 February 2015, Brugge, Belgium

Participation in working groups and workshops:

ICES DIG meeting, May 2018, Copenhagen

Webinar Delft3D Phytoplankton modelling: concepts of bloom, March 2016

BEDIC workshop, 7 December 2015, Brussels

LifeWatch Data Analyses Workshop, 26-27 November 2015, Ostend

World Ocean Day, RBINS, 7 June 2015, Brussels

ICES working Group on Marine Sediments in relation to pollution (WGMS), 2 March 2015

OSPAR-ICES Study Group on Ocean Acidification, 6-9 October 2014

Marine Chemistry Working Group (ICES), 5 March 2014

Publications:

Scientific journals

Nature Scientific Reports

Ecosystems

Science Data

Informative journals and press

De Grote Rede

3.2. Data management

4DEMON is a data rescuing project submitted in the frame of the Brain.be axis 3: “Cultural, historical and scientific heritage”. As the amount of expected data to process from multiple (historic) projects was overwhelming, it was important to ensure the most efficient dataflow. Therefore, the methodologies involved in the project mainly related to data management and analyses. Two out of five partners were specialized marine data managers (BMDC and VLIZ), supporting the research partners in all data related issues (general data management, data integration and dissemination). To streamline the tasks within this WP, a dataflow scheme (Fig. 2) was worked out, including the provided data management tools, the processes and the databases used for centralizing and safeguarding all compiled data. Thanks to the project scheme (Fig. 1), it was clear that there was a close interaction with the other WPs. Both data management partners are involved in renowned international data infrastructure networks, ensuring the most adequate data dissemination activities by integration into various systems and making the data world-wide accessible via multiple platforms.

3.2.1. Data flow: data compilation, integration and dissemination

The dataflow scheme, worked out by the data managers at the start of the project is shown in Fig. 2. Each step is explained in more detail below. The scheme was shown very complete and efficient and can be used as such as an example and dataflow guideline for future projects on data rescuing. Furthermore, this scheme can be included as part of the data management plan of any other research and monitoring project in the BPNS, as they practically always involve data compilation and are thus dealing with all included steps.

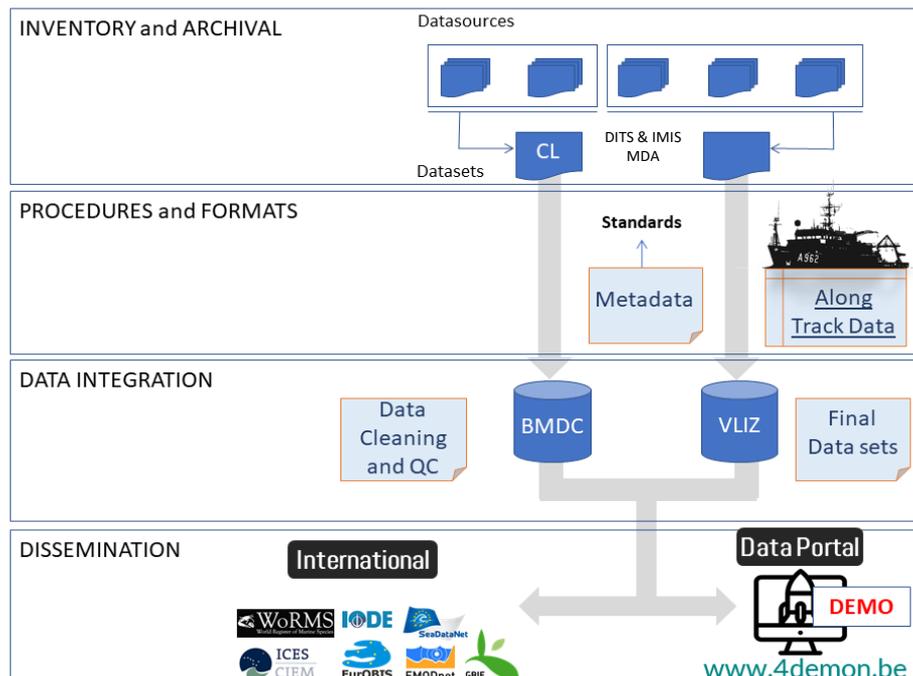


Figure 2: Data flow scheme linking all steps confided to the data managers (from inventory to dissemination) including all data management tools and databases of both datacentres.

3.2.1.1. Data compilation: archival and inventory

The initial and major task within each WP was focussed on data compilation, involving archival and inventory. First, all possible data sources related to the projects themes needed to be recovered from archives, computers and databases available at the partners offices or external institutes. Secondly, the data sources needed to be digitized (e.g. reports in paper format were scanned and handwritten logbooks were typed in spreadsheet format), processed and prepared for integration. The data managers provided a data tracking tool to inventory and follow-up the digitization and further integration process (DITS) and an archival environment (MDA):

- The Data Inventory and Tracking System (DITS) (<http://dits.bmdc.be/>) was developed by BMDC within the scope of 4DEMON to inventory all datasources, submit data, generate ingestion and public datasets, identify missing (meta-)data and gaps, and track the import status in the databases. Today the tool is also used for MSFD data submission. More information on the technicalities are available in Annex 6.
- A file archival environment within the Marine Data Archive (MDA) (<http://mda.vliz.be/>) was set up by VLIZ. This platform was used both to share data files and project documents among project partners.

3.2.1.2. Procedures and formats

- Data exchange formats to the datacentres

To transfer the in-situ data compiled by the partners from the identified datasources to the datacentre, the data reporting formats or templates were revised and simplified if possible. These are spreadsheets that vary per datatype (e.g. water, sediment, biota) and make sure that all data together with its (mandatory) metadata (see below) is structured in a way that it can be used for further integration in the central databases.

- Data exchange routine for 'en route' data

The data portal contains links to the en-route data of the MIDAS system (<http://rshiny.lifewatch.be/Underway%20data/> for the RV Simon Stevin) and ODAS system (<https://odnature.naturalsciences.be/belgica/nl/index> for the RV Belgica).

The complete data of the ODAS system is available on request to info@bmdc.be. A batch routine harvests the ODAS database for BMDC in a more practical format, ready for querying, making the ODAS RV Belgica data more accessible. More details on the procedure are available in Annex 4.

3.2.1.3. Data integration

3.2.1.3.1. Data cleaning: quality control and standardization of metadata

Before the raw data could be integrated into the central databases of VLIZ and BMDC, common quality control checks (see Table 2) were performed by R-scripts. Most partners built for this purpose a more flexible intermediate work data base where all compiled data were initially integrated in its raw form with sometimes incomplete however required metadata (e.g. without geographical coordinates, analyses methods, units). From this point, some required conversions could be performed (see below) and the mandatory (meta)data could be further completed and standardized. If needed, the data-originators were contacted or more information was gathered, until all data were ready for final integration in the central databases. An overview of such metadata fields is provided in Table 3. Marine data management standards used are *i.a.* common vocabularies of NERC (as adopted by the EmodNet partnership and the SDC consortium - <https://www.seadatanet.org/About-us/SeaDataCloud>), ICES platform codes and WoRMS taxonomy codes. These standards ensure that the data is interoperable in an international context.

Table 2: conversions and standard quality checks required before integration in the central databases.

Basic conversions of fields	Standard quality checks
- Conversion of units	- Duplicate checks
- Geographical coordinates	- Range checks
- Taxonomy mapping and AphiaID retrieval (http://www.marinespecies.org/)	- Ensure correct datatype and format of the fields (e.g. number, date...)
	- Exclude missing data

Table 3: Required metadata and standards before integration into central databases.

Subject	What metadata	Standard lists
Project	Project Name	EDMERP
Organization	Organization Name	EDMO
Campaign	Platform start- and end date Geographical coverage or bounding box Water depth	CSR ICES platform code
Sampling	Start- and end date Duration Sample depth Start and end latitude and longitude Station Sampling gear Volume	CDI ICES station list SDN-NERC Vocabulary
Sediment	Grain size fraction Upper and lower depth	
Species	Biodiversity: taxonomy Tissue Body length and weight	ERMS/WORMS
Values	Parameters Units (incl. dry weight vs wet weight) Matrix Replicates Detection limit	CDI SDN-NERC Vocabulary
Analyses methods	Laboratory Description Instrument Date Sample handling (pre-treatment, separation, preservation)	

3.2.1.3.2. Data integration in central databases

Contaminants in sediment were directly imported in the central database of BMDC (IDOD). Contaminants in biota and all abiotic data, including the data already in IDOD, were compiled in an intermediate database before integration of the quality controlled datasets in IDOD. The advantages of these intermediate databases were that, because of the high variety in projects with sometimes overlapping data, duplicate checks and identification of erroneous data could first be performed

Figure 3 gives an overview of the count of values compiled during 4DEMON. Monitoring data (MONIT) and public research data (OTHER) are data that were already mainly accessible. Data of the Bagger monitoring (BAGGER) were centralized but not yet accessible. In red (4DEMON) are data that have been centralized and made accessible. Figures 4-6 give an overview of the parameters in biota, sediment and water compiled in 4DEMON.

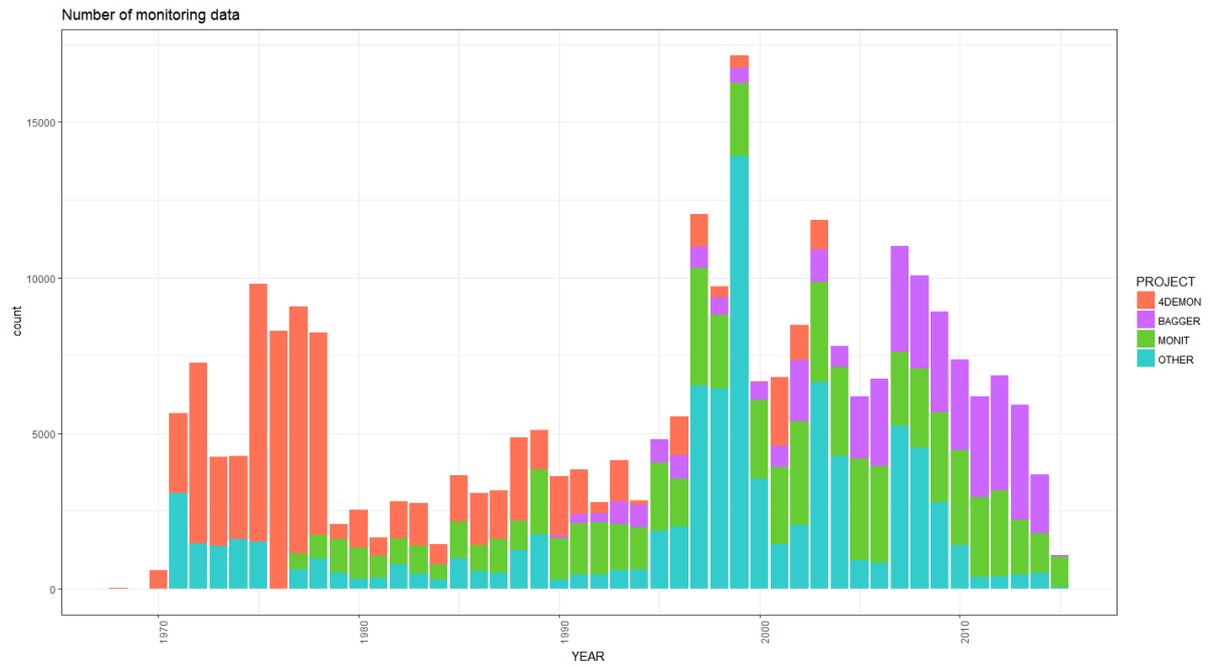


Figure 3: Overview of the count of values compiled during 4DEMON.

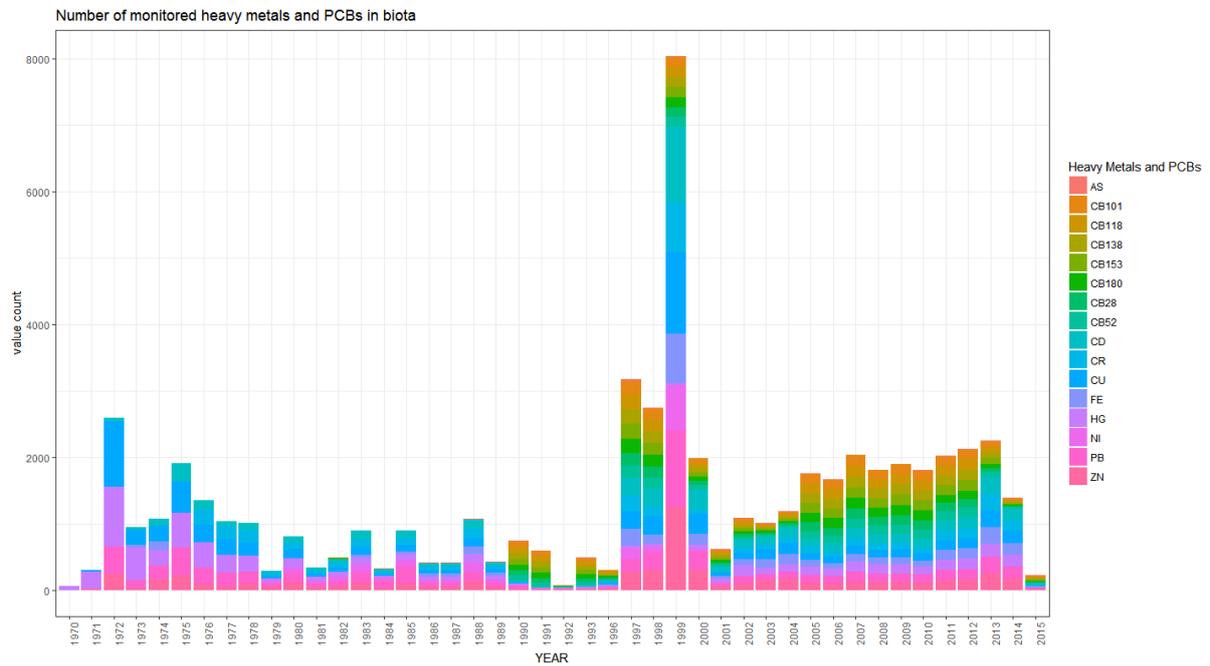


Figure 4: Overview of parameters in biota compiled during 4DEMON.

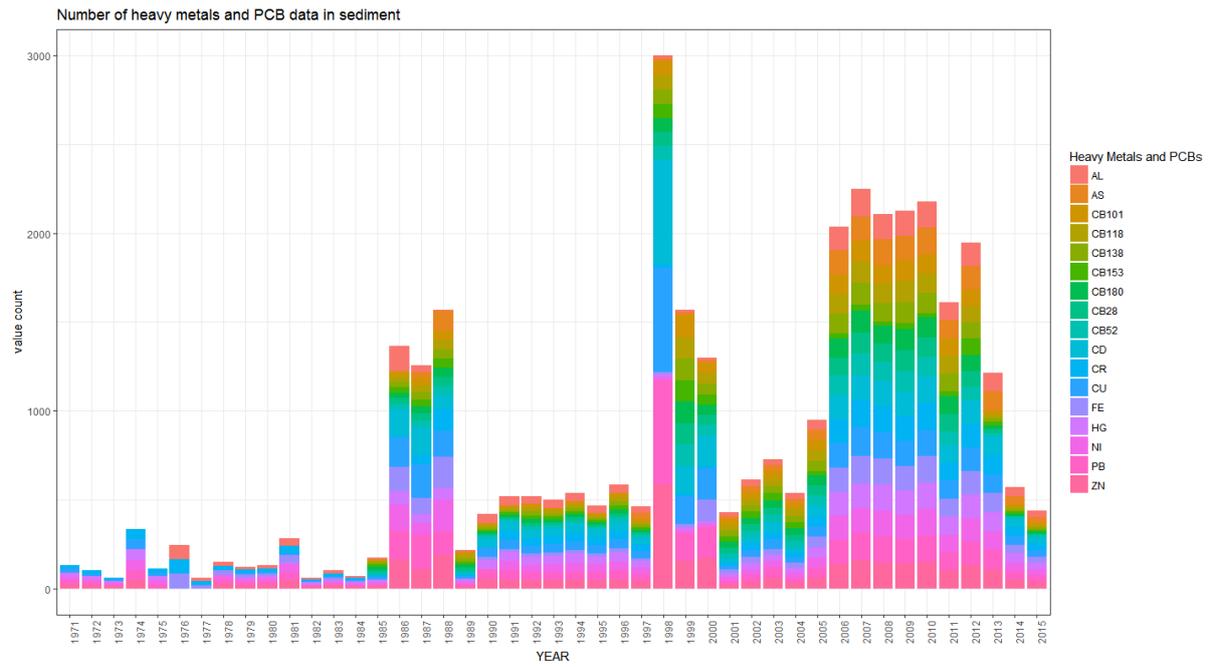


Figure 5: Overview of parameters in sediment compiled during 4DEMON.

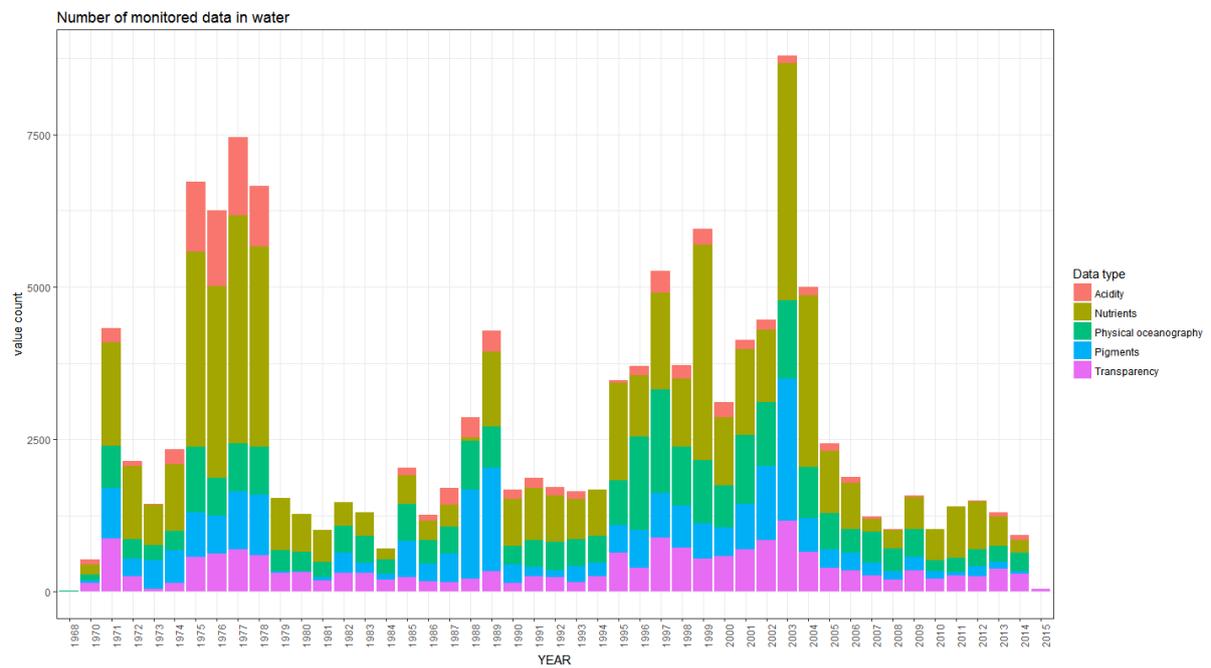


Figure 6: Overview of parameters in water compiled during 4DEMON.

3.2.1.3.3. Final (public) datasets

The final datasets are online available via the 4DEMON catalogue (see 3.2.1.4.1.) and the data itself can be accessed via the data portal (see 3.2.1.4.2). An overview of the projects, datasets and data-contributors is given in Annex 1. An overview of the Parameters can be found in Annex 2.

3.2.1.4. Data dissemination

3.2.1.4.1. Website and catalogue

- Public catalogue on projects and datasets

A special collection '4DEMON' was added to the Integrated Marine Information System (IMIS) hosted by VLIZ. This catalogue was populated with metadata of relevant 4DEMON publications, datasets and projects. A plugin of this system is used on the website.

- Data product portal

A product gallery was set up on the 4DEMON website (Fig. 7). This gallery enables to highlight project outcomes.



Figure 7: Data product portal.

3.2.1.4.2. Public central data portal

A dedicated dataportal was developed by VLIZ. This portal allows access to public datasets, where the data is dynamically queryable from the central databases at both VLIZ and BMDC. Technical specifications can be found in Annex 5.

The data portal offers following functionalities:

- Selection of data based on different criteria: Sources, Geographical area, Time, Taxonomy, Abiotic parameters.

- Tabular view of selected data.
- Different download formats possible.
- Save data selection URL (all selection criteria included).
- Create webservice URL for integration in workflows (e.g. R).
- View selected data points on Map Viewer.

3.2.1.4.3. Dissemination via international marine data management initiatives

Both BMDC and VLIZ are National Oceanographic Data Centres (NODCs) designated by the International Oceanographic Data and Information Exchange (IODE) (<https://www.iode.org/nodc>). These NODCs are active partners of renown international marine data initiatives ensuring the international data dissemination via following initiatives:

- SeaDataCloud (SDC, <https://www.seadatanet.org/About-us/SeaDataCloud>) is a standardized infrastructure for managing the large and diverse datasets collected by the oceanographic fleets and the automatic observation systems. It connects over 100 datacentres aiming at preserving and making re-useable marine observations ranging from ocean physics to chemistry and biology. The metadata resulting from 4DEMON (mainly Organizations via EDMOs, cruises via CSRs and projects via EDMERP) will be incorporated in this infrastructure. Also, the data will be disseminated via the Common Data Index (CDI) service, which provides a unique interface for requesting access, and if granted, for downloading datasets from the distributed datacentres across Europe.
- The European Marine Observation and Data Network (EMODnet, <http://www.emodnet.eu>) is a network of organisations supported by the EU's integrated maritime policy. These organisations work together to observe the sea, process the data according to international standards and make that information freely available as interoperable data layers and data products. The EMODnet portals provide easy access to marine chemical data, standardized harmonized validated data collections and reliable data products. The resulting datasets from 4DEMON will be disseminated via:
 - Emodnet Chemistry (<http://www.emodnet.eu/chemistry>): all data on nutrients, pigments, heavy metals, physical measurements (via the abovementioned CDIs).
 - EmodNet Biology (<http://www.emodnet-biology.eu/>): all phytoplankton biodiversity data.
- Ocean Biogeographic Information System (OBIS, <http://www.iobis.org/>) is a global open-access data and information clearing-house on marine biodiversity for science, conservation and sustainable development.
- The World Register of Marine Species (WoRMS, www.marinespecies.org) is the official taxonomic reference list for OBIS: All required taxonomic information are be mapped with WoRMS taxonomic reference list. New species will be integrated.
- Global Biodiversity Information Facility (GBIF, <https://www.gbif.org/>) is an international network and research infrastructure funded by the world's governments and aimed at providing anyone, anywhere, open access to data about all types of life on Earth.

- The Ocean Data Interoperability Platform (ODIP, (<http://www.odip.eu/>)) aims at the effective sharing of data across scientific domains and international boundaries by disseminating best practices and transferring knowledge and technology. The developments within 4DEMON regarding data sharing and standardization (DITS, ODAS data accessibility, central dataportal) will be further disseminated via this platform.

3.2.2. An example: The Belgian Phytoplankton Database

As an example of the 4DEMON data flow, the workflow is given (Fig. 8) for compiling the BPD or Belgian Phytoplankton Database (*cf.* Chapter 3.4.3), containing phytoplankton count data and its metadata (Nohe et al., 2018). After inventorying all relevant data sources from multiple projects and studies, the non-digital data were digitized and recovered. Several quality checks were conducted on the dataset, among which the taxonomic intercalibration. During the last decades, there were many extensive nomenclatural and other taxonomic revisions of phytoplankton taxa. For this reason, species names needed to be referenced. This was done using the taxon match option available in WoRMS. The database can be downloaded via this link: <http://www.vliz.be/en/imis?dasid=5717&doiid=320>.

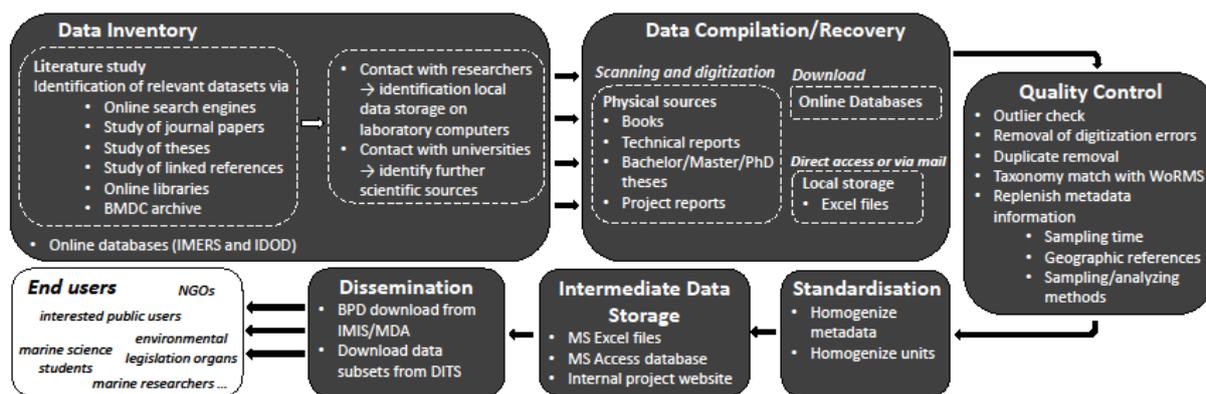


Figure 8: Representation of the workflow starting from the data source identification up to the final dissemination of the Belgian Phytoplankton Database (BPD).

3.2.3. Obstacles encountered in compiling data sources

3.2.3.1. Varied accessibility of compiled data before 4DEMON

The accessibility level of the data compiled in 4DEMON varied over time, but also the context in which the data were gathered. The main bulk of data that were not yet accessible date back from before 1993. Some datasets needed more processing effort than others, e.g. written logbooks had to be digitized from scratch.

- Public marine monitoring and research data funded by the Belgian Federal Science Policy Office (Belspo) follow the open data policy imposed by the PSI Directive. Data management has always been a concern, however often only for the duration of the

project. In the nineties, the need to have a permanent structure for managing the data collected by means of public finance. In 1997, the central database of BMDC was launched to manage monitoring data (e.g. in the frame of OSPAR and WFD) and data resulting from Belgian Federal North Sea Research programs like Science for a Sustainable Development (SSD), <http://www.belspo.be/ssd/>). This is also the reason why historic data (< 1997) were often not yet centralized. Furthermore, data without reporting obligations can be centralized but not disseminated (e.g. Bagger data), while some are only available in excel formats on individual computers.

- Data from individual research projects or data collected by institutes of neighbouring countries: more dispersed availability. For example, (historic) research data gathered in the frame of PhD and master theses or by private funding, are often stored in excel tables and on individual computers, or still only available on paper reports (grey literature) or on old floppy disks. Often hidden and inaccessible (dark data). RWS (Netherlands) data were received upon request.
- Data from industry: often restricted access, not explored under 4DEMON.

3.2.3.2. Evolution in data storage methods and management infrastructure

The marine data management infrastructure became much more organized over the years thanks to the effort of European data management and standardization initiatives like SeaDataCloud, EmodNet and ODIP (See Chapter 3.2.1.4.3.). For historic data however, not always all 'currently' mandatory or required metadata are available, leaving you with 'unknown' information or gaps, even if the data were digitized and stored, for example in old database structures or Access format (e.g. the old monitoring database before IDOD at BMDC).

3.3. Contaminants in biota and sediment

Background information

Until today, heavy metals and PCBs are still pollutants of concern. Monitored heavy metals include lead (Pb), mercury (Hg), zinc (Zn), copper (Cu), chromium (Cr), cadmium (Cd), nickel (Ni) and arsenic (As). Heavy metal concentrations are determined by natural background as well as anthropogenic impact. They may enter the marine environment through atmospheric deposition as well as by water transport. Stringent pollution control measures within industrial combustion processes, metal production, transport and waste streams are reported by OSPAR (2010) in the eighties and nineties. Large amounts of PCB were manufactured between 1930 and 1983 and reach the environment through disposal, leakage, evaporation and accidents (Roose et al., 2005). Although PCB were banned in 1985 ([Directive 96/59/EC](#) on the disposal of PCBs and PCTs), sources still remain, e.g. within waste disposal, PCB-containing equipment, by remobilisation from sediments or by formation of by-products in thermal and chemical processes (OSPAR, 2012).

After all data and metadata were collected and compiled (See Chapter 3.2.), direct use of contaminant data for trend analysis was still hampered. Analytical values differed depending on the matrix selected for analysis: PCB and heavy metal concentrations are grain size dependent while different concentrations can also be measured within different parts of an organism. Changes in analytical method may have induced systematic and/or random errors. Since sampling locations and sampling time have changed throughout 40 years of marine monitoring, data binning is needed to obtain coherent time series. Within WP 3, these issues were tackled by applying a sediment normalization procedure, spatial clustering and the use of appropriate models. An additional principle component analysis (PCA) could identify correlations between contaminants.

3.3.1 Sediment normalization

Contaminant concentrations in sediment depend on the level of pollution but also on the natural variability of the sediment granulometry (i.e. grain size distribution) and mineralogy (i.e. mineralogical composition). To compare contaminant levels irrespective of natural variability, normalization should be performed. Two normalization approaches are widely used: a granulometric and a geochemical approach. The first approach consists in sieving and isolating the clay fraction to reduce the differences in granulometric composition such as isolating the <63µm fraction which is the most widespread monitoring fraction used (OSPAR, 2015). The second approach relies on the use of a proxy to reflect the binding capacity of the sediment mineralogy as well as grain size changes. This proxy should be a conservative element, like Al, which reflects the clay mineral content (Loring et al., 1991). Since the optimal normalization can be regional dependent, geochemical normalization was optimized for the Belgian Part of the North Sea (BPNS). Ideally, a linear relationship exists between contaminant C and normalizer or cofactor N for equally polluted samples. The standardized contaminant content can be calculated by the expression (Smedes, 2002):

$$C_{ss} = (C_s - C_x) \frac{N_{ss} - N_x}{N_s - N_x} + C_x \quad \text{Equation 1}$$

With:

N_{ss} a standard reference co-factor content, C_{ss} the standardized contaminant concentration, C_s the contaminant content, N_s the cofactor content and C_x and N_x the contaminant and cofactor content in pure sand.

Contaminant – cofactor linearity can be tested by analyzing equally polluted samples. Analogously to Smedes & Nummerdor (2003), this was done by fractionation of sediment samples into 9 subsamples of different granular fractions by sieving. All subsamples of the same sediment sample are equally polluted. Based on granulometry, 9 locations on the BPNS were selected for sediment sampling. Detailed information on sample selection and sieving procedure can be found at Le et al. (in preparation, a).

Figure 9 gives examples of contaminant-co-factor relationships, whereas average R^2 values are presented at Table 5. Within OSPAR, Al is used for heavy metal normalization and TOC for PCB normalization (OSPAR, 2015). Other commonly applied co-factors are Li, lutum content or Fe (Kersten & Smedes, 2002). Within 4DEMON, Li and lutum could not be applied as co-factor since data on these parameters were available for only a limited number of historical samples. As can be seen at Table 3.3.1 and Figure 3.3.1(a), normalization could be done with Al, Fe as well as TOC. Lowest linearity for heavy metals was obtained for As ($R^2 = 0.37-0.59$), but correlation was good for all other heavy metals, with R^2 -values varying from 0.68 to 0.91. From Table 5, it can be seen that highest R^2 -values for As, Cd, Ni, Zn and Hg were obtained for co-factor TOC. However, a detailed look at heavy metal data collected within 4DEMON learned that TOC data are frequently missing. Moreover, method switches for TOC are at different moments in time than heavy metal method switches, increasing the number of method changes for time trend modelling. TOC was therefore not withheld for heavy metal normalization. Comparing Al with Fe normalization for sediment at the BPNS, Fe was found to be a better co-factor than Al. Within a first normalization exercise, data was normalized to Al as well as to Fe.

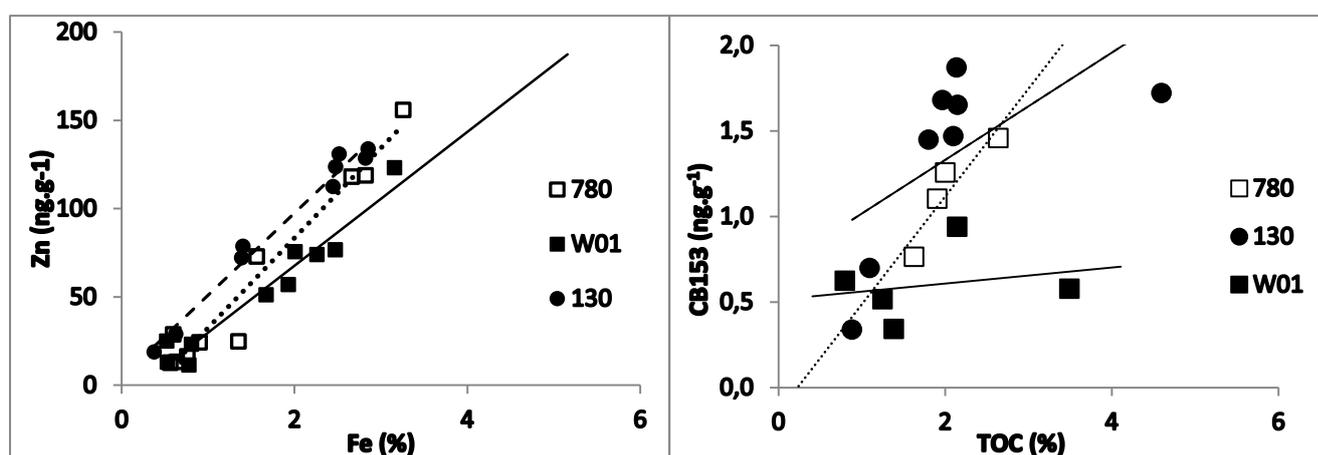


Figure 9: Contaminant – co-factor linearity for (a) Zn-Fe and (b) CB153-TOC for 3 selected sediment samples, fractionized in equally polluted subsamples.

Table 4: R^2 average of contaminant vs. potential co-factors regressions for all samples locations. Highest values are presented in bold.

	As	Cd	Cr	Cu	Ni	Pb	Zn	Hg
Al	0.37	0.68	0.75	0.74	0.71	0.80	0.81	0.77
Fe	0.51	0.79	0.88	0.80	0.75	0.80	0.87	0.82
TOC	0.59	0.90	0.78	0.88	0.84	0.77	0.89	0.91
NI	0.54	0.80	0.84	0.90		0.71	0.88	0.84

To apply the normalization procedure for Al or Fe, “pivot values” N_x and C_x and “standard seafloor value” N_{ss} has to be determined to apply Equation 1. N_x and C_x can be deduced from equally polluted samples. This can be done by (1) plotting co-factor versus TOC to identify the y-axis intercept (Y_1) at TOC=0 and (2) plotting heavy metal concentration versus co-factor to identify the average $Y_{intercept}$ (Y_2) at cofactor $x=Y_1$ (Smedes et al., 2002). Another

approach is to consider historical samples in pure sand, *i.e.* from sampling locations remotely from the coast at the BPNS. Within Table 6, values were compared with N_x values determined by Smedes *et al.* (2002) for the Dutch Part of the North Sea as well as those determined by OSPAR for comparable total digestion (OSPAR, 2008). Determining C_x and N_x from equally polluted samples suffered from issues as a high intercept deviation between the different samples, resulting in large standard deviations and negative intercepts (Table 6). In contrast, N_x and C_x values derived from 20 pure sands sampled from 2008-2014 revealed less deviation and were in line with Smedes N_x and C_x values and were therefore used further. For Cd, C_x values could not be derived from pure sand samples since too many values were below limit of detection. In these cases, the values for equally polluted samples were used.

Table 5: N_x and C_x values derived from equally polluted samples and from pure sand, compared to Smedes (2002) and OSPAR values (OSPAR, 2008). Final selection is presented in bolt.

	Equally polluted samples		Pure sand		Smedes		OSPAR	
	Average	STDEV	Average	STDEV	Average	STDEV	Average	STDEV
Cd (mg/kg)	0.02	0.02			0.03	0.06	0.03	0.06
Pb (mg/kg)	4.2	3.7	5.45	1.01	2.00	2.20	9	3
As (mg/kg)	4.7	2.2	4.78	2.11	3.00	1.50	5	3
Cr (mg/kg)	9.7	9.3	4.99	2.27	13.00	6.00	13	6
Cu (mg/kg)	-0.8	1.6	1.06	0.41	1.00	1.00	3	1
Ni (mg/kg)	0.6	1.5	2.41	0.56	2.50	1.10	4	2
Zn (mg/kg)	-1.2	7.8	5.52	1.54	8.00	9.00	13	5
Hg (mg/kg)	-10.2	11.5	1.67	0.45	0.00	0.04	0	0.04
Al (%)	1.1	0.3	0.99	0.22			1.4	6
Fe (%)	0.4	0.4	0.31	0.10				

For the determination of standard seafloor values N_{ss} , linear regressions of possible co-factors were plotted against TOC. The reference value was determined as the average of co-factor concentrations deduced from each set of 9 regressions at TOC equal to 2.5%, set by OSPAR as TOC reference value (OSPAR, 2015).

Once C_x , N_x and N_{ss} values were determined, Equation 1. could be applied to all data, applying Al and Fe as normalizers. Where in most cases, heavy metal data normalized to Al and Fe were in good agreement with each other, large difference could occur for many samples, hampering the choice between Al or Fe since one out of the two values could be considered outlier. This is clarified with some examples in Table 7. It was therefore decided that only heavy metal data could be taken into account for time trend analysis if Al and Fe-normalization results differed to maximum 50%. All data were normalized to 50% Al and 50% Fe and was considered as the best approach for trend analysis. Heavy metal in sediment time series were modelled, applying this approach, for data from the eighties until 2016.

Table 6: Normalization of Hg data to Fe, Al and Fe/Al for some sediment samples from 1990. Normalized Hg concentrations are expressed as $\mu\text{g}\cdot\text{kg}^{-1}$.

Location name	Latitude	Longitude	Fe normalization	Al normalization	Criterion	Fe-Al normalization
800	51.847	2.867	391	424	OK;<50%	408
330	51.433	2.808	64	66	OK;<50%	65
435	51.581	2.790	125	119	OK;<50%	122
120	51.185	2.701	31	2684	NOK;>50%	
315	51.323	2.464	65	-296	NOK;>50%	

Due to lack of Fe or Al data of sediment samples from the seventies, a second approach was selected for trend analyses since beginning of the seventies. Data were normalized to Ni instead of Al or Fe. The choice for Ni resulted from the analysis of equally polluted samples, in which Ni also revealed good linearity with other heavy metals (Table 5). Moreover, Ni values in marine sediments of the BPNS are limitedly impacted by anthropogenic sources. For example, at point ZEB (N 51.357, E 3.152) at the BPNS, annual analyses of sediment show very high variability for most heavy metals. *E.g.* for Hg, concentrations in the <63 μm fraction varied between 22 to 192 $\mu\text{g}\cdot\text{kg}^{-1}$ within samples from 2009-2013. This is due to the sampling moment. Depending on the tide, fresh deposited sediment or holocene consolidated mud was sampled (Fettweis et al., 2009), revealing clear effects of industrial pollution. However, Ni was one of the contaminants with little variation, from 21 to 26 $\text{mg}\cdot\text{kg}^{-1}$ in the <63 μm fraction, indicating limited influence of anthropogenic pollution. Ni can therefore be applied as normalizer.

Whereas appropriate co-factors could be found for heavy metals in sediment, the analysis of equally polluted samples resulted in poor results for PCB co-factors. As can be deduced from Figure 10. for CB153, linearity between PCB-concentrations and co-factors as TOC is poor and no clear intercept can be derived from the different regression lines. It was therefore not considered appropriate to apply a co-factor normalization to PCB-analysis. For PCB modelling, regression was restricted to a single sediment fraction, hence only applying granulometric normalization.

3.3.2 Cluster analysis

To define spatial zones for trend modelling, a Ward hierarchical clustering was performed, which consists of an ascending agglomerative algorithm applying successive aggregations of similar individuals (Ward, 1963). It outputs a dendrogram from which the samples can be grouped in a desired number of cluster groups with similar level of contamination (Table 8). The clustering was performed based on samples from a 5 years period (2007-2011). Only samples with granular fraction <63 μm were considered. Variables included were the concentrations of heavy metals and PCBs. Clustering analysis was performed on centered scaled concentrations (the difference between individual values and the mean value was divided by the standard deviation). A hierarchical clustering with 5 groups was finally selected.

Table 7: Cluster characterization with -- very low level of contaminant, - low level of contaminant, o medium level of contaminant, + high level of contaminant, ++ very high level of contaminant.

Contaminant	Cluster 1	Cluster 2	Cluster 3	Cluster 4	Cluster 5
As	o	-	--	+	++
Cr, Ni	+	o	--	o	++
Cu, Pb, Zn	-	o	--	o	++
Cd, Hg	--	++	+	o	-
PCBs	-	o	+	++	--

Based on these 5 cluster groups, the BPNS was divided in 5 zones according to the similarities in cluster groups of the samples present (Fig. 10).

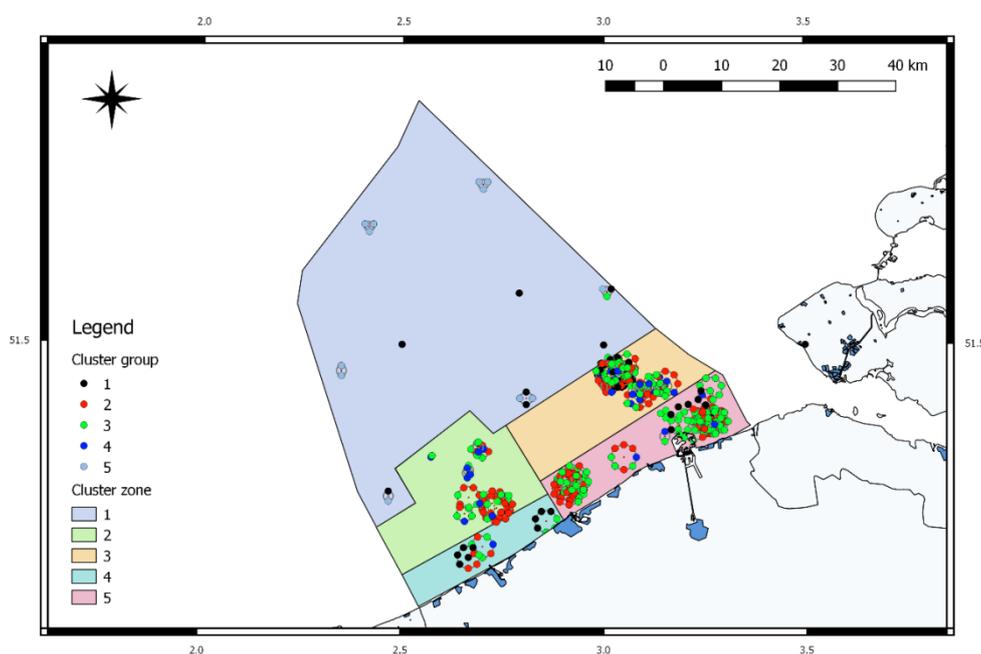


Figure 10: Spatial zonation of the BPNS, based on the cluster analysis.

As can be deduced from Fig. 10, the offshore zone 1 is grouping samples especially from cluster group 5. Zones 2 and 5 have dominance of samples from cluster groups 2 and 3, whereas cluster group 1 is more dominant within zone 4. Zone 3 is the most diverse spatial zone, containing samples of all 5 cluster groups. This diverse pollution profile at zone 3 may be caused by the mixed presence of sandy sediments with a sludge disposal zone.

3.3.3 Principle compound analysis

A Principal Component Analysis (PCA) was applied to non-normalized PCB and heavy metal data on the <63 μ m fraction as well as on normalized heavy metal data. The closer contaminants are positioned in the correlation circle, the more correlated they are between each other. Results are given in Figure 11.

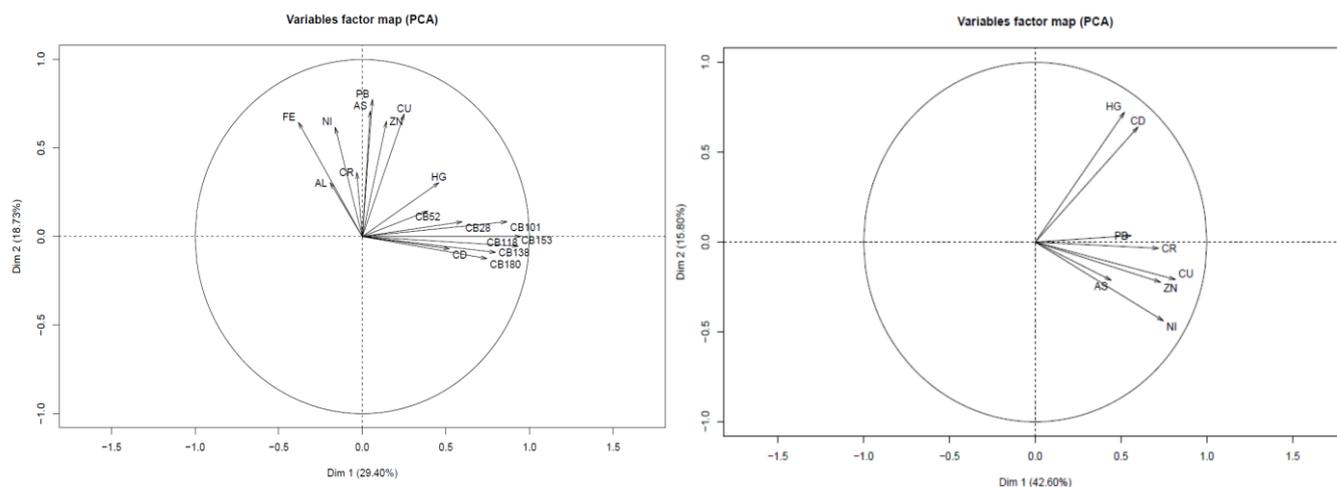


Figure 11: Correlation circles from principle compound analysis on heavy metal and/or PCB data. Left: granulometric normalized data (<63µm fraction, heavy metals and PCBs), right: Al-Fe normalized data (heavy metals).

As can be seen from Fig. 11, PCBs clustered well together, indicating large similarities in distribution patterns between individual PCBs. For heavy metals, Hg and Cd behave differently from the other heavy metals and show to be more similar to PCBs. Hg and Cd are not or only at very limited concentration present in a coarse mineral matrix (OSPAR, 2015). They also tend to bind more to organic material (Loring et al., 1991). Related to the organic material distribution at the BPNS, this leads to low off-shore concentrations and high near-shore concentrations of Hg and Cd, similar to PCBs but clearly distinct from other heavy metals.

3.3.4 Time trend modelling

A linear mixed effect model is applied to assess temporal and spatial distribution of contaminants in sediment and biota. A linear mixed effect model is a linear regression model with fixed effect variables, *i.e.* explanatory variables or predictors, but also including a random effect term.

In a linear mixed effect model (Zuur et al., 2009) the random effect term ε can be decomposed in two parts: the real noise component but also random effect terms which give a structure to the correlations existing between individuals of same attributes. Including such random terms in a linear mixed effect model can allow to overcome modelling issues (*e.g.* independence/nested data/heterogeneity, temporal and spatial correlation) and improve goodness of fit. In this study, a random term was included to capture spatial correlation between contaminant levels in samples from the same location. Concentrations in samples from same locations are expected to be more similar between each other compared to concentrations at other sampling locations. For trend modelling on sediment, shrimp and

swimming crab data, the variables “time”, “time²”, “cluster zone”, “season”, “laboratory analytical method” and interaction term between “time” and “cluster zone” were selected. The last allows to have different trends/slopes per cluster zone. For mussel data, the variable “cluster zone” was altered by the variable “groyne”, indicating the location where the mussel sample was taken (Nieuwpoort, Oostende, Blankenberge or Knokke). For flounder, time trend modelling did not consider a spatial clustering of the BPNS. However, an additional variable “tissue” was included since analysis was done on liver or muscle.

As laboratory analytical methods changed several times over time, observed shifts of concentrations trends may be caused by laboratory effects. Since laboratory analysis do not overlap over time, time trend shifts due to laboratory effects should be distinguished from real changes in environmental concentration. Because laboratory methods are generally applied for multiple years, time trends can be derived over covered time periods for each analytical method. It was observed that slopes of contaminant concentrations between individual methods were similar, indicating that laboratory effects especially imply level shifts rather than trend changes. Therefore, analytical method was included as a fixed term in the model without interaction term between time and analytical method which would make the model more complex.

For each model, log transformation was done to obtain a normal distribution and to comply to normality conditions necessary for the validity of the fitted model. Cook's distance was used to identify outliers. Cook's distance measures how much an observation influences the overall model fitting of predicted values. Data with a Cook's distance value exceeding the cut-off value of 0.2 was discarded one by one. Model assumptions, *i.e.* normality, homogeneity and independence, were checked. In some cases, independence and homogeneity could be improved by adding a temporal correlation structure AR-1-auto-correlation (auto-regressive model of order 1 modelling the residual at time s as a function of the residual of time $s - 1$ (Zuur et al., 2009). However, this made the model more complex without affecting the model outcome, except for CB52, and was therefore only withheld for CB52.

Variable selection was done by single term deletions based on likelihood ratio tests on a Chi² statistic. All terms remaining were significantly different from zero at the 5% level.

3.3.5 Historical sample analysis

Since heavy metals can neither degrade nor evaporate from sediment samples, concentrations will be constant in well stored samples. Moreover, analysis of historical samples by one method allows to exclude the variable analysis method when comparing historical data. Historical samples, stored at RBINS, ILVO and CODA, were collected from five time periods, including 3 samples for each cluster zone, *i.e.* 15 samples for each time series. Details on sampling locations can be found in Le et al. (in preparation, a). Collected historical samples from the BPNS were “Gilson” samples (1901-1911), samples from

“Project Sea”/PMPZ (1973), sediment cores sampled in 1986-1988, and samples from the sludge disposal project from 2001-2002 and from 2012.

Since selected samples were from different grain size fractions, normalization was essential. Al-Fe normalization was applied, only retaining samples for which the result of independent Al- and Fe-normalization differed to at maximum 50%. Results are presented in Chapter 3.6.1.

3.4. Eutrophication

3.4.1. Nutrients and water clarity

a. Nutrients

The nutrients data derived from many projects with various analyses methods. Data originators were contacted and interviewed. Because there was shortness of overlap in time for the different data sources, the nutrients were not intercalibrated. According to the data-origimators (oral communication M. Knockaert, K. Parmentier, W. Baeyens, M. Elskens, 2015) no big changes in analyses methodology have occurred, but probably more in sampling methodology. These should not have an impact on the BPNS data and there should be no need for intercalibration. This is confirmed by Hager *et al.* (1972) who did a comparison at sea of manual and autoanalyzer analyses of phosphate, nitrate and silicate. Today, the detection limits are somehow better and the standard deviation is smaller. The analyses methodology for Ammonium of the OSPAR monitoring data by RBINS (Fig. 12) has changed from spectrophotometry (phenol method) to technicon Auto Analyzes (AA) in 1995 and Skalar AA in 2001 (salicylate method), with a higher accuracy. The higher values for the spectrophotometric data (< 1995) are no surprise, as the 1980s are marked for its high nutrient concentrations, with the biggest source of Ammonium coming from the agriculture. Followed by a decrease after the 1990s, following the multiple European Directives that came into force.

Below: for MUMM data.

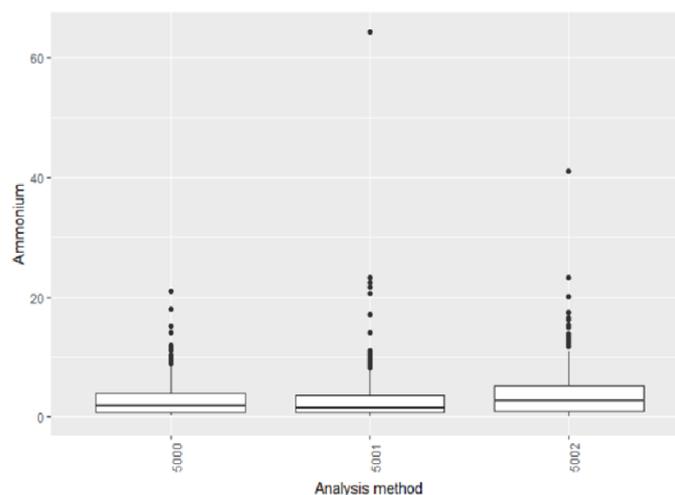


Figure 12: Comparison of the OSPAR monitoring data for Ammonium (5002: Spectrophotometry (> 1995); 5001: Technicon II AA (1995-2000); 5000: Skalar AA (>2001)).

b. Water clarity

Water clarity data, TSM and secchi, were compiled since the 1970s. Outliers and extremes are difficult to identify, especially with lack of quality flags (e.g. suspect data) provided by the data originator. While the result of a secchi sampling is straightforward, the analyses procedure for TSM has a high impact on the result; the filtration method has an error of 10-30%. Furthermore, intercomparing the data before 1997 was not possible, as there is no overlap in time between the different data sources. Standard deviation decreases with higher count of samples per period (Fig. 13). Especially in our high turbid area the data needs good spatio-temporal coverage. Plotting the TSM data versus the secchi data clearly shows the inverse relationship of both parameters for coastal and offshore waters (see Annex 6) Regrettably, no secchi data were recovered between 1985 and 1990.

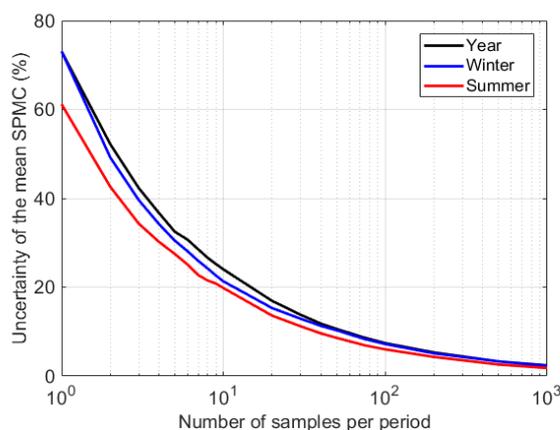


Figure 13: Uncertainty of the mean TSM over the year, winter and summer.

Data validation on water clarity based on plotting IS against EO is possible from 1997 onwards, making these data more reliable. TSM IS versus EO show the same dynamism, however the signal for EO data is systematically lower because of atmospheric correction. Comparing the trends of different parameters, shows the TSM IS dynamism is following the dynamism observed for the chlorophyll EO data. Finally, the chlorophyll a IS data (*excl.* trichromatic data) match with the EO data.

3.4.2. Phytoplankton biomass

Background information

In the coastal waters of Belgium and The Netherlands, high biomass phytoplankton blooms occur systematically in spring. The onset of the spring biomass bloom is determined by the light availability (Peperzak, 1993; Peperzak et al., 1998), while the magnitude of the spring maximum of chlorophyll a – usually calculated as the 90th percentile of CPHL, i.e. CPHL P90 – is commonly considered to be limited by the nutrient availability (Desmit et al., 2015; Muylaert et al., 2006). In the Belgian and Dutch parts of the North Sea (respectively BPNS and DPNS), nutrient inputs result from the combination of two end-members: the continental freshwater discharges and the Atlantic water inflow (Brion et al., 2006; de Vries et al., 1998; Ruddick and Lacroix, 2006). The nutrient-enriched river waters are the cause of coastal eutrophication, causing nutrient imbalance and ecological disturbances (Jickells, 1998; Lancelot et al., 2009, 1987; Philippart et al., 2007). The spatial distribution of the CPHL P90 in the BPNS and the DPNS typically shows high values in the coastal zone and a decreasing gradient towards the offshore (de Vries et al., 1998; Los and Borkhorst, 1997; Rousseau et al., 2006; Schaub and Gieskes, 1991). The near-shore spring bloom occurs between March and May (usually around mid-April) and is most of the years dominated by the colonial haptophyte *Phaeocystis globosa* (Rousseau et al., 2013) after the early diatom bloom has been limited by silica (Baretta-Bekker et al., 2009; Lancelot et al., 1987; Rousseau et al., 2006). In recent years, the increase in Sea Surface Temperature (SST) in the north west Atlantic has been reported to cause an increase in the cell division rate of the coastal phytoplankton species *Synechococcus*, with earlier blooms and a timing varying by 4 weeks (Hunter-Cevera et al., 2016). It is therefore not excluded that an increase in SST may have changed the CPHL phenology in the Southern North Sea. The objectives within 4DEMON were to gather nutrient (N, P, Si), water clarity and CPHL data across the period 1970-2015, subsequently to select the subset of reliable data, and to examine the spatial and temporal patterns of variability based on integration with Earth Observation (EO) data .

3.4.2.1. Intercalibration of in situ CPHL

The objective of the intercalibration is to gather the many datasets of chlorophyll a concentration (CPHL) measured in the period 1970-2015 in the Belgian waters, and re-build a dataset suitable for analysis. The samples have been taken in different places at different times and, therefore, data had to be lumped within waterbodies based on expert knowledge to draw time series (Fig. 14). In this period, several techniques have been used by different laboratories and by different technicians, probably introducing some errors that could not be

corrected. Most importantly, the comparison between the methods of CPHL measurement remains a challenge (Baretta-Bekker et al., 2015; Noklegaard et al., 2005). Typically, the CPHL measured by trichromatic spectrophotometry is problematic and does not well compare to samples measured with any other method (Neveux et al., 1990). In our case, comparisons showed deviations, probably because the trichromatic method is not suitable to identify the pigment phaeophytin *a*, a degradation product of chlorophyll *a*. Also, the CPHL dataset from trichromatic showed peculiar values with aberrant seasonal profiles and high winter values. This resulted in the exclusion of the CPHL data before 1985. The monochromatic spectrophotometry and the fluorimetric methods are much more comparable when they are adapted to measure phaeopigments through the acidification step and the use of Lorenzen's equations (Lorenzen, 1967). They are also generally comparable to HPLC determination of CPHL (Murray et al., 1986), even though Latasa et al. (1996) have shown that monochromatic spectrophotometry may overestimate CPHL by 6-9% in comparison to HPLC. Regarding the determination of CPHL by HPLC, these authors also showed that a variability of 10-20% could be expected depending on the protocol used. Therefore, we have estimated that the Belgian dataset would include CPHL measured by monochromatic spectrophotometry (Lorenzen), by fluorimetry (adapted to Lorenzen) and by HPLC without any additional transformation, acknowledging the fact that errors of 20% are possible.

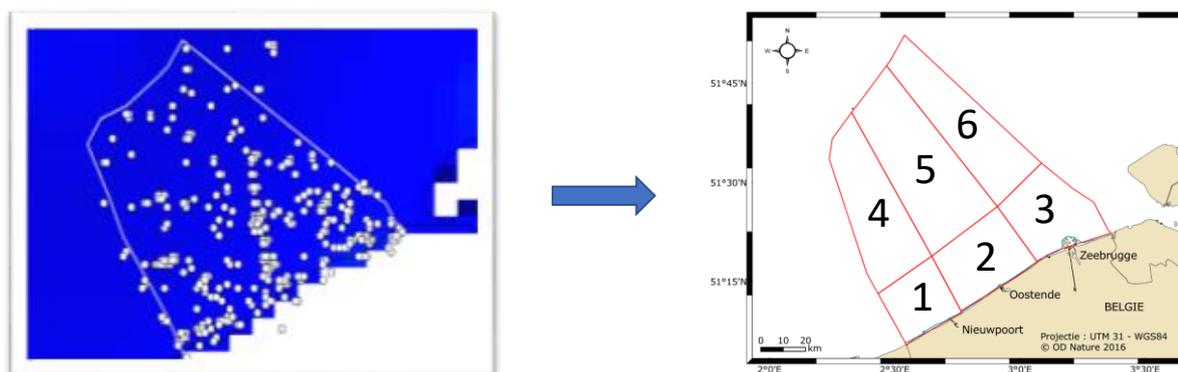


Figure 14: Left – Sampling sites in the BPNS during the period 1970-2015. Right – Lumping of the sampling sites within six areas designed through expert knowledge.

3.4.2.2. Satellite-based chlorophyll *a* product for the North Sea

Satellite data from ocean colour sensors (i.e. SeaWiFS, MODIS, MERIS, VIIRS, Sentinel-3) can provide spatially coherent data on chlorophyll concentrations using chlorophyll retrieval algorithms. Results presented here are obtained from a collaboration between the 4DEMON project and the EU JMP-EUNOSAT project. The satellite-based chlorophyll products were developed within the JMP-EUNOSAT project and analysed for the Belgian Coastal Zone within the 4DEMON project (Van der Zande et al., 2018). There has been considerable success with blue/green-ratio algorithms in case 1 waters where the variation of optical properties (absorption and scattering) is dominated by phytoplankton and associated material (O'Reilly et al., 1998; Gohin et al., 2002). In contrast, the optical complexity in coastal waters often poses many challenges to the accurate retrieval of biogeochemical parameters using satellite remote sensing (IOCCG, 2000, 2006). Chlorophyll retrieval by

blue green ratio algorithms tends to fail when applied to coastal waters whose optical properties are strongly influenced by non-covarying concentrations of Total suspended matter (TSM) and coloured dissolved organic matter (CDOM). Such waters are defined as case 2 waters. Several constituent retrieval algorithms for use in case 2 waters have been developed: 1) red-edge algorithms (Gons et al., 2002) taking advantage of the chlorophyll absorption peak near 670 nm and 2) artificial network approaches trained to varying parameter concentrations and optical property ranges specifically developed for use with MERIS data, such as the MERIS Ground Segment Processor (MEGS, Doerffer & Schiller, 2007) and the FUB/WeW (Schroeder, Schaale & Fisher, 2007). For each of these products we determined for which water types, described in terms remote sensing reflectance (Rrs) spectra, they provided the most accurate chlorophyll estimations (i.e. relative error < 50%) based on a variety of reference datasets from the Coast Colour Round Robin project (CCRR; <http://www.coastcolour.org>) (Nechad et al., 2015). Next, the quality controlled chlorophyll-a datasets are merged together based on best suited algorithm/water type combination, with special attention to the transition zones between different water types to ensure a gradual merge. This process resulted in a data archive of 20 years of coherent satellite-based chlorophyll-a products starting from 1997. The quality controlled and merged satellite-based chlorophyll-a observations are compared to *in situ* observations that have been collected in national monitoring programs. Figure 14 shows 90-percentile map of chlorophyll a for the growing season (March-Oct incl.) of 2003 providing a spatial interpretation of the intensity of the algal blooms in the North Sea. Additionally, chlorophyll-a time series are provided for the national monitoring stations Stonehaven (Schotland), Rottumerplaat 50 (the Netherlands), 330 (Belgium) and Boulonge (France) for the year 2003 showing the ability of the satellite data to capture the temporal chlorophyll dynamics. The *in situ* measured CPHL was analyzed using the HPLC-method. For the time series of satellite data, we extracted a 3 x 3 macro-pixel and the 1 x 1 km center pixel containing the monitoring station location. The resulting time series are presented in monthly bins as *in situ* data is mostly collected monthly in these stations. The satellite data is presented as boxplots to demonstrate the increased availability of satellite data compared to *in situ* sampling, i.e. 20-50 observations per growing season depending on the location, cloud cover and water conditions.

3.4.3. Phytoplankton taxonomic composition

Background information

Phytoplankton drives the ocean carbon pump and global cycles of nutrients and oxygen, and fuels marine food webs, affecting both lower and higher trophic levels from microbiota to zooplankton to pelagic fish and seabirds (Beaugrand, 2009; Edwards and Richardson, 2004; Maso and Garces, 2006; Tréguer et al., 2017; Turner and Tester, 1997). Primary production thus constrains commercial fish stocks, with important economic consequences (Chassot et al., 2010). Phytoplankton production is affected by several environmental factors, most notably nutrient availability, light and temperature, but also by biotic interactions with

symbionts, parasites and grazers (Lima-Mendez et al. (2015)). Natural variability in most of these factors is increasingly modified by local to global anthropogenic activities, from dredging over eutrophication and pollution to climate change and the effects of over-fishing (Cloern, 2001, Colijn et al., 2002; Prins et al., 2012), with important consequences for phytoplankton dynamics and production.

The dataflow for compiling and quality controlling the Belgian Phytoplankton Database (BPD) is given in Chapter 3.2.2.

3.4.3.1. Clustering

Sampling took place in a large number of different stations depending on the research project. In order to analyse the spatio-temporal variation in the datasets the high amount of different sampling stations was homogenized by clustering them in a coastal and an offshore zone (Figure 15).

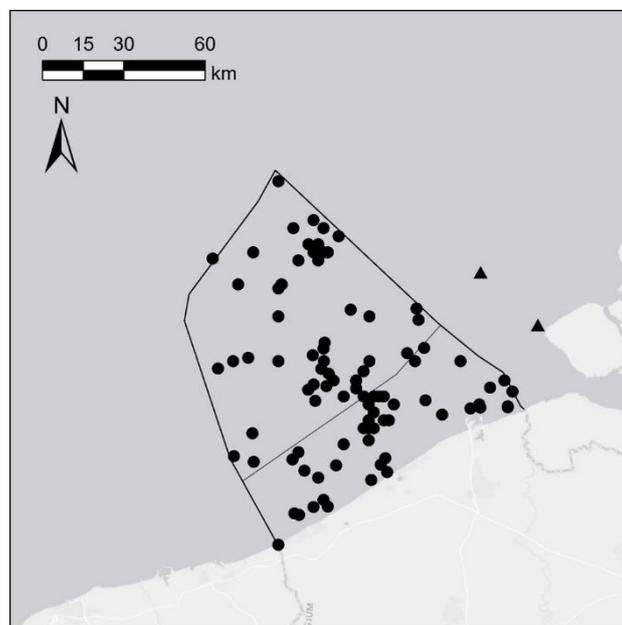


Figure 15: Belgian Part of the North Sea (BPNS) with sampling locations and division in on- and offshore area. The Walcheren stations (RWS monitoring) in the Dutch coastal area are indicated as triangles.

3.4.3.2. General Additive Mixed Models

To unravel the seasonal trends in the datasets, the total coastal diatom and total coastal dinoflagellate dataset were analysed with an additive mixed modelling approach. The *mgcv* package (Wood, 2017) in the open-source software R was used (RStudioTeam, 2017). Previously, the phytoplankton data were aggregated on the total diatom and total dinoflagellate level. The data were split in four data subsets: diatom abundance data in the 1970s (1970-1978, no data in 1975), diatom abundance data from 2003 to 2010, dinoflagellate abundance data in the 1970s (1970-1978, no data in 1975 and 1976) and dinoflagellate abundance data from 2003 onwards (2003-2010).

Space (sampling location and methodology set as random effect) and time (seasonal smoother) were incorporated in the models. By incorporating 'method' as a random effect, a possible taxonomist dependent identification bias was excluded from the models. The data were $\log_{10}(x)$ -transformed prior to the analysis and a Gaussian distribution was assumed in the models. In addition, a log-link function was used to ensure that the fitted values are always positive (Philippart et al., 2010). One cubic regression spline (cc) was used to model the seasonal trend. This type of smoother ensures that the value of the smoother at the far left point of the gradient is the same as at the far right point which is convenient to model an annual cycle (Zuur et al., 2009). The models were expanded with different residual auto-correlation structures and the best model for each dataset was identified by the lowest Akaike information criterion (AIC), which takes into account the model fit versus the complexity of the model calculation (Zuur et al., 2009).

3.4.3.3. *Fulcrum Analysis*

The relative annual cumulative abundance was calculated for the total diatom and total dinoflagellate data. The day of the year on which 50 % of the total annual phytoplankton abundance has been reached, was identified by fitting a binomial *glm* smoother on the data.

3.4.3.4. *Diatom biovolume analyses*

Based on geometrical shape and size measurements taken from literature and online sources, diatom biovolumes were calculated on the lowest taxonomic level available, mostly on species level (Hillebrand et al., 1999; Hoppenrath et al., 2009; Horner, 2002; McQuoid and Nordberg, 2003; Naz et al., 2013; Throndsen et al., 2007). According to Terseleer Lillo (2014) the diatom taxa were combined into three size classes, small cells ($< 6,000 \mu\text{m}^3$), cells of intermediate cell biovolume ($6,000 \mu\text{m}^3 - 4.9 \cdot 10^4 \mu\text{m}^3$) and large diatom cells ($> 4.9 \cdot 10^4 \mu\text{m}^3$). These data were used to follow the seasonal size class distribution.

3.4.3.5. *Ordination analysis*

In order to investigate the seasonal distribution of the phytoplankton genera, a Principal Component Analysis (PCA) was conducted on a relative abundance dataset using CANOCO. Relative phytoplankton abundances were $\log_{10}(x+1)$ transformed prior to the analysis.

3.4.3.6. *CHEMTAX analysis of HPLC pigment dataset (2003-2016)*

During the period October 2002 to December 2016, monthly sampling cruises were performed in the BPNS, by the research vessel (RV) 'Zeeleeuw' and subsequently the RV 'Simon Stevin'. Pigments were extracted from filter seawater samples and analysed by means of reverse phase high-performance liquid chromatography (HPLC). Marker pigments of key phytoplankton groups were identified based on their retention time and absorption spectra. During the whole sampling period three different HPLC protocols were applied: Wright (1991) (2002-2003, 2005-2008), Zapata (2000) (2004) and Van Heukelem (2001) (2008-2016). The 2004 data were not used in the analyses, because of biased results due to the application of the Zapata (2000) methodology. Also 2002 and 2008 data were not used, due to missed spring peaks.

With a priori knowledge on pigment ratios of the phytoplankton groups, the relative contribution of each phytoplankton group to the total phytoplankton biomass was estimated using CHEMical TAXonomy (CHEMTAX v1.95) software (Mackey, 1996). CHEMTAX requires three matrixes, the first matrix contains the marker pigments to CPHL ratios from the sampled stations. In this study the following marker pigments were used: fucoxanthin (diatoms), chlorophyll *c3* (*Phaeocystis*), peridinin (dinoflagellates), zeaxanthin (cyanobacteria), chlorophyll *b* (chlorophytes, and euglenophytes) and alloxanthin (cryptophytes). The second matrix contains the theoretical values of the marker pigments to CPHL ratios. For this study the theoretical matrix is an adaptation on the matrix used in Muylaert (2006), which was based on published accessory pigment to CPHL ratios (Schlüter et al., 2000; Antajan et al., 2004; Muylaert et al., 2006). In the third matrix the limits on the theoretical marker pigment to CPHL ratios are set. CHEMTAX optimizes the contribution of the phytoplankton groups using a steepest descent algorithm to find the lowest pigment content unexplained (lowest root mean square) of the theoretical and the limit matrix on the natural sample matrix.

3.5. Ocean acidification

Background information

The accumulation of anthropogenic CO₂ in surface waters of the ocean has altered carbonate chemistry in surface waters since pre-industrial times (Caldeira and Wickett, 2003). This corresponds to ocean acidification meaning an increase of [CO₂] and of [H⁺], and a decrease of pH, [CO₃²⁻], and the saturation states of calcite (Ω_{ca}) and aragonite (Ω_{ar}), all related to shifts in thermodynamic equilibria. Ocean acidification can alter the rates and fates of primary production and calcification of numerous marine organisms and communities (Kleypas et al., 2006; Doney et al., 2009). Such changes can change the ocean's carbon sequestration capacity, marine biodiversity and marine ecosystem services and goods. Indeed, some of these calcifying organisms such as bivalves are important economic resources (fishery) and constitute important resources for marine birds (Gutiérrez et al., 2003). The detection of complex and intricate interactions require long time series of quality checked and uniform format variables related to carbonate chemistry such as pH.

Methane (CH₄) is the second most important greenhouse gas (GHG) after CO₂ (IPCC 2013), but has a shorter residence time in the atmosphere (10 yrs). This means that alleviating CH₄ emissions could represent an efficient option for mitigation of climate change, since CH₄ accounts for 32% of the anthropogenic global radiative forcing by well-mixed GHGs in 2011 relative to 1750 (IPCC, 2013). This requires a full account with a reasonable accuracy of the sources and sinks of CH₄. Yet, there are large uncertainties in the quantification of natural and anthropogenic CH₄ sources and sinks (Saunois *et al.*, 2016). The open ocean is a very modest source of CH₄ to the atmosphere (<2 TgCH₄ yr⁻¹) compared to other natural (300 TgCH₄ yr⁻¹) and anthropogenic (330 TgCH₄ yr⁻¹) CH₄ emissions (Saunois *et al.*, 2016). Coastal regions, and in particular estuarine zones, are more intense sources of CH₄ to the atmosphere (Borges and Abril, 2011) than open oceanic waters. The CH₄ emission to the atmosphere from coastal areas is sustained by riverine inputs and methanogenesis in the

sediments due to high organic matter (OM) deposition (Borges and Abril, 2011). Additionally, natural gas seeps are sources of CH_4 leading to high dissolved CH_4 concentrations in bottom waters (from dozens of nmol L^{-1} up to several $\mu\text{mol L}^{-1}$). Sediments of the BPNS have been documented as containing gas pockets, possibly of CH_4 (Missiaen et al., 2002), although the impact of these structures on the emission of CH_4 to the atmosphere has not been documented so far.

Data on pH, water temperature, and salinity were compiled from available public databases and digitalized from publications and grey literature. The dataset spans at from 1970 to 2015, and totals > 8,800 values (Fig. 16). Three main contributors to the datasets are: Rijkswaterstaat (RWS), Royal Belgian Institute of Natural Sciences (RBINS), and University of Liège (ULg). A direct comparison of the 3 datasets is not possible since the data were obtained at different dates and locations, but for some periods data is available simultaneously from the 3 main records (Fig. 17). This shows that the data from the 3 main records are comparable numerically. Reassuringly, the data-sets also show a seasonal variation that is consistent with the know biological cycles: increase of pH in spring and summer resulting from primary production, decrease of pH in fall and winter with a dominance of respiration over primary production (Fig. 17). Data on partial pressure of CO_2 and total alkalinity were compiled mainly from data-sets generated by ULg over the last couple of decades. As part of data rescue, methane (CH_4) data in surface waters of the BPNS generated by the BELCOLOUR-II project in spring, summer and fall 2010 and 2011 were recovered, quality checked and formatted. In addition unpublished data obtained by the Stony Brook University were recovered and collocated with salinity and water temperature derived from the Belgica cruise report. These CH_4 data were analyzed in conjunction with recently acquired data in 2016.

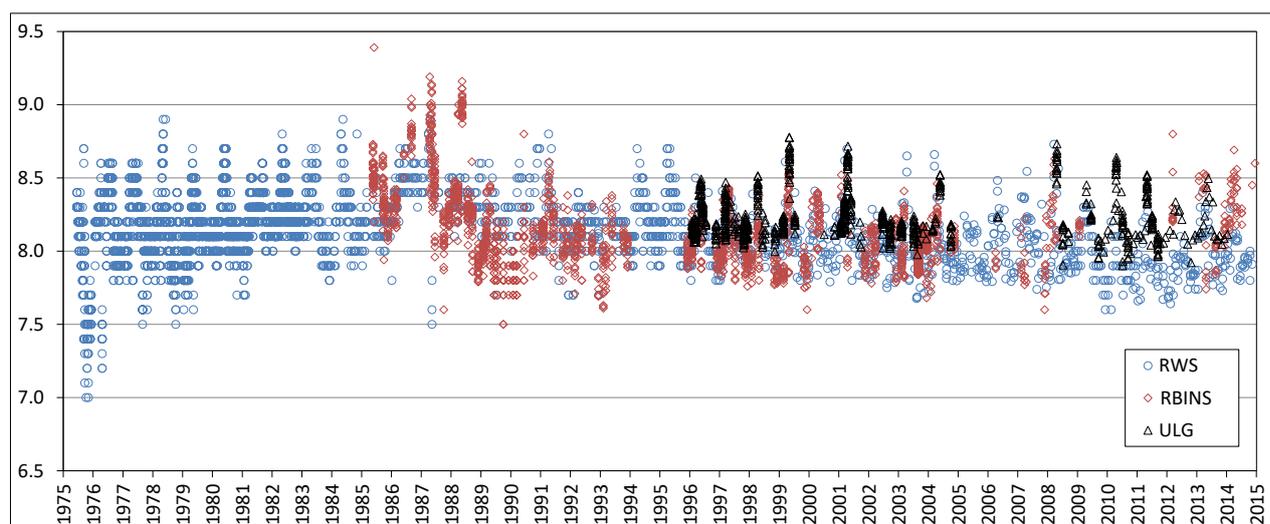


Figure 16: Available pH data records in the Belgian coastal zone as function of time from Rijkswaterstaat (RWS), Royal Belgian Institute of Natural Sciences (RBINS), and University of Liège (ULg).

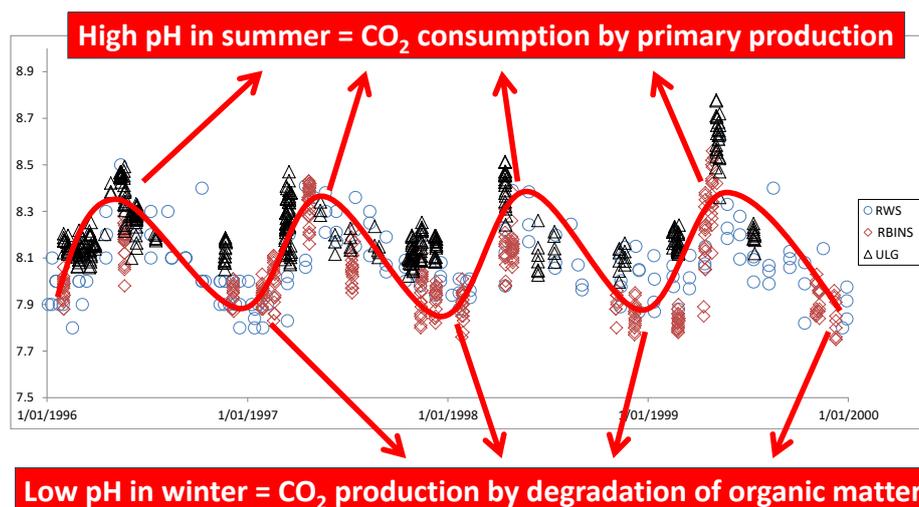


Figure 17: Sub-set from 1996 to 2000 of pH data records in the Belgian coastal zone as function of time from Rijkswaterstaat (RWS), Royal Belgian Institute of Natural Sciences (RBINS), and University of Liège (ULg).

3.6. Trends and analyses of long-term environmental change in the BPNS

3.6.1 Trends on contaminants in sediment and biota

A selection of model results is given within Figures 18 and 19. Figure 18 presents time trends in sediment for geochemical normalized heavy metal data, including Al-Fe- as well as Ni-normalized metal data. It also contains PCB model results on the <63 μm grain size fraction. Models on heavy metal data based on granulometric normalization are presented at Le et al. (in preparation, b). Biota results are presented within Fig. 19 for mussels, fish and shrimp. All models and more details on the models, including model coefficients, confidence intervals and detailed model descriptions are given within Le et al. (in preparation, b) and Bekaert et al. (in preparation).

For heavy metal data in marine sediments, trends applying Al-Fe-normalization are in line with trends applying Ni-normalization or granulometric normalization, which is an indication that the applied models are robust (Fig. 18). Most heavy metals, i.e. As, Cr, Cu, Pb, Cd and Hg reveal a decreasing trend throughout 40 years of marine monitoring. As, Cu, Pb and Cd reveal decreasing trends for both, Al-Fe- and Ni-normalization. For Cr and Hg, decreasing trends are found for Al-Fe-normalization. For Ni-normalization, Hg data is decreasing at zone 1 since 1971 (-1.58% per year) while other zones reveal no clear trend for Hg. Cr-data with Ni-normalization also reveals no clear trend. Decreasing trends for most heavy metals are as expected, since different time trends on a more limited time frame all give decreasing trends for heavy metals at the BPNS or the North Sea. Guns et al. (2015) evaluated the 1979-1995 time period, Gao et al. (2013) 1978-1998, De Witte et al. (2016) 2005-2014. Also within

OSPAR modelling, decreasing heavy metal time trends were noted for 1998-2007 (OSPAR, 2010). In the most recent OSPAR intermediate assessment (OSPAR, 2017), a decrease in Hg concentrations was noted but no statistical changes occurred for Cd and Pb for the 2005-2015 period at the southern part of the North Sea.

In contrast to these decreasing trends for heavy metal concentrations in marine sediments, an increase of Zn concentrations was noted at the BPNS from 1970 until 2016 varying from 2.7 to 6.7% since 1979 (Al-Fe normalization) to 10.6% since 1971 (Ni normalization). At restricted locations of the BPNS and on a more limited timeframe, a Zn increase was already noted from 2005 to 2014 at cluster zone 2 (sludge disposal site Nieuwpoort) and the west side of cluster zone 5 (nearby sludge disposal site Oostende) by De Witte et al. (2016). This work indicates that also on a larger time frame and within the different cluster zones of the BPNS, this increasing trend in Zn concentrations is appearing. Zn is a heavy metal with multiple sources within the marine environment. Within the seventies and eighties, large amounts of Zn were dumped at the BPNS, coming from titanium dioxide industry waste (Baeteman et al., 1987). Next to Cu, Zn is also used in marine antifouling products, especially since the ban on TBT (Turner, 2010). It is also used as anode on ships and marine constructions such as wind mill parks. Considering the expansion of wind mill parks and combined with trends in shipping traffic, it will be of special interest to follow up Zn concentrations in the upcoming years.

Heavy metal concentrations in mussel (Fig. 19) and flounder generally confirm sediment trends. Trends for Pb, Hg and Cd in flounder are decreasing by on average 2.4% (since 1971), 1.6% (since 1970) and 2.2% (since 1975) per year, respectively. Mussel heavy metal data was available from 1980 in Blankenberge and Oostende, from 1973 in Knokke and from 1974 in Nieuwpoort. In mussel tissue, trends in Cu (1.9-3.1% per year), Pb (2.3-8.7% per year), Cd (2.1-2.2% per year) and Hg (1.4-2.3% per year) were also decreasing. For Zn and As, trends in mussels are different from trends in sediment. An annual decrease of 1.6-1.7% was found for Zn in mussels while for As, the annual increase was 3.8-5.3%. This may be related to contamination differences between the tidal zones where groyne mussels are sampled and the open sea where sediment samples were taken. For Zn, input from new and historical sources at open sea such as wind mill parks, shipping or TiO₂ industry dumping sites may be counteracted at the coastal zone by reduced inputs through reductions of heavy metal emissions on land (OSPAR, 2010). The latter led to reduced inputs from the Schelde and by atmospheric deposition (Anon., 2016) which may impact concentrations of Zn in the water column near coast and hence affect groyne mussels. For As, it is still unclear what source may have affected the increase in mussel tissue.

Within latest OSPAR intermediate assessment (OSPAR, 2017), a status quo or downward trend was noted for Pb in fish and shellfish of the North Sea, no statistical change was found for Hg and even an increase for Cd over the 2005-2015 period has been observed. 4DEMON time trends provide a view on a larger time scale. Time trends for fish and shellfish reveal a decreasing trend for Hg, Cd and Pb in the seventies to nineties (Fig. 19) in line with publications from that period (Guns et al., 1992; De Clerck et al., 1995; Vyncke et al., 1996; Guns et al., 1999) However, concentrations levelled off over the last ten years which may

result in a steady state or even increase if a more recent, smaller time scale is considered, in agreement with OSPAR intermediate assessment (OSPAR, 2017).

Heavy metal data in swimming crab confirms mussel data, except for Cd, for which an increasing trend was found at the off-shore cluster zones 1 and 3. For shrimp, the concentration versus time modelling results in a maximum for Cu, Zn and Cd, followed by a decrease. It is remarkable that shrimp is the only species for which these maxima in time were identified. Due to its shorter lifetime and restricted habitat compared to swimming crab or flounder, concentrations in shrimp may be more affected by temporal or local differences. Since maxima in Cu, Zn and Cd concentrations occur in the nineties, there might be a relationship with heavy metals emissions from titanium dioxide industry dumping in the seventies and eighties (Baeteman et al, 1987).

For PCB trends in sediments at the BPNS and, extended, the southern part of the North Sea, literature information seems to give contradictory results. In sediments, Roose et al. (2005) did not find a significant decrease from 1991 to 2001 for CB153, the OSPAR quality status report (OSPAR, 2010) indicated no significant decrease for more than 80% of the PCB time series in the OSPAR zone II from 1998 to 2010. De Witte et al. (2016) found steady state or even an increase from 2005 to 2014. In contrast, Everaert et al. (2014) found a 50-66% decrease from 1991 to 2010. With the advantage of considering a larger timeframe and taking into account method switches within 4DEMON, it was found that most PCB's revealed a decreasing trend from 1991 to 2016 at the BPNS. E.g. a decrease in the different cluster zones of 0.28 to 3.6% per year was found for CB101 and 2.2 to 3.7% for CB180. Exception was CB52, for which a steady state was noted. The overall decrease in PCB concentrations resulted from a strong decrease in the nineties, followed by a levelling off or even an increase over the last 10-15 years. This is especially true for cluster zone 5 which reveals the strongest increase for all cluster zones. As can be deduced for CB180 in Fig. 18, PCB concentrations have increased in this zone since 2005, which may be related to inputs from the nearby mouth of the Westerschelde and/or dredge disposal sites (Everaert et al., 2014). In biota (Fig. 19, these levelling off is not or not yet seen in the data, with clear decreases since beginning of the eighties in flounder (2.9% per year for the sum of 7 OSPAR PCBs), mussel (3.0-3.5%/year) and shrimp (1.2-3.2%/year).

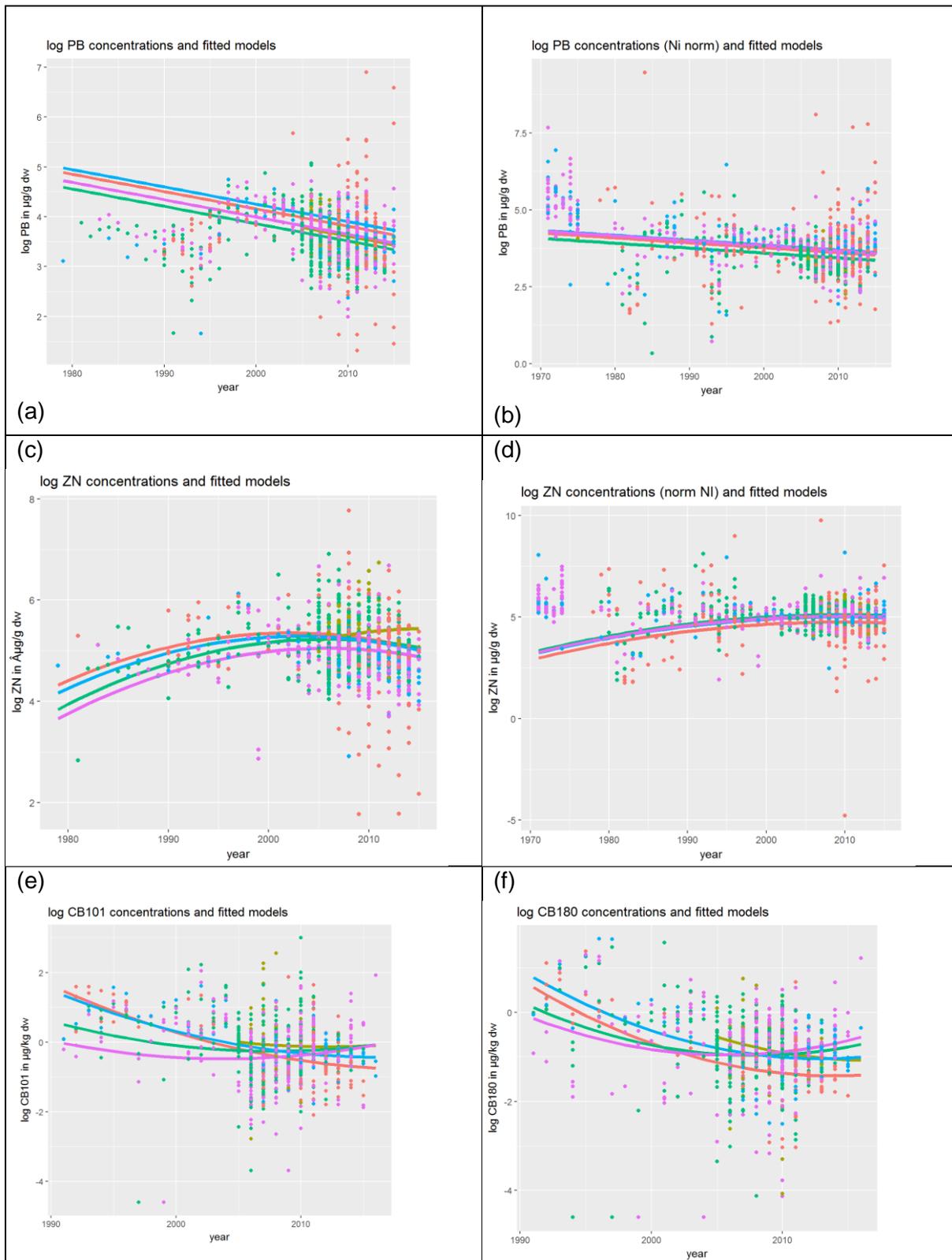
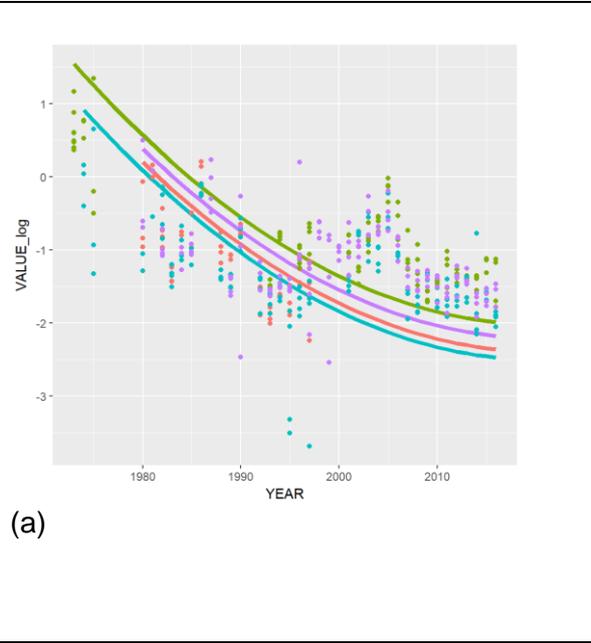
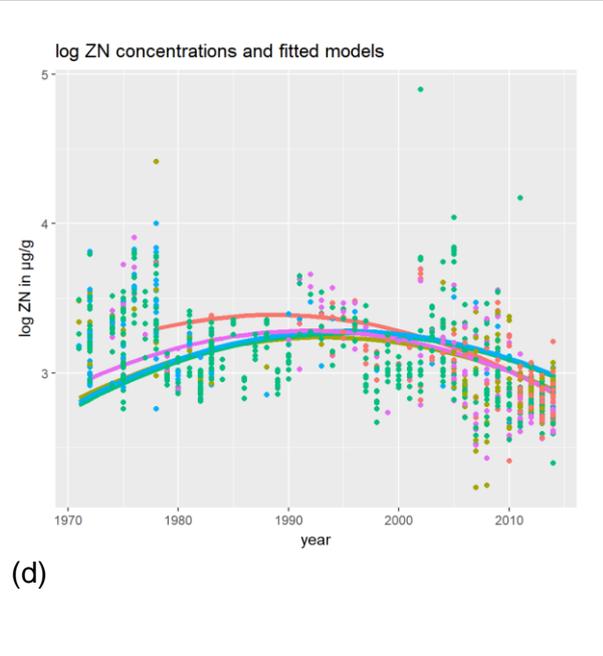
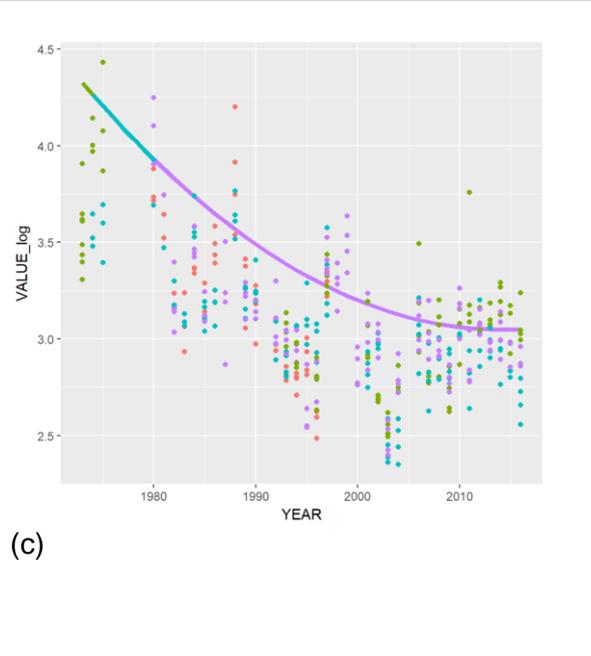
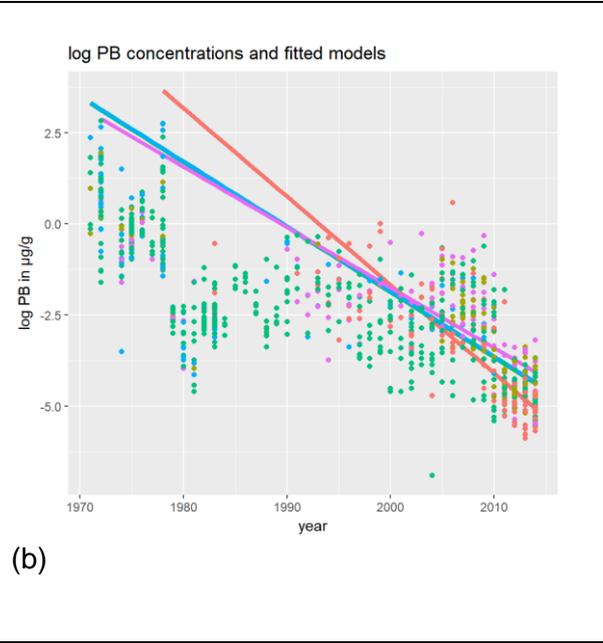


Figure 18: Log scale example results of sediment modelling, with (a) Pb, Al-Fe normalized, (b) Pb, Ni-normalized, (c) Zn, Al-Fe normalized, (d) Zn, Ni normalized, (e) CB101 and (f) CB180; Different zones are identified by color: zone 1 (red), zone 2 (olive green), zone 3 (green), zone 4 (blue) and zone 5 (purple).

Mussel



Shrimp



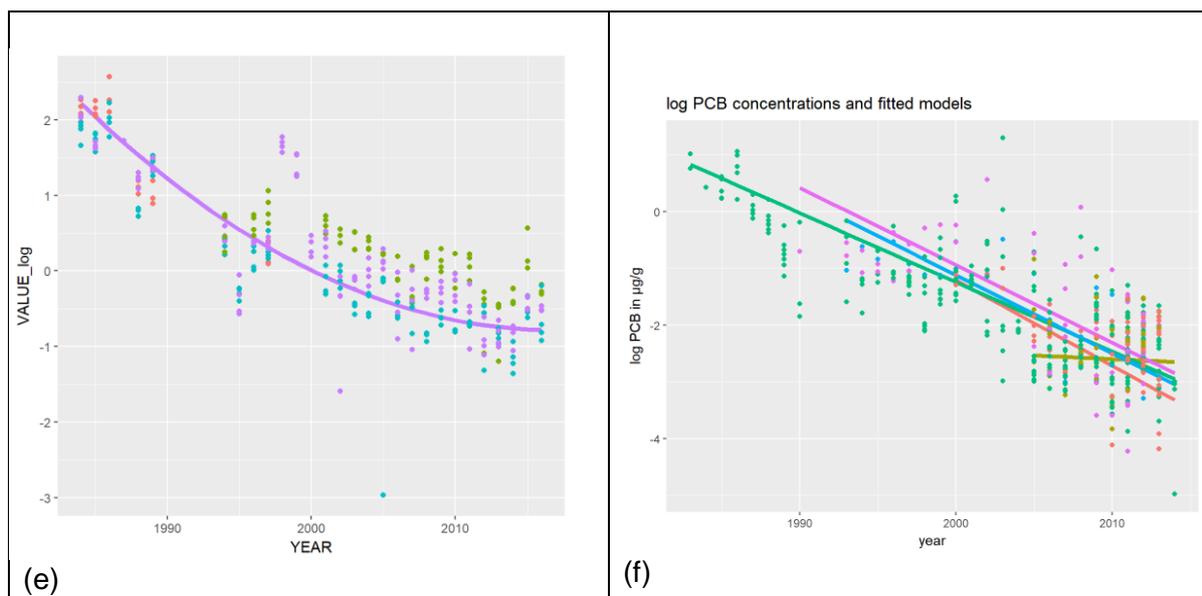


Figure 19: Log scale example results of biota modelling, with (a) Pb in mussel, (b) Pb in shrimp (c) Zn in mussel (d) Zn in shrimp (e) PCB in mussel and (f) PCB in shrimp. For mussels, different groynes are identified by color: Blankenberge (red), Knokke (green), Nieuwpoort (blue), Oostende (purple). For shrimp, different zones are identified by color: zone 1 (red), zone 2 (olive green), zone 3 (green), zone 4 (blue) and zone 5 (purple).

For heavy metals in sediment, time trend results could be compared with the analysis results of historical samples (WP3). Since only 5 time points were included, with a limited number of maximum 15 data points at each time point, no trend modelling was performed. Relative differences between time points were considered.

Selected results are presented at Fig. 20. Detailed results for all heavy metals are presented by Le et al. (in preparation, a). From 1970 to 2012, concentrations of Pb and As (Fig. 20) confirm heavy metal time trend modelling, revealing lowest values in 2012 and hence reduction in heavy metal concentrations. For Cu, Zn and Hg, no clear trend could be determined (Fig. 20), which is probably linked to the limited number of data points compared to the trend modelling results. No trends could be deduced for Cd and Cr. For Cd, too many values were below the quantification limit of the analytical method, for Cr, differences between duplo analysis were too large to give trustworthy results.

Focusing on the “Gilson” samples (1901-1911), Zn, Cu and Pb revealed low concentrations compared to the 1970-2012 period. For As and Hg, however, relatively high concentrations were noted (Fig. 20). This indicates that there were important sources of As and Hg in the 19th century. The use of coal, e.g. for domestic heating, may be an important source, since both compounds belong to the most volatile heavy metals in coal (Otero-Rey et al., 2003) and may enter the marine environment by atmospheric transport. As was also used in the 19th century as pesticide, pigment, wall paper glue, etc. Hg was used in the production of felt, within electrochemistry, as a medicine, amongst other applications. Results on Hg concentrations also confirm its tendency to strongly bind to fine sediments (Loring et al., 1991), as concentrations of Hg are lowest at cluster zone 1, medium at cluster zones 2 and 3, and highest at coastal zones 4 and 5 (Fig. 3.6.3).

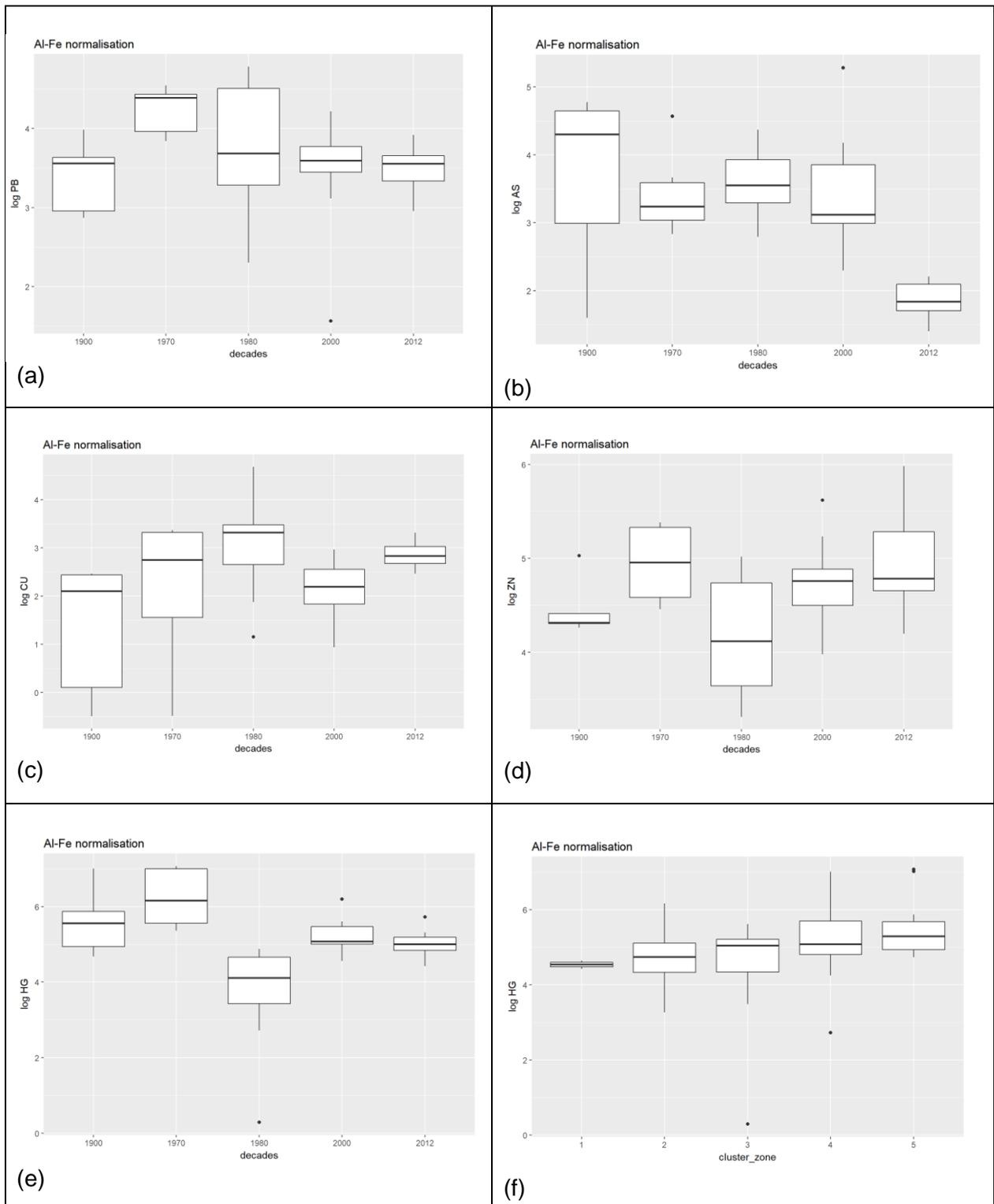


Figure 20: Log scale boxplots of historical sample analysis, normalized to Al-Fe. (a)-(e) Concentrations in function of time points for respectively Pb, As, Cu, Zn and Hg and (f) Hg concentrations in function of cluster zone.

3.6.2. Results on eutrophication

3.6.2.1. Trends in eutrophication and CPHL patterns of variability (Results WP4 Xav)

3.6.2.1.1. Trends in nutrients: river loads and marine concentrations

N and P loads from the rivers Scheldt and Rhine/Meuse have reached a maximum in the 1980's and followed a decreasing trend since the mid-1980's (Brion et al., 2006). This de-eutrophication (or oligotrophication) process is visible in the time series of riverine N and P concentrations (Fig. 21, left). The decrease in riverine P is mainly due to the ban of polyphosphate from washing powder in the early 1990s, and to waste water treatment (WWT) from urbanized and industrialized areas (Urban Waste Water Treatment Directive, UWWTD, Directive 91/271/EEC). The decrease in riverine N is linked to a combination of measures: the operation of WWT that removed organic matter and the NH_4^+ loads of the Scheldt and the increasing implementation of good agricultural practices (moderate use of fertilizers following the Nitrate Directive, Directive 91/676/EEC). That rapid decrease in riverine nutrient concentrations was not followed by such a rapid decrease in marine nutrient concentrations (Fig.X2, right), which suggests a coastal accumulation of nutrients through benthic-pelagic coupling. The dissolved silica (DSi) concentration in rivers has been increasing during the de-eutrophication because of a lesser diatom production in rivers and estuaries. As a result, the export of DSi to the coastal zone has increased, probably fostering diatom production (Prins et al., 2012).

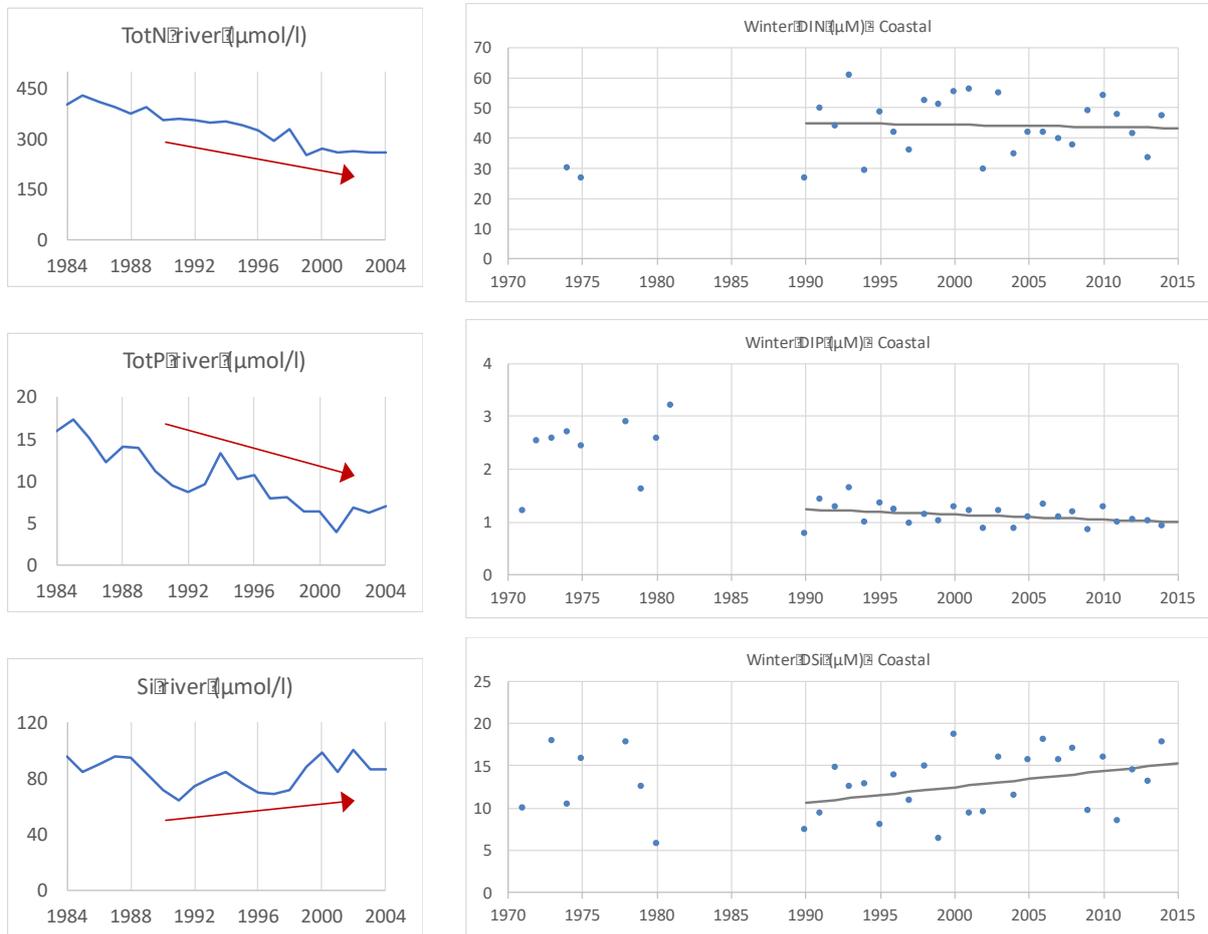


Figure 21: Left – Nutrient concentrations lumped in the Scheldt [51.37N, 4.208E], Rhine/Meuse [51.98N, 4.12E] and North Sea Canal [52.47N, 4.6E]. Right – Coastal marine nutrient concentrations lumped in the Zones 1-2-3 of BPNS. The regressions are Generalized Least Squares functions taking account of the heterogeneity of residuals (performed in R).

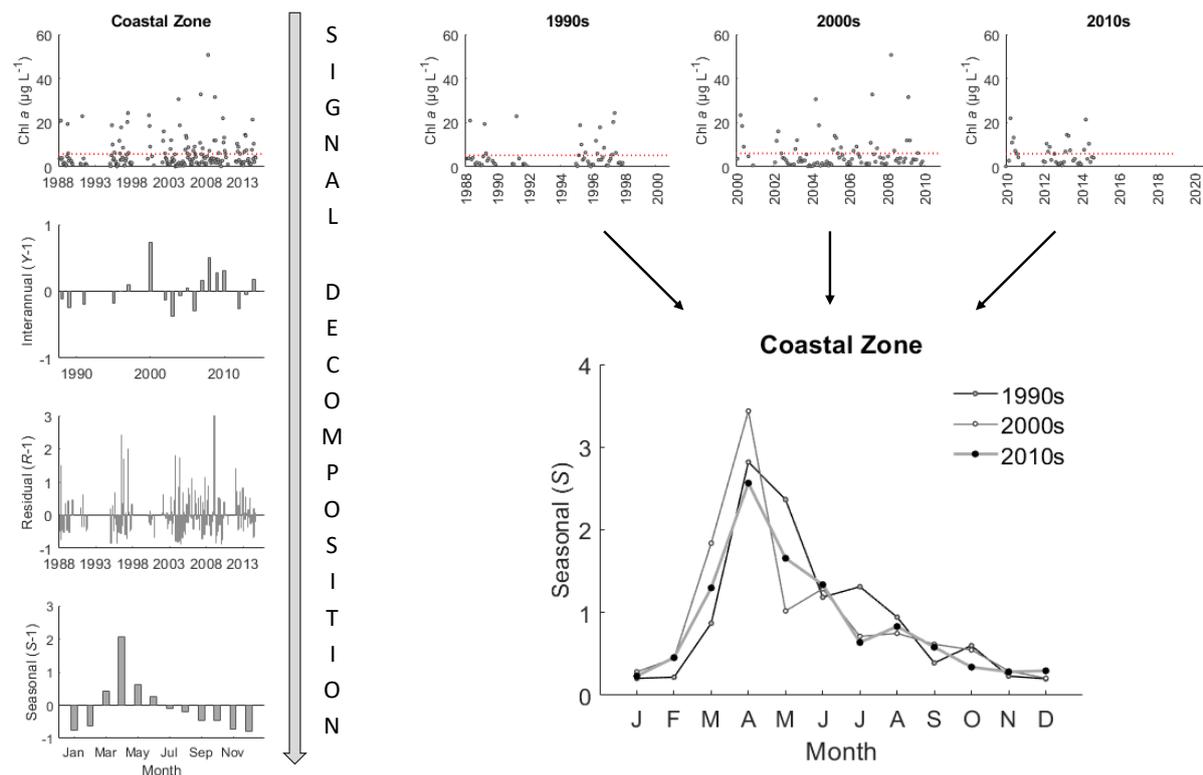


Figure 22: Left – Example of a multiyear CPHL signal decomposition into its seasonal component of variability in the coastal zone of the BPNS (Zones 1-2-3) over the period 1988-2013. Right – The CPHL signal decomposition is made decade per decade in the coastal zone, keeping only the years with sufficient seasonal coverage. The outcome is a CPHL seasonal component of variability per decade.

The Fig. 22 illustrates such a decomposition of the CPHL signal in the coastal BPNS and the resulting seasonal signals of CPHL per decade. There is a visible change in the CPHL phenology with an earlier bloom in more recent years. The analysis was repeated in every Zone (1 to 6; not shown) and indicated the same trend everywhere, also in the offshore parts of the BPNS less exposed to freshwater influence. The change in CPHL phenology is clearer in less eutrophicated areas (Z1, Z4; not shown). Earlier CPHL blooms in recent years were also observed in Dutch coastal and offshore stations of the Walcheren transect (closest to BE; Fig. 23, left-top). In the last decades, marine N and P concentrations have decreased in the coastal zone and that may have influenced the phytoplankton succession, but the nutrient decrease is marginal offshore (not shown). Therefore, nutrients cannot explain the ubiquitous change in CPHL phenology. However, SST has increased in the last four decades in the Belgian waters, especially in spring and summer (Fig. 23, right). Following the observations of Hunter-Cevera et al. (2016), our hypothesis is that the SST increase in the Southern North Sea caused an increase in phytoplankton cell division rates in the spring. The earlier and more rapid formation of CPHL blooms has also an impact on the nutrient consumption (Fig. 23, left-bottom).

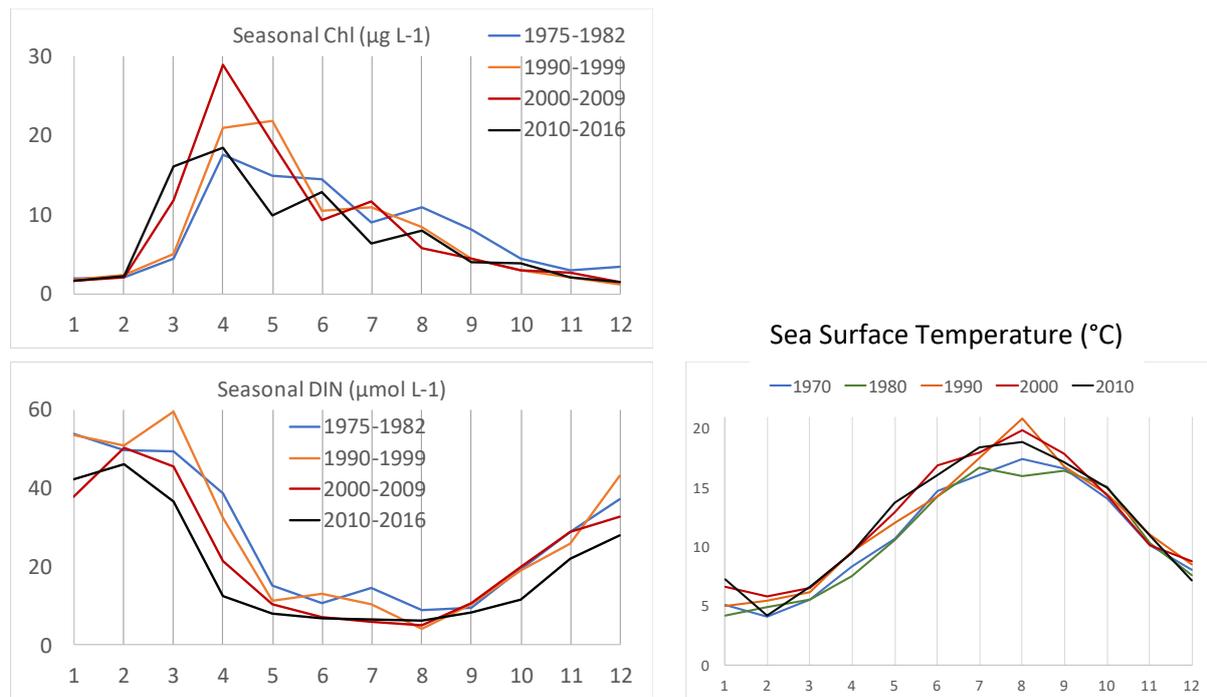


Figure 23: Left – Seasonal signal of CPHL and DIN at the Dutch site Walcheren 2 in the period 1975-2016. Right – Seasonal signal of SST in the BPNS (spatial aggregation) in the period 1970-2010.

3.6.2.1.2. Signature patterns of chlorophyll *a* variability in the Greater North Sea

The satellite-based CPHL archive provided 20-year long CPHL time series per 1km x 1km pixel describing the phytoplankton dynamics for that location (Desmit X. and Van der Zande D., in preparation). Understanding of phytoplankton variability is a key to understanding variability of biogeochemical processes, ecosystem metabolism, and production in food webs. To identify patterns in phytoplankton variability, the CPHL time series were decomposed into an annual effect, mean seasonal pattern, and residual “events” following the model described by Cloern and Jassby (2010). Additionally, the grand mean was calculated at pixel level resulting in four components describing the algae dynamics in the considered pixels. The annual component measures the deviation of mean CPHL for an individual year from the long-term mean. Similarly, the seasonal component describes the deviation of mean CPHL for a given month from the annual mean; it provides us with an “average” pattern of seasonal variability. Finally, the residual component measures deviations from the average seasonal pattern. The four components describing phytoplankton variability were subsequently used as input into an unsupervised k-means classification algorithm to cluster the pixels into an undefined number of groups with a minimized intra-cluster variance. The results provide a classification map (Fig. 24) where pixels of the same class are characterized with similar phytoplankton variability enabling the determination of ecologically relevant regions in the North Sea, the Celtic Seas and the BPNS.

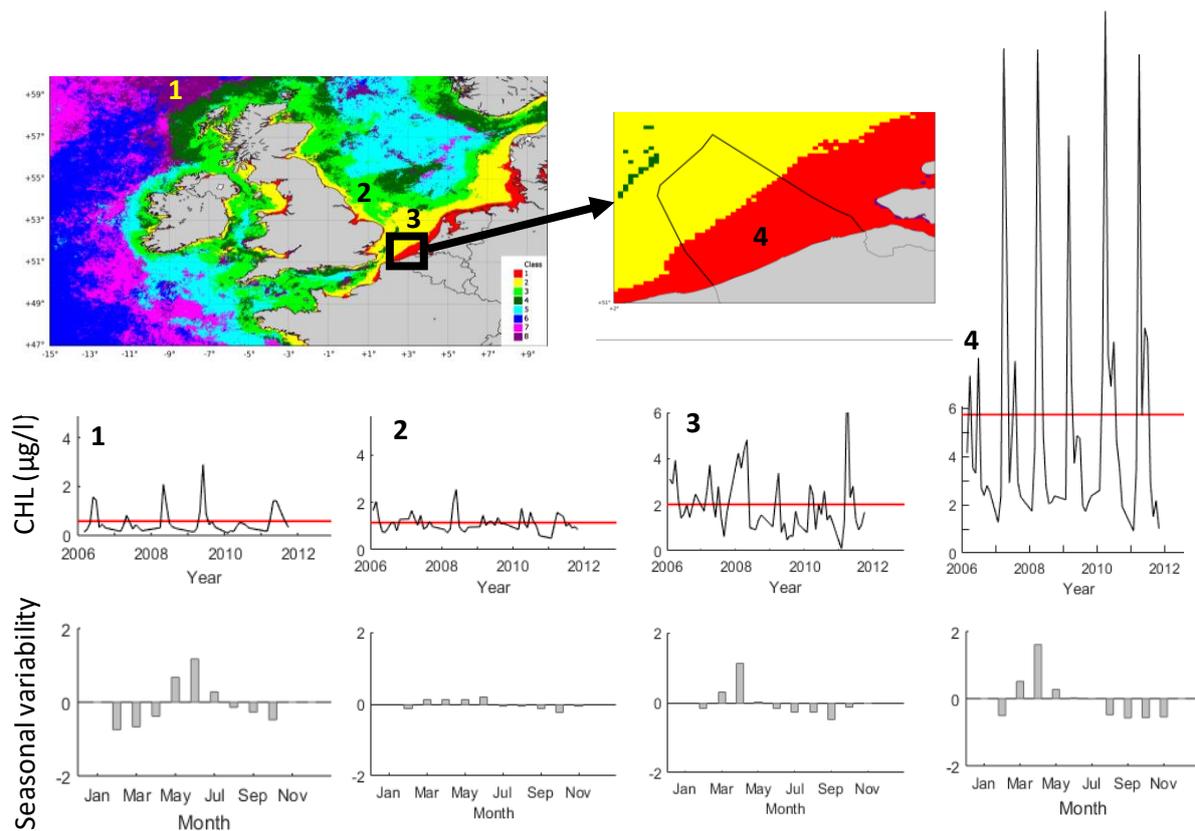


Figure 24: Top left – Classification map for the North Sea showing eight classes in which CPHL exhibits different patterns of variability, with a zoom on the Belgian coastal zone. Example of CPHL time series are shown for pixels in four different classes: the North Atlantic front (1), the eastern UK coast (2), the Southern North Sea tidal front (3) and the Belgian coastal zone (4). These CPHL time series were decomposed to show their mean seasonal pattern (bottom row).

Figure 24 illustrates the classes drawn from the signature patterns of CPHL variability. The map reflects the strong influence of bathymetry on CPHL variability, but also the influence of other physical properties of the basin: the stratification regimes, the fronts, the deep-water ascent at the margin. For instance, the CPHL at the North Atlantic front shows relatively high concentrations compared to the ocean (not shown) and a regular seasonal pattern with a bloom in June. In contrast, the eastern UK coastal waters exhibits a high irregularity in the CPHL signal with a bloom timing varying from year to year and depending on the intermittent stratification in that area (van Leeuwen et al., 2015). The seasonally stratified waters of the Central North Sea exhibit a slightly more regular seasonal pattern (not shown). At the tidal front between the Southern and the Central North Sea, the CPHL time series also shows irregularity although the main CPHL bloom occurs every year in April. The CPHL signal there is perturbed by events causing irregular conditions in nutrient inputs or stratification regimes. The Belgian coastal waters shelter very intense CPHL blooms. The regularity of the spring bloom from year to year suggests that the temporal variability of CPHL is more depending on astronomical processes, such as the solar irradiance and the daylength, than on local processes despite the obvious influence of freshwater nutrient inputs. Some years,

like 2006, show a small spring peak of CPHL and a lower annual mean, suggesting that those years large colonies of *Phaeocystis globosa* are not found.

3.6.2.2. Changes in phytoplankton taxonomic composition

3.6.2.2.1. Long-term trends in diatom and dinoflagellate abundance, biomass, community structure and seasonality: comparison between 1970s and 2000s

Phytoplankton is mainly dominated by diatoms, dinoflagellates and the colonial haptophyte *Phaeocystis*, but unfortunately no 1970s data were available for the latter. Marked changes in the abundance, biomass, structure and seasonality of diatom and dinoflagellate communities were observed between the 1970s and 2000s. Distinct changes in all parameters were observed between the 1970s and 2000s, with (1) a pronounced increase in diatom biomass from late winter to summer, but a decrease in autumn, resulting in a more continuous and intense growing season; (2) a year-round but especially summer increase in dinoflagellate biomass; (3) a 3-week and 2-month forward shift of the diatom and dinoflagellate fulcrum respectively; (4) marked changes in community structure, with a trend towards seasonal homogenization in the diatom community (i.e. a blurring of the distinction between three regionally typical seasonal community types); (5) increased occurrence of harmful diatom (*Pseudo-nitzschia*) and dinoflagellate (e.g. *Prorocentrum*) genera.

Diatom and dinoflagellate abundances have increased between the two periods with overall higher means and medians in the 2000s (Figure 25).

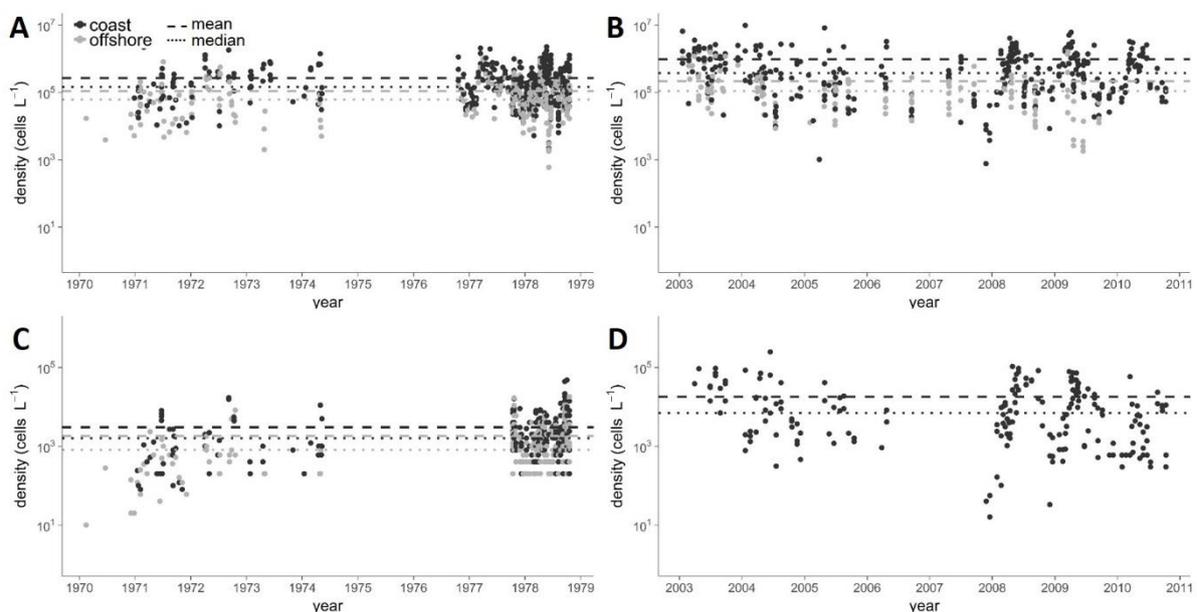


Figure 25: Phytoplankton count data. (A) Diatom data in 1970s, (B) diatom data in 2000s, (C) Dinoflagellate data in 1970s and (D) sampling in 2000s. Coastal (grey) and offshore (black) stations are distinguished.

In the 1970s large diatom species dominated the diatom community twice a year (spring and late summer/autumn, Figure 26A). In the 2000s, large diatoms almost continuously dominated the community from late winter to summer (Figure 26B), but were generally low in autumn and early winter.

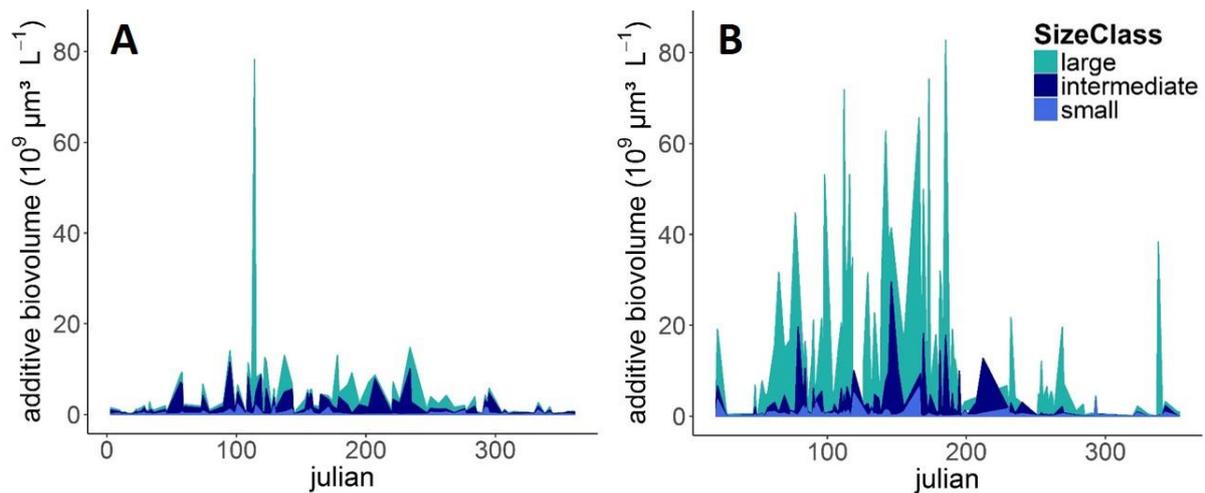


Figure 26: Annual cycle of small, intermediate and large diatoms in the onshore stations of the BPNS. A 1970s and B 2000s.

The diatom bloom pattern in 1970s typically had two annual peaks in spring and summer (Figure 27A), but only one defined peak in the 2000s (Figure 27B). Dinoflagellates in both periods bloom twice a year (Figure 27C and Figure 27D), but in the 1970s the first smaller peak occurs around May and the second one in late summer to early autumn (Sep-Oct), while in the 2000s the peaks are not very sharply distinguished due to an overall increase in dinoflagellate biomass between spring and summer, and the second peak occurs earlier during the year (July/Aug).

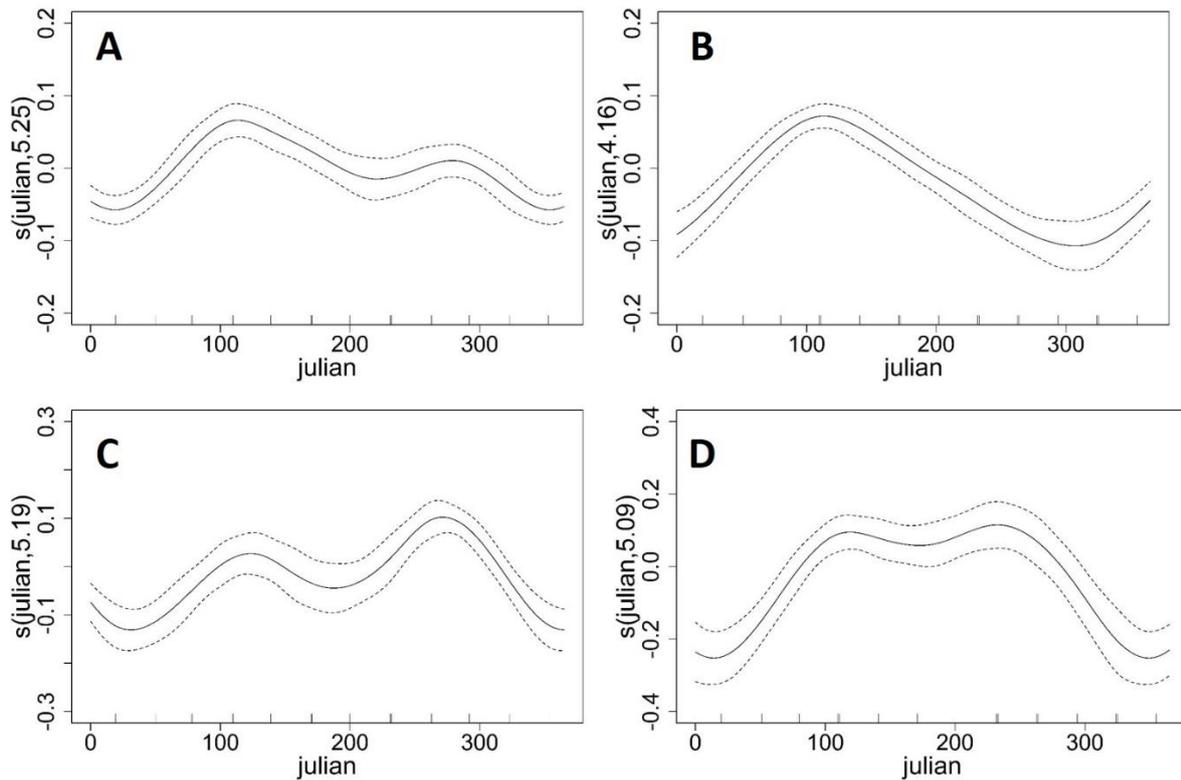


Figure 27: Seasonal trends of the diatom and dinoflagellate abundances. The solid line is the smoothing curve fitted by the GAMM model and the dotted lines represent the 95% confidence bands. A Diatom abundances in the 1970s, B diatoms in the 2000s, C dinoflagellates in the 1970s and D dinoflagellate abundances in the 2000s.

This annual forward shift of the diatom and dinoflagellate maximum abundances and the higher diatom winter abundances are confirmed by the forward move of the day on which the fulcrum is reached, which is for the diatoms about three weeks and for the dinoflagellates even two months (Figure 28).

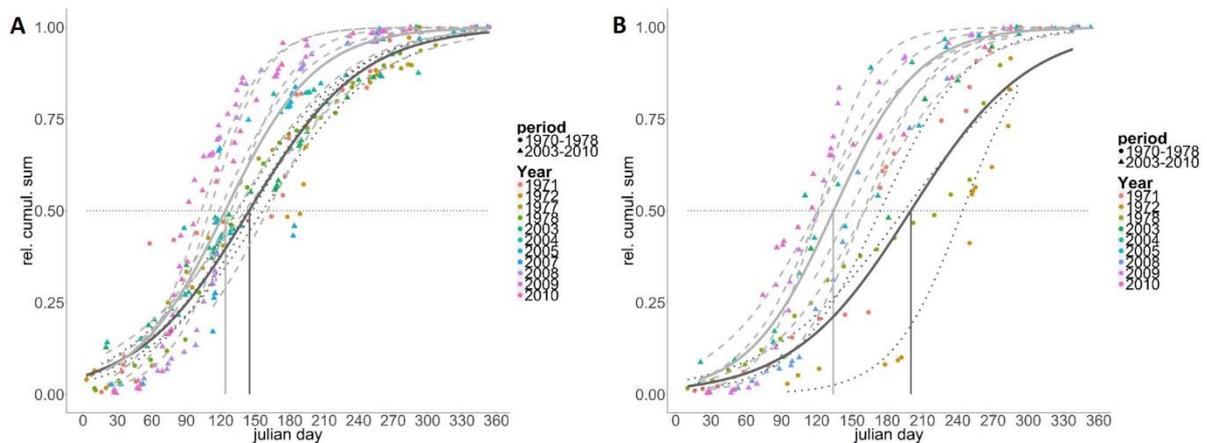


Figure 28: Fulcrum analysis of the relative cumulative sum of the annual cell abundances. A binomial glm smoother function is fitted on each annual data subset. In addition, a 'mean' smoother per period is added (in bold). Dark grey: 1970s, light grey: 2000s. The Julian day on which half of the annual phytoplankton cell abundance (the fulcrum) is reached is indicated. A Diatoms, B dinoflagellates.

In the BPNS three phytoplankton communities have traditionally been distinguished (Figure 29) (Rousseau et al. (2008)): (1) a late winter-early spring community with mainly by benthopelagic diatom taxa such as *Paralia*, *Rhaphoneis*, *Thalassionema* and also benthic diatoms; (2) an 'intermediate' community dominated by *Chaetoceros* but also containing *Pseudonitzschia* spp. and (3) a spring-early summer community dominated by large-sized diatoms such as *Guinardia* and *Rhizosolenia* spp., which often co-occur with *Phaeocystis* colonies. The most striking difference between the 1970s and 2000s is that the distinction between these communities becomes more blurred as the diatom community becomes seasonally more homogenized: both the autumn-winter and summer communities become more similar to the intermediate community due to a general increase of diatom genera belonging to the latter community, and especially *Chaetoceros*.

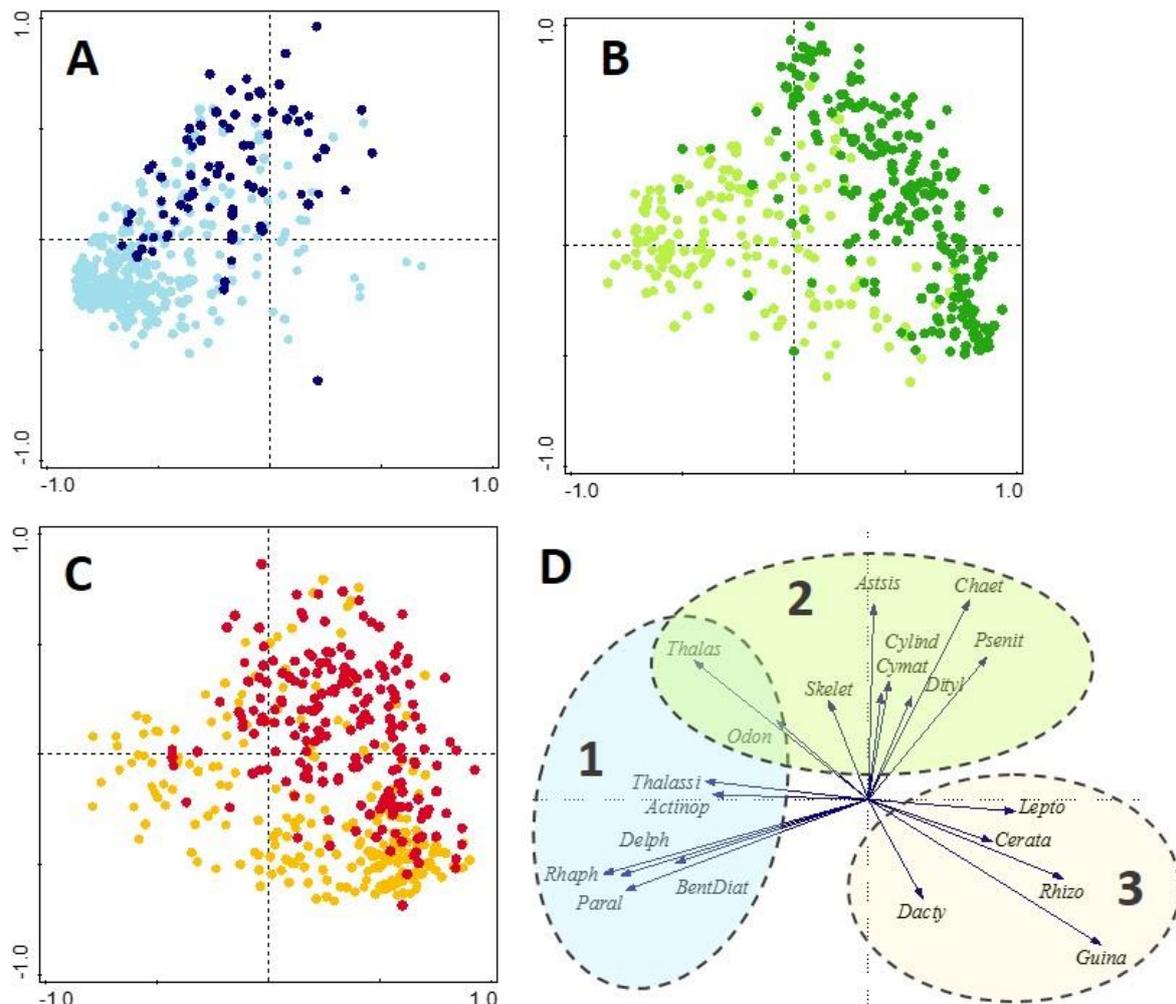


Figure 29: Principal Component Analysis (PCA) of the logarithmic transformed relative phytoplankton abundances. A Autumn-winter, B spring, C summer, D genera. Ellipses indicate the three typical annual communities in the BPNS. Darker colors represent p2 (A-C). Autumn/winter: September-February, spring: March-May, summer: June-August.

3.6.2.2.2. *Phaeocystis* spp. and its relation to environmental conditions

Generally, there are big differences in the development of *Phaeocystis* blooms even in areas relatively close to each other like the French, Belgian and Dutch coastal areas (Figure 30A). In the BPNS, there are years with dense *Phaeocystis* blooms (e.g. 2007, 2008, 2015 and 2016) and years in which *Phaeocystis* does not form blooms (e.g. 2011-2013) (Figure 30B).

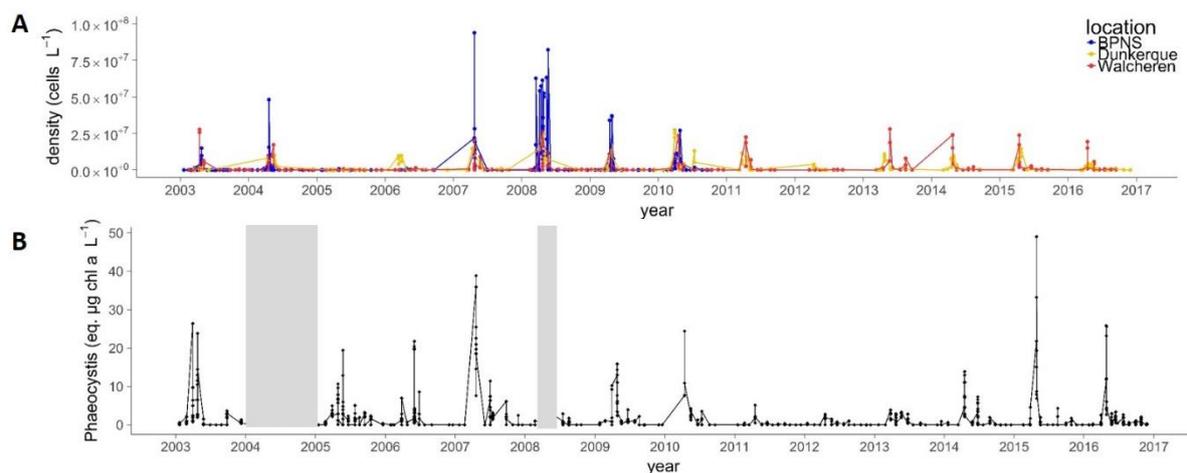


Figure 30: Long-term trend of A Phaeocystis cell counts in the BPNS, the French (Dunkerque transect) and the Dutch (Walcheren transect) coastal zone and B the Phaeocystis chlorophyll equivalent deriving from HPLC-CHEMTAX analysis in the BPNS. Source French SRN datasets: <http://www.seanoe.org/data/00397/50832/>. Dutch Phaeocystis count data has been provided by the service desk of the RWS.

The link of the *Phaeocystis* spring peak values with the preceding environmental winter conditions reveals a negative correlation with the nutrient and TSM concentrations and a positive correlation with the DIN:DIP ratio, SST and winter NAO index (Figure 31).

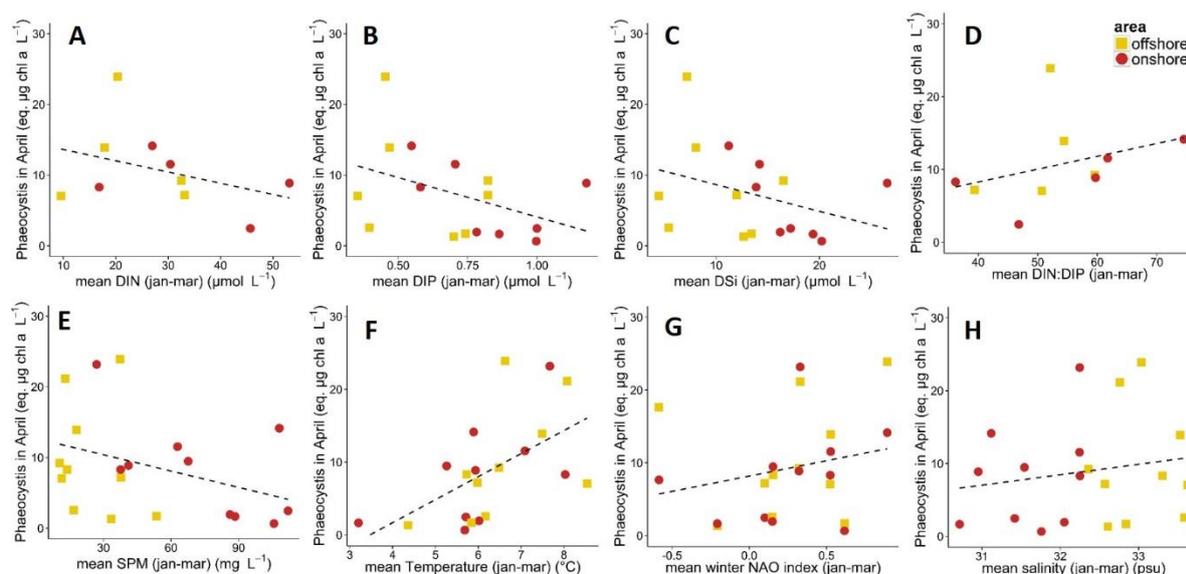


Figure 31: Correlation Phaeocystis in April determined by HPLC-CHEMTAX and mean winter environmental parameters (January- March). A DIN, B DIP, C DSi, D DIN:DIP ratio, E TSM, F temperature, G winter NAO index and H salinity.

These observations could be confirmed by the performance of a correlation test (Table 10). A higher SST in the preceding months stimulates the *Phaeocystis* spring bloom. Nutrients were most of the time negatively correlated with the *Phaeocystis* peak. As further as we go back in time, most correlations get less strong with only one significant relationship of the January to March conditions left, the correlation of temperature to the *Phaeocystis* spring peak.

Table 8: Correlation of April Phaeocystis (HPLC-CHEMTAX, 2003-2016, without 2004, 2006 and 2008) with means of April, March, February-March and January-March of other HPLC-CHEMTAX phytoplankton groups, in-situ abiotics, Remote Sensing chlorophyll a and TSM and winter NAO index. Spearman rank correlation is used.

	<i>Phaeocystis</i> - variables (April)	<i>Phaeocystis</i> - variables (March)	<i>Phaeocystis</i> - variables (Feb-Mar)	<i>Phaeocystis</i> - variables (Jan-Mar)
Chlorophytes	-0.28	-0.14	-0.31	-0.28
Cyanobacteria	0.4	0.02	-0.08	-0.06
Diatoms	-0.54**	0.29	-0.05	-0.01
Dinoflagellates	0.02	-0.16	-0.35	-0.24
Euglenophytes	-0.49*	-0.26	-0.4	-0.31
NO ₂	-0.12	-0.42	-0.14	-0.18
NO ₃	-0.54*	-0.67**	-0.48	-0.33
NO ₂ NO ₃	-0.49	-0.65**	-0.48	-0.33
NH ₄	0.36	0.22	0.12	0.1
DIN	-0.52	-0.49	-0.3	-0.2
DIP	-0.1	-0.67**	-0.44	-0.39
DSi	0.61*	-0.71**	-0.4	-0.37
DIN:DIP	-0.32	0.52	0.08	0.62
DSi:DIP	0.81***	-0.16	-0.11	-0.03
CPHL a (in situ)	0.78***			
CPHL a (Remote Sensing)	0.52*			
TSM (in situ)	0.12	-0.43	-0.47*	-0.32
secci	0.01	0.31	0.22	0.13
TSM (Remote Sensing)	0.19	-0.11	-0.22	-0.15
temp	0.4	0.54*	0.61**	0.57**
salinity	-0.1	0.28	0.1	0.11
NAO index	-0.31	0.51*	0.24	0.31

3.6.3. Trends in pH and CH₄

Annual means show an increase of pH from the early 1970's to the mid 1980's, and then a decrease on-wards (Fig. 32). This pattern could be related to changes in primary production related to eutrophication and nutrient reduction policies, in response to European legislation (reduction of phosphate emissions to rivers and coastal zones implemented from the mid

1980's onwards), in agreement with the MIRO model (Lancelot *et al.*, 2007). The seasonal signal of pH remained the same during the time-series with an increase during spring during the phytoplankton bloom (Fig. 32). However, the seasonal amplitude of pH changed according to the decade, and was highest during the 1980-1989 decade indicating a more intense phytoplankton biomass, as the seasonal amplitude of the pH signal can be interpreted as a integrative proxy of annual primary production.

Data in surface waters of the BPNS in spring, summer and fall 2010 and 2011 show very high CH₄ concentrations (up to 1,100 nmol L⁻¹) were observed in surface waters of the BPNS compared to open oceanic conditions (<5 nmol L⁻¹) due to release of CH₄ from sediments (in-situ production and leakage from gassy sediments) and the well-mixed water column that allows an efficient transfer of CH₄ from bottom waters to surface waters (Fig. 33). Our data suggest that further warming of surface waters could increase CH₄ emissions (Fig. 34) and provide a positive feedback on warming climate. This feedback will be expected to be acute in shallow gassy areas such as the BCZ since they are natural hotspots of CH₄ emission, and the well-mixed water column will allow an efficient propagation of additional heat to the sediment that will be buffered by seasonal thermal stratification in deeper seep areas. The increase of temperature will stimulate the biogenic CH₄ production, as well as, decrease Henry's constant promoting bubbling from sediments. Dissolved CH₄ concentrations were measured at five stations in the BPNS in March 1990 (Scranton and McShane, 1991) and were compared to data acquired at the same stations in March 2016 (Fig. 35). The average water temperature in March 1990 (8.7±0.5°C) was similar to the one in March 2016 (8.3±0.2°C), as well as average salinity (33.5±1.4 in 1990 versus 32.8±2.0 in 2016) and average monthly wind speed (6.2±2.4 m s⁻¹ in 1990 versus 5.5±2.1 m s⁻¹ in 2016). This suggests that the two cruises were comparable with regards to the seasonal biological and physical cycles. The CH₄ concentrations in March 1990 ranged between 26 and 454 nmol L⁻¹, and were systematically higher than in March 2016 when they ranged between 8 and 41 nmol L⁻¹. The same spatial gradients as present-day (Borges *et al.*, 2017) were observed in March 1990 with higher concentrations in near-shore stations (120, 700 and 710) than off-shore stations (215, 330). The largest difference between March 1990 and 2016 was observed at a near-shore station (26.8 times higher at station 120 in 1990 than 2016) and the lowest difference at a more off-shore station (3.5 times higher at station 330 in 1990 than 2016). The differences of CH₄ concentration between 1990 and 2016 can be due to long-term trends or inter-annual variability. We do not have additional data in March of other years to evaluate inter-annual variability, but data from April 2010 (Borges *et al.*, 2016) at three stations (120, 700 and 710), are also much lower than the March 1990 values (Fig. 33). The tendency of a decrease of CH₄ concentrations in the BCZ between 1990 and 2016 is consistent with a strong decrease in primary production in the area since the mid-1980's (Lancelot *et al.* 2007). The decrease since the mid-1980's of primary production resulted from river nutrient load reduction policies, and has been shown to also strongly affect the CO₂ and DMS concentrations and air-sea fluxes in the BCZ (Gypens *et al.*, 2009; Gypens and Borges, 2014). A decrease of primary production would result in a decrease of OM delivery to the sediments that drives the benthic CH₄ production (Fig. 4). Although the water temperature during the two cruises in

1990 and 2016 were comparable, the long-term trend in the North Sea for that period is a fast increase in temperature at a rate of $0.035^{\circ}\text{C yr}^{-1}$ (Høyer and Karagali, 2017). This would imply that despite the warming of nearly 1°C from 1990 to 2016, the change primary production had a much more important effect on CH_4 concentrations in the BCZ. The BCZ has experienced during the last decades a decrease in eutrophication, that probably resulted in a decrease of CH_4 production in sediments and CH_4 concentrations in surface waters (Fig. 35). The corollary implies an increase of CH_4 production and emission might be expected from eutrophication that is on-going in numerous coastal marine areas worldwide (e.g. Díaz and Rosenberg, 2008). The positive relationships between dissolved CH_4 concentration and water temperature and sediment organic matter (OM) content we found in the study area suggests that an increase of emissions of CH_4 from coastal areas may occur in response to warming of surface waters and an increase in OM deposition due to eutrophication. Warming of superficial sediments should be more acute in permanently well-mixed continental shelves, which account for about one third of total continental shelf areas. In addition, in permanently well-mixed continental shelves the transfer of CH_4 from bottom waters to surface waters (hence emission to atmosphere) is much more efficient than in seasonally thermally stratified continental shelves (Borges *et al.*, 2016). This scenario contrasts with the one of Naqvi *et al.* (2010) who postulated that in open oceanic regions the CH_4 emissions should not change in response to climate warming and expanding oxygen minimum zones, because an enhancement of methanogenesis was assumed by these authors to be compensated by an increase of methane oxidation.

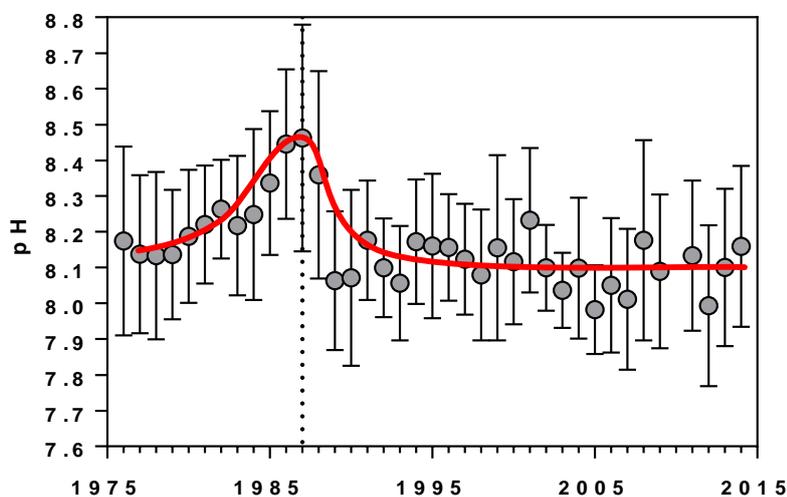


Figure 32: Annual average of pH in the BPNS.

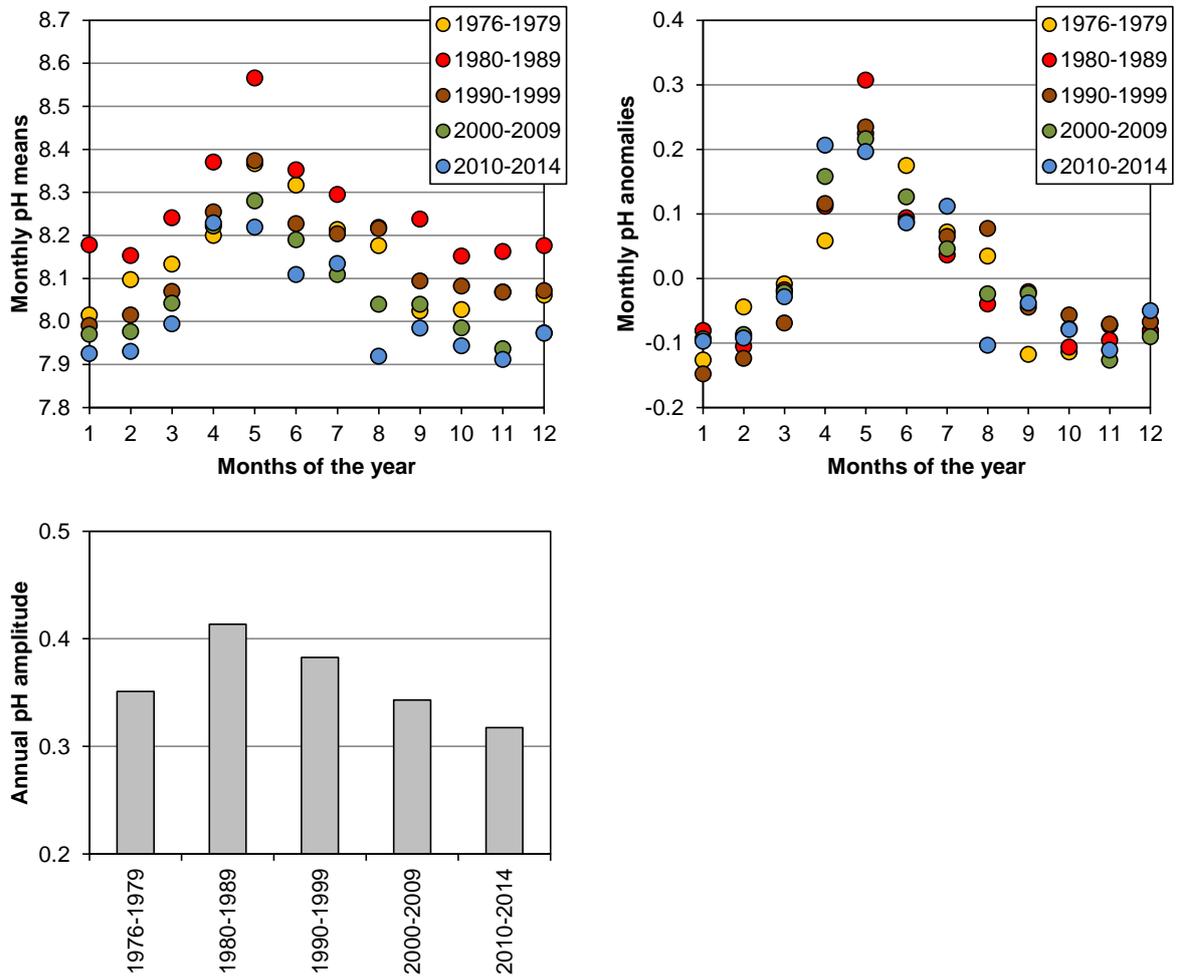


Figure 33: Seasonal evolution of pH, monthly pH anomalies, and annual pH amplitude in the BPNS per decade.

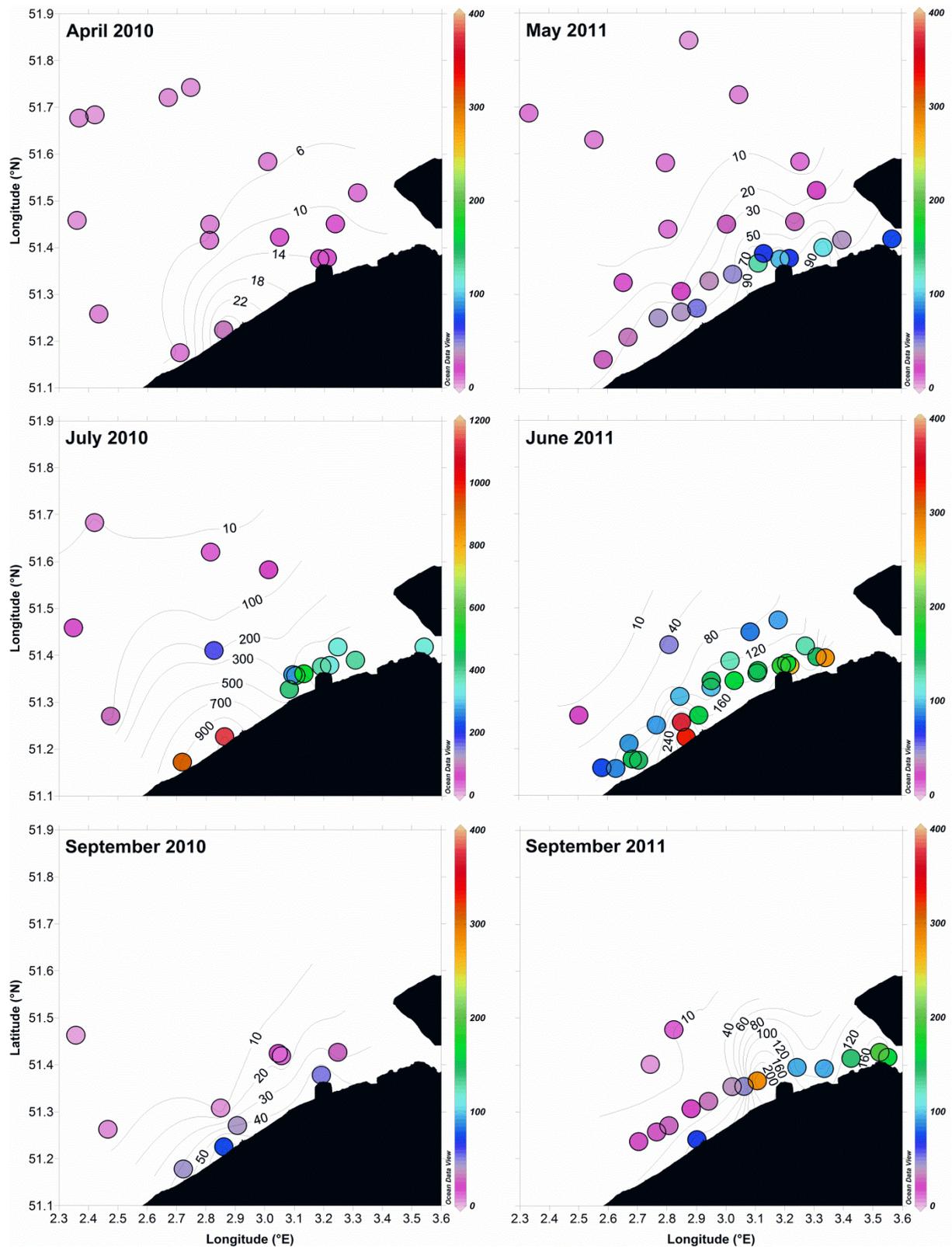


Figure 34: Concentration of dissolved CH₄ in surface waters of the BPNS in spring, summer and fall 2010 and 2011. Note the different color scale in July 2010 compared to the other cruises.

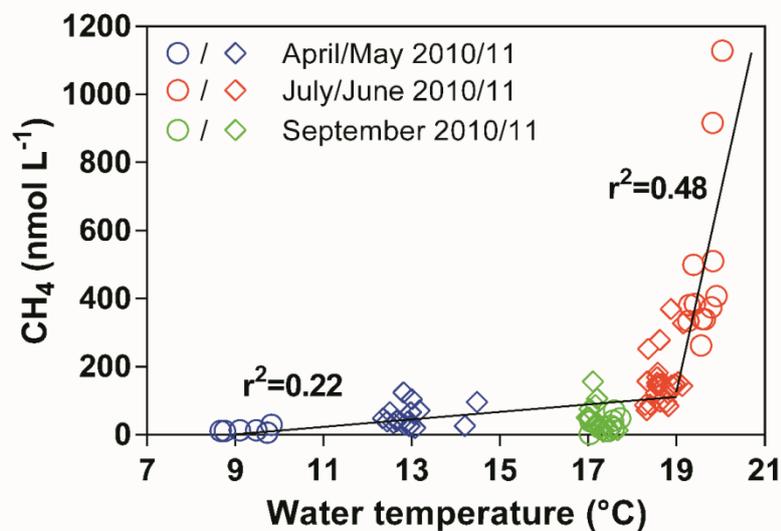


Figure 35: Concentration of dissolved CH₄ in surface waters of the BPNS as a function of temperature in spring, summer and fall 2010 and 2011. Solid lines indicate the linear regressions for data < and > 19°C.

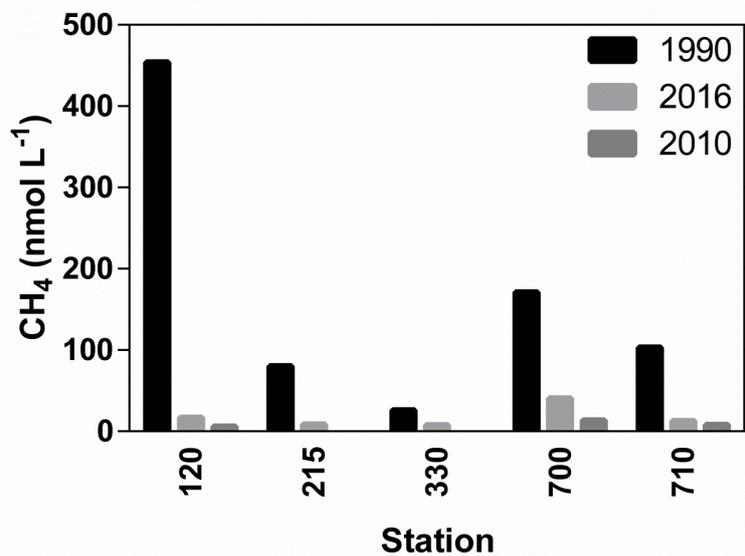


Figure 36: Comparison of dissolved CH₄ concentrations at five stations in the Belgian coastal zone obtained in 12-14/03/1990 and 30-31/03/2016, and at three stations in 22-23/04/2010.

3.6.4. Summary of the long-term trend analyses

Table 9 shows a summary of the results of the trend analyses performed on the compiled datasets within 4DEMON.

Table 9: Summary of state of the art for each parameter group of the long-term trend analyses performed in 4DEMON.

parameter	Trends 1970-2014 (or later)
Heavy metals (sediment, biota)	Most heavy metals reveal a decreasing trend in sediment and biota, however, some exceptions should be closely monitored, such as Zn in sediment and As in mussels.
PCBs (sediment, biota)	Decreasing in biota. In sediment, there is an overall decrease although local increases are again noticed over the last 10 years, especially within contaminant cluster zone 5.
Nutrients (DIN, DIP, DIN:DIP)	Pronounced decrease in DIP and also DIN (but not winter), increase in Si (except in winter), all resulting in significant changes in nutrient ratios (DIN:DIP and DSi:DIP higher > 2000 from October to March).
TSM	Pronounced decrease in winter TSM values (December-March) but not the rest of the year.
Chlorophyll a (phytoplankton biomass)	Changes in phenology (earlier blooms, from February onwards) and seasonal dynamics (with increases > 2000 from February-May and lower values throughout the rest of the year).
Phytoplankton community structure	Pronounced shift in community structure, seasonal dynamics and biomass between the 70s and > 2000 (no information in primary production). In general, the spring bloom peak (April) mainly related to the harmful alga <i>Phaeocystis</i> has become more defined and probably also more pronounced. Diatoms (as the second most important phytoplankton group) have undergone dramatic changes, with higher biomass > 2000 from December-August and lower biomass from September-November, a significant shift in community structure with communities becoming more similar throughout the year (seasonal homogenization) and with an earlier start to the spring bloom (probably related to a combination of lower turbidity, higher temperature and changing nutrient ratios in winter). Dinoflagellate biomass has increased throughout the whole year and especially in summer. Toxic diatom (<i>Pseudo-nitzschia</i>) and dinoflagellate (e.g. <i>Prorocentrum</i>) species have increased > 2000.
pH	1970-1985 increase of annual mean and seasonal amplitude, 1985-2014 decrease of annual mean and seasonal amplitude. Annual mean and seasonal amplitude peaked in 1985.
Sea Surface Temp	Increase with ca. 1°C throughout the year.
CH4 emission	Decrease

4. DISCUSSION AND RECOMMENDATIONS

Within 4DEMON, datasets on change indicators of contamination, eutrophication and acidification in the Belgian Part of the North Sea (BPNS), dating back to 1968, were compiled, integrated and made publicly accessible via the central data portal. At the start of the project, the accessibility level of the data compiled in 4DEMON was highly variable, with data derived from both research projects and monitoring programs. The main bulk of recovered data dates back from before 1993. This timing is not a coincidence, as it coincides with the increased number of marine data management initiatives and European legislation making it mandatory to make governmental funded data freely accessible. The initially defined data flow by the datacentres ensured efficient data centralisation, safeguarding and dissemination. Because many data are still undiscoverable, similar projects should be encouraged. For example, at European level, the Emodnet Data Ingestion Portal (DIP) project should result in a vast amount of newly accessible data (See Recommendations Pillar 1). The data flow can be used as a guideline and can be included in the data management plan as suggested by the data policy of the funding body. The development of the central project data portal, with the data remaining physically stored at VLIZ and BMDC, was a challenge as there were technical issues and parameters needed to be mapped. To ensure data sharing at worldwide level, meta data standards are available which are continuously subject to improvement via multiple international initiatives in which both NODCs are actively involved. They will ensure that the data is further disseminated at international level

Even after compiling the above datasets, several obstacles were encountered during the trend analyses, and made it a challenge to select the best methodology. First, there were temporal gaps: important data gaps remain, for example for phytoplankton counts and taxonomy only 2 periods (70s and > 2000) have more or less complete data, with a big gap in between. Secondly, spatial gaps: the compiled data were not always evenly spread over the whole BPNS zone. Thirdly, the changes in methodology: sometimes it remains difficult to compare data (e.g. chlorophyll a for which monochromatic versus trichromatic analyses do not measure the exact same parameters) or to intercalibrate them (e.g. when there is no overlap in time between different methods). Even changes in co-factor analytical method may also affect contaminant concentrations. For the phytoplankton dataset, we found that, depending on the research focus of the different past projects, the taxonomic detail in the phytoplankton community composition datasets was variable (e.g. sometimes focus was exclusively on specific taxa, or the data were not gathered at the same taxonomic resolution (e.g. species to phylum).

Co-factors used for contaminant normalization in sediment were not systematically analysed, leading to time gaps because of missing metadata and to the use of multiple modelling approaches, applying different co-factors. To this end, a novel approach to normalize data and to model multiple decades of pollutant data is proposed (See Recommendations Pillar 2). This approach consists of spatial clustering, normalization and modelling. For spatial clustering, the BPNS was divided in several zones depending on pollution profile. This

clustering was based on a Ward hierarchical clustering. Several normalization procedures were applied for heavy metal data in marine sediments, with the use of Al-Fe as cofactors as best approach. A double co-factor, with quality check between the single cofactor normalization results, protects the data set from outliers created by the normalization procedure. For time trend assessment, a linear mixed effect model was applied. To correct for method switches which occurred over 4 decades of pollutant monitoring, the factor "method" was included as a variable in the model. Decreased trends in heavy metal and PCB concentration were noted which can be related to efficient emission reductions for heavy metals and the ban of PCBs. However, opposing trends should be followed with care. The last decade, PCB concentrations are again slightly increasing near Zeebrugge and the Wester-Scheldt, possibly due to inputs from the Scheldt. Increasing Zn concentrations at the BPNS may be related to multiple Zn sources at open sea, such as historical dumping sites of TiO₂ industry waste, wind mill parks or shipping. As concentrations in mussel were also found increasing. Since heavy metals do not degrade in marine sediments, historical samples from several decades were analyzed by one single method. Within the oldest samples, originating from 1901-1911, concentrations of As and Hg were higher than within samples of recent decades.

In the last decades there has been a decrease in export of terrestrial N and P, which was followed by a small decrease in marine nutrient concentrations. The benthic-pelagic coupling probably causes nutrient retention in coastal zone. The phenology of CPHL has changed in coastal and offshore waters, with earlier and rapid bloom formation. The observed increase in SST is most probably the cause of an increase in phytoplankton spring cell division rate. Clustering some aspects of CPHL variability allows drawing geographical areas in the North Sea. These areas picture physical properties of the basin: bathymetry, stratification regimes, fronts, deep water ascent. Each area shows characteristic patterns of CPHL variability. Belgian waters feature relatively intense blooms showing a strong stability from year to year. At the same time, the observed changes in phytoplankton taxonomy correlate well with overall increases in temperature, pronounced changes in nutrient loads and ratios, with especially more balanced DIN:DIP and DSi:DIP ratios during winter and early spring, and lower water suspended matter in winter-early spring. In addition, higher summer silica values, competition with other phytoplankton (e.g. dinoflagellates) and changes in grazing pressure could have contributed to the observed changes. Our observations agree with other studies reporting increases in phytoplankton biomass after the 1980s North Sea regime shift, but disagree with earlier reports of decreasing trends in diatom abundance and/or no changes in phenology in the southern North Sea. As such, they underscore the importance of regionally idiosyncratic responses of phytoplankton to global change phenomena, and the absolute necessity of maintaining local long-term monitoring programs for predicting regional ecosystem responses to global change.

Temperature seems to be one important factor controlling the spring peak maximum of *Phaeocystis* spp., while the NAO index seems to be less important than the overall climate change effect. Even though there is a negative correlation with the preceding nutrient concentrations, we should not assume that less nutrients lead to a higher *Phaeocystis* peak.

It is more likely the case that there are enough nutrients available anyway and that possibly nutrient ratios play an important role in the *Phaeocystis* spring bloom development.

It is likely that the change in phytoplankton bloom structure and dynamics (with earlier spring bloom) is related to (1) the continuous increase in SST over the period 1970-2015 (esp. in winter and spring); (2) a decrease in turbidity (~ TSM) in winter and (3) changing nutrient loads and ratios. The influence of SST increase seems to have been larger than that of the nutrient decreases over the period, even if both are expected to cause changes in phytoplankton bloom dynamics. pH data collected from 1975 to 2015 reveal an increasing trend of pH from the mid-70's to the mid-80's and a decrease of pH from the mid-80's to nowadays that seems consistent with changes in primary production patterns based on ecological models of the Belgian coastal zone. The comparison of CH₄ concentrations at five stations in the Belgian coastal zone obtained in March 1990 and 2016, showed a decreasing trend consistent with alleviation of eutrophication in the area. The changes in the amplitude of pH seasonal changes seems to indicate a maximum of primary production during the 1980-1989 decade and progressive alleviation of eutrophication during the following decades. Further research is required to evaluate the impact of changes in food web structure (zooplankton grazing, fish community, dynamics of parasites and viruses, etc.) on phytoplankton dynamics and structure.

The long-term datasets within 4DEMON resulted in an increased availability of data since the 1970s. They are highly valuable in the assessment of the definition of the GES within the BPNS as stated under the MSFD for various descriptors, and can thus serve further research to study the ecosystem. More specifically, data related to descriptors 5 (Eutrophication is minimised) and 8 (Concentrations of contaminants are at levels not giving rise to pollution effects), 9 (Contaminants in seafood are below safe levels), 1 (Biodiversity is maintained) and 4 (Elements of food webs ensure long-term abundance and reproduction), with the latter including ocean acidification, are now centrally accessible via the 4DEMON data portal. A complete list of parameters subject to the 4DEMON quality control and intercalibration exercises is available in Annex 2. The data will not only serve studies of the BPNS, but as they are disseminated at international level, e.g. via ICES, SeaDataCloud and EMODnet, they can also be included in regionally larger scaled ecosystem studies and assessments.

Assessing a GES to implement the MSFD implies various challenges, like defining the indicators and the best fitted procedure to study its health status (Van Hoey, 2010; McQuatters-Gollop, 2012). On top of that, working with historic data of multiple origin encompasses the additional factor of the high amount of alterations in measuring over time. As such, methods had to be developed to make the historic data comparable with today's values. The historic phytoplankton taxonomy data of the 1970s were carefully screened and mapped against the current nomenclature as registered in WoRMS. The difference in sediment fractions over time was resolved by granulometric normalization, while the unavailability of the standard co-factor for heavy metals (TOC), lead to the best available alternative co-factor (Fe/Al). Furthermore, spatial gaps were overcome by clustering zones with similar patterns (see 3.3.2. and 3.4.2.1.).

Based on the 4DEMON results of the trend analyses, we can confirm various findings regarding the GES. For example, metals and PCB values in mussels are within the safe levels for consumption (related to descriptor 9). However, some metals exceed the acceptable concentrations (related to descriptor 8) in sediment (CB118) and biota (Hg). On top of that, the shift in phytoplankton species that has been observed (related to descriptor 1), could have an impact on the species composition at higher level. As compared to the latest MSFD assessments (Belgian State, 2018), the added value of 4DEMON is without doubt the extended timeframe enabling specialists to make even more correct interpretations. Likewise, in the frame of other monitoring programs such as the surveillance of windfarms and dredging activities, the 4DEMON results are of high interest. Finally, the available data enable us to directly relate the impact of the taken policy measures on the health of the BPNS, like the European directives adopted since the 1990s. For example, the ban of PCBs and metals resulted in a marked reduction in concentrations of sediment and biota, while the reduction of winter DIP can be linked to the ban of polyphosphate from washing powder. However, there are some exceptions that need to be closely monitored and require further research.

To understand the interaction between natural and human-induced ecosystem change in the BPNS, which forms the basis for efficient and cost-effective mitigation, remediation and restoration measures, it is essential that standardized and rigorous monitoring of the main ecosystem components and environmental parameters is rigorously sustained in the future (See Recommendations Pillar 3). Without these datasets it is not possible to understand how human activities affect the BPNS ecosystem and to design effective management strategies. The intensified and diverse use of the BPNS (wind farms, aquaculture, fisheries, dredging, sand extraction, nature conservation and recreation) makes the need for continuous monitoring even more urgent and pressing. Data gaps resulting from short-term policy decisions should be absolutely avoided.

GENERAL CONCLUSION

The datasets rescued, compiled and analyzed in the 4DEMON project have revealed some surprising insights into the long-term ecosystem evolution of the BPNS. As such, the project has been very successful. The datasets are especially valuable from a European (and probably also global) perspective as it contains data from the 1970s, which is extremely valuable as many monitoring programs only started in the 1990s.

RECOMMENDATIONS

Pillar 1: Safeguard (historic) research data and increase public availability of and accessibility to quality controlled long-term datasets.

Recommendations on data handling
1. Digitize ' <u>dark data</u> ' before it is too late. There are still a lot of hidden marine data around that are at risk of getting lost. It will become increasingly difficult to contact the data-originators who hold the main bulk of information. Funding data recovery and analysis projects and cooperation between data managers, scientists, statisticians etc. is of crucial importance.
2. Increase the cost-efficiency of research and monitoring programs and results from industry through <u>data sharing initiatives</u> and encourage <u>open data policy</u> as much as possible (e.g. via projects like Emodnet Data Ingestion).
3. Importance of a <u>data management plan</u> . All research projects and monitoring projects should have a data management plan ensuring efficient data treatment. The data flow used in 4DEMON can be included as a guideline for such projects in the BPNS.
4. Importance of <u>standardized meta information</u> (e.g. campaign, project, geographical coordinates, depth, date, methods). To make the data internationally re-usable all this information should be made accessible together with the actual results. An overview of these fields is given in the dataflow as mentioned above.
5. <u>Storage of samples</u> . Importance of adequate sampling storage and long-term preservation as they can be reused for future reanalyses (e.g. heavy metals in sediments are persistent). Samples should be well labelled and inventoried, so the metadata can be easily retrieved.

Pillar 2: Adapt or improve methodologies and protocols to study and monitoring long-term environmental changes in the complex ecosystem of the BPNS.

Recommendations on monitoring procedures and trend detection methodology

1. A novel approach was developed to normalize and model long term data on contaminants. The applied normalization approach for heavy metal concentrations in marine sediments, combining Al-Fe as co-factors, increases reliability on contaminant data. This is inherently linked to the quality check mechanism within this normalization approach. The applied model, including method as a factor within a linear mixed-effect model, allows long term data comparison. The 4DEMON approach can be of value in contaminant assessments within an international framework and will therefore be reported to OSPAR.
2. The use of new methods and technologies is unavoidable and should be encouraged (e.g. changes of analysis instruments). Samples should always be simultaneously analysed with both methods for some time to make an intercomparison possible. For example, DNA-based methods and high-throughput imaging and counting techniques (e.g. (imaging) flow cytometry) are rapidly becoming the standard tool to assess changes in pelagic and benthic biomass and community structure, especially for microbial organisms. These methodologies are still being optimized so it is essential that more traditional tools (e.g. microscopic counts) are for some time used alongside these new techniques to make intercomparison unambiguous. It is recommended however that regular analyses (of selected samples) of phytoplankton using traditional microscopic remains an inherent part of monitoring programs.
3. Some components in the food web are currently not (or not sufficiently) monitored. Techniques, like DNA-based and high-throughput imaging allow monitoring of viruses and bacteria and can be installed on passive samplers like buoys and tripods for continuous measurement.
4. Monitoring procedures and protocols (like sampling frequency, timing, location, duration, species, methodology and gear) must be optimized to have full coverage of the study area within a specific timeframe to study the complex ecosystem of the BPNS. For example, the effect (vertical and horizontal) of the tide on the sampling of CPHL should be tested as it remains largely misunderstood. For this, there is need for well-defined sampling locations (e.g. gradient coast-offshore), timeseries (e.g. 24h measurements), and time window of in situ measurements must be mapped with earth observation data. As data from different sampling methodologies (e.g. with research vessel, earth observation, continuous and passive samplers) can complement each other, sampling campaigns need to be planned accordingly so these data can be mapped in time and space.

Pillar 3: Policy support and relevance of selected themes (Eutrophication, contamination and ocean acidification)

Recommendations towards policy and monitoring strategy

1. The understanding of long-term trends in phytoplankton seasonal accumulation absolutely requires continuous monitoring programs at high frequency (at least monthly) of CPHL and associated parameters at fixed sites. The application of 'new' techniques needs to be embraced as these will allow acquiring data at high temporal, spatial and taxonomic resolution of phytoplankton but also other major groups in the food web (bacteria to fish). Such techniques include DNA-based methods (metabarcoding and metagenomics), high-throughput imaging (e.g. flow cytometry) and remote sensing (e.g. based in the new Sentinel data). All of these methods are already being implemented in monitoring (e.g. in the framework of the LifeWatch campaigns). However, for reasons of rigorous intercalibration with older data and because some of the methods (e.g. metabarcoding) are still immature and need to be fine-tuned, it is essential that at least a monthly sampling campaign aimed at collecting data using the traditional, standardized methods (for measuring chlorophyll and phytoplankton community composition) for at least a few carefully selected sites (representing surrounding water bodies).
2. It is recommended to study the release of Zn to the marine environment in detail and to identify the impact of different sources. Also, the impact of increased Zn concentrations at the BPNS on the marine ecosystem requires more research.
3. Although banned for already several decades, PCB concentrations may still have negative effects on the marine ecosystem while concentrations may locally increase over the last decades. This stresses the importance for continuing monitoring.
4. Given the huge value of long-term datasets to realistically assess the impact of long-term global change on marine systems the full continuation and elaboration of ongoing monitoring campaigns is essential and should under no circumstances be discontinued, even temporarily. Groups that are currently not monitored (e.g. bacteria) should be included in future monitoring programs (using DNA-based tools).
5. It is essential that data such as nutrients are carefully and accurately collected with standardized methods: even for the most recent period (> 2000) there are large gaps in the nutrient data series. This jeopardizes the usefulness of the whole monitoring effort.

REFERENCES

Anon., 2016. Ontwerp van het stroomgebiedsbeheersplan voor de Belgische kustwateren voor de implementatie van de Europese kaderrichtlijn Water (2000/60/EG). Federale Overheidsdienst Volksgezondheid, veiligheid van de voedselketen en leefmilieu, dienst marien milieu, 80p.

Antajan E., Chrétiennot-Dinet M. J., Leblanc C., Daro M. H., Lancelot C., 2004. 19'-hexanoyloxyfucoxanthin may not be the appropriate pigment to trace occurrence and fate of Phaeocystis: the case of *P. globosa* in Belgian coastal waters. *Journal of Sea Research*, 52(3), 165-177. Doi: 10.1016/j.seares.2004.02.003.

Baeteman M., Vyncke W., Gabriels R., Guns M., 1987. Heavy metals in water, sediments and biota in dumping areas for acid wastes from the titanium dioxide industry. International Council for the exploration of the Sea, Marine Environmental Quality Committee, C.M.1987/E:7.

Baretta-Bekker H., Sell A., Marco-Rius F., Wischnewski J., Walsham P., Malin Mohlin L., Wesslander K., Ruiter H., Gohin F., Enserink L., 2015. The chlorophyll case study in the JMP NS/CS project. Document produced as part of the EU project: 'Towards joint Monitoring for the North Sea and Celtic Sea' (Ref: ENV/PP 2012/SEA).

Baretta-Bekker J.G., Baretta J.W., Latuhihin M.J., Desmit X., Prins T.C., 2009. Description of the long-term (1991-2005) temporal and spatial distribution of phytoplankton carbon biomass in the Dutch North Sea. *J. Sea Res.* 61, 50–59. <https://doi.org/10.1016/j.seares.2008.10.007>.

Belgische Staat, 2018. Herziening van de initiële beoordeling voor de Belgische mariene wateren. Kaderrichtlijn Mariene Strategie - Art 8 lid 1a & 1b. BMM, Federale overheidsoverheidsdienst Volksgezondheid, Veiligheid van de Voedselketen en Leefmilieu, Brussel, België, 228 pp.

Beaugrand G., 2009. Decadal changes in climate and ecosystems in the North Atlantic Ocean and adjacent seas. *Deep Sea Research Part II: Topical Studies in Oceanography*, 56(8-10), 656-673. doi: 10.1016/j.dsr2.2008.12.022.

Bekaert K., Le H.M., Lagring R., Ampe B., Ruttens A., Waegeneers N., De Witte B. Modelling 4 decades of biota contaminant data from the Belgian Part of the North Sea (BPNS). *Chemosphere*, in prep.

Borges A.V., Abril G., 2011. Carbon Dioxide and Methane Dynamics in Estuaries. In: *Treatise on Estuarine and Coastal Science, Volume 5: Biogeochemistry* (eds Wolanski E, McLusky D), pp. 119-161, Academic Press, Waltham.

Borges A.V., Champenois W., Gypens N., Delille B., Harlay J., 2016. Massive marine methane emissions from near-shore shallow coastal areas, *Scientific Reports*, 6, 27908, doi:10.1038/srep27908.

Borges A.V., Speeckaert G., Champenois W., Scranton M.I., Gypens N., 2017. Productivity and temperature as drivers of seasonal and spatial variations of dissolved methane in the Southern Bight of the North Sea, *Ecosystems*, DOI: 10.1007/s10021-017-0171-7.

Boyce D. G. and Worm B., 2015. Patterns and ecological implications of historical marine phytoplankton change. *Marine Ecology progress series*, 534, 251-272. doi: 10.3354/meps11411.

Brion N., Jans S., Chou L., Rousseau V., 2006. Nutrient loads to the Belgian Coastal Zone, in: Rousseau, V., Lancelot, C., Cox, D. (Eds.), *Current Status of Eutrophication in the Belgian Coastal Zone*. Presses Universitaires de Bruxelles, Brussels, pp. 17–43. Caldeira K

& ME Wickett 2003. Oceanography: Anthropogenic carbon and ocean pH, *Nature* 425, 365, doi:10.1038/425365a.

Caldeira K. and Wickett M.E., 2003. Oceanography: Anthropogenic carbon and ocean pH, *Nature* 425, 365, doi:10.1038/425365a.

Capuzzo E., Stephens D., Silva T., Barry J., Forster R. M., 2015. Decrease in water clarity of the southern and central North Sea during the 20th century. *Glob Chang Biol*, 21(6), 2206-2214. doi: 10.1111/gcb.12854.

Chassot E., Bonhommeau S., Dulvy N. K., Melin F., Watson R., Gascuel D., Le Pape O., 2010. Global marine primary production constrains fisheries catches. *Ecol Lett*, 13(4), 495-505. doi: 10.1111/j.1461-0248.2010.01443.x.

Cloern J. E., 2001. Our evolving conceptual model of the coastal eutrophication problem. *Marine Ecology progress series*, 210, 223-253. doi: Doi 10.3354/Meps210223.

Cloern J. E. and Jassby A. D., 2008. Complex seasonal patterns of primary producers at the land-sea interface. *Ecol Lett*, 11(12), 1294-1303. doi: 10.1111/j.1461-0248.2008.01244.x.

Cloern J.E., Jassby A.D., 2010. Patterns and Scales of Phytoplankton Variability in Estuarine-Coastal Ecosystems. *Estuaries and Coasts* 33, 230-241. <https://doi.org/10.1007/s12237-009-9195-3>.

Colijn F., Hesse K. J., Ladwig N., Tillmann U., 2002. Effects of the large-scale uncontrolled fertilisation process along the continental coastal North Sea. *Hydrobiologia*, 484(1-3), 133-148. doi: Doi 10.1023/A:1021361206529.

De Clerck R., Vyncke W., Guns M., van Hoeyweghen P., 1995. Concentrations of mercury, cadmium, zinc and lead in sole from Belgian catches (1973-1991). *Mededelingen van de Faculteit Landbouw, Universiteit Gent*, 60, 1-6.

de Vries I., Duin R., Peeters J., Los F., Bokhorst M., Laane R., 1998. Patterns and trends in nutrients and phytoplankton in Dutch coastal waters: comparison of time-series analysis, ecological model simulation, and mesocosm experiments. *ICES J. Mar. Sci.* 55, 620-634. <https://doi.org/10.1006/jmsc.1998.0399>.

Desmit X., Ruddick K., Lacroix G., 2015. Salinity predicts the distribution of chlorophyll a spring peak in the southern North Sea continental waters. *J. Sea Res.* 103, 59-74. <https://doi.org/10.1016/j.seares.2015.02.007>.

De Witte B., Ruttens A., Ampe B., Waegeneers N., Gauquie J., Devriese L., Cooreman K., Parmentier K., 2016. Chemical analyses of dredged spoil disposal sites at the Belgian part of the North Sea, *Chemosphere*, 156, 172-180.

Díaz R.J., Rosenberg R., 2008. Spreading dead zones and consequences for marine ecosystems. *Science*, 321, 926-929.

Directive 91/676/EEC of 12 December 1991 concerning the protection of waters against pollution caused by nitrates from agricultural sources. *Official Journal L*. 375 (31), 12 (Dec 31).

Directive 91/271/EEC, d. Urban Waste Water Treatment Directive - EEA (2010) Waterbase. Version of November 2014 <http://www.eea.europa.eu/data-and-maps/data/waterbase-uwatd-urban-waste-watertreatment-directive>.

Doerffer R. and Schiller H., 2007. The MERIS case 2 water algorithm. *International Journal of Remote Sensing*, 28, 517-535.

Doney S.C., Mahowald N., Lima I., Feely R.A., Mackenzie F.T., Lamarque J.-F., Rasch P.J., 2007. Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system, *P. Natl. Acad. Sci. USA* 104: 14580-14585.

Edwards M. and Richardson A. J., 2004. Impact of climate change on marine pelagic phenology and trophic mismatch. *Nature*, 430(7002), 881-884. doi: 10.1038/nature02808.

Everaert G., De Laender F., Deneudt K., Roose P., Mees J., Goethals P.L.M., Janssen C.R., 2014. Additive modelling reveals spatiotemporal PCB trends in marine sediments. *Marine Pollution Bulletin*, 79, 47-53.

Fettweis M., Houziaux J.-S., Du Four I., Van Lancker V., Baeteman C., Mathys M., Van den Eynde D., Francken F., Wartel S., 2009. Long-term influence of maritime access works on the distribution of cohesive sediments: analysis of historical and recent data from the Belgian nearshore area (southern North Sea). *Geo-Marine Letters*, 29, 321-330.

Gao Y., De Brauwere A., Elskens M., Croes K., Baeyens W., Leermakers M., 2013. Evolution of trace metal and organic pollutant concentrations in the Scheldt River Basin and the Belgian Coastal Zone over the last three decades. *Journal of Marine Systems*, 128, 52-61.

Gohin F., Druon J.N., Lampert L., 2002. A five channel chlorophyll concentration algorithm applied to SeaWiFS data processed by SeaDAS in coastal waters. *Int. J. Remote Sens.* 23 (8), 1639–1661.

Gons H., Rijkeboer M., Ruddick K., 2005. Effect of a waveband shift on chlorophyll retrieval from MERIS imagery of inland and coastal waters. *Journal of Plankton Research* 27(1): 125–127.

Guns M., Van Hoeywegen P., Baeten H., Hoenig M., Vyncke W., Hillewaert H., 1995. Evolutie van de gehalten van zware metalen in sedimenten van het Belgisch Continentaal Plat (1979-1995). Medelingen van het Rijksstation voor Zeevisserij, Publicatie nr. 242, D/1997/0889/1.

Guns M., Va, Hoeywegen P., Vyncke W., Hillewaert H., 1999. Trace metals in selected benthic invertebrates from Belgian coastal waters (1981-1996). *Marine Pollution Bulletin*, 38(12), 1184-1193.

Guns M., Vyncke W., De Clerck R., 1992. Mercury concentrations in plaice, flounder and dab from Belgian continental shelf water (1971-1990). *Landbouwtijdschrift*, 45, 959-693.

Gutiérrez J.L., Jones C.G., Strayer D.L., Iribarne O.O., 2003. Mollusks as ecosystem engineers: The role of shell production in aquatic habitats. *Oikos* 101: 79-90.

Gypens N., Borges A.V., Lancelot C., 2009. Effect of eutrophication on air-sea CO₂ fluxes in the coastal Southern North Sea: a model study of the past 50 years. *Global Change Biology*, 15, 1040-1056.

Gypens N, Borges A.V., 2014. Increase in dimethylsulfide (DMS) emissions due to eutrophication of coastal waters offsets their reduction due to ocean acidification. *Frontiers in Marine Science - Marine Ecosystem Ecology*, 1, 4, doi: 10.3389/fmars.2014.00004.

Hager S.W., Atlas E.L., Gordon L.I., Mantyla A.W., Park P.K., 1972. A comparison at sea of manual and autoanalyzer analyses of phosphate, nitrate and silicate. *Limnology and Oceanography* 17(6):931-937. DOI: 10.4319/lo.1972.17.6.0931.

Hillebrand H., Dürselen C. D., Kirschtel D., Pollinger U., Zohary, T., 1999. Biovolume calculation for pelagic and benthic microalgae. *Journal of Phycology*, 35(2), 403-424. doi: DOI 10.1046/j.1529-8817.1999.3520403.x.

Hoppenrath M., Elbrächter M., Drebes G., 2009. *Marine Phytoplankton - Selected microphytoplankton species from the North Sea around Helgoland and Sylt*. Stuttgart: E. Schweizerbart'sche Verlagsbuchhandlung (Nägele u. Obermiller).

Horner R. A., 2002. *A taxonomic guide to some common marine phytoplankton*: Biopress Ltd.

Høyer J, Karagali I., 2017. Sea surface temperature climate data record for the North Sea and Baltic Sea. *Journal of Climate*, in press, doi:10.1175/JCLI-D-15-0663.1.

Hunter-Cevera K.R., Neubert M.G., Olson R.J., Solow A.R., Shalapyonok A., Sosik H.M., 2016. Physiological and ecological drivers of early spring blooms of a coastal phytoplankton. *Science* 354, 326–329. <https://doi.org/10.1126/science.aaf8536>.

IOCCG, 2000. Remote sensing of ocean colour in coastal, and other optically-complex waters. In S. Sathyendranath (Ed.), *Reports of the International Ocean-Colour Coordinating Group, No. 3*, IOCCG, Dartmouth, Canada (140 pp.).

IOCCG, 2006. Remote sensing of inherent optical properties: Fundamentals, tests of algorithms, and applications. In Z. P. Lee (Ed.), *Reports of the International Ocean-Colour Coordinating Group, No. 5*, IOCCG, Dartmouth, Canada (122 pp.).

IPCC, 2013. *Fifth Assessment Report of the Intergovernmental Panel on Climate Change* (eds Stocker, T.F., Qin, D., Plattner, G.-K., Tignor, M., Allen, S.K., Boschung, J., Nauels, A., Xia, Y., Bex, V. & Midgley, P.M.) 1535 pp. Cambridge University Press, Cambridge.

Jickells T.D., 1998. Nutrient Biogeochemistry of the Coastal Zone. *Science* 281, 217–21. <https://doi.org/10.1126/SCIENCE.281.5374.217>.

Kersten M., Smedes F., 2002. Normalization procedures for sediment contaminants in spatial and temporal trend monitoring. *Journal of Environmental monitoring*, 4, 109-115.

Kleypas J.A., Feely R.A., Fabry V.J., Langdon C., Sabine C.L., Robbins L.L., 2006. *Impacts of Ocean Acidification on Coral Reefs and Other Marine Calcifiers: A Guide for Future Research*, report of a workshop held 18–20 April 2005, St. Petersburg, FL, sponsored by NSF, NOAA, and the U.S. Geological Survey.

Kromkamp J. C. and Van Engeland T., 2009. Changes in Phytoplankton Biomass in the Western Scheldt Estuary During the Period 1978–2006. *Estuaries and Coasts*, 33(2), 270-285. doi: 10.1007/s12237-009-9215-3.

Lagring R., De Cauwer K., Devolder M., Scory S., 2012 a. Resuscitation of the data collected during the first years of modern oceanography in Belgium (PMPZ-DBII). Final report. Brussels: Belgian Science Policy Office 2012 - 69 p. (Research Programme Science for a Sustainable Development).

Lancelot C., Billen G., Sournia A., Weisse T., Colijn F., Veldhuis M.J.W., Davies A., Wassman P., 1987. Phaeocystis Blooms and Nutrient Enrichment in the Continental Coastal Zones of the North Sea. *Ambio* 16, 38–46. <https://doi.org/10.2307/4313316>.

Lancelot C., Gypens N., Billen G., Garnier J., Roubex V., 2007. Testing an integrated river–ocean mathematical tool for linking marine eutrophication to land use: the Phaeocystis-dominated Belgian coastal zone (Southern North Sea) over the past 50 years. *J. Mar. Sys.* 64: 216-228.

Lancelot C., Rousseau V., Gypens N., 2009. Ecologically based indicators for Phaeocystis disturbance in eutrophied Belgian coastal waters (Southern North Sea) based on field observations and ecological modelling. *J. Sea Res.* 61, 44–49. <https://doi.org/10.1016/j.seares.2008.05.010>.

Latasa M., Bidigare R.R., Ondrusek M.E., Kennicutt M.C., 1996. HPLC analysis of algal

pigments: a comparison exercise among laboratories and recommendations for improved analytical performance. *Mar. Chem.* 51, 315–324. [https://doi.org/10.1016/0304-4203\(95\)00056-9](https://doi.org/10.1016/0304-4203(95)00056-9).

Le H.M., Bekaert K., Lagring R., Ampe B., Ruttens A., Waegeneers N., De Witte B. Sediment normalisation at the Belgian Part of the North Sea: an approach to intercalibrate 40 years of marine contaminant data. *Chemosphere*, in preparation, a.

Le H.M., Bekaert K., Lagring R., Ampe B., Ruttens A., Waegeneers N., De Witte B. spatio-temporal trend assessment of 4 decades of contaminants in sediment from the Belgian part of the North Sea using multivariate analysis and mixed models. *Chemosphere*, in preparation, b.

Lewandowska A. and Sommer U., 2010. Climate change and the spring bloom: a mesocosm study on the influence of light and temperature on phytoplankton and mesozooplankton. *Marine Ecology progress series*, 405, 101-111. doi: 10.3354/meps08520.

Lima-Mendez G., Faust K., Henry N., Decelle J., Colin S., Carcillo F., Chaffron S., Ignacio-Espinosa J. C., Roux S., Vincent F., Bittner L., Darzi Y., Wang J., Audic S., Berline L., Bontempi G., Cabello A. M., Coppola L., Cornejo-Castillo F. M., d'Ovidio F., De Meester L., Ferrera I., Garet-Delmas M. J., Guidi L., Lara E., Pesant S., Royo-Llonch M., Salazar G., Sanchez P., Sebastian M., Souffreau C., Dimier C., Picheral M., Searson S., Kandels-Lewis S., Tara Oceans coordinators, Gorsky G., Not F., Ogata H., Speich S., Stemmann L., Weissenbach J., Wincker P., Acinas S. G., Sunagawa S., Bork P., Sullivan M. B., Karsenti E., Bowler C., de Vargas C. and Raes J., 2015. Ocean plankton. Determinants of community structure in the global plankton interactome. *Science*, 348(6237), 1262073. doi: 10.1126/science.1262073.

Lorenzen C.J., 1967. Determination of chlorophyll and phaeo-pigments: spectrophotometric equations. *Limnol. Oceanogr.* 12, 343–346. <https://doi.org/10.4319/lo.1967.12.2.0343>.

Loring J., 1991. Normalization of heavy-metal data from estuarine and coastal sediments. *ICES Journal of Marine Science*, 48, 101-115.

Los F., Borkhorst M., 1997. Trend analysis Dutch coastal zone. New challenges North Sea Res. Zent. Meeres- und Klimaforschung, Univ. Hamburg. 161–175.

Mackey M. D., Mackey D. J., Higgins H. W. and Wright S. W., 1996. CHEMTAX - A program for estimating class abundances from chemical markers: Application to HPLC measurements of phytoplankton. *Marine Ecology Progress Series*, 144(1-3), 265-283. doi: 10.3354/Meps144265.

Maso M. and Garces E., 2006. Harmful microalgae blooms (HAB); problematic and conditions that induce them. *Marine Pollution Bulletin*, 53(10-12), 620-630. doi: 10.1016/j.marpolbul.2006.08.006.

McQuatters-Gollop A., 2012. Challenges for implementing the Marine Strategy Framework Directive in a climate of macroecological change. *Phil. Trans. R. Soc. A* 370, 5636–5655. doi:10.1098/rsta.2012.0401.

McQuoid, M. R. and Nordberg K., 2003. The diatom *Paralia sulcata* as an environmental indicator species in coastal sediments. *Estuarine, Coastal and Shelf Science*, 56(2), 339-354. doi: 10.1016/s0272-7714(02)00187-7.

Missiaen T., Murphy S., Loncke L., Henriët J.P., 2002. Very high-resolution seismic mapping of shallow gas in the Belgian coastal zone. *Continental Shelf Research*, 22, 2291-2301.

Murray A., Gibbs C., Longmore A., Flett D., 1986. Determination of chlorophyll in marine

waters: intercomparison of a rapid HPLC method with full HPLC, spectrophotometric and fluorometric methods. *Mar. Chem.* 19, 211–227.

Muylaert K., Gonzales R., Franck M., Lionard M., Van der Zee C., Cattrijsse A., Sabbe K., Chou L. and Vyverman, W., 2006. Spatial variation in phytoplankton dynamics in the Belgian coastal zone of the North Sea studied by microscopy, HPLC-CHEMTAX and underway fluorescence recordings. *Journal of Sea Research*, 55(4), 253-265. doi: 10.1016/j.seares.2005.12.002.

Muylaert K., Gonzales R., Franck M., Lionard M., Van der Zee C., Cattrijsse A., Sabbe K., Chou L., Vyverman W., 2006. Spatial variation in phytoplankton dynamics in the Belgian coastal zone of the North Sea studied by microscopy, HPLC-CHEMTAX and underway fluorescence recordings. *J. Sea Res.* 55, 253–265. <https://doi.org/10.1016/J.SEARES.2005.12.002>.

Naqvi S.W.A., Bange H. W., Farías L., Monteiro P.M.S., Scranton M.I., Zhang J., 2010. Marine hypoxia/anoxia as a source of CH₄ and N₂O. *Biogeosciences*, 7, 2159-2190.

Naz T., Burhan Z. U. N., Munir S. and Siddiqui P. J. A., 2013. Biovolume and Biomass of Common Diatom Species from the Coastal Waters of Karachi, Pakistan. *Pakistan Journal of Botany*, 45(1), 325-328.

Nechad B., Ruddick K., Schroeder T., Oubelkheir K., Blondeau-Patissier D., Cherukuru N., Brando V.E., Dekker A.G., Clementson L., Banks A., Maritorea S., Werdell P.J., Sá C., Brotas V., Caballero de Frutos I., Ahn Y-H., Salama S., Tilstone G., Martinez-Vicente V., Foley D., McKibben M., Nahorniak J., Peterson T.D., Siliò-Calzada A., Röttgers R., Lee Z.P., Peters M., 2015. CoastColour Round Robin datasets: a database to evaluate the performance of algorithms for the retrieval of water quality parameters in coastal waters. *Earth System Science Data*, 7(2), 319-348.

Neveux J., Delmas D., Romano J., Algarra P., Ignatiades L., Herbland A., Morand P., Neori A., Bonin D., Barbe J., Sukenik A., 1990. Comparison of chlorophyll and phaeopigment determinations by spectrophotometric, fluorometric, spectrofluorometric and HPLC methods. *Mar. Microb. Food Webs* 4, 217–238.

Nohe A. Knockaert C., Goffin A., Dewitte E., De Cauwer K., Desmit X., Vyverman W., Tyberghein L., Lagring R., Sabbe K., 2018. Marine phytoplankton community composition data from the Belgian part of the North Sea, 1968-2010. *Sci. Data* 5:180126 doi: 10.1038/sdata.2018.126.

Noklegaard T., Cusack C., Silke J., McDermott G., O'Boyle S., 2005. METRIC REPORT - Marine Ecological Tools for Reference, Intercalibration and Classification: Intercomparison of Chlorophyll a measurement methods by the Marine Institute and the Environmental Protection Agency.

O'Reilly J. E., Maritorea S., Mitchell B. G., Siegel D. A., Carder K. L., Garver S. A., Kahru S. A., McClaim, C., 1998, Ocean colour chlorophyll algorithms for SeaWiFS. *Journal of Geophysical Research*, 103, 24 937–24 953.

OSPAR, 2012. CEMP Assessment Report. Monitoring and Assessment Series. OSPAR Publication 563/2012. ISBN 978-1-907390-68-5.

OSPAR, 2008. CEMP assessment manual. Co-ordinated Environmental Monitoring Programme Assessment Manual for contaminants in sediment and biota. Monitoring and assessment series. commission, London, p.31.

OSPAR, 2010. Quality Status Report 2010. OSPAR commission. London. 176 pp.

OSPAR, 2015. JAMP Guidelines for monitoring contaminants in sediment, revision 2015. OSPAR commission, London, p.111.

OSPAR, 2017. OSPAR Intermediate Assessment 2017. OSPAR commission. www.ospar.org/assessments.

Otero-Rey J.R., López-Vilariño J.M., Moreda-Piñeiro J., Alonso-Rodríguez E., Muniategui-Lorenzo S., López-Mahía P., Prada-Rodríguez D., 2003. As, Hg, and Se flue gas sampling in a coal-fired power plant and their fate during coal combustion. *Environmental Science and Technology*, 37(22), 5262-5267.

Paerl H. W., Hall N. S. and Calandrino E. S., 2011. Controlling harmful cyanobacterial blooms in a world experiencing anthropogenic and climatic-induced change. *Sci Total Environ*, 409(10), 1739-1745. doi: 10.1016/j.scitotenv.2011.02.001

Paerl H. W., Hall N. S., Peierls B. L. and Rossignol, K. L., 2014. Evolving Paradigms and Challenges in Estuarine and Coastal Eutrophication Dynamics in a Culturally and Climatically Stressed World. *Estuaries and Coasts*, 37(2), 243-258.

Peperzak, L., 1993. Daily irradiance governs growth rate and colony formation of *Phaeocystis* (Prymnesiophyceae). *J. Plankton Res.* 15, 809–821. <https://doi.org/10.1093/plankt/15.7.809>.

Peperzak L., Colijn F., Gieskes W.W.C., Peeters J.C.H., 1998. Development of the diatom- *Phaeocystis* spring bloom in the Dutch coastal zone of the North Sea: the silicon depletion versus the daily irradiance threshold hypothesis. *J. Plankton Res.* 20, 517–537. <https://doi.org/10.1093/plankt/20.3.517>.

Philippart C.J.M., Beukema J.J., Cadée G.C., Dekker R., Goedhart P.W., van Iperen J.M., Leopold M.F., Herman P.M.J., 2007. Impacts of Nutrient Reduction on Coastal Communities. *Ecosystems* 10, 96–119. <https://doi.org/10.1007/s10021-006-9006-7>.

Philippart C. J. M., van Iperen J. M., Cadée G. C. and Zuur A. F., 2010. Long-term Field Observations on Seasonality in Chlorophyll-a Concentrations in a Shallow Coastal Marine Ecosystem, the Wadden Sea. *Estuaries and Coasts*, 33(2), 286-294. doi: 10.1007/s12237-009-9236-y.

Prins T. C., Desmit X. and Baretta-Bekker J. G., 2012. Phytoplankton composition in Dutch coastal waters responds to changes in riverine nutrient loads. *Journal of Sea Research*, 73, 49-62. doi: 10.1016/j.seares.2012.06.009.

Prins T.C., Desmit X., Baretta-Bekker J.G., 2012. Phytoplankton composition in Dutch coastal waters responds to changes in riverine nutrient loads. *J. Sea Res.* 73, 49–62. <https://doi.org/10.1016/j.seares.2012.06.009>.

Roose P., Raemaekers M., Cooreman K., Brinkman U.A.T., 2005. Polychlorinated biphenyls in marine sediments from the southern North Sea and Scheldt estuary: a ten-year study of concentrations, patterns and trends. *J. Environ. Monit.* 7, 701-709.

Roose P., Raemaekers M., Cooreman K., Brinkman U.A.T., 2005. Polychlorinated biphenyls in marine sediments from the southern North Sea and Scheldt estuary: a ten-year study of concentrations, patterns and trends. *Journal of Environmental Monitoring*, 7, 701-709.

Rousseau V., Lantoin F., Rodriguez F., LeGall F., Chrétiennot-Dinet M.-J., Lancelot C., 2013. Characterization of *Phaeocystis globosa* (Prymnesiophyceae), the blooming species in the Southern North Sea. *J. Sea Res.* 76, 105–113. <https://doi.org/10.1016/j.seares.2012.07.011>.

Rousseau V., Park Y., Ruddick K., Vyverman W., Parent J.-Y., Lancelot C., 2006. Phytoplankton blooms in response to nutrient enrichment, in: Rousseau, V., Lancelot, C., Cox, D. (Eds.), *Current Status of Eutrophication in the Belgian Coastal Zone*. Presses Universitaires de Bruxelles, Brussels, pp. 45–59.

Rousseau V., Lancelot C., Cox, D., 2008. Current Status of Eutrophication in the Belgian Coastal Zone. Belgian Science Policy. Presses Universitaires de Bruxelles, Avenue Paul Héger 42, 1000 Bruxelles, Belgique.

RStudioTeam, 2017. RStudio: Integrated Development for R (version 3.4.0 (2017-04-21)).

Ruddick K., Lacroix G., 2006. Hydrodynamics and meteorology of the Belgian Coastal Zone, in: Rousseau, V., Lancelot, C., Cox, D. (Eds.), Current Status of Eutrophication in the Belgian Coastal Zone. Presses Universitaires de Bruxelles, Brussels, pp. 1–15.

Rühland K., Paterson A. M., Smol J. P., 2008. Hemispheric-scale patterns of climate-related shifts in planktonic diatoms from North American and European lakes. *Global Change Biology*, 14(11), 2740–2754. doi: 10.1111/j.1365-2486.2008.01670.x.

Saunois M. et al., 2016. The global methane budget. *Earth System Science Data*, 8, 697–751.

Schaub B., Gieskes W., 1991. Eutrophication of the North Sea: The relation between Rhine River discharge and chlorophyll-a concentration in Dutch coastal waters., in: Elliott, M., Ducrotoy, J.-P. (Eds.), Estuaries and Coasts: Spatial and Temporal Intercomparisons. Olsen & Olsen, Fredensborg, pp. 85–90.

Schlüter L., Mohlenber F., Havskum H., Larsen S., 2000. The use of phytoplankton pigments for identifying and quantifying phytoplankton groups in coastal areas: testing the influence of light and nutrients on pigment/chlorophyll a ratios. *Marine Ecology Progress Series*, 192, 49–63.

Schroeder T., Schaale M., Fisher J., 2007. Retrieval of atmospheric and oceanic properties from MERIS measurements: A new Case-2 water processor for BEAM. *International Journal of Remote Sensing* 28, 5627–5632. van Leeuwen, S., Tett, P., Mills, D., van der Molen, J., 2015. Stratified and nonstratified areas in the North Sea: Long-term variability and biological and policy implications. *J. Geophys. Res. Ocean.* 120, 4670–4686. <https://doi.org/10.1002/2014JC010485>.

Suikkanen S., Pulina S., Engstrom-Ost J., Lehtiniemi M., Lehtinen S., Brutemark A., 2013. Climate change and eutrophication induced shifts in northern summer plankton communities. *PloS one*, 8(6), e66475. doi: 10.1371/journal.pone.0066475.

Smedes F., 2002. Zand, slib en zeven: standaardisering van contaminantgehaltenes in mariene sedimenten. Rapport RIKZ-96.043. Rijksinstituut voor Kust en Zee\RIKZ, p.137.

Smedes F., Nummerdor G.A.N., 2003. Grain-size correction for the contents of butyltin compounds in sediment. Report RIKZ\2003.035. National Institute of coastal and marine management\RIKZ, p.40.

Terseleer Lillo N., 2014. *Bottom-up and Top-down Controls of Diatoms in the Belgian Coastal Zone (Southern North Sea): Combining Plankton Functional Type Modelling and Trait-based Approaches*. (PhD), Université libre de Bruxelles, Brussels.

Throndsen J., Hasle G. R., Tangen K., 2007. *Phytoplankton of Norwegian Coastal Waters*: Almatel Forlag AS.

Tréguer P., Bowler C., Moriceau B., Dutkiewicz S., Gehlen M., Aumont O., Bittner L., Dugdale R., Finkel Z., Iudicone D., Jahn O., Guidi L., Lasbleiz M., Leblanc K., Levy M., Pondaven P., 2017. Influence of diatom diversity on the ocean biological carbon pump. *Nature Geoscience*, 11(1), 27–37. doi: 10.1038/s41561-017-0028-x.

Turner A., 2010. Marine pollution from antifouling paint particles. *Marine Pollution Bulletin* 60, 159–171.

Turner J. T. and Tester P. A., 1997. Toxic marine phytoplankton, zooplankton grazers, and pelagic food webs. *Limnol. Oceanogr.*, 42, 12. doi: 10.4319/lo.1997.42.5_part_2.1203.

Van der Zande D., 2018. JMP-EUNOSAT User case: Joint Monitoring Programme of Eutrophication of the North-Sea with satellite data. CMEMS Ocean State Report 2018. In prep.

Van Heukelem L. and Thomas C. S., 2001. Computer-assisted high-performance liquid chromatography method development with applications to the isolation and analysis of phytoplankton pigments. *Journal of Chromatography A*, 910(1), 31-49. doi: 10.1016/s0378-4347(00)00603-4.

Van Hoey G., Angel Borja, Silvana Birchenough, Lene Buhl-Mortensen, Steven Degraer, Dirk Fleischer, Francis Kerckhof, Paolo Magni f,g, Iñigo Muxika, Henning Reiss, Alexander Schröder, Michael L. Zettler, 2010. The use of benthic indicators in Europe: From the Water Framework Directive to the Marine Strategy Framework Directive. *Marine Pollution Bulletin* 60 (2010) 2187–2196.

Vyncke W., Guns M., Roose P., Cooreman K., De Clerck R., Van Hoeyweghen P., 1996. Contaminants in Belgian fish and shellfish (1971-1993). In: Dialogue between scientists and users of the sea. Federal Office for Scientific and Cultural Affairs, Brussel, 57-66.

Winder M. and Cloern J. E., 2010. The annual cycles of phytoplankton biomass. *Philos Trans R Soc Lond B Biol Sci*, 365(1555), 3215-3226. doi: 10.1098/rstb.2010.0125.

Wood S. A., 2017. Mixed GAM Computation Vehicle with GCV/AIC/REML Smoothness Estimation.

Worm B., Barbier E. B., Beaumont N., Duffy J. E., Folke C., Halpern B. S., Jackson J.B.C., Lotze H.K., Micheli F., Palumbi S.R., Sala E., Selkoe K.A., Stachowicz J.J., Watson R., 2006. Impacts of biodiversity loss on ocean ecosystem services. *Science*, 314(5800), 787-790. doi: 10.1126/science.1132294.

Wright S. W., Jeffrey S. W., Mantoura R. F. C., Llewellyn C. A., Bjornland T., Repeta D., Welschmeyer N., 1991. Improved Hplc Method for the Analysis of Chlorophylls and Carotenoids from Marine-Phytoplankton. *Marine Ecology Progress Series*, 77(2-3), 183-196. doi: 10.3354/Meps077183.

Zapata M., Rodríguez F., Garrido J. L., 2000. Separation of chlorophylls and carotenoids from marine phytoplankton: a new HPLC method using a reversed phase C8 column and pyridine-containing mobile phases. *Marine Ecology Progress Series*, 195, 29-45. doi: 10.3354/meps195029.

Zuur A. F., Ieno E. N., Walker N. J., Saveliev A. A., Smith G. M., 2009. *Mixed effect Models and Extensions in Ecology with R*.

Zuur A.F., Ieno E.N., Walker N.J., Saveliev-Graham A.A., Smith M., 2009. Mixed effects models and extensions in ecology with R. Springer, P.580, ISBN 978-0-387-87458-6.

5. DISSEMINATION AND VALORISATION

Below, an overview is given of the dissemination activities in chronological order.

Oral presentations

Nohe, A. The impact of decades of environmental change on phytoplankton communities in the Belgian part of the North Sea. European Marine Biology Symposium (EMBS), Oostende, Belgium, 17-21 September 2018.

Le H.M., De Witte B., Lagring R., Bekaert K, A novel approach to model 4 decades of sediment contaminants, 50th International Liege colloquium on Ocean Dynamiscs, Liège, 28 May -1 June 2018.

Nohe, A. The impact of decades of human-induced environmental change on phytoplankton communities in the Belgian part of the North Sea. VLIZ Marine Science Day 2018, Bredene, Belgium, 21st March 2018.

De Witte Bavo, H.M. Le, R. Lagring, K. Bekaert, 4DEMON: a novel approach to integrate 4 decades of pollution data, Ocean Sciences Meeting, 11-16 February 2018, Portland, USA.

Borges AV, G Speeckaert, W Champenois, MI Scranton & N Gypens, Temperature, productivity and sediment characteristics as drivers of seasonal and spatial variations of dissolved methane in the near-shore coastal areas (Belgian coastal zone, North Sea), EGU General Assembly, Vienna, Austria, 23–28 April 2017

Lagring R., 2015. Data management of 4DEMON. BEDIC workshop, 7 December 2015.

De Cauwer, K., Borges, A.V., Deneudt, K., Desmit, X., De Witte, B., Gauquie, J., Goffin, A., Lagring, R., Nohe, A., Sabbe, K., Stojanov, Y., Strobbe, F., Tyberghein, L., Van der Zande, D., 2015. 4 decades of Belgian marine monitoring: uplifting historical data to today's needs – 4DEMON. 47th International Liege colloquium: Marine Environmental Monitoring, Modelling and Prediction. Liège, Belgium, 4th – 8th May 2015.

De Cauwer K., 4DEMON: 4 decades of Belgian marine monitoring. IODE Scientific Conference. Brugge, 16 March 2015.

Monteyne E., presentation at the ICES Working Group on Marine Sediments in Relation to Pollution (WGMS), 2 March 2015.

Cooreman K., presentation at the OSPAR-ICES Study Group on Ocean Acidification, 6-9th October 2014.

De Witte B., presentation at the Marine Chemistry Working Group (ICES), 5th of March 2014.

Posters

Nohe A. et al, 2018. Long-term anthropogenically-induced change in North Sea phytoplankton: changes in diatom and dinoflagellates biomass, community structure and seasonality in the Belgian part of the North Sea between 1970 and 2010. 50th International Liège Colloquium on Ocean Dynamics, Liège, Belgium, 28 May – 1 June 2018.

Borges AV, W Champenois, MI Scranton, F Strobbe, T Vandenberghe, R Lagring, N Gypens, Productivity and temperature as drivers of seasonal, spatial and long-term variations of dissolved methane in the Southern Bight of the North Sea, 50th International Liege Colloquium on Ocean Dynamics, 28 May – 1 June 2018, Liège, Belgium.

Borges AV, T Vandenberghe, F Strobbe, R Lagring, Ocean acidification in the Belgian coastal zone, a contribution to the BELSPO project “4 decades of Belgian marine monitoring” (4DEMON), 50th International Liege Colloquium on Ocean Dynamics, 28 May – 1 June 2018, Liège, Belgium.

Lagring R. Vander Zande D., Fettweis M., Le H.M., Nohe A., Tyberghein L., Sabbe S., Desmit X. Long-term dataset on water clarity in the BPNS (1971-2016). 50th International Liege Colloquium on Ocean Dynamics, 28 May – 1 June 2018, Liège, Belgium.

Le Hong Minh, R. Lagring, K. Bekaert, B. De Witte, A. Nohe, Y. Stojanov, F. Strobbe, L.Tyberghein, F. Waumans, B. Wydooghe T. Vandenberghe, Uplifting 4 decades of Belgian historical data to today's needs: Harmonization is key, Ocean Sciences Meeting 11-16 February 2018, Portland, USA.

Bekaert K. et al, 2016. Determination of the best normalizing parameter for heavy metals in sediments of the Belgian Part of the North Sea (BPNS). North Sea Open Science Conference, 7-10 November 2016, Ostende, Belgium.

Borges A.V. et al, 2016. Ocean acidification in the Belgian coastal zone, a contribution to the BELSPO project “4 decades of Belgian marine monitoring” (4DEMON). North Sea Open Science Conference, 7-10 November 2016, Ostende, Belgium.

Borges A.V. et al, 2016. Methane dynamics in the Belgian coastal zone, a contribution to the BELSPO project “4 decades of Belgian marine monitoring” (4DEMON). North Sea Open Science Conference, 7-10 November 2016, Ostende, Belgium.

Lagring R. et al, 2016. The implementation of a data management plan to uplift historical data: long-term change detection in the Belgian Continental Shelf. IMDIS 2016, 11-13 October 2016, Gdansk, Poland.

Strobbe F. et al, 2016. Dealing with (historical) data and making it accessible: Data Inventory and Tracking System (DITS) applied in the scope of the “4 decades of Belgian marine monitoring” project (4DEMON), IMDIS 2016, 11-13 October 2016, Gdansk, Poland.

Nohe A. et al, 2016. Long-term phytoplankton monitoring data (1970 - 2010) from the Belgian North Sea reveal shifts in community composition and seasonal dynamics. VLIZ Marine Science day, 12 February 2016.

Gauquie, J., De Cauwer, K., Lagring, R., De Witte, B., 2015. Challenges to combine 40 years of marine contamination data within 4DEMON. 47th International Liege colloquium: Marine Environmental Monitoring, Modelling and Prediction. Liège, Belgium, 4-8 May 2015.

Lagring, R., Strobbe, F., Borges, A.V., Desmit, X., De Witte, B., Gauquie, J., Goffin, A., Nohe, A., Sabbe, K., Stojanov, Y., Tyberghein, L., Van der Zande, D., De Cauwer, K., 2015. 4 Decades of Belgian Marine Monitoring: uplifting historical data to today's needs. IODE Scientific Conference. Brugge, 16-20 March 2015.

Tyberghein, L., Borges, A.V., Deneudt, K., Desmit, X., De Witte, B., Gauquie, J., Goffin, A., Lagring, R., Nohe, A., Sabbe, K., Strobbe, F., Van der Zande, D. & De Cauwer K. 4DEMON – 4 Decades of Belgian Marine Monitoring. VLIZ Young Scientist Day, 20 February 2015.

Nohe, A., Tyberghein, L., Goffin, A., De Cauwer, K., Lagring, R., Vyverman, W. & Sabbe, K. Phytoplankton composition and biomass changes during the last four decades in the Belgian Coastal Zone. VLIZ Young Scientist Day, 20 February 2015.

Participation workshops and webinars

VIB training 'Introduction to the analysis of NGS data', Leuven, Belgium, January 2018.

Following the webinar "Delft3D Phytoplankton modelling: Concepts of BLOOM", March 2016.

Participation in the "LifeWatch Data Analysis Workshop", November 2015.

4DEMON participated with an information stand to the World Ocean Day at the Royal Belgian Institute of Natural Sciences on 7 June 2015.

Unfortunately, due to major technical problems with the RV Belgica, the planned 4DEMON participation to the opendoors in Temse was cancelled.

Other

The BMDC-team became member of the Technical Task Group in the Data Ingestion Project of EMODnet (European Marine and Observation Data Network), launched in May 2016, where a Data Ingestion Portal will be developed (with reference to DITS).

6. PUBLICATIONS

Scientific journals

Nohe A., Knockaert C., Goffin A., Dewitte E., De Cauwer K., Desmi, X., Vyverman W., Tyberghein L., Lagring R. and Sabbe K., 2018. Marine phytoplankton community composition data from the Belgian part of the North Sea, 1968-2010. Scientific Data. (accepted)

Borges A.V., Speeckaert G., Champenois W., M.I. Scranton M.I., Gypens N., 2017. Productivity and temperature as drivers of seasonal and spatial variations of dissolved methane in the Southern Bight of the North Sea, Ecosystems, doi: 10.1007/s10021-017-0171-7.

Borges A.V. et al., 2016. Massive marine methane emissions from near-shore shallow coastal areas. Nature Scientific Reports, 6:27908, (doi:10.1038/srep27908).

Informative journals and press

Anon. 1, 2016. Mer du Nord: d'importantes émissions de méthane. Techno-Science.net, 20 June 2016. Online press publication (<http://www.techno-science.net/?onglet=news&news=15271>).

Anon. 2, 2016. Pics de méthane dans les eaux côtières belges. Daily Science, 12 July 2016. Online press publication (<http://dailyscience.be/2016/07/12/pics-de-methane-dans-les-eaux-cotieres-belges/>).

Anon. 3, 2016. 300 fois plus de méthane en mer du Nord. L'avenir, 28 July 2016, p. 6-7. Written press publication (http://www.lavenir.net/cnt/dmf20160727_00858447).

Anon. 4, 2016. The North Sea: high levels of methane emissions at Réflexions web site (<http://reflexions.ulg.ac.be/en/MethaneNorthSea>)

Anon. 5, 2016. Une mer du Nord fiévreuse va rejeter 300 fois plus de méthane (https://www.rtf.be/info/societe/detail_une-mer-du-nord-fievreuse-va-rejeter-300-fois-plus-de-methane?id=9352403)

Lecrenier P., 2016. The North Sea: high levels of methane emissions at Réflexions website, 14 June 2016. Online press publication (<http://reflexions.ulg.ac.be/en/MethaneNorthSea>).

Louvigny A., 2016. Une mer du Nord fiévreuse va rejeter 300 fois plus de méthane. rtbf.be, 16 July 2016. Online press publication (https://www.rtf.be/info/societe/detail_une-mer-du-nord-fievreuse-va-rejeter-300-fois-plus-de-methane?id=9352403).

Sabbe K. and Vyverman W., 2016. Moet de zee niet weer bemest worden? Grote Rede 44: 18. Vulgarising publication.

Master Theses

Aththanayaka T., 2016. Temporal and spatial phytoplankton dynamics in the Southern Bight of the North Sea, MSc Thesis. Oceans & Lakes, Interuniversity Master in Marine and Lacustrine Science and Management: Antwerpen, Gent and Brussel. 96 pp.

Labatt C.K., 2016. Long-term phytoplankton trends in the Belgian North Sea (2002-2015): patterns and potential causes. MSc Thesis. Oceans & Lakes, Interuniversity Master in Marine and Lacustrine Science and Management: Antwerpen, Gent and Brussel. 59 pp.

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ANNEXES

1. Inventory projects, datasets and data-originators by theme
2. Inventory parameters
3. DITS
4. ODAS
5. Data portal
6. Water clarity parameter mapping

ANNEX 1. Inventory of projects, datasets and data-originators by theme

Projects	Datasets	Services	Contributors	Theme
AFVALWATEREN (1970-1972) - Ekologische en biologische studie van de kustwateren ter hoogte van Nieuwpoort in verband met het lozen van afvalwateren by IHE				
	Verontreiniging van de Noordzee aan de Belgische Kust van Koksyde tot Middelkerke: Fysico-chemisch onderzoek, by IHE, 1970-1972	IHE		Contaminants
	Ekologische en biologische studie van de kustwateren ter hoogte van Nieuwpoort in verband met het lozen van afvalwateren	IHE		
AMORE-I (1997-2001) - Advanced modelling and research on eutrophication linking eutrophication and biological resources				
		ULB-ESA	Lancelot C.	Eutrophication, Acidification
AMORE-II (2002-2006) - Advanced modelling and research on eutrophication linking eutrophication and biological resources				
		VUB-ECOL	Daro M.H., Breton E.	Eutrophication
		ULB-ESA	Lancelot C., Rousseau V.	Eutrophication
AMORE-III (2006-2009) - Advanced modelling and research on eutrophication linking eutrophication and biological resources				
		ULB-ESA	Lancelot C., Rousseau V.	Eutrophication
BAGGER: Heavy metals and PCBs in biota (1990-2014)				
	BAGGER-Bagger sediments data 2004-2015			Contaminants
	Heavy metals and PCBs in biota by ILVO (2002-2014)	ILVO		Contaminants
	Heavy metals and PCBs in biota by ILVO (1990-2003)	ILVO		Contaminants
BELCOLOUR (2010-2011)				
	CH4 dataset	ULg	A. V. Borges	
BIOCHEMISTRY (1997-2001) - Biochemistry of nutrients, metals and organic micropollutants in the North Sea				
		UA-MITAC	Van Grieken R.	Contaminants
		VUB-ANCH	Baeyens W.	Eutrophication
CANOPY (2002-2006) - Biogeochemical carbon, nitrogen and phosphorus fluxes in the North Sea				
		ULg-UOC	Borges A.V.	Acidification
		VUB-ANCH	Baeyens W., Brion N.	Eutrophication
		ULB-LOCGE	Chou L., Van der Zee C.	Eutrophication

Various Phytoplankton studies in the Southern Bight of the North Sea

Micro- and nannoplankton in Belgian coastal waters near Nieuwpoort between 1970 and 1972	Laboratorium voor Ekologie. Rijksuniversitair Centrum Antwerpen	De Pauw N.	Eutrophication
Phytoplankton community and environmental variables in the North Sea between 1970 and 1971	KUL	Louis A.; Petes J.; Ramboer T.; Vanderveken L.	
Total phytoplankton inventory in the North Sea and the river Scheldt in 1972			Eutrophication
Fytoplankton van de Belgische kustwateren: samenstelling, seizoenale dynamiek en ruimtelijke verspreiding			
Historical phytoplankton and environmental research data from the Laboratory for Hydrobiology, KULeuven.	Laboratory for Hydrobiology, KULeuven.		Eutrophication
PROJECT SEA - Projet Mer - Projekt Zee (PMPZ) (1970-1976)			
PMPZ: Distribution of phytoplankton pigments by ULB-OC, 1971-1976	ULB-OC		Eutrophication
PMPZ: Inventory of phytoplankton by KUL-SYTO, 1971-1976	KUL-SYTO		Eutrophication
PMPZ: Zooplankton studies in the Southern Bight of the North Sea between 1971 and 1974	ULB-LCI, ULB-OC, VUB-ECOL		
PMPZ: Study of nutrients by VUB-ANCH and ULB-LCI, 1971-1976	ULB-LCI, VUB-ANCH		Eutrophication
PMPZ: Heavy metals in fish and other marine organisms, by IRC	CLO-BIOMET, CODA-CERVA, ICES, IRC-ISO, KUL-SED, RVS, RVZ, RVZ-CLO		Contaminants
	Gent, ULB, VUB		
PMPZ: Study of hydrology by ULg and VUB, 1971-1976	CDCD, RBINS, ULg-CHIM, ULg-UOC, VUB-ANCH		
PMPZ: Inventory of water pollution in the Belgian coastal zone, 1971-1976	IHE, IHE-MUMM, IRC-ISO, RBINS		Eutrophication
PMPZ: First trophic level: analyse of phytoplankton by ULB, 1971-1976	ULB-OC		
PMPZ: Primary production in the Southern Bight of the North Sea between 1971 and 1975, by VUB	ULB-LCI, VUB-ECOL		Eutrophication
PMPZ: Salinity measurements by FN-ZM	FN/ZM-CHIM		Acidification
PMPZ: Pesticides in marine organisms, by VUB (1971-1976)	VUB-FARM,		Contaminants
PMPZ: Heavy metals in mussels and other from quayside (Van der Ben)	IHE, IRC-ISO		Contaminants
PMPZ: PCBs in biota by Station de Phytopharmacie	CES, IRC-ISO, KUL-SED, RVZ, RVZ-CLO		Contaminants
	Gent, ULB, VUB		
PMPZ: Study on sediment by ULB and KUL: metals in sediment 1000P	FN-ZM-DER, ULB-LOCGE		Contaminants
PMPZ: R�el�eve des donn�ees m�et�eorologiques et de navigation by FN-ZM, 1971-1976	FN-ZM		Acidification
PMPZ: Total diatom abundance in the Southern Bight of the North Sea in January and February 1971	ULB-OC	Steyaert J.; Bouillon J.	Eutrophication
PMPZ: Total phytoplankton inventory in the Southern Bight of the North Sea between 1973 and 1974	KUL-SYTO	Huys L.	Eutrophication
PMPZ: Total phytoplankton inventory in the Scheldt estuary on the 19th of March 1974	KUL-SYTO	Huys L.	Eutrophication

PMPZ: Total phytoplankton inventory in the Southern Bight of the North Sea in 1971	KUL-SYTO	Robijns J.; Rabijns M.; Huys L.; Louis A.	Eutrophication
PMPZ: Total hydrozoa abundance in the Southern Bight of the North Sea between 1971 and 1972	Laboratoire d'Océanologie. ULB	Houvenaghel G	Eutrophication
PMPZ: Benthic communities in the Southern Bight of the North Sea 1970-1976	RUG-MORF, RVZ, RVZ-CLO Gent, VUB- ECOL		Eutrophication
PMPZ: Phytoplankton in the Southern Bight of the North Sea 1970-1976			Eutrophication
PMPZ: Primary production in the Southern Bight of the North Sea 1971-1975	ULB-LCI, ULB-OC, VUB-ECOL, VUB- ETOX		Eutrophication
PMPZ: Phytoplankton inventory in the Southern Bight of the North Sea 1971-1975	KUL-SYTO, RBINS, VUB-ECOL		Eutrophication
PMPZ: Diatom abundance in the Southern Bight of the North Sea 1971-1976	RBINS		Eutrophication
PMPZ: Zooplankton in the Southern Bight of the North Sea 1970-1976	ULB-OC, VUB-ECOL		Eutrophication
PMPZ: Hydrozoa abundance in the Southern Bight of the North Sea 1971-1972			Eutrophication
PMPZ: Copepoda in the Southern Bight of the North Sea 1971-1974			Eutrophication
PMPZ: Meteorological and hydrological observations in the Southern Bight of the North Sea 1970-1976			Acidification
PMPZ: Alkalinity measurements in the Southern Bight of the North Sea 1970-1976	ULg-UOC		Acidification
PMPZ: Meteorological data in the Southern Bight of the North Sea 1970-1976			Acidification
PMPZ: Salinity measurements in the Southern Bight of the North Sea 1970-1976	FN/ZM-CHIM, FN-ZM, FN-ZM-DER		Acidification
PMPZ: Temperature and dissolved oxygen measurements in the Southern Bight of the North Sea 1970-1976	BINS, RVZ-CLO Gent, ULg-CHIM, ULg- UOC, VUB-ANCH, VUB-ECOL		Eutrophication, Acidification
PMPZ: Contaminants in the Southern Bight of the North Sea 1970-1976			Contaminants
PMPZ: Heavy metals in the Southern Bight of the North Sea 1970-1976	CODA-CERVA, IRC-ISO, MUMM, RBINS, RVZ, RVZ-CLO Gent, UIA-SCHEIK, ULB- LCI, ULB-ZOO, ULg-CHIM, ULG-HECQ, VUB-ANCH		Contaminants
PMPZ: Marine organisms in a dumping area or organic industrial waste along the Belgian coast 1970-1976	RVZ-CLO Gent		Contaminants
PMPZ: Pesticides in the Southern Bight of the North Sea 1970-1976	IHE, VUB-FARM		Contaminants
PMPZ: Eutrophication data in the Southern Bight of the North Sea 1970-1976			Eutrophication
PMPZ: Chlorophyll a and particulate organic matter in the Southern Bight of the North Sea 1970-1976	ICES, RBINS, RVZ-CLO Gent, ULB, ULB- OC		Eutrophication
PMPZ: Nutrients in the Southern Bight of the North Sea 1970-1976	ULB-ESA, ULB-LCI, VUB-ANCH, VUB- ECOL		Eutrophication
PMPZ: Suspended matter in the Southern Bight of the North Sea 1970-1976	KUL-SED, ULB-LCI		Eutrophication

PMPZ: Inventory of pollutants in the Belgian hydrographic network and coastal zone 1970-1976	IHE, IHE-MUMM, IRC-ISO, RBINS	Contaminants
PMPZ: Contamination of marine organisms - physiology: Laboratory Studies 1970-1976	RUG, ULB, ULG-HECQ	Contaminants
CRA (1976-1982) - Concerted Research Actions Oceanology		
CRA: Dosage van chlorofyl- en pheophitinepigmenten in de Belgische kustwateren, by VUB, ULB and ULg (1977-1982)	VUB	Eutrophication
CRA: Nutrients in the Southern Bight of the North Sea, by VUB, ULB and MUMM (1976-1982)	Labo Analytische Scheikunde, VUB	Eutrophication
CRA: Zooplankton studies at a fixed station (West-Hinder) in the North Sea, by VUB (1977 and 1979)	Laboratory of Ekologie en Systematiek, VUB (remark: original documents from BMDC transfered to VLIZ)	
CRA: Working Group on Organic Matter (1977-1982)		
CRA: Zooplankton study in the North Sea, by ULg (1976-1982)	Laboratory of general Biology, ULg	
CRA: Spatial variation of the zooplankton community in the Belgian part of the North Sea on 18-19 April 1978	Laboratorium voor Ekologie en Systematiek. VUB	
CRA: Physical measurements, by MUMM (1977-1982)	MUMM	Eutrophication, Acidification
CRA: Heavy metals in marine organisms, by RVZ	RVZ	
CRA: Heavy metal contamination in sediments from the Belgian Coast and Scheldt Estuary		
DREDGING (1978-) - Effects of dredged material dumping on the marine environment		
Research on the effects of dredged material dumping on the marine environment	OD nature, MOW, Flanders Hydraulics Research, RVZ	Contaminants
DUMP (1976-1988) - Effects of disposal of waste from the Belgian titanium dioxide industry		
Research on the effects of dumping of waste from the Belgian titanium dioxide industry	RVZ, Sciensano (Coda-Cerva)	Contaminants
GILSON (1899-1926) - Exploration of the Southern North Sea by G. Gilson		
		G. Gilson
GOBY (2003-2005) - Goby Dataset		
IJSLANDVAARTEN (1970-1971)		
IPMS-PHAEO (1992-1995) - Dynamics of coastal eutrophicated ecosystems		
	ULB-ESA	Lancelot C.
LIFEWATCH (2002-2015) - LifeWatch observatory data: nutrient, pigment, suspended matter and secchi measurements in the Belgian Part of the North Sea		
	VLIZ	Eutrophication
MARBIOL (1993-1999) - Hyperbenthos and Meiobenthos		
Hyperbenthos and Meiobenthos compiled by MARBIOL	MARBIOL	Eutrophication

MULTICOLOR-1 (1996-1999) - The multicoloured North Sea		Pichot G.	Eutrophication
MULTICOLOR-2 (1998-2000) - The multicoloured North Sea			
REMSEM dataset (1996-2014) - Remote Sensing and Ecosystem Modelling	ODNATURE-REMSEM		Eutrophication
In Situ data	RBINS-REMSEM		Eutrophication
Earth Observation data	RBINS-REMSEM	Dimitry Van der Zande	Eutrophication
REVAMP (1997-2003) - Remote sensing for eutrophication monitoring			
	RBINS-MUMM	Ruddick K., De Cauwer V.	Eutrophication
	PML	Tilstone V., Vicente V.	Acidification
SISCO (2002-2006) - Silica retention in the Scheldt continuum and its impact on coastal eutrophication			
	ULB-LOCGE	Chou L.	Eutrophication
TROPHOS (2002-2006) - Higher trophic levels in the Southern North Sea			
	UGent-MARBIO	Vincx M., Vanaverbeke J.	Eutrophication
	UGent-MARBIO, VLIZ		
UA (1987-1988) Heavy metals study in North Sea and Schelde by Van Grieken			
	UA	Van Grieken, PhD Van Alsenoy, Araujo	
	VUB-ECOL		
VUB_ECOL_data (1987-1990)			
Diskette	VUB-ECOL		Eutrophication
MONIT_WFD (2002-) - Monitoring in the frame of the Water Framework Directive (WFD)			
	ODNATURE_GULLEDELLE		Contaminants
	ODNATURE_GULLEDELLE		Eutrophication
MONIT WFD (2004-2010)			
Datacompilation thesis Sara Denayer		Denayer	Eutrophication

ANNEX 2. List of parameters tackled within 4DEMON

Parameter Name	Parameter code	Category	Theme
Ammonia	AMONA	Nutrient	Eutrophication
Ammonium (NH ₄ -N)	AMON	Nutrient	Eutrophication
Chlorophyll-a	CPHL	O-MAJ	Eutrophication
Dissolved Inorganic Nitrogen	DIN	Nutrient	Eutrophication
Nitrate (NO ₃ -N)	NTRA	Nutrient	Eutrophication
Nitrate + Nitrite (-N)	NTRZ	Nutrient	Eutrophication
Nitrite (NO ₂ -N)	NTRI	Nutrient	Eutrophication
Phosphate (PO ₄ -P)	PHOS	Nutrient	Eutrophication
Secchi depth	SECCI	Physical	Eutrophication
Secchi depth (black)	SECCIB	Physical	Eutrophication
Secchi depth (white)	SECCIW	Physical	Eutrophication
Silica (SiO ₂)	SIO2	Nutrient	Eutrophication
Silicate (SiO ₄ -Si)	SLCA	Nutrient	Eutrophication
Silicon	SI	Metal	Eutrophication
Suspended solids	SUSP	Physical	Eutrophication
Turbidity	TURB	Physical	Eutrophication
Total alkalinity	ALKY	I-MAJ	Acidification
Salinity	PSAL	I-MAJ	Acidification
Water Temperature	TEMP	Physical	Acidification
pH - Hydrogen ion concentration	PH	I-MAJ	Acidification
Methane	CH4		Acidification
Chromium	CR	Metal	Contaminants
Mercury	HG	Metal	Contaminants
Cadmium	CD	Metal	Contaminants
Lead	PB	Metal	Contaminants
Arsenic	AS	Metal	Contaminants
Copper	CU	Metal	Contaminants
Maximum weight	WTMAX	Biological	Contaminants
Weight	WEIGHT	Biological	Contaminants
Mean weight (or individual weight)	WTMEAN	Biological	Contaminants
Minimum weight	WTMIN	Biological	Contaminants
Standard deviation of the weight	WTSD	Biological	Contaminants
Zinc	ZN	Metal	Contaminants
Extractable lipids	EXLIP	O-MAJ	Contaminants
Sum 7 PCBs	SUM7PCB	PCB	Contaminants
Maximum length	LNMAX	Biological	Contaminants
Mean length (or individual length)	LNMEAN	Biological	Contaminants
Minimum length	LNMIN	Biological	Contaminants
Standard deviation of the length	LNSD	Biological	Contaminants
2,2',4,5,5'-pentachlorobiphenyl	CB101	PCB	Contaminants
2,3,3',4,4'-pentachlorobiphenyl	CB105	PCB	Contaminants
2,3',4,4',5-pentachlorobiphenyl	CB118	PCB	Contaminants
2,2',3,4,4',5'-hexachlorobiphenyl	CB138	PCB	Contaminants

2,2',4,4',5,5'-hexachlorobiphenyl	CB153	PCB	Contaminants
2,3,3',4,4',5-hexachlorobiphenyl	CB156	PCB	Contaminants
2,2',3,4,4',5,5'-heptachlorobiphenyl	CB180	PCB	Contaminants
2,4,4'-trichlorobiphenyl	CB28	PCB	Contaminants
2,4',5-trichlorobiphenyl	CB31	PCB	Contaminants
2,2',5,5'-tetrachlorobiphenyl	CB52	PCB	Contaminants
Sum 7 PCBs	SUM7PCB	PCB	Contaminants
Aroclor 1254	PCBAROCHLOR1254	PCB	Contaminants
Chlorine	CL	I-MAJ	
Dissolved organic carbon	DOC	O-MAJ	
Dissolved organic nitrogen	DON	O-MAJ	
Dissolved organic phosphorus	DOP	Nutrient	
Dissolved oxygen	DOXY	I-MAJ	
Fluorine	F	I-MAJ	
Organic nitrogen	ON	Nutrient	
Oxygen	OXY	I-MAJ	
Oxygen saturation	DOXYSAT	I-MAJ	
Particulate inorganic carbon	PIC	Nutrient	
Particulate nitrogen	PN	O-MAJ	
Particulate organic carbon	POC	O-MAJ	
Particulate organic nitrogen	PON	O-MAJ	
Total carbon	CTOT	O-MAJ	
Total organic carbon	TOC	O-MAJ	

ANNEX 3: Data Inventory and Tracking System (DITS)

1. Summary

The Data Inventory and Tracking System (DITS) (www.dits.bmdc.be) was designed by BMDC in the frame of 4DEMON. It was developed to improve the data flow between our partners and BMDC. DITS has a 3-tier architecture based on the open standards allowing any of the three tiers to be upgraded or replaced independently.

2. Architecture

The three tiers in the DITS architecture are:

- a. **Presentation Tier:** Users operate on this tier and need no knowledge of the underlying layers. Each user needs to register and receives a user role with specific privileges. After logging in, (s)he has access to the dynamic XHTML pages via different tabs:
 - *Submit:* adding new datasources, datasets, projects, services and platforms.
 - *Explore or Edit:* browsing through the data and update records if required.
 - *Database Statistics:* showing readymade views for the users with numbers and lists.
 - *Status Tracking:* consulting and managing the status of datasources (e.g. original file, processed, imported) and datasets.

The user can upload files (any format) during submission and during each status update. These files are saved on a separate server and can be downloaded at any given moment. All results can be exported in three formats (XLS, CSV, XML). A FAQ page and a manual are available via 'Help and Hints'. Specific web services can be developed to communicate with other systems or, in our case, to show information on the 4DEMON project website.

- b. **Application Tier:** Developed in Java, the Java EE and Java Persistence API reside here. This tier is the mediator between the user and the database. All views are also generated here. An open-source application server is coordinating all actions.
- c. **Database Tier:** The Database Tier is housing an Oracle database. This can easily be replaced by any other type of database (e.g. MySQL database).

This 3-tier database architecture is highly convertible, as almost all its components are independent and can be modified independently. We are now using this architecture for the inventory and reporting for the Marine Strategy Framework Directive (MSFD).

3. Objectives

DITS has been developed with the specific objectives below (a-g). In the frame of 4DEMON, the main concerns were creating an inventory of available sources (a), identification of missing data (e) and providing a follow-up system of import in the database (f). Files that are uploaded need to be well managed (c). The data manager can combine data sources in datasets (d), decide whether the dataset can be imported and document the status of import. The system will be used for reporting data to the BMDC by all data-originators, where the original data files can be uploaded (b).

- a. **Inventory of sources (eg. Publications, databases, data files...)**
 - *Submit:*

- Metadata: the user can enter a new datasource (e.g. publication, original common layout) and enter relevant metadata (e.g. author, service, project, dates).
- Upload file (not mandatory): the user can upload the original file (all formats: pdf, excel, access, text, etc.) which will be stored on a secure separate server.
- External link: in case the file is digitally available on another website.
- *View*: the user can explore the list of datasources available, select them and browse through the detailed information for each datasource
- *Edit*: the user can select a datasource to update the information or to upload a new file in case the previous loaded file needs to be replaced.

b. Reporting data to BMDC

- The data file can be uploaded when creating a new datasource, where it is linked to the metadata (project, service, etc. see (a)).

c. Archival of files

- Uploaded files are put on a separate secure server.
- Versioning of files: the first upload is version 1 (v1). If the user notices an error and uploads a new file via the interface, v2 is created. In this system, both files are stored. In the database on the other hand, only the latest version (v2) is referred to.
- The path of uploaded files is organised in a logical way (e.g. Data acquisition Project\Service)

d. Identification of datasets

- The data manager can combine data sources in a comprehensive dataset, a logical group of data that will be imported together in the database (cfr. Common Layout or internal reporting template).
- Processed files can be uploaded here (e.g. Processed Common Layout, imported txt file)

e. Identification of missing data

- For each dataset, the data manager can indicate what metadata is missing (e.g. Sampling gear, analysis method, units), this information can help to determine the completeness of the dataset.

f. Follow-up: status tracking of files and datasets

- Datasources: For each original file, the import status can be entered (which is imported together with information on date and user).
- Datasets: during each step of the import process, the status can be entered and the processed file can be uploaded (together with the date and the user).
- The chronological process of import can be followed by the user and the data manager.

g. Exploration of database and export of data

- Here, the database can be explored. External users only have restricted view on sources and sets (their sources, specific projects), but they can follow their data at any moment in time.
- Links to specific tables (services, projects, platforms) in the central BMDC database are set up to re-use the already existing information.

Annex 4: Processing procedures for continuous along track data (ODAS)

The oceanographic underway data of the Belgica is continuously stored in the Ocean Data Acquisition System (ODAS) since 1984. A program has been written in the Perl programming language to copy all existing data from the ODAS database to PostgreSQL; this program now operates as a nightly job to copy over all data. During the years 560 different physical properties/sensor combinations have been created (in what follows, we refer to this broad concept as a 'parameter'). The central table is the values table, which is a single table containing 180 million individual observations.

As new and better sensors are acquired all the time and might operate concurrently, the start and stop dates of all parameters should be stored.

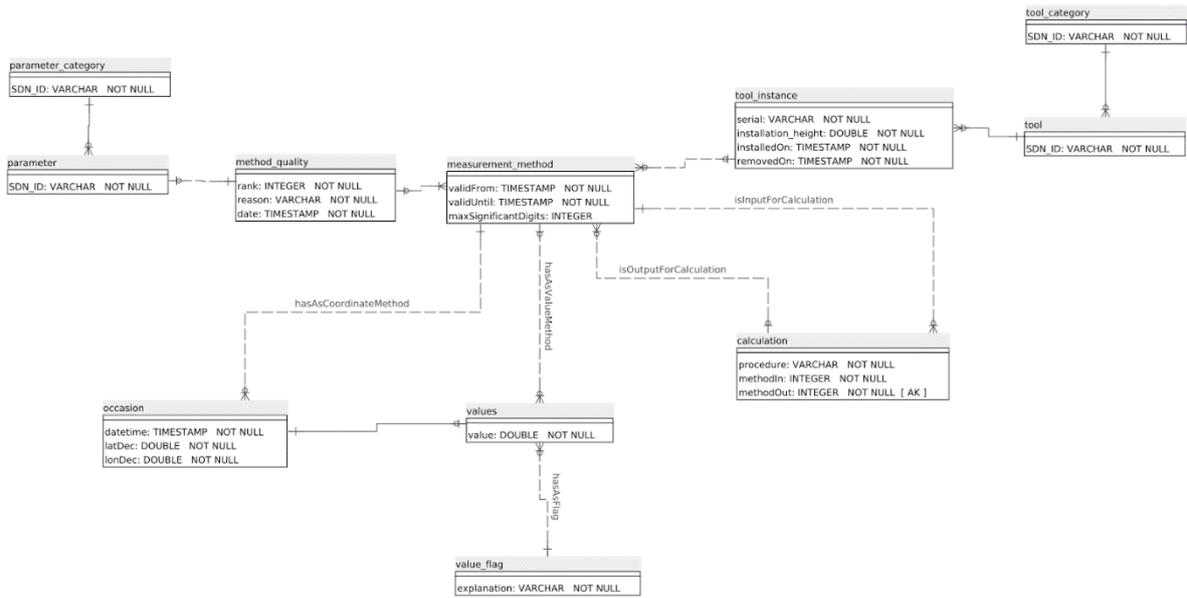
Before building the database, an entity-relationship (ER) model has been created in order to explore of what an acquisition system should be capable of:

- Storing information on sensor installation details (date installed, date removed, height, serial number);
- Making a clear distinction between measured and imputed parameters; Imputed parameters are calculated from different other (imputed or measured) parameters;
- For imputed parameters: storing how (as a string) and from which parameters the parameter is calculated;
- Information on the measurement methods used for measuring a specific physical property ('parameter' in the strict sense) and ranking the quality of each method for one parameter;
- Splitting values from measurement occasions. A measurement occasion denotes the unique combination of the time and location of the vessel (for this reason, it is advisable that it only contains the most accurate GPS localisation). Value has been separated from occasion because an occasion is not necessarily associated with a physical measurement value and because the location information in itself has a measurement method and quality.

The ideas from this ER-model have been implemented partially, in five tables, each table denoting the parameter*measurement method*sensor information, the quality ordering, the values, the occasion and the parameter category. The quality table denotes for parameters measuring the same physical property a quality ordering, so that it is possible to select the most precise physical property value when it is measured by different sensors at the same time. This is crucial to retrieve accurate location info.

A set of functions have been created to extract data in a transposed format (one row for each timestamp, with the most correct location info), and to query by year and by parameter category. No web service or user interface has been built to expose this data; it can be requested by email to info@bmdc.be. Further dissemination of this data should make use of the Observations and Measurements and Sensor ML standards.

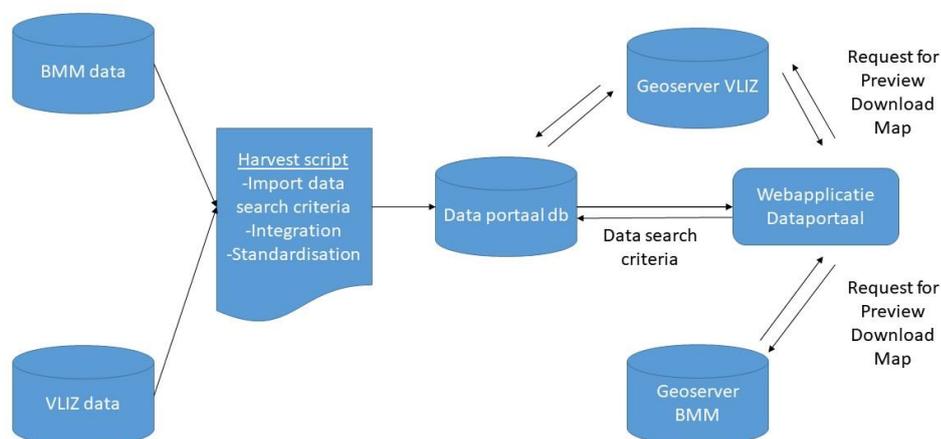
The entity-relationship model of the ODAS data and parameter metadata.



Annex 5: Technicalities data portal

An important task was the creation of a data portal capable of viewing the data subsets relevant to 4DEMON present in IDOD and DATAPORTAL DB. The data portal makes use of the OGC Web Feature Service standard (WFS), which provides an interface to allow data requests on spatial data; this approach can be used in conjunction with showing the data on a map (by using a WMS). Both BMDC and VLIZ implemented their own WFS endpoints using GeoServer; those provide interoperable APIs for data querying. The data portal itself is based on the existing VLIZ data portal template. The VLIZ data portal has two domains, and as a first step provides the user data with a choice on biotic (ecology, populations and morphology) and abiotic (nutrients and contaminants in the environment and in biota) data.

Data flow scheme



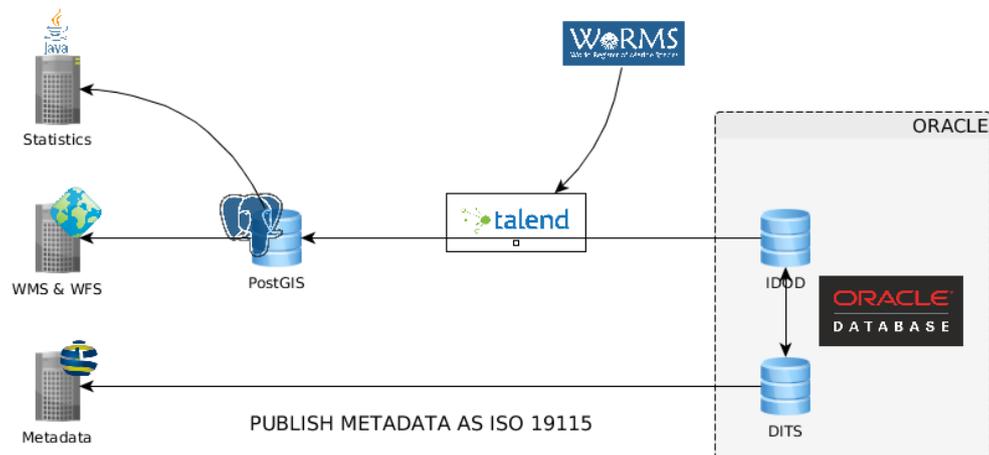
General map of the data flow

The 4DEMON data portal makes use of a faceted search to drive the interface, with an estimate of the number of results a query will result. To calculate the number of results, BMDC has created a web service that calculates the count of each combination (ie. substrate, parameter code, location box, project (identifier and name), dataset (identifier, name and url) and season). VLIZ harvests this web service periodically, stores the results in a database, combines it with their counts, and builds an index from this. As a crucial part of interoperability, VLIZ and BMDC have matched their parameters. This parameter matching allows to query both systems concurrently. When a user of the data portal selects query arguments provided by the faceted search, these are passed on as query arguments for the WFS and the results shown.

Data sharing scheme - BMDC perspective

The IDOD database of BMDC is an Oracle 10 database. In order to allow GIS capabilities and expose the data through a WFS, it was found most feasible to transform the data to a PostGIS database and make use of GeoServer, a server solution to sharing geographical data using multiple protocols (among others WFS and WMS). This data warehouse strategy

has the additional benefit of speeding up queries as the data of each domain (data on biota vs. abiotic data) is shown as two separate tables. The data transformation is performed using a Talend ETL procedure. The below graph shows the complete workflow. A matching with World Register of Marine Species is performed by the ETL procedure.

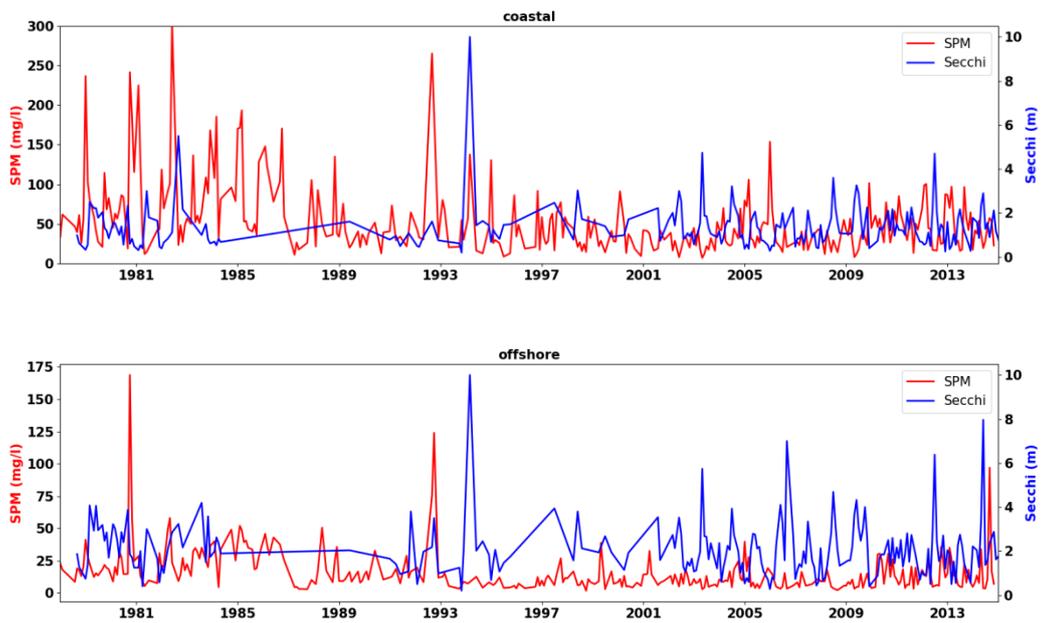


Integration of DITS into the IDOD data

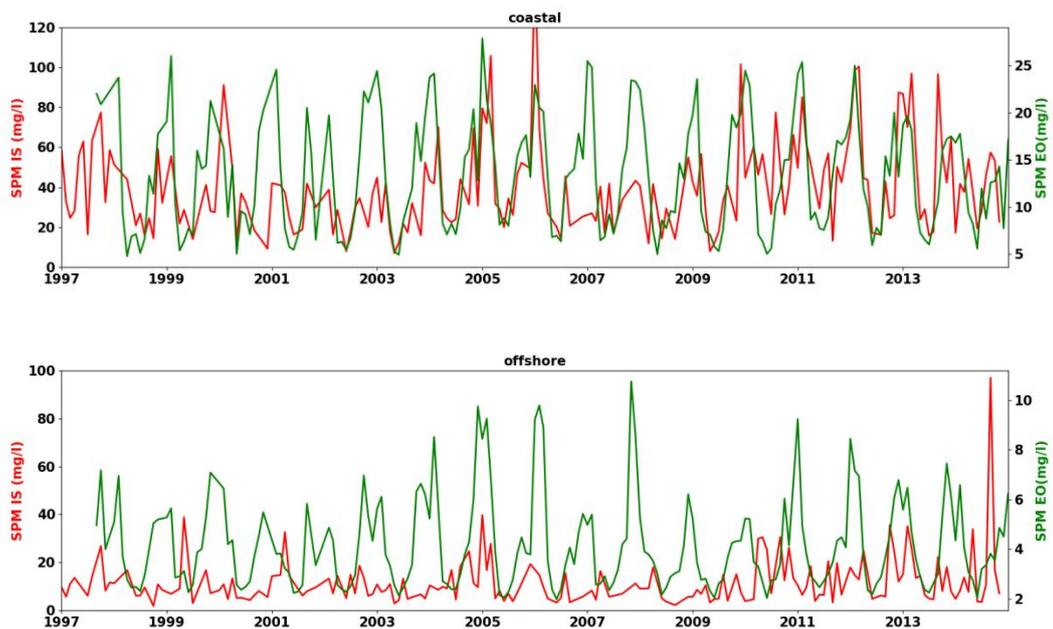
DITS has been implemented to fulfill the requirements of task 2.1, in order to assist in compiling an integrated inventory and management tool for all BMDC datasets. DITS has been extended to capture the concept of hierarchical datasets and has been linked with IDOD so that assigning measurement values to datasets is easier. For this, the concepts of 'ingestion dataset' and 'collection dataset' have been defined. Both need to be defined in DITS, and this necessity has been entered into the Standard operating Procedure of BMDC to import data. The ingestion dataset is the dataset originally used to enter the data into IDOD, i.e. the "Common Layout" dataset that BMDC prepares from one or more primary sources. These primary sources and the ingestion dataset concepts have been part of (the goal of) DITS since the beginning, but have been formalized a bit more. Datasets have also received a flag of whether they can be published, i.e. that the metadata description can be read by an external user and (possibly) that the data itself can be read. Only the collection datasets are public. Combined with a standardized API (WFS) to access the data, it is easy to go from a metadata description to the actual data.

Annex 6: Water clarity plots

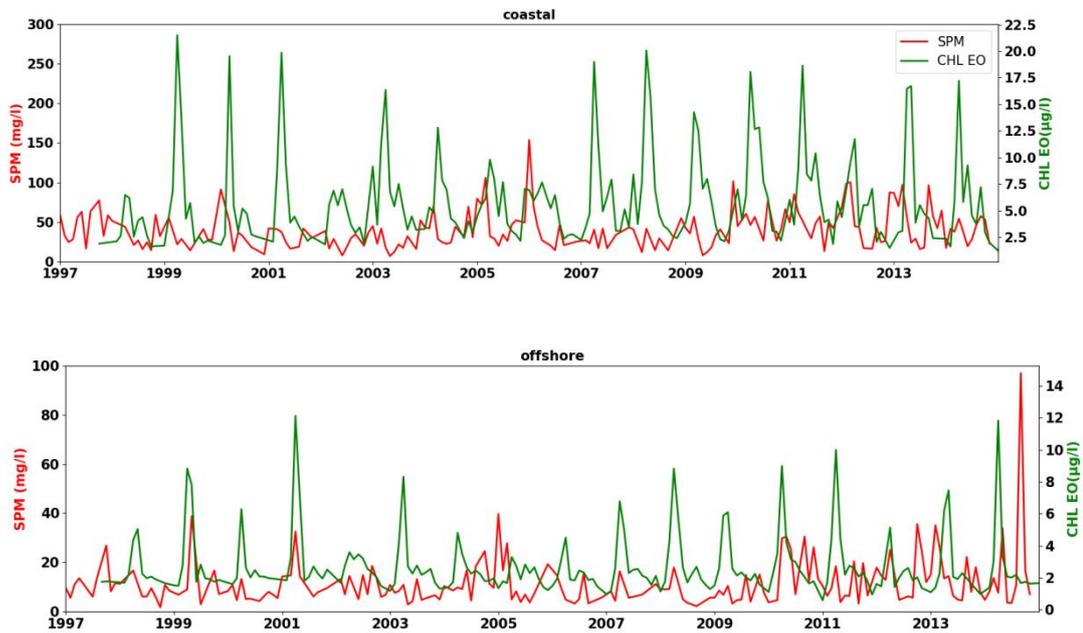
TSM IS versus secchi data



TSM IS data versus EO



TSM IS data versus CPHL EO



CPHL IS versus CPHL EO (exclusive trichromatic CPHL)

