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The EcoQO on Mercury and Organohalogens in Coastal Bird Eggs

Report on the Pilot Study 2008 – 2010

Tobias Dittmann, Peter H. Becker, Joop Bakker, Anders Bignert, Elisabeth Nyberg, M. Glória Pereira, Ursula Pijanowska, Richard Shore, Eric W.M. Stienen, Geir Olav Toft and Harald Marencic

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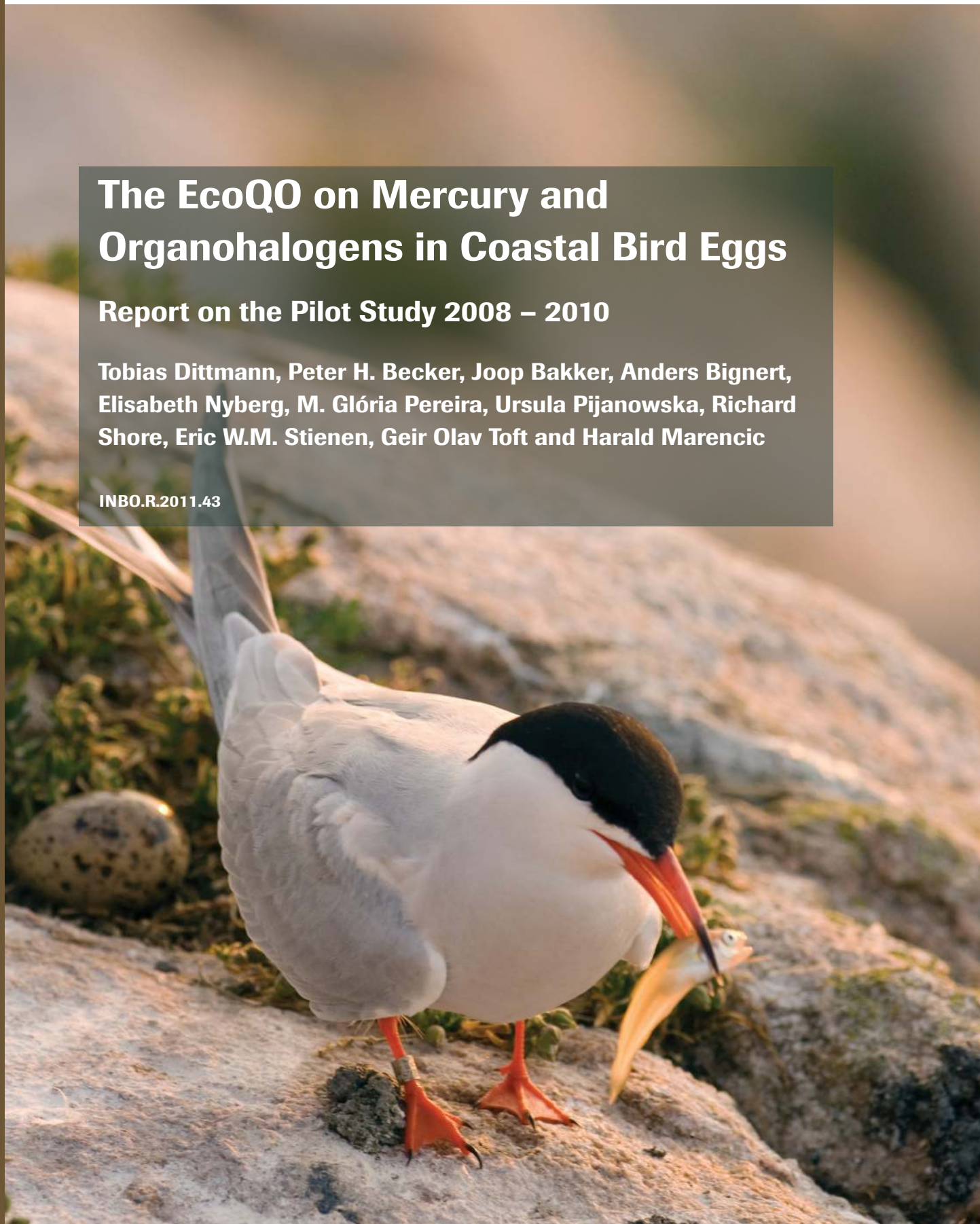

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Editorial foreword

On the one hand the coastal areas of the North Sea hold important breeding populations of marine birds but on the other hand these areas are also under pressure from pollution with environmental chemicals. Due to biomagnification effects through the food chain, the bird egg is an ideal matrix to get a consistent and reliable picture of the pollutant level of the marine environment over time and space.

With this report, we are pleased to give, for the first time, a North-Sea-wide overview about the spatial patterns of mercury and organochlorine pollution in coastal bird eggs, presenting data from Oystercatcher, Common Tern and Arctic Tern in the period 2008-2010. We are able to show how the recently defined Ecological Quality Objectives (EcoQOs) for these substances in coastal bird eggs have been fulfilled in the different parts of the North Sea. This report aims to provide input for the further development of EcoQOs in the North Sea area and for their potential role in the implementation of the EU Marine Strategy Directive.

We would like to thank all those who contributed to the report by coordinating and organizing the field sampling, by participating in workshops and providing funding for the project. A large part of the work was carried out in the framework of the long established Trilateral Monitoring and Assessment Program (TMAP) of the Trilateral Wadden Sea Cooperation which provided resources and data for the Wadden Sea sites. A cooperation with institutes in Belgium, Norway, Sweden and the UK made it possible to extend the monitoring of pollutants from the Wadden Sea to the entire North Sea. This was achieved thanks to the enormous commitment of many colleagues.

We hope to be able to continue this fruitful cooperation in the future with the aim to further support the protection of the North Sea ecosystem with integrated approaches.

The authors

Abstract

To categorize the current environmental health status of the Northeast Atlantic and the North Sea, ecological quality objectives (EcoQOs) have been formulated by OSPAR in recent years for different ecological quality elements such as the contamination of the marine environment with anthropogenic heavy metals and organochlorines. To measure it, coastal bird eggs have proven to provide a favorable matrix. In 2005, following advice from ICES, OSPAR agreed on the threshold concentrations proposed as EcoQOs for these two major substance groups in bird eggs. In a pilot study, residues of the heavy metal mercury (Hg) and the organochlorines PCBs (62 congeners summarized = ∑PCB), DDT and metabolites (∑DDT), HCB and the different HCH isomers (∑HCH) were analyzed in Common Tern Sterna hirundo or Arctic Tern Sterna paradisaea and Oystercatcher Haematopus ostralegus eggs collected at in total 21 sites in seven nations surrounding the North Sea. For the majority of sites, sampling took place in 2008, 2009 and 2010. The methods of sampling and analyzing were performed according to internationally recognized standards (JAMP, OSPAR). The major aims of this report are:

- to suggest an actualized threshold concentration for Hg for which only preliminary target values had been proposed
- to test the suitability of the Arctic Tern as a potential alternative species to replace the Common Tern at sites where the latter does not occur in sufficient numbers
- to present spatial contamination patterns and temporal developments
- to address whether the hitherto existing EcoQO objectives are fulfilled
- to give recommendations for monitoring, assessment and management of this EcoQO in the future
- to mention the advantages of species and matrix as well as the limitations of the application of this EcoQO
- to discuss how the preconditions for a potential inclusion of the chemical monitoring with seabird eggs into a coordinated environmental monitoring program (CEMP) can be fulfilled (see above)
- to give suggestions for combining the EcoQO program with other existing or planned monitoring programs for the marine environment.

Interspecific variation

Contamination levels of Common Tern and Arctic Tern eggs were similar for most chemicals, indicating the suitability of the Arctic Tern to replace the Common Tern in areas where the latter is rare. The contamination of the terns was in most cases higher than of the Oystercatcher, which can be explained by different feeding, breeding and migration strategies.

Actualized EcoQO for Hg

Measurements of Hg in the new reference areas resulted in an actualized target threshold concentration of 160 ng/g in the tern species. For the Oystercatcher, the study has confirmed the preliminary threshold concentration of 100 ng/g.

Spatial contamination patterns

In Common and Arctic Tern eggs, concentrations of Hg, ∑PCB, HCB and ∑DDT peaked at the inner Elbe estuary. In case of ∑PCB, a second peak was recorded at Terneuzen, followed by further comparatively high values at Delfzijl and Zeebrugge. HCB and ∑DDT showed a second, but considerably lower peak at Middlesbrough. ∑HCH showed a clear peak at Middlesbrough.

In the Oystercatcher, the spatial pattern of contamination varied stronger between substance groups than in the terns. However, for Hg, ∑PCB, ∑DDT and ∑HCH, highest mean concentrations were measured at the Elbe estuary or in its immediate surroundings (Trischen). HCB reached its second highest concentration at the inner Elbe estuary following a very prominent maximum value at Delfzijl. For Hg, a further concentration peak was measured at Balgzand and for ∑PCB at Delfzijl and Dollart.

In summary, in the southern North Sea, egg contamination was characterized by comparatively high levels of industrial chemicals whereas in the western North Sea and the Elbe estuary, insecticides reached peak concentrations.

Temporal contamination patterns

In the Common and, respectively, the Arctic Tern, contamination with Hg, HCB and ∑HCH was increasing in the three study years 2008-2010 at most sites. ∑PCB and ∑DDT were increasing at Stroemstad and Presteskjaer, and, respectively, at one or several German sites whereas they were decreasing at several Dutch sites.

In the Oystercatcher, HCB and ∑HCH were also increasing at more than the half of the sites. As in the terns, ∑PCB was increasing at Elbe and ∑DDT was increasing at several German sites. In contrast, Hg concentrations were increasing only at two sites in Germany whereas decreases were recorded at Griend, Elbe, Trischen and Stroemstad. For the other substance groups, concentration decreases were only found at single sites.

Fulfillment of the hitherto existing EcoQO objectives

In the terns, Hg, ∑PCB and ∑DDT exceeded the EcoQO objectives in all three study years. HCB remained below the target threshold value of 2 ng/g only in 2008 at Zeebrugge (the only study year at that site) and in one year (2008) at Presteskjaer. At all sites except Middlesbrough and Elbe, ∑HCH fulfilled the EcoQO at least in some study years.

In the Oystercatcher, Hg exceeded the target concentration of below 100 ng/g at all study sites in all study years except at Presteskjaer in 2008. ∑PCB exceeded the target concentration of below 20 ng/g in all years at all sites. HCB fulfilled the EcoQO in some or even all three study years at most sites except at those sites situated in or near the estuaries of the large rivers Ems and Elbe where the values exceeded the EcoQO in all study years. ∑DDT remained below the target threshold in some years at Hallig Hooge and in the Danish Wadden Sea. At all other sites, concentrations remained higher. ∑HCH concentrations fulfilled the EcoQO at all sites in some years. However, ∑HCH concentrations have increased at 10 of 12 sites.

Suitability of the matrix bird egg and of the study species

Coastal birds accumulate contaminants effectively in their eggs and enable the monitoring of substances even if their absolute concentrations in water, sediment or soil are very low. In comparison to those measured in other matrices, contaminant levels measured in bird eggs show a low variability, they enable a high statistical power of results and provide a high information density. Compared to other sampling methods, collecting bird eggs requires only a low logistical effort.

Due to their feeding, breeding and migration ecology, tern species are particularly effective accumulators of pollutants ingested in a short period at the breeding site and are easy to sample, but some uncertainties persist due to their long-distance migrations and possible migrations of their prey. The Arctic Tern can replace the Common Tern as study species.

In contrast, the much more sedentary Oystercatcher integrates environmental pollution over a larger period of time and feeds on sedentary prey but may forage to a higher degree than the terns in non-marine habitats. It is favorable to monitor both the Oystercatcher and a tern species, as the specific advantages and disadvantages complement each other.

Generally, the spatial pattern of sites where the EcoQO was fulfilled or, respectively, not fulfilled, identifies the large rivers as main input sources of several pollutants. The actual values indicate that, in case of a concentration decrease, the EcoQO will probably be fulfilled first for most chemicals in the northern and northeastern part of the study area. The findings of the study confirm the suitability of the EcoQO for drawing the pollutant patterns of the coastal North Sea in an easily understandable manner.

Role of bird eggs in the CEMP

The network of monitoring is based on many connections between e. g. the OSPAR EcoQO approach, the relevant EU Directives and the Trilateral Wadden Sea Cooperation. Presently, the definition of specific objectives and reference values, as well as several monitoring programs are under development. Therefore, there are opportunities to tune these activities across the countries in order to address both the requirements under the various EU Directives and the objectives of OSPAR and the Trilateral Wadden Sea Cooperation. The opportunity arises to harmonize the definition of a good environmental state with the EcoQO targets. With the pollution data collected within the TMAP program, a time series of several decades exists already for a substantial part of the study area. The distribution of the study species also allows sampling in the Northeast Atlantic and the Baltic Sea. The inclusion of UK monitoring data has confirmed the suitability of the current EcoQO target values, and the inclusion of further areas into the EcoQO program for bird eggs is not expected to question the currently determined threshold values. The study design could be easily adapted to the current CEMP design. In consequence, this pilot study confirms that pollutants measured in bird eggs fulfill the prerequisites for an inclusion into a general Coordinated Environmental Monitoring Program (CEMP).

Recommendations for conception and monitoring

To guarantee a sufficient statistical power to detect changes of the pollutant levels in the environment and to be able to react also on short-time events, we recommend maintaining the hitherto conducted annual sampling scheme and to treat each egg as an individual sample.

To avoid a difficult and costly intercalibration of values and a potentially doubtful comparability of results, we recommend to analyze each substance group completely in the same lab and to sub-divide egg samples according to the number of labs involved. The participating labs need to work under recognized standards of quality assurance. We present a provisional budget to finance this EcoQO monitoring around the North Sea.

We propose the inclusion of further regions into the EcoQO concept. Among the countries surrounding the North Sea, these are in particular further sites in UK but also a site at the Rhine delta. The inclusion of further European regions within the OSPAR region and beyond is highly recommended. On a larger spatial scale, it is desirable to include the Baltic Sea into the EcoQO program.

In addition to new areas, the emergence of a variety of new environmental pollutants may require the monitoring of these in addition to the actual substances or, in case of a disappearance of the latter, instead of these. Of major importance are polybromates and new persistent organic pollutants (POPs) such as the dioxins Polychlorinated dibenzodioxins (PCDD) and polychlorinated dibenzofurans (PCDF) as well as perfluorinated compounds (PFCs). These substances have been determined as part of the pre-CEMP and tools for quality assurance procedures and assessment criteria. Their inclusion into the coordinated monitoring is dependent on resolving the status of the EcoQO.

Contents

1 Introduction

1.1 General aims of this report

Following recommendations by OSPAR to formulate ecological quality objectives (EcoQOs) for pollutants in coastal bird eggs, a pilot study has been initiated, based on the experiences of an international monitoring program running since the 1980ies in the Wadden Sea (TMAP program; Becker & Dittmann 2009). In this pilot study, data have been analyzed which were collected between 2008 and 2010 at 21 sites around the North Sea, thus covering an area of an up to now unique spatial extension within the OSPAR area. The superior aims of this report are to present the results of the pilot study and to develop the basics for a future continuation of the EcoQO monitoring.

1.2 The development of EcoQOs by the OSPAR commission

The Oslo and Paris (OSPAR) commissions are responsible for the monitoring, assessment and regulation of pollution in the Northeast Atlantic and the North Sea (Stagg 1998). Established in 1992, the OSPAR convention is committed to prevent and to eliminate pollution of the marine environment (from dumping, land-based and offshore sources) and to conduct quality assessments of the marine environment (Stagg 1998, Hagger et al. 2006). Under the framework of the Joint Assessment and Monitoring Program (JAMP) an upgrade of the Coordinated Environmental Monitoring Program (CEMP) is currently under development and will incorporate various chemical parameters. Current environmental health is evaluated using Ecological Quality Objectives (EcoQOs). These have been formulated in recent years by experts (OSPAR 2009a, b, c). EcoQOs are specific targets defined for different ecological quality elements and considered as the status of e. g. plankton, benthos, fish, birds and marine mammals that approximates the expected status without or after a complete stop of any further input of anthropogenic pollutants. EcoQOs formulated also include parameters describing the state of the marine ecosystem more holistically such as species community composition, population health, nutrient concentrations and oxygen consumption.

The EcoQOs have been developed as tools to help OSPAR and the North Sea Conference process to fulfill their commitments to manage human activities that may affect the marine ecosystem. They are intended to represent clear environmental indicators within the concept of a "healthy and sustainable marine ecosystem" for present and future generations stating aspirations for a healthy North Sea as part of an ecosystem approach. Thus, a description has been developed of what constitutes a good EcoQO. It needs to have a clear scientific basis, to enable data to be collected effectively and economically, to have a clear reference level or target, and to be generally accepted by all stakeholders (OSPAR 2006).

The definition of corresponding aims with those developed in the framework of the Marine Strategy Framework Directive (MSFD) could open the chance to use synergisms between both approaches and to conduct the monitoring on an even larger spatial scale.

1.3 EcoQOs for contaminants in coastal bird eggs

One aspect of major importance is the contamination of the marine environment with anthropogenic heavy metals and organochlorines which may affect the marine life in a variety of ways (Lozán & Kausch 1996, Lozán et al. 2003). The eggs of coastal breeding

birds have proven to be a favorable matrix for monitoring such pollution. As top predators, coastal birds effectively bioaccumulate many contaminants in their eggs. This enables the analysis of substances even if their absolute concentrations in water, sediment or soil are very low (e. g. Furness 1993, Becker 2003, Becker et al. 2003). In contrast to contaminant levels measured in sediment and water, bird eggs represent the actual bio-uptake of contaminants. Contaminant levels in bird eggs also have relatively low variability (compared with concentrations in abiotic and other biotic matrices), partly because eggs can be clearly assigned to date and site of laying. This low variability confers a high statistical power to detect inter-site and inter-year variation in contamination (OSPAR 2007b). Supported by the fact that the ecology of most bird species in question is well studied, the results from analyses of bird egg contamination provide a high information density. Furthermore, compared to other sampling methods such as ship-based sampling, collecting bird eggs requires only a little logistical effort, making sampling cost-effective. The removal of eggs from a population of breeding birds is less damaging than that of adults, having only a minor impact on the breeding success of the studied population. Table 1 gives an overview on how the contaminant levels in coastal bird eggs fulfill the above mentioned ICES criteria for the suitability as an EcoQO.

Thus, mercury (Hg) and organochlorines in coastal bird eggs have been among the 10 issues considered when developing the EcoQO-system for the North Sea (OSPAR 2009b). In 2005, following advice from ICES (2003, 2004), OSPAR agreed on the EcoQOs for Hg and organochlorines, derived from a trilateral monitoring of their concentrations in the Wadden Sea which has been conducted for several decades (TMAP program; Becker et al. 1991, 1998, 2001, Becker & Muñoz Cifuentes 2004, Becker & Dittmann 2009).

1.4 Indicator species and preliminary target values

As indicator species, Eurasian Oystercatcher *Haematopus ostralegus* and the Common Tern *Sterna hirundo* have been selected. OSPAR (2007a, 2009c) formulated as EcoQO:

That the average concentrations of Hg in the fresh mass of ten eggs from separate clutches of Common Tern and Oystercatcher breeding adjacent to the estuaries of the Rivers Elbe, Weser, Ems, Rhine/Scheldt, Thames, Humber, Tees, and Forth, should not significantly exceed concentrations in the fresh mass of ten eggs from separate clutches of the same species breeding in similar (but not industrial) habitats in south-western Norway and in the Moray Firth (Scotland). So the target concentration should approximate as closely as possible the pristine state of the environmental concentration of this element which is also occurring naturally in low concentrations. The inclusion of new sampling sites in the hardly industrially affected areas mentioned above offered the opportunity to refine the provisional proposal for an EcoQO metric for Hg which was set at 170 ng/g fresh weight (FW) in Common Tern and 100 ng/g FW in Oystercatcher eggs, based on the minimum concentrations measured in earlier years (OSPAR 2009c). In particular, this means that the EcoQO metric for Hg should be lowered if Hg concentrations in the Norway and/or the Moray Firth fall below 170 or, respectively, 100 ng/g fresh egg mass.

For the concentration of organochlorines, specific maximum target values have been proposed (OSPAR 2007a, 2009c). Although for these exclusively anthropogenic substances, concentrations of zero would be desirable in the environment, values above zero have been proposed as EcoQO based on the specific detection limits and on the assumption which targets can be realistically achieved during the next decades, considering e. g. the long half-value periods for DDT derivates. These are 20 ng/g for the sum of 62 PCB-congeners which have been analysed during a Wadden Sea monitoring since the 1980s (∑PCB; Becker & Dittmann 2009), 10 ng/g for the sum of six forms of DDT-derivates, i. e. p,p´-DDT, o,p´-

DDT, p,p´-DDD, o,p'-DDD, p,p'-DDE and o,p'-DDE (=∑DDT), and 2 ng/g for HCB and the sum of α-, β- and γ-isomers of HCH (=∑HCH) in the Oystercatcher and Common Terns. Ten eggs of each species should be sampled from separate clutches of birds breeding adjacent to the estuaries of the Rivers Elbe, Weser, Ems, Rhine/Scheldt, Thames, Humber, Tees, and Forth, and in similar (but not industrial) habitats in south-western Norway and in the Moray Firth.

1.5 Pilot study 2008-2010

Whereas several detailed monitoring programs on bird egg contamination have been running for decades in different species in different countries of the OSPAR area (see Table 2 and Chapter 5), a coordinated monitoring program in the whole OSPAR region has been lacking until now. Based on the data and financial support of the TMAP program mentioned above, of the Norwegian Directorate for Nature Management, Norway, of the Centre of Ecology & Hydrology (CEH), Lancaster, United Kingdom, Institute for Nature and Forest (INBO), Belgium, and of the Swedish Environmental Protection Agency, Sweden, this pilot study has been conducted between 2008 and 2010 to measure the contamination of coastal bird eggs with Hg and organochlorines in seven countries surrounding the North Sea and to compare these values with the EcoQO targets formulated. The core of the spatial range was the Wadden Sea area covered by the TMAP, supplemented by in total six additional sites at the Scheldt estuary, in eastern UK as well as three reference areas in Sweden and Norway. The countries concerned were Great Britain, Belgium, The Netherlands, Germany, Denmark, Sweden and Norway with in total 21 sampling sites which were selected according to the following criteria:

- to address hot spots of anthropogenic contamination, especially the estuaries (Marine Strategy Directive)
- to include sites with an expected lower degree of contamination as reference sites
- to include Important Bird Areas such as the German Bight, which are in the focus of the EU Birds and Habitat Directives
- to consider logistics of sampling (number of breeding pairs available for sampling per site, also in the future prospect)
- to select an appropriate number of monitoring stations along the North Sea coast to assess the EcoQO on a larger scale.

This approach has been faced with difficulties to sample sufficient numbers of Oystercatcher and Common Tern eggs at all sites. Therefore, for sites where the Common Tern does not occur in sufficient numbers a closely related and ecologically similar species, the Arctic Tern *Sterna paradisaea*, has been proposed as alternative indicator species.

The pilot study has been promoted, prepared and attended by a group of experts from the participating countries surrounding the North Sea under the lead of the CWSS, Germany. This group met in Hull, UK (November 2007) and in Bremen, Germany (February 2009; March 2011). During the last meeting beyond other points the group discussed the critical aspects of coastal bird egg monitoring and the EcoQO on Hg and organochlorines in coastal bird eggs before the background of a potential inclusion into the CEMP. Participants also agreed on preparing a comprehensive report about the pilot study of the EcoQO.

Table 1: ICES criteria on the suitability of a parameter as an EcoQO and comments how they are fulfilled in coastal bird eggs (according to OSPAR 2007a)

ICES criteria	Evaluating comments
Relatively easy to understand by non-scientists and those who will decide on their use.	There is a clear link between the anthropogenic input of mercury and organochlorines into the environment and the concentration of these substances in bird eggs. Their level in bird eggs provides an indication of their level and trends in the ecosystem. Common Tern, Arctic Tern and Eurasian Oystercatcher are coastal birds which are well known to the public.
Sensitive to a manageable human activity	Most of these substances enter the ecosystem entirely through human activities, which can be controlled by management intervention.
Relatively tightly linked in time to that activity	Bioaccumulation and persistence in ecosystems mean that some linkage will occur, but not always. Mercury and organochlorines in the environment are very persistent, and tend to increase up food chains. Because of this persistence, a time lag would exist between applying management measures and the response in seabird eggs.
Easily and accurately measured, with a low error rate	Eggs are readily available and the analytical methods are well established. The ability to integrate pollutant signals over time and space of bioaccumulating contaminants in tissues means that to obtain a given level of accurate measurements, a smaller number of animal samples is required than of physical samples thus increasing the power of trend analyses.
Responsive primarily to a human activity, with low responsiveness to other causes of change	Fully responsive to human activity. However, due to the persistence of many of these compounds, it will take many years before they disappear from the environment.
Measurable over a large proportion of the area to which the EcoQO metric is to apply	Common Tern and Eurasian Oystercatcher are abundant and widely distributed throughout the North Sea area. Alternatively, eggs of Arctic Tern can by analyzed instead of Common Tern. As these species occur also on coasts of the west Atlantic and comparable species even on the coasts of other oceans there is potential to expand the EcoQO to other seas of the world.
Based on an existing body or time-series of data to allow a realistic setting of objectives	The combination of long time series of data for the Wadden Sea (since 1980's) and the current pilot project (2008-2010) confirm the existing EcoQO metrics and values.

Consequently, the aims of this report are to present and to discuss the results of the pilot study conducted with focus on:

- to suggest an actualized threshold concentration for Hg for which only preliminary target values of 100 ng/g in the Oystercatcher and 200 ng/g in the Common Tern had been proposed
- a test of the suitability of the Arctic Tern as a potential alternative species to replace the Common Tern at sites where the latter does not occur in sufficient numbers
- the presentation of spatial contamination patterns and temporal developments
- to address whether, where and when the hitherto existing EcoQO objectives are fulfilled
- to give recommendations for monitoring, assessment and management of this EcoQO in the future
- to mention the advantages of species and matrix as well as the limitations of the application of this EcoQO
- to discuss how the preconditions for a potential inclusion of the coastal bird egg monitoring of pollutants into the CEMP can be fulfilled (see above)
- to give suggestions for combining the EcoQO program with other existing or planned monitoring programs for the marine environment

2 Methods

2.1 The Environmental Chemicals Under Study

Environmentally adverse chemicals are substances of toxicological relevance for organisms which are emitted by man into the environment but (e. g. in case of some heavy metals) may also occur naturally. Among the chemicals studied and addressed by this EcoQO is the heavy metal Hg, environmental concentrations of which result largely from anthropogenic inputs although there are low baseline concentrations as this element is a naturally-occurring micropollutant. The other analyzed chemicals are xenobiotics, i.e., they are man-made and exclusively introduced into the environment by human activities (Koch 1991). In this report, we classify the analyzed substances in industrial chemicals (Hg, ∑PCB and HCB) and pesticides (ΣDDT, ΣHCH).

Hg

Hg is used by man in many products (thermometers, barometers, energy-saving compact fluorescent light-bulbs etc.; during former decades, it was also used in button-cell batteries and as a fungicide on seeds) and as a catalyst in many industrial processes (paper manufacturing, production of vinyl chloride, urethane foam, etc). Being an element, Hg that is released into the environment will remain there indefinitely. In addition, Hg occurs naturally in the environment ("background values"; Koch 1991, Haarich 1994, Schlüter 2000). Hg is transformed into a very toxic form (methylmercury) by bacteria and chemical processes. In its organic form, this heavy metal is readily bio-accumulated through the food-chain. For this reason, relatively low levels of Hg in aquatic ecosystems can lead to toxic contamination in organisms with a high position within the food-chain (e.g. predators such as coastal birds). In man, a daily oral intake of 4 ng/g is assumed to be toxic. In birds, Hg is enriched in growing feathers and eggs (e.g. Furness 1993, Gochfeld 1997) and threshold oral intakes and tissue and egg concentrations associated with adverse effects on reproduction and survival have been proposed (Shore et al. 2011).

PCBs

Polychlorinated biphenyls (PCBs) are industrial products or by-products formed in industrial processes (Holoubek et al. 1994, Kočan et al. 1994, 1996), and are composed of 209 individual congeners with varying levels of toxicity. Because of their physical-chemical properties (inert and lipophilic), PCBs were widely applied in industry. The excellent properties of PCBs for industrial use also make them hazardous to the environment. PCBs are highly persistent to metabolic breakdown, promoting their accumulation in the food-chain. The toxicity of the PCBs depends on two factors: the chlorination degree (the toxicity increases with rising chlorine number) and the number of substituting chlorine atoms in ortho-positions where, the smaller their number, the greater the toxicity of the congener (e.g. Parkinson & Safe 1987). Therefore, the coplanar congeners "nonortho PCBs" (without substitution in ortho-position of the phenyl ring) have a higher toxicity than mono-ortho PCBs (one chlorine atom in orthoposition) or di-ortho-PCBs (two chlorine atoms in ortho-position), because of a completely flat (planar) conformation which is similar to that of dioxins. The coplanar congeners strongly induce activity of cytochrome-P-450s and have similar effects to 2,3,7,8-TCDD (tetrachloro-dibenzo-p-dioxin). A propeller-like conformation of orthochlorines prevents the planar conformation to a varying degree, and therefore ortho-

Table 2: Overview of the actually existing monitoring programs for measuring pollutants in seabird eggs that are conducted by Contracting Parties (OSPAR 2007b)

Country	Program type	No. of stations	Frequency	Since (year)	Species	Analyzed Parameters*
	Monitoring					
DK	Monitoring TMAP	2	annual	1998	Common Tern, Oystercatcher	Hg, 62 PCB, HCB, DDTs, HCHs, Chlordanes
D	Monitoring TMAP	7	annual	1981/86	Common Tern, Oystercatcher	Hg, 62 PCB, HCB, DDTs, HCHs, Chlordanes
D	Specimen Banking	3	annual	1988	Herring Gull	Hg, As, Se, Tl, Cu, Pb, 7 PCBs, DDTs, HCHs, HCB, OCS, Dieldrin, PeCBZ
NL	Monitoring TMAP	4	annual	1993/97	Common Tern, Oystercatcher	Hg, 62 PCB, HCB, DDTs, HCHs, Chlordanes
S	Specimen Banking	1	annual	1969	Guillemot	Hg, Pb, Cd, Ni, Cr, Cu and Zn; 7 PCBs, HCB, HCHs, DDTs, PFCs, dioxins and furans, PBDEs, HBCD
UK	Predatory Bird Monitoring Scheme	1	biennial	1973	Gannet	Hg, 35 PCBs, HEOD, DDTs, HCHs, HCB
	Research					
NL	Research project RIKZ	1	-		Common Tern	PCBs, PBDEs, HBCD, PFOS dioxins, furans, PAHs, PFOA, others

*In case of specimen banking, only parameters are listed which are analyzed continuously (see text).

congeners are less dioxin-like than non-ortho-congeners. Chlorines in ortho-positions are poorly biodegraded, and consequently are present in the environment at higher concentrations than non-ortho congeners (Fiedler & Lau 1998). The production and use of PCBs was banned in western Europe during the 1980s, but PCBs present in closed systems, e.g. transformers, condensers, can still be released into the environment.

HCB

Hexachlorobenzene (HCB) is a chlorinated aromatic hydrocarbon with moderate volatility. It is highly lipid-soluble and bioaccumulative and is a byproduct in the production of chlorine gas and chlorinated compounds, including several pesticides and solvents. It is emitted to the atmosphere in the flue gas from waste incineration, and is also formed in metallurgical processes. It had a limited use as a fungicide in the past. HCB was banned in the Netherlands, Germany and Denmark during the 1980s. HCB enters the environment for example as a contaminant of other chemical products, for example from the reduction of PCBs, as a metabolite of Lindane or during the production of pentachlorophenol (Becker et al. 1991) or is present in historical sediment-bound waste deposits. HCB is a long-range transport POP (persistent organic pollutant), listed under the Stockholm Convention, and as such is globally circulated by the atmosphere and can be deposited in remote areas, such as Arctic latitudes (Bakker et al. 2009).

DDT

The insecticide p,p'-DDT is probably the best known pesticide because of its well documented toxic effects on certain biota (Koch 1991). DDT is a mixture of six forms, p,p´-DDT, o,p´-DDT, p,p´-DDD, o,p'-DDD, p,p'-DDE and o,p'-DDE. The main metabolite of DDT is p,p´-DDE, which is associated with shell thinning in bird eggs (see reviews, e.g. Moriarty et al. 1986, Furness 1993). This pesticide and its metabolites are relatively stable under most environmental conditions and are resistant to complete breakdown by the enzymes present in soil micro-organisms and higher organisms. DDT and its metabolites are very soluble in lipids and organic solvents. DDT was banned in western Europe during the 1970s but was still used in several countries of eastern Europe during the 1980s and is still used in some African countries to combat malaria. Similarly to HCB, DDT is also a long-range transport POP, listed under the Stockholm Convention, and is also transported to high latitudes via the atmosphere (Bakker et al. 2009).

HCH

The technical mixture of hexachlorcyclohexane (HCHs) was banned in western Europe during the 1980s, but the gamma isomer (γ-HCH), known as lindane, is still in use as insecticide. Lindane is a chlorinated hydrocarbon with a relatively long residual activity, is transported over large distances by the atmosphere, and is listed under the Stockholm Convention (Bakker et al. 2009). Due to the long-lasting systematic application, the hexachlorocyclohexanes are widespread in the environment and will remain in soils for some decades. The β-isomer, present in the lindane technical mixture, is considered a greater environmental problem than the γ-isomer.

2.2 Study species

Eggs of Common Tern *Sterna hirundo* and Eurasian Oystercatcher *Haematopus ostralegus* have been selected for the definition of EcoQOs concerning their contamination with Hg and organochlorines. These species are widespread and common and breed in coastal areas of Europe (Goss-Custard 1996, Becker & Ludwigs 2004). However, due to difficulties in sampling sufficient numbers of Common Tern eggs at some sites, eggs from both the Common Tern and a closely related and ecologically similar tern species, the Arctic Tern *Sterna paradisaea*, were sampled at Hallig Hooge, Germany, to compare the pollutant levels and to check the suitability of the Arctic Tern as an alternative study species to the Common Tern in areas where the latter is rare.

Both tern species are considered income breeders, i. e. substances forming the eggs do largely originate from nutrients incorporated by the female in the two weeks of courtship feeding by the male mate immediately before egg-laying (Wendeln & Becker 1996, Wendeln 1997). In the breeding season, foraging of Common Terns takes place in comparatively small distances mostly within 10 km of the breeding colony (Becker et al. 1993), in Arctic Terns, most feeding takes place within 3 km of the breeding colony (Cramp 1985) characterizing both tern species as inshore feeders. Both species feed mainly on small fish and crustaceans taken by plunge-diving and are considered top-predators in the marine food-chain. The terns are long-distant migrants: Common Terns are wintering in west/southwest Africa, Arctic Terns in the Antarctic (Cramp 1985, Becker & Ludwigs 2004).

Compared with the terns, the Oystercatcher is more a capital breeder, producing eggs also from substances stored in the body over longer time periods. The species is a resident breeder over large parts of the North Sea area (Koffijberg et al. 2006). It is feeding on macrozoobenthic organisms such as mussels and worms, which makes it another favorable model species and may have a slightly smaller feeding range that is mostly less than 5 km from the breeding site (Cramp et al. 1983; Exo 1992).

The extensive knowledge of the ecology of these selected indicator species, their large populations, wide geographical distribution of breeding sites, high trophic position in marine food chains and capacity to accumulate persistent contaminants make them especially suitable monitors of contamination of the local marine environment with environmental pollutants and to be included in the EcoQO concept

2.3 Sampling sites

To study spatial patterns in coastal bird pollution, in total 21 coastal sampling sites have been chosen in seven countries surrounding the North Sea. According to the requirements of OSPAR (2007a, 2009c) site choice aimed to include the estuaries of large rivers draining industrial areas as potential sources for several environmental pollutants but also sites which were hardly affected by industrialization. However, in contrast to the initial aims to cover five sites in GB, eggs were sampled at only one site (Middlesbrough) due to logistic reasons.

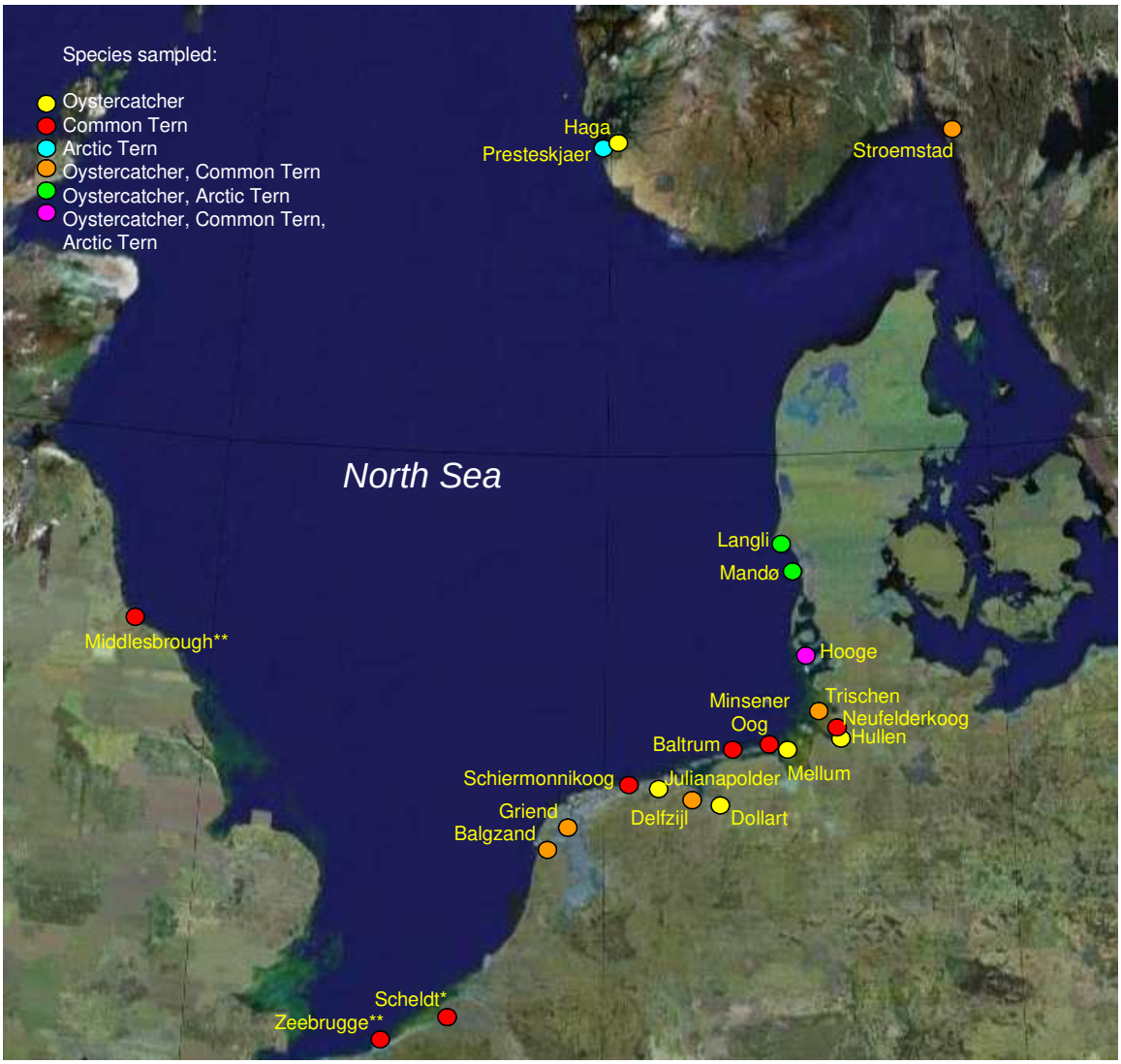

Fig. 1: Sampling sites of Oystercatcher, Common Tern and Arctic Tern eggs between 2008 and 2010. Colour of the dots indicates sampled species (see legend). At the sites marked with *, eggs were sampled only in one year, ** eggs were sampled only in two years. For more details on the sampling sites see Table A.1. Map source: Google Earth.

Actually, the sampling sites were from the western to the northeastern part of the North Sea as follows: Middlesbrough (UK), Zeebrugge (B), Terneuzen, Balgzand, Griend, Julianapolder, Delfzijl (NL), Dollart, Baltrum, Minsener Oog, Mellum, Hullen, Neufelderkoog, Trischen, Hallig Hooge (D), Langli, Mandoe (DK), Stroemstad (S), Haga (N) and Presteskjaer (N; Fig. 1). For an overview, which species was sampled at which sites and which sites are

The sites from Balgzand to Langli have been subject of the Trilateral Monitoring Program for pollutants in the Wadden Sea conducted since 1998 (TMAP; Becker & Muñoz Cifuentes 2004, Becker & Dittmann 2009). Historical data of contaminant concentrations in coastal bird eggs from the German Wadden Sea are available from 1981 and 1985 to 1997, previous to the start of the TMAP (see above; Becker et al. 1985, 1998, 2001).

2.4 Collection of Egg Samples

Eggs were sampled according to the guidelines of JAMP (OSPAR 1997) and VDI (2009). Ten fresh eggs per species, site and year were taken under license. Since in general, intraclutch variation is low compared to interclutch variation, one egg per clutch was chosen randomly (e.g. Becker et al. 1991). Because egg levels reflect the contamination of the egg-laying female (Becker et al. 1989, Lewis et al. 1993), the ten eggs collected per site and species indicate the current contamination of ten females breeding at the respective site and year. In contrast, in the breeding season, males - which are unable to excrete pollutants into eggs - exhibit higher concentrations of Hg – and probably also other environmental pollutants - in their primary feathers which are moulted at the end of the breeding period (Lewis et al. 1993). The eggs were kept frozen at -18 °C until they were analysed. Total egg weight (to the nearest 0.1 g), length, and width (0.01 mm) were recorded. The eggshells were air-dried and weighed (0.01 g) and the shell thickness was measured with a micrometer (0.01 mm). The egg's content was homogenized using an Ultra-Turrax, filled into suitable polypropylene cups, and frozen at -18 °C until chemical analysis.

2.5 Chemical Analyses

All egg samples from continental Europe were analysed in one lab, the ICBM-Terramare Wilhelmshaven. The lab participated in an intercalibration with two other labs, and regularly in an international quality assurance (QUASIMEME project), whose results were ranked as satisfactory in most analyses. Besides the heavy metal Hg, 62 PCBs and further organochlorine substances (see below) were determined (Becker et al. 1991, 1998). Most of the PCBs were baseline separated during the gaschromatographic separation, but 21 PCBs coeluted in nine peaks. The selection of the 62 PCB congeners (abbreviated to ∑PCB in the following text) was made due to their concentration in coastal bird eggs and their toxicology. The further organochlorine substances analysed were Hexachlorobenzene (HCB), the insecticide p,p'-DDT (dichlorodiphenyltrichloroethane), the metabolites p,p'-DDD (dichlorodiphenyldichlorethane), and p,p'-DDE (dichlorodiphenyldichlorethene; ∑DDT = sum of all metabolites), as well as the alpha-, beta- and gamma-isomers of hexachlorocyclohexane (∑HCH). The methods were in agreement with the OSPAR guidelines (OSPAR 1997). Sample preparation coincided with the method used by the Chemical Institute of School and Veterinary Medicine, Hanover (Heidmann 1986). For further details of the chemical analysis see Becker et al. (2001) and Appendix A.1.

The egg samples from Middlesbrough were analysed at the Centre of Ecology & Hydrology, Lancaster, UK. Besides Hg, this lab analysed HCB, a total of 37 PCB congeners (of which 26 congeners were also analysed at Wilhelmshaven), p,p'-DDT, p,p'-DDD, p,p'-DDE as well as α- and β-HCH. In addition, HEOD (dieldrin) concentration was determined but HEOD concentrations are not presented here due to a lack of EcoQO for this substance. For details of the chemical analyses see Appendix A1, and Pereira et al. (2009). For a comparison of the methodological approaches performed at Wilhelmshaven, and, respectively, Lancaster, see Appendix A.1 and Table A.3. An overview which chemicals were analyzed in Wilhelmshaven and, respectively, Lancaster is given in Table 3, all PCB congeners analyzed are listed in Table A.5. To be able to compare the pollutant levels measured in UK to those in continental Europe and to the EcoQOs defined, an intercalibration of both methodological approaches was conducted. Substance group specific calibration and conversion factors were calculated for pollutant levels measured in UK where 20 of the 720 eggs had been analysed. Due to the low proportion of the UK eggs compared to the total sum of eggs collected, because the EcoQO was defined on the basis of eggs analysed on the mainland, and because the EcoQOs

for the summarized values for several substance groups based on specific sets of components analyzed in Wilhelmshaven, we multiplied the values measured in UK (and not those from the mainland) with specific factors to achieve comparability despite different labs and, partly different, but overlapping composition of congeners analyzed. For details see Appendix.

According to the intercalibration, the concentrations of eggs from Middlesbrough were multiplied by specific calibration factors of 1.0 (Hg), 1.21 (ΣPCB, sum of 26 congeners; calibration factor based on 23 congeners analysed in both labs), 0.696 (HCB), 0.789 (ΣDDT), and, respectively 1.0 (γ-HCH). Mean contaminant concentrations of identical samples analyzed in both Lancaster and Wilhelmshaven are shown in Table A.4. To be able to compare the summarized PCB-levels of British eggs with those on the mainland and with the EcoQO determined for the continent, the summarized PCB-levels of British eggs were multiplied by a conversion factor of 1.29, accounting for the fact that a lower number of PCB congeners was analyzed in UK than in continental Europe and allowing a comparison to the 62 PCB compounds for which the EcoQO has been defined in continental Europe. However, predominant PCB congeners were identical. The concentrations of chemicals measured are always given in ng/g fresh weight of egg content in this report.

Table 3: Overview over environmental pollutants analyzed in Lancaster and/or in Wilhelmshaven. For specification of PCB-congeners see Table A.5.

Contaminant	Lancaster	Wilhelmshaven
Hg	X	X
∑PCB, 62 congeners		X
∑PCB, 37 congeners	X	
∑PCB, 26 congeners	X	X
HCB	X	X
ppDDT	X	X
ppDDD(=TDE)	X	X
ppDDE	X	X
α-HCH	X	X
β-HCH		X
γ-HCH	X	X

2.6 Statistical Methods

Contaminant values were log-transformed (log n + 1) to achieve homogeneity of variances and normal distribution. A GLM model was used to analyse effects of the main factors species (Oystercatcher or tern, Common and Arctic Tern eggs pooled), site and year (2008-2010). Year effects at specific sites were tested for with ANOVA. For interyear-comparisons, Scheffé tests and, in case of only two comparable years, t-tests were done. Results were considered as significant at p-values < 0.05 (*), < 0.01 (**, highly significant), and < 0.001 (***, very highly significant). All tests were two-tailed. The statistics were performed by SPSS 18.0 for Windows.

When reporting temporal changes in the results, results from the three (or, depending on the study site, two) study years are presented. Year effects at specific sites were tested for with ANOVA and Scheffé tests or, when there were only two comparable years, t-tests were done. It is not really possible to detect a consistent time trend from pilot data based on at most three years of data. However, based on the inter-year variation that we observed, we considered that there may be an increase in contamination over the study period 2008-2010 if pollutant concentrations were significantly higher in at least one later year compared to an earlier year. A decrease was complementarily defined as the case that pollutant concentrations were significantly lower in at least one later year compared to an earlier year. If no significant differences were recorded between years or if both an increase and a decrease were observed during the three study years, it was considered that there was no upward or downward temporal trend.

3 Results

3.1 The Arctic Tern as a possible alternative for the Common Tern as indicator species

Towards the north of the study area, the Common Tern becomes increasingly rare and is replaced by the related Arctic Tern in similar coastal habitats (BirdLife International 2004). Where both species occur syntopically, they often breed in mixed colonies although the Arctic Tern may prefer slightly lower vegetation at the nest site (Cramp 1985, Grave 2010, Heckroth 2010). Analysis of eggs of both tern species at one site (Hallig Hooge, Germany) from 2008 (Chapter A.2, Table A.6) revealed no differences between species for Hg, ∑PCB and HCB concentrations in eggs. Concentrations of ∑DDT and ∑HCH were slightly higher in Arctic Tern than in Common Tern eggs. However, the difference between species in ∑DDT concentration was small and absolute values were in the same order of magnitude. The measured concentration for ∑HCH was close to the overall determination limit (as well as to the EcoQO) and analytical accuracy tends to decrease as the detection limit is approached. Hence, the difference between species in ∑HCH concentrations may have been spurious. Overall therefore, the data indicate that Common Tern and Arctic Tern are effectively mutually replaceable as EcoQO monitors (cf. Chapter A.2, Table A.6).

3.2 The derived EcoQO for Hg

Concerning tern eggs, minimum mean Hg concentrations of 160.1 ng/g were measured in Arctic Tern eggs sampled in 2009 at Presteskjaer, Norway. In Oystercatcher eggs, minimum mean Hg concentrations of 97.4 ng/g were recorded in 2008, again at Presteskjaer. In line with the concept that the EcoQO for Hg should be based on the minimum concentration measured in the study area and that the area includes sites with very low levels of industrialization (see above), a value of 160 ng/g Hg is now proposed as the actual EcoQO for Common and Arctic Tern eggs and 100 ng/g for Oystercatcher eggs (Table 4).

Table 4: Proposed EcoQOs for environmental chemicals in Common/Arctic Tern and Oystercatcher eggs (concentrations in ng/g fresh egg wet weight) in the North Sea (ICES 2004, OSPAR 2007a), and lowest average levels measured at various sites in 2008 - 2010.

Substance	Species	Minimum concentration (ng/g)	Site	**Proposed EcoQO (ng/g)**
Hg	Oystercatcher	97	Haga/Presteskjaer, N	**< 100**
	Arctic Tern	160	Presteskjaer, N	**< 160**
∑ 62 PCB congeners	Oystercatcher	130	Hallig Hooge, D	**< 20**
	Arctic Tern	137	Presteskjaer, N	
HCB	Oystercatcher	0.7	Hallig Hooge, D	**< 2**
	Arctic Tern	1.7	Presteskjaer, N	
DDT and metabolites	Oystercatcher	4.8	Hallig Hooge, D	**< 10**
	Arctic Tern	12.2	Presteskjaer, N	
HCH isomers	Oystercatcher	0.0	several sites	**< 2**
	Common/Arctic Tern	0.0	several sites	

3.3 Influence of species, site and year on contaminant concentration

A statistical analysis of how species, site and year affect contaminant concentrations in eggs was assessed using a generalized linear model. All three factors, and their interactions, significantly affected the concentrations of all five contaminant groups. Comparing the mean contamination of eggs sampled between 2008 and 2010, mean pollutant levels measured in tern eggs were 1.7-times (∑PCB) to 2.6-times (HCB) higher than those in Oystercatcher eggs at the same or adjacent sites (cf. also Fig. 2 and Fig. A.2). Interspecific differences were in particular clear at sites where concentrations in eggs were relatively high. However, for most substance groups, site proved to be the strongest source of variation in concentrations (Table A.7). The effects of each single factor are considered below.

3.4 Spatial patterns in contamination

Despite some inter-annual concentration differences, overall spatial patterns in contaminant concentrations were generally consistent across years for both species. So sites with high and, respectively, low concentrations of pollutants remained generally the same sites throughout the three study years. Variation between years was low, also indicated by the low variation between samples of most sites (Fig. 3). For a comparison of mean concentration values between years, see, for example, the HCB concentrations given in Fig. 2. For completeness, the further substances are given in Fig. A.2 in the Appendix. Although statistically significant, annual variation was relatively low and considered spatial patterns in contamination using the mean values of samples taken across the three study years (Fig. 3).

3.4.1 Common/Arctic Tern

Concentrations of Hg, ∑PCB, HCB and ∑DDT in Common and Arctic Tern eggs were greatest in the Elbe estuary. The next highest ∑PCB concentration was in eggs from Terneuzen and concentrations were also relatively high at Delfzijl and Zeebrugge. The second highest levels of HCB and ∑DDT were in eggs from Middlesbrough, although concentrations were considerably lower than those in eggs from the Elbe estuary. ∑HCH concentrations were by far the highest in eggs from Middlesbrough even though only two isomers (α-HCH and γ-HCH) were measured in those samples. The lowest mean concentrations for four out of the five contaminant groups were in eggs from Presteskjaer (Hg) or Stroemstad (∑PCB, ∑DDT and ∑HCH) while the lowest mean HCB concentration was measured in eggs from Terneuzen (Fig. 3). Overall, there was a clear, continuous decrease in Hg, ∑PCB, HCB and ∑DDT concentrations as distance away from the Elbe estuary increased towards the north, whereas concentrations decreased abruptly towards the west (Fig. 3, Fig 4.2, 4.4, 4.6, 4.8).

3.4.2 Oystercatcher

When comparing the spatial contamination patterns of both species, it should be remembered that no Oystercatcher eggs were collected from the UK or Belgium. Overall, in Oystercatcher eggs, the spatial contamination patterns varied stronger between the different substance groups than in tern eggs. However, the highest mean concentrations of Hg, ∑PCB, ∑DDT and ∑HCH were measured in eggs from a site in the Elbe estuary or in its immediate surroundings (Trischen). The HCB concentration in eggs was also relatively high from the Elbe estuary although it was particularly high at Delfzijl. Concentrations in eggs were also relatively high for Hg at Balgzand and ∑PCB at Delfzijl and Dollart. ∑HCH concentrations in eggs varied markedly but over a small spatial scale. Sites with the lowest contaminant concentrations in eggs were Presteskjaer (Hg), Hallig Hooge (∑PCB and ∑DDT), the Danish Wadden Sea (HCB) and Stroemstad (∑HCH). There was a decrease in Hg, ∑PCB and HCB concentrations with increasing distance from the river Elbe or the island of Trischen (Fig. 3, Fig. 4.1, 4.3, 4.5) but this was less pronounced than in the Common Tern.

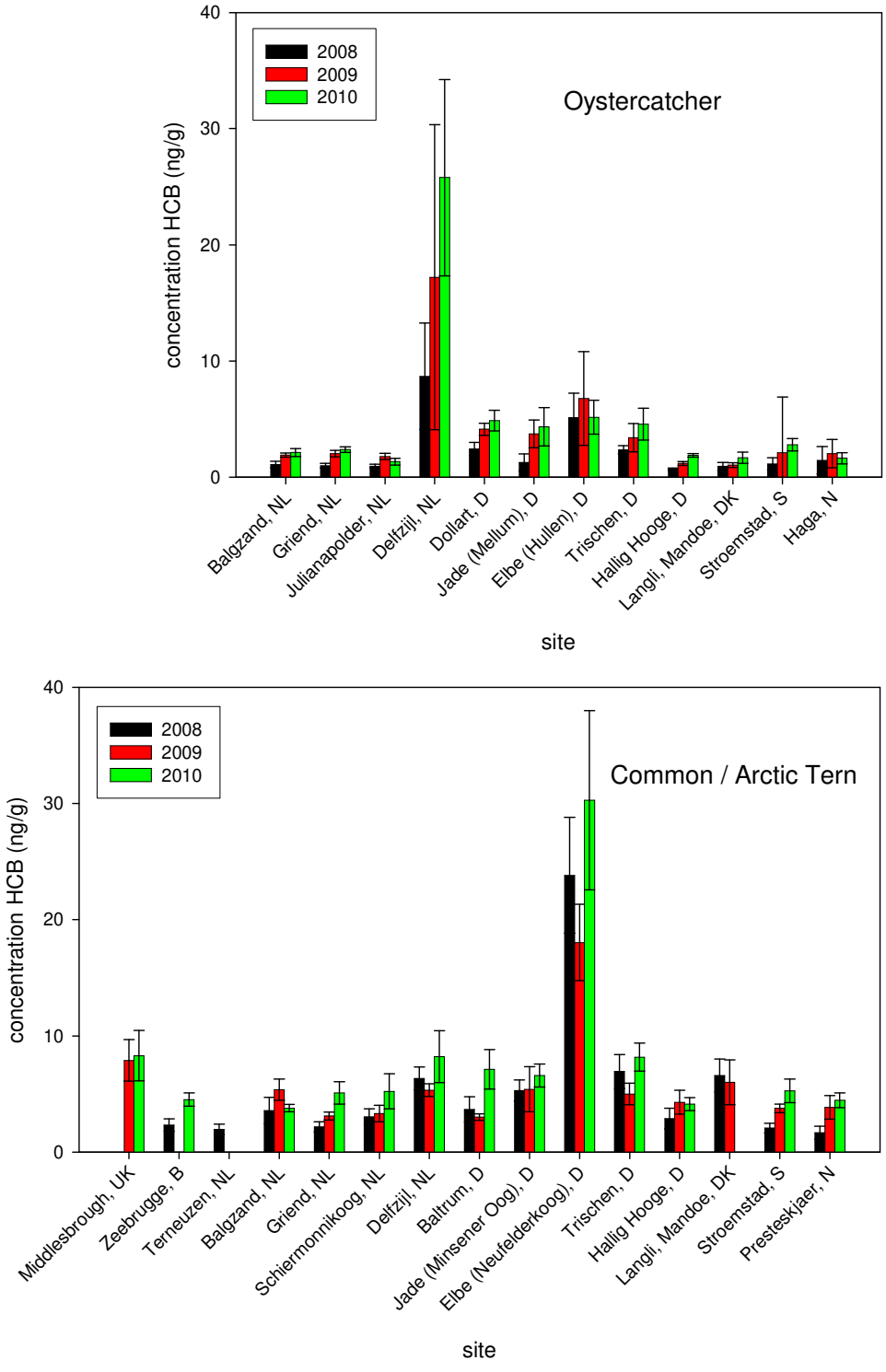

Fig. 2: Mean contamination ± 95%-confidence interval of Oystercatcher and Common Tern with HCB in the studies years 2008-2010 at different sampling sites around the North Sea. For an overview of sample sizes per year, see Table A.2.

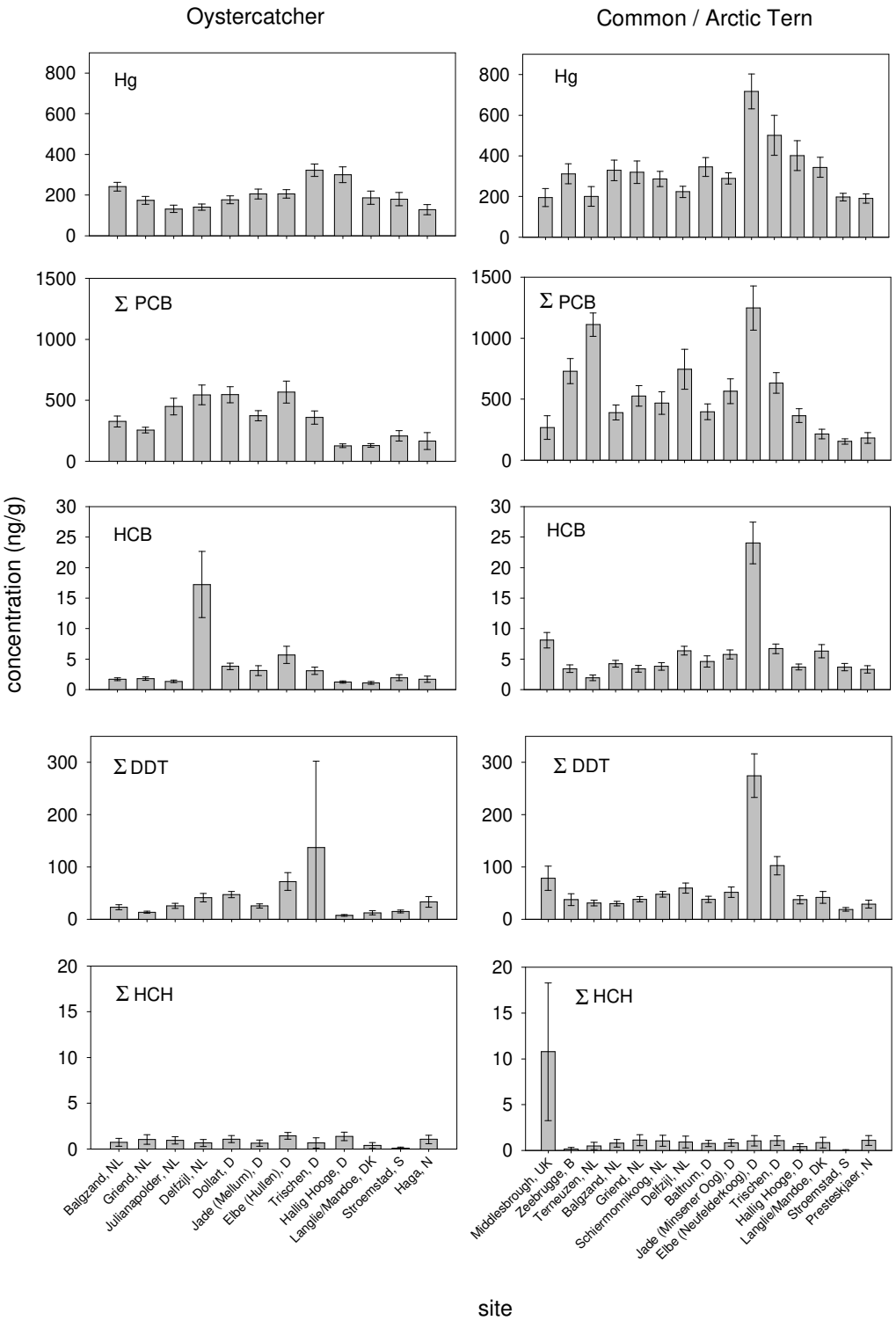

Fig. 3: Mean contamination ± 95%-confidence interval of Oystercatcher and Common Tern with environmental pollutants in the years 2008-2010 at different sampling sites around the North Sea. For an overview of sample sizes per year, see Table A.2. ∑PCB comprises 26 congeners, ∑HCH summarizes α-, β- and γ-HCH, but only α- and γ-HCH at Middlesbrough.

3.5 Temporal differences and trends: 2008-2010

An overview of the temporal differences in pollutant concentrations in the tern species and the Oystercatcher eggs is given in Table 5. In the following, the findings for the different substance groups are described for the three species studied (cf. Fig. 4.1-4.10):

3.5.1 Common/Arctic Tern

Hg concentration was increasing over the three study years at nine of the 14 study sites. These were all Dutch sites, all German sites except Jade, and Presteskjaer. At Middlesbrough, Zeebrugge, Jade, the Danish Wadden Sea and at Stroemstad, no significant changes were detected.

∑PCB was increasing at three sites (Elbe, Stroemstad and Presteskjaer) and decreasing at three of four Dutch sites (Balgzand, Schiermonnikoog and Delfzijl) and at the Jade. At Griend and Trischen, both positive and negative changes were detected. At the other sites (Middlesbrough, Zeebrugge, Baltrum, Hallig Hooge and the Danish Wadden Sea) no significant changes were recorded.

HCB was increasing at ten of the 14 study sites. These were all Dutch sites, all German sites except Jade, Stroemstad and Presteskjaer. At the remaining sites, no significant changes could be recorded.

∑DDT was increasing at the three German insular sites (Baltrum, Trischen, Hallig Hooge) as well as at Stroemstad and Presteskjaer. Decreases were observed only in The Netherlands at Schiermonnikoog and Delfzijl. At the remaining sites, no significant changes were recorded.

∑HCH was increasing at all four Dutch sites, at three German sites (Baltrum, Elbe and Trischen), in the Danish Wadden Sea and at Presteskjaer. The only decrease was observed at Jade. At the remaining sites, no changes were detected.

3.5.2 Oystercatcher

Hg concentrations were increasing only at two sites in Germany (Dollart and Hallig Hooge). Decreases were recorded at Griend, Elbe, Trischen and Stroemstad. At the further sites, no significant changes were measured.

∑PCB was increasing at Elbe and decreasing at Hallig Hooge. At Dollart, both positive and negative changes were measured. At the other sites, no significant changes were found.

HCB was increasing at seven of 11 sites. At Julianapolder, both positive and negative changes were recorded. At the other sites, no changes were detected.

∑DDT was increasing at Jade, Elbe and Hallig Hooge. At Balgzand and Julianapolder, both positive and negative changes were found. At the further sites, no changes were found.

∑HCH was increasing at eight of 11 sites. At Dollart, both positive and negative changes were found. Only at Stroemstad, no changes were detected.

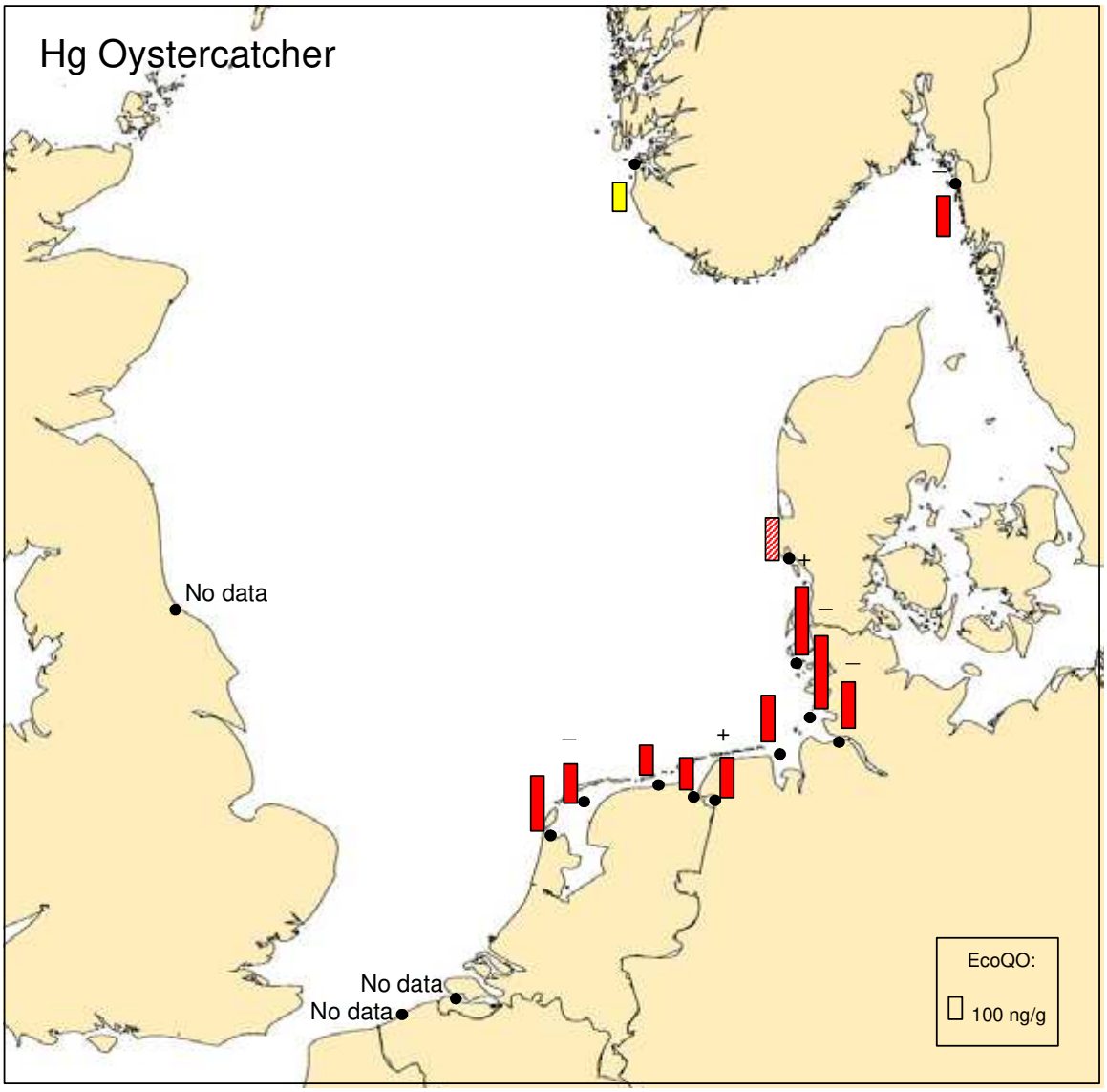

Fig. 4.1: Mean Hg contamination of Oystercatcher eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO fulfilled in some of the study years. ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2.

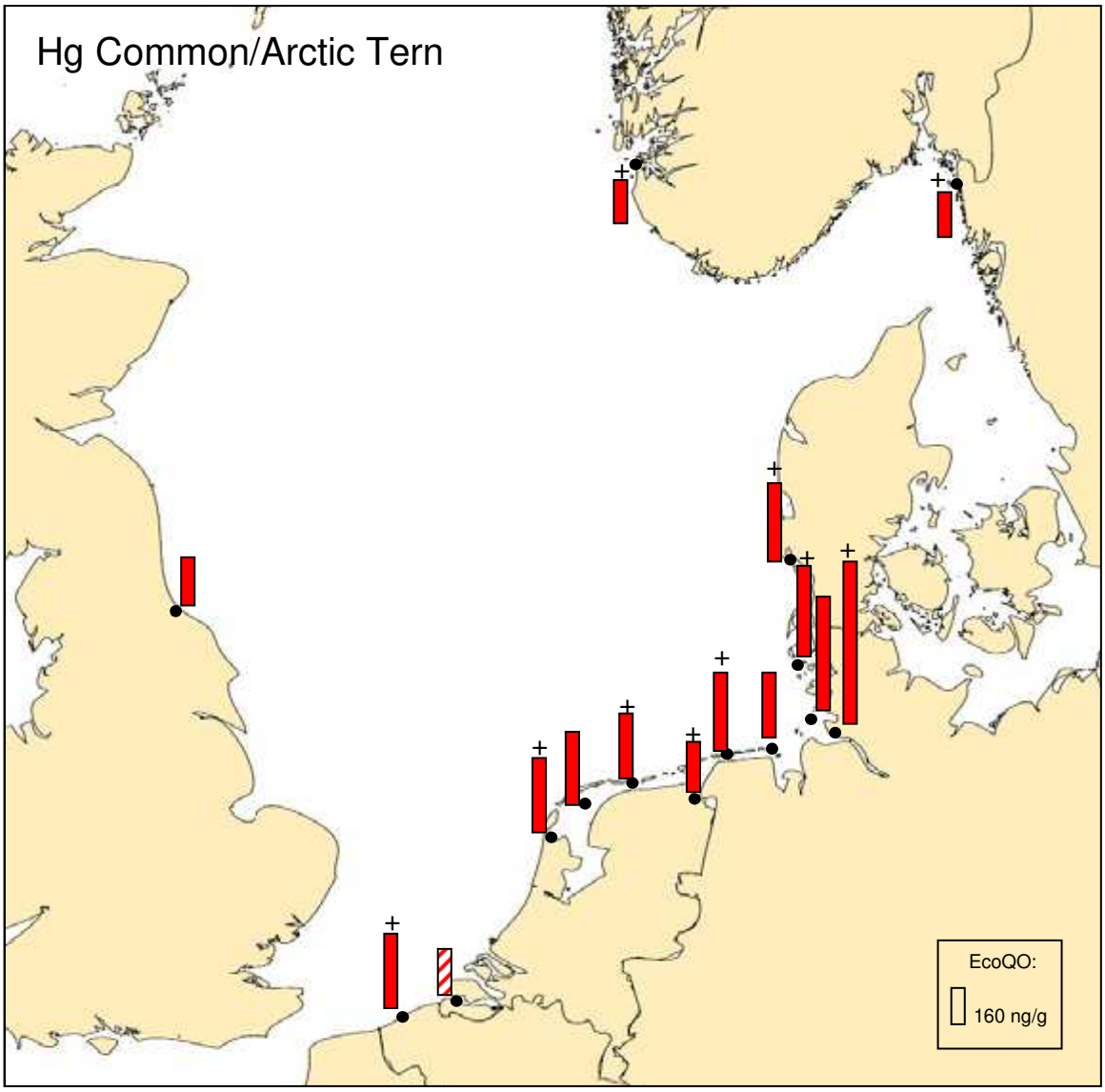

Fig. 4.2: Mean Hg contamination of Common/Arctic Tern eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2.

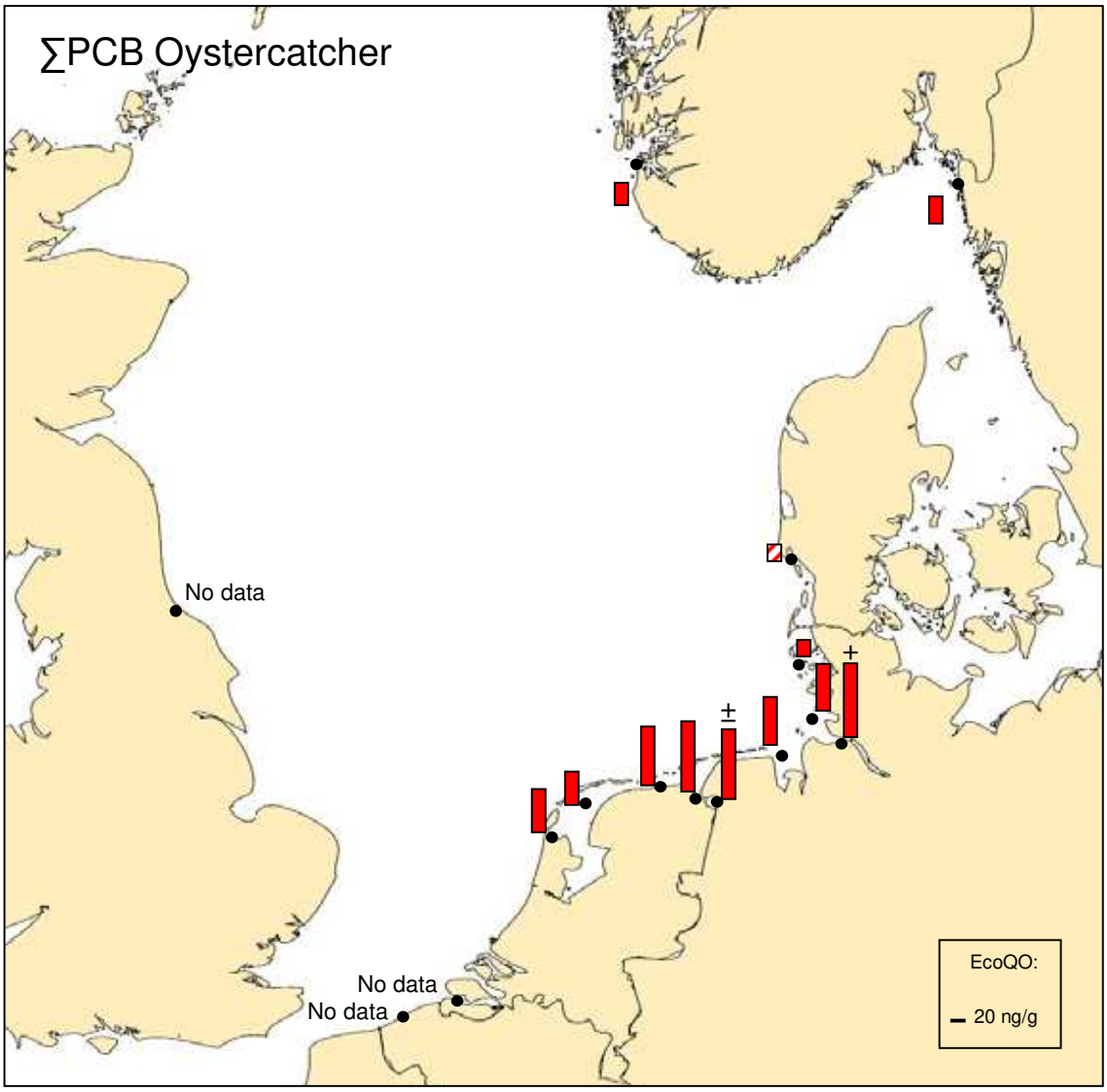

Fig. 4.3: Mean ∑PCB contamination of Oystercatcher eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2.

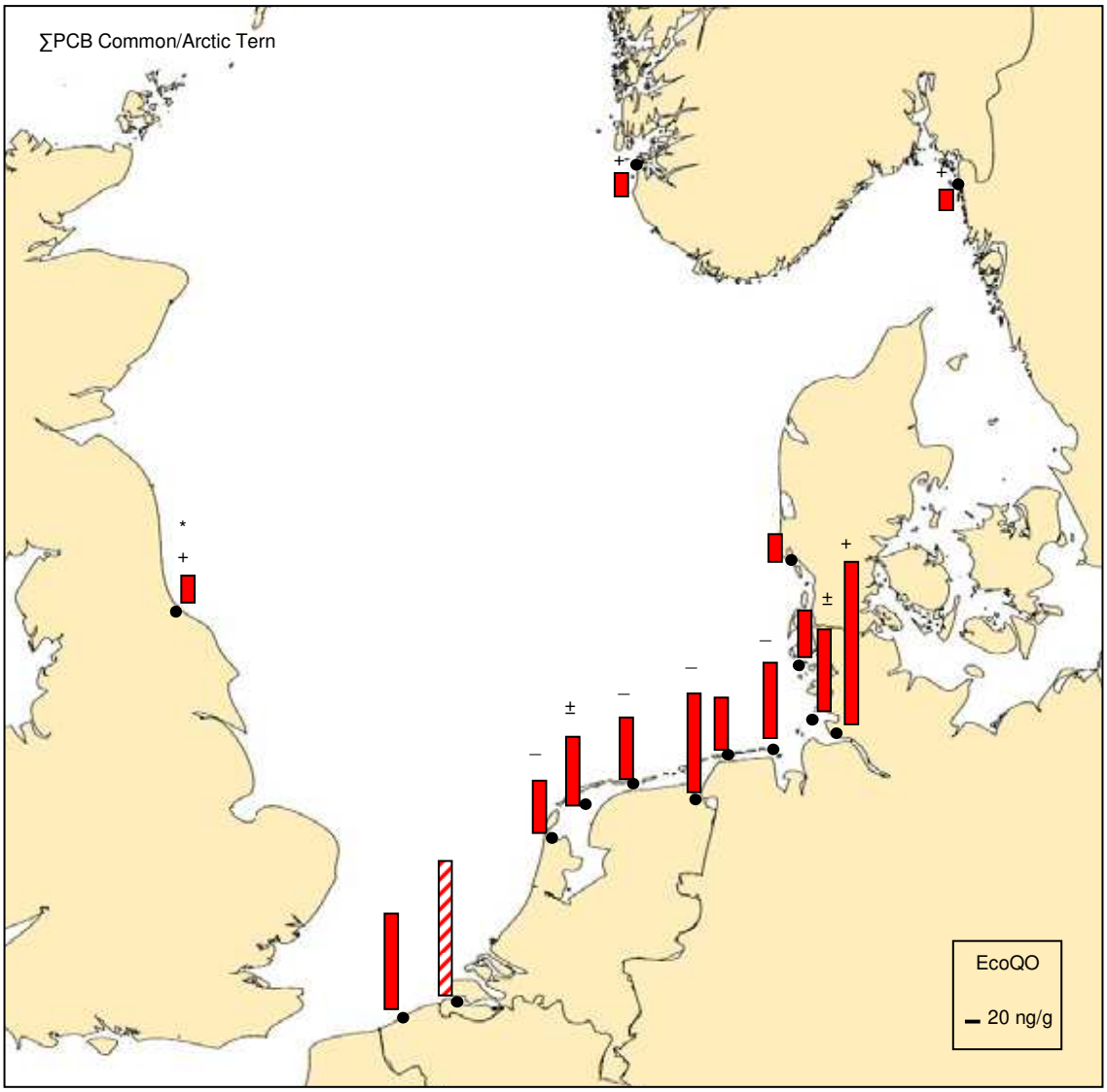

Fig. 4.4: Mean ∑PCB contamination of Common/Arctic Tern eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2. *: Values of GB were multiplied with a specific conversion factor to be comparable with the EcoQO.

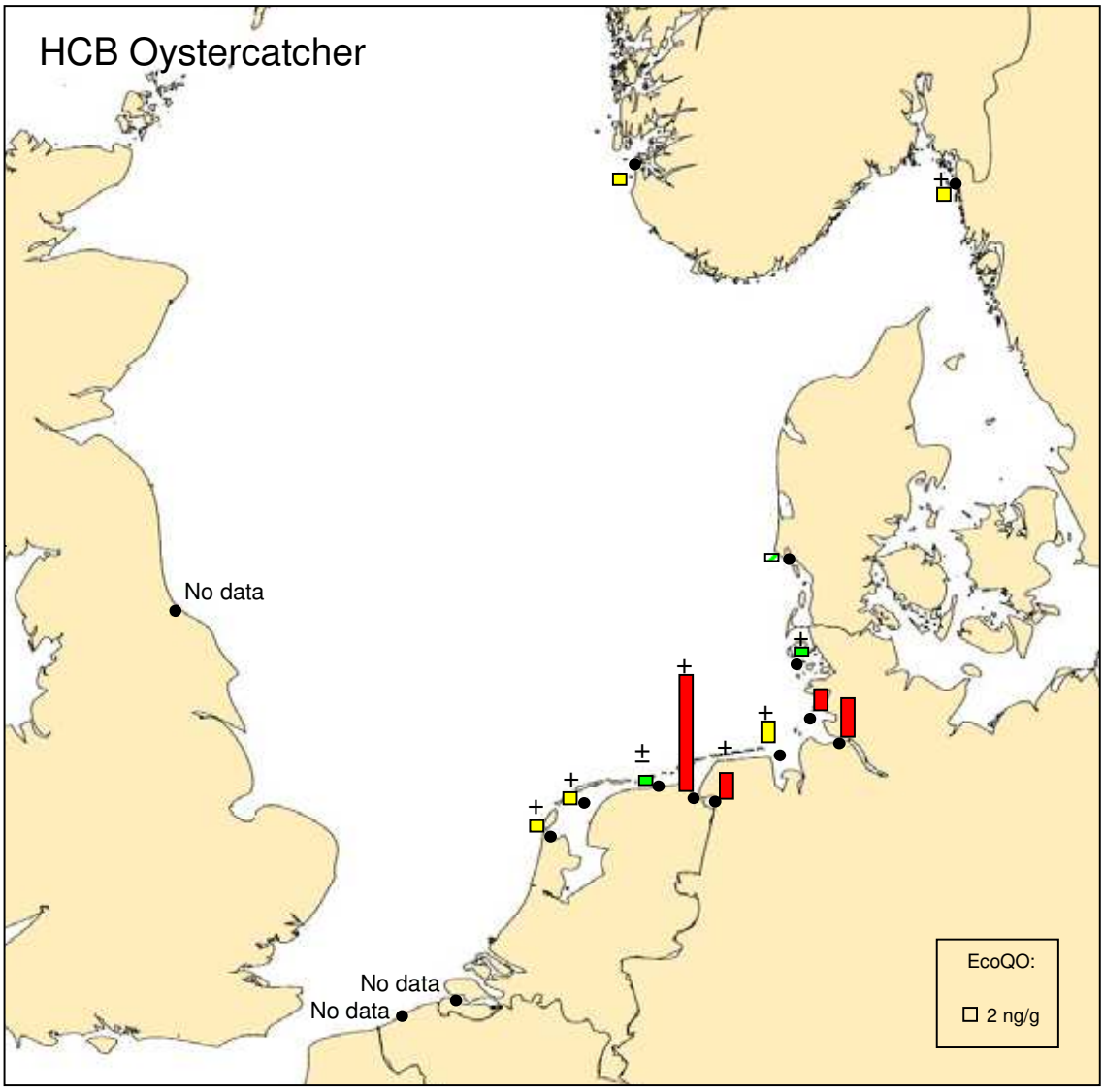

Fig. 4.5: Mean HCB contamination of Oystercatcher eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ■: EcoQO fulfilled in all study years, ■: EcoQO fulfilled in some of the study years, ■: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2.

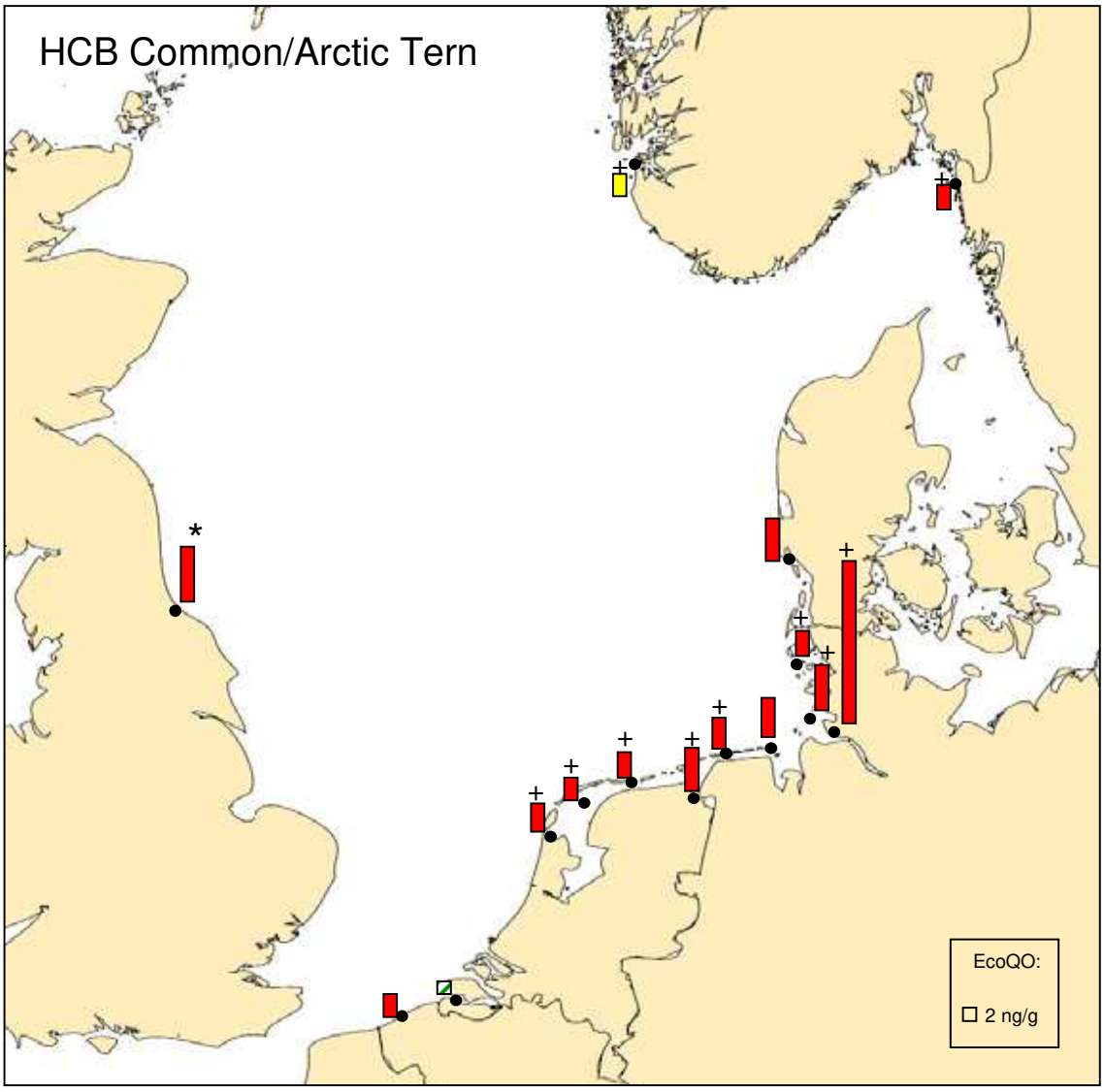

Fig. 4.6: Mean HCB contamination of Common/Arctic Tern eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO fulfilled in some of the study years, ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2. *: Values of GB were multiplied with a specific conversion factor to be comparable with the EcoQO.

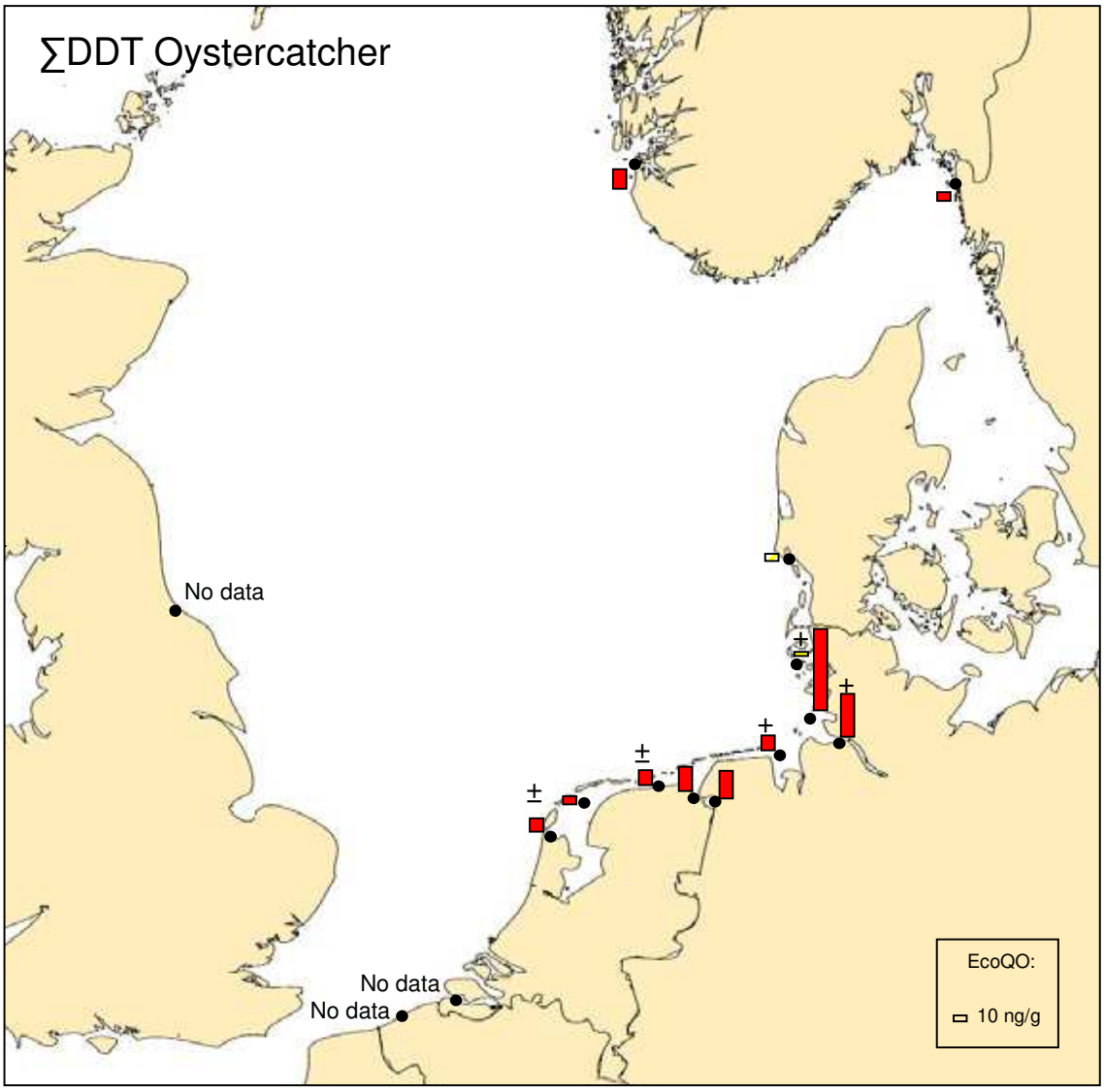

Fig. 4.7: Mean ∑DDT-derivate contamination of Oystercatcher eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. : EcoQO fulfilled in some of the study years, : EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2. *: Values of GB were multiplied with a specific conversion factor to be comparable with the EcoQO.

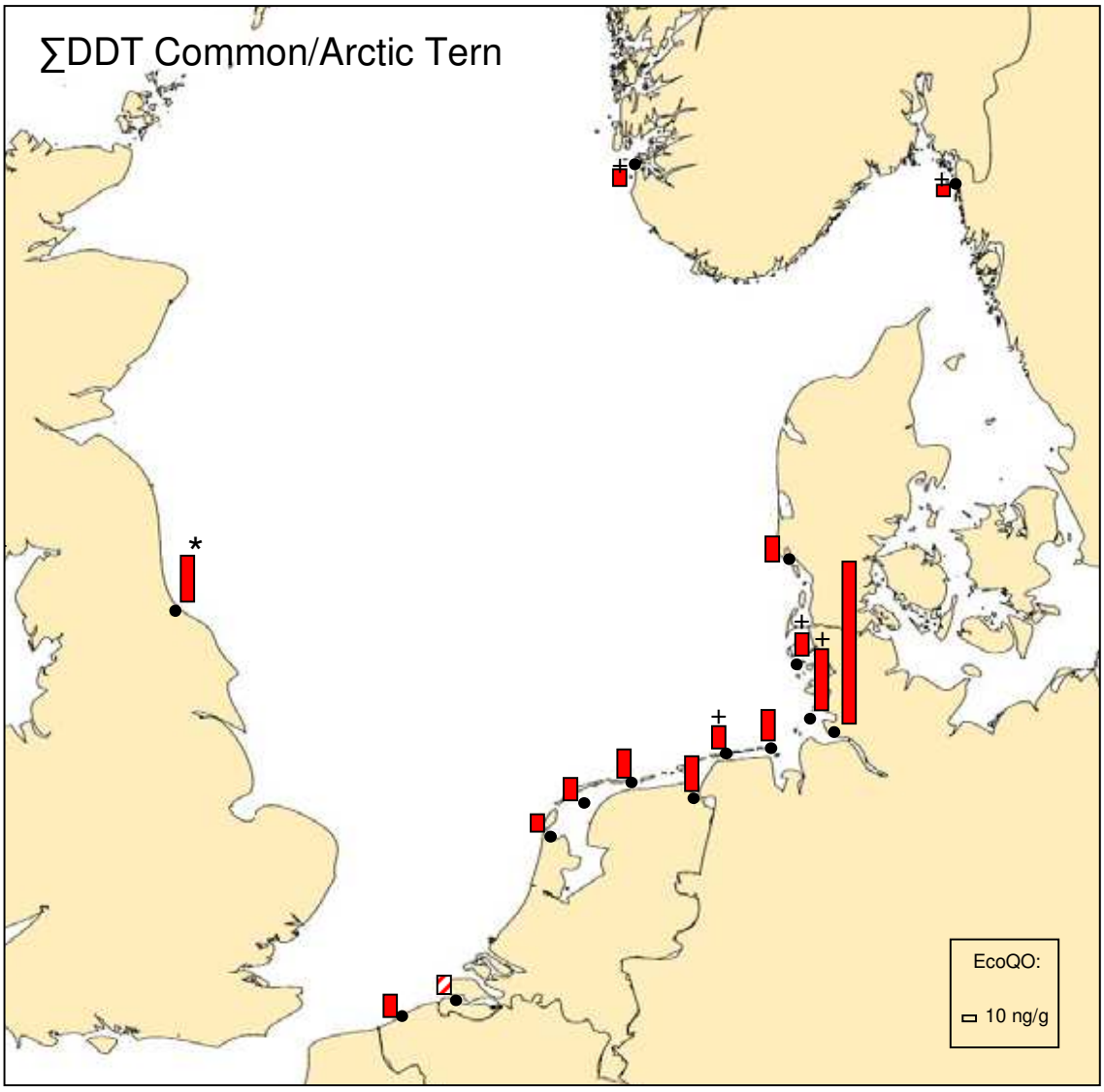

Fig. 4.8: Mean ∑DDT-derivate contamination of Common/Arctic Tern eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2. *: Values of GB were multiplied with a specific conversion factor to be comparable with the EcoQO.

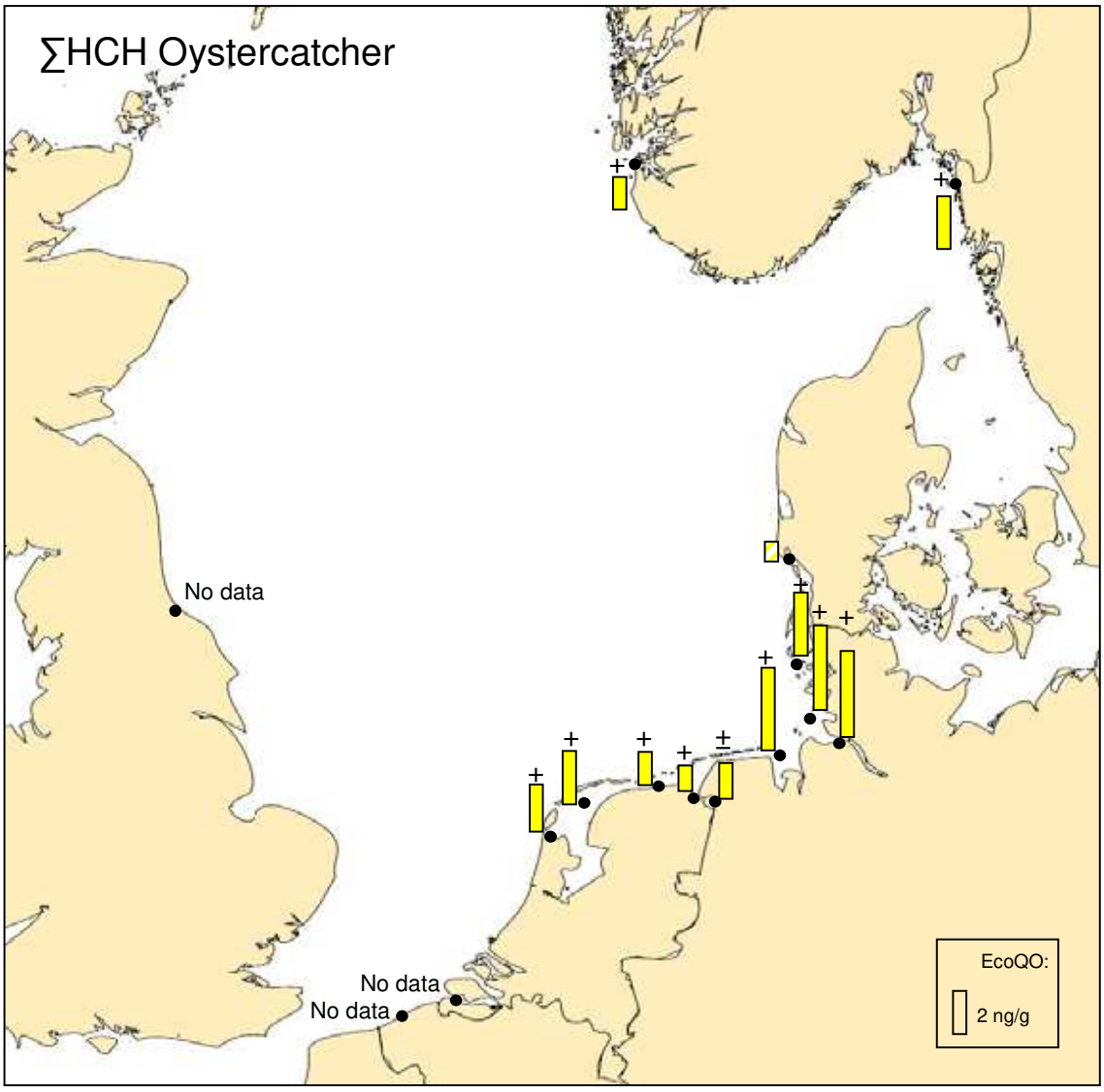

Fig. 4.9: Mean ∑HCH contamination of Oystercatcher eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO fulfilled in some of the study years Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. For sample size per year, see Table A.2.

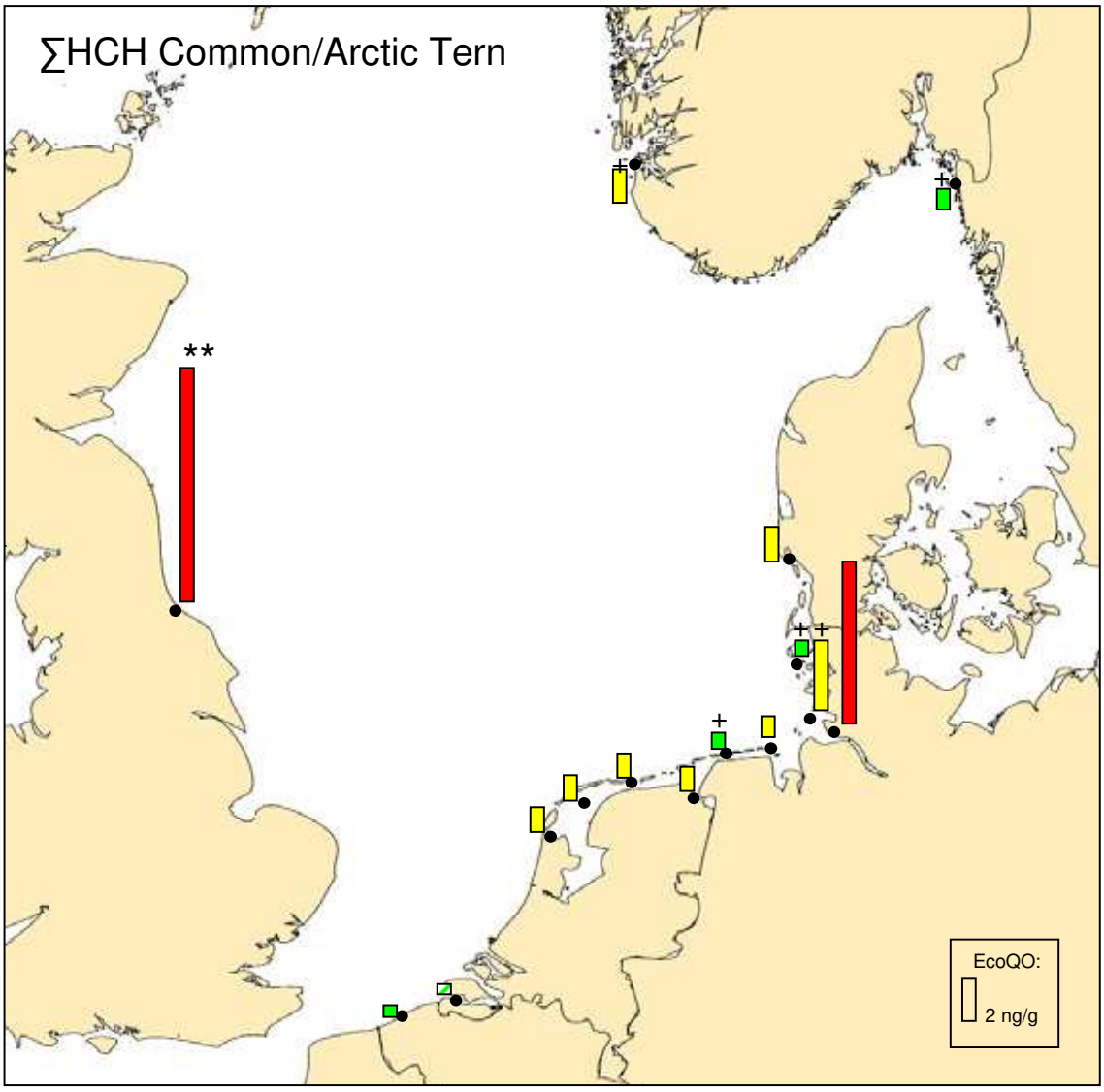

Fig. 4.10: Mean ∑HCH contamination of Common/Arctic Tern eggs in comparison to the EcoQO level, indicated by bar length, and temporal development. Study years 2008-2010. ▮: EcoQO fulfilled in all study years, ▮: EcoQO fulfilled in some of the study years, ▮: EcoQO not fulfilled in any study year. Dashed bar: Sample size too low to determine a temporal change. +: increase, -: decrease of pollutant level. ** In GB, only values of α- and γ-HCH were measured and summarized (Table 3). For sample size per year, see Table A.2.

Table 5: Temporal changes in pollutant concentrations in Common and, respectively, Arctic Tern eggs in the period 2008-2010. + (orange): increase, - (turquoise): decrease of concentrations between the years compared, white: no difference between years (Scheffé Tests and, respectively, T-tests, based on logarithmic values). Grey: no samples or samples too small for comparison. *: Arctic Tern sampled instead of Common Tern.

			Middlesbrough, GB	Zeebrugge, B	Balgzand, NL	Griend, NL	Julianapolder, NL	Schiermonnikoog, NL	Delfzijl, NL	Dollart, D	Baltrum, D	Jade, D	Elbe, D	Trischen, D	Hallig Hooge, D	Langli, Mandø, DK*	Stroemstad, S	Presteskjaer, N*
Oystercatcher																		
Hg	2008	2009								+			-	-				
		2010				-							-		+		-	
	2009	2010												+	+			
HCB	2008	2009			+	+	+			+		+			+			
		2010			+	+	+		+	+		+		+	+	+	+	
	2009	2010					-								+			
ΣPCB	2008	2009								-			+		-			
		2010											+					
	2009	2010								+						-		
ΣDDT	2008	2009			+		+						+			+		
		2010										+						
	2009	2010			-		-								+	-		
ΣHCH	2008	2009			+	+	+			-		+		+	+	+		+
		2010			+	+	+		+	+		+	+	+	+	+	+	+
	2009	2010							+	+		+			+	+		+
Common /Arctic Tern																		
Hg	2008	2009							+							+		
		2010		+	+	+		+	+		+		+	+	+			+
	2009	2010				+		+			+		+		+			+
HCB	2008	2009			+	+									+		+	+
		2010		+		+		+	+		+				+		+	+
	2009	2010				+		+			+		+	+			+	
ΣPCB	2008	2009			-	-		-	-			-		-		+		
		2010						-	-			-						+
	2009	2010	+			+							+	+			+	+
ΣDDT	2008	2009							-						+	+		+
		2010												+	+		+	+
	2009	2010						-			+			+			+	
ΣHCH	2008	2009				+					+	-		+	+	+	+	+
		2010			+	+		+	+		+	-	+	+			+	+
	2009	2010	+		+	+		+	+		+		+	+				+

Summarizing, it can be stated that contamination decreases were patchy on sites (terns) and somewhat more evident for PCBs in oystercatchers, increases were generally evident across sites for HCB and HCH (both species) and Hg (Oystercatcher). For other contaminants, no such general pattern of change was detectable across sites for other contaminants. These findings indicate that there is little evidence for current declines (Table 5). It has to be mentioned that in the Danish Wadden Sea, sample size of Oystercatcher eggs was below 5 in 2008 and 2010, so no statistical comparison of years was possible to detect temporal changes.

3.6 Comparison of the actual pollution with the EcoQOs

Fig. 4.1-4.10 present the mean values for the five contaminant groups over the three study years in relation to the corresponding EcoQO. In Fig. A.1, the fluctuations of contamination values of the single eggs around the mean and the EcoQO targets are shown with two examples.

3.6.1 Common/Arctic Tern

Hg and **∑PCB** concentrations in eggs at all sites exceeded the target EcoQOs (Hg: 160 ng/g; ∑PCB: 20 ng/g) by 1.03-5.8 fold and 7.8- 99 fold, respectively.

HCB concentrations were only as low or lower than the EcoQO target of 2 ng/g at Zeebrugge (2008 - the only study year at that site) and in one year (2008) at Presteskjaer.

∑DDT on all sites exceeded by 1.2 to 35.3 fold the EcoQO target of 10 ng/g.

∑HCH were as low or lower than the target EcoQO of 2 ng/g in all study years at Zeebrugge, Terneuzen, Baltrum and Stroemstad and in some study years at all other sites except Middlesbrough and Elbe. At Middlesbrough and Elbe, concentrations were up to 7.7 and 5.9 times higher, respectively, than the target value.

3.6.2 Oystercatcher

Hg exceeded (by 1.2-4.3 times) the target EcoQO of 100 ng/g at all sites in all years except Presteskjaer in 2008.

∑PCB exceeded the EcoQO concentration of 20 ng/g by 6.5-42 times in all years and sites.

HCB met or was below the target EcoQO of 2 ng/g in all study years at Julianapolder and Hallig Hooge and in some study years at Balgzand, Griend, Jade, Stroemstad and Presteskjaer. HCB concentrations exceeded the EcoQO in all years at all of the sites situated in the estuaries of the large rivers Ems and Elbe (Delfzijl, Dollart, Elbe and Trischen).

∑DDT was at or below the EcoQO of 10 ng/g in some years at Hallig Hooge and in the Danish Wadden Sea. At all other sites, concentrations exceeded the EcoQO by factors of between 1.2 and 25.0.

∑HCH concentrations met the EcoQO of 2 ng/g in some years at all sites. However, concentrations had increased at 10 of 12 sites over the study period.

3.6.3 Differences between species

At several sites where both the Oystercatcher and a tern species were sampled, the EcoQO for HCB and ∑DDT were met at least in some years in the Oystercatcher but not in the terns (Fig. 5). Only in the Common Tern, the EcoQO was not met by any substance group in any study year at the sites Middlesbrough and Elbe.

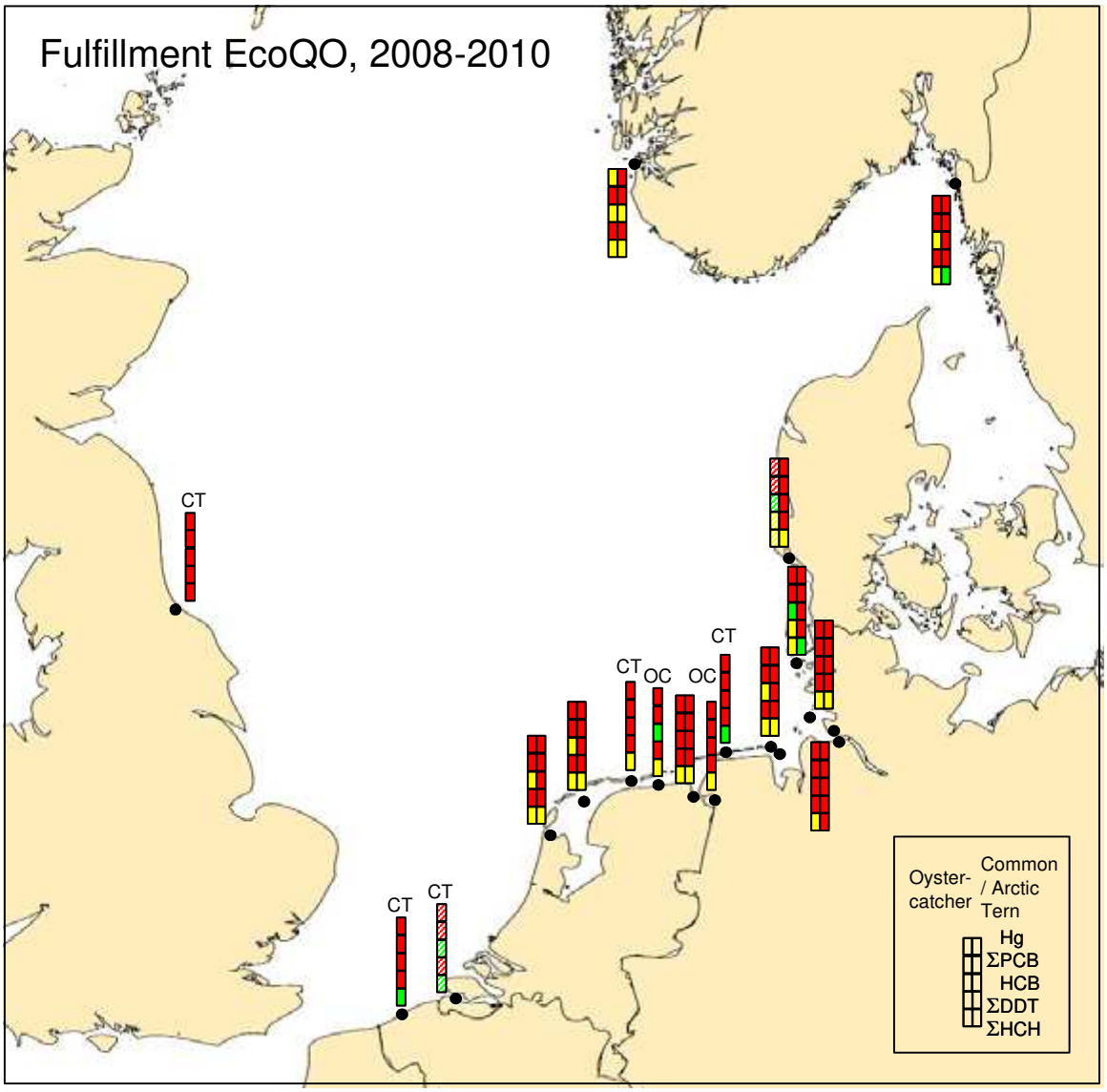

Fig. 5: Fulfillment of EcoQOs for five groups of environmental pollutants in 2008-2010. ■: EcoQO fulfilled in all study years, ■: EcoQO fulfilled in some of the study years, ■: EcoQO not fulfilled in any study year. Dashed bar: Sample size only in one year larger than four. For a complete overview of sample sizes per year, see Table A.2.

4 Discussion

This report presents data on coastal bird egg pollution with Hg and organochlorines from an, up to date, unique number of sites and countries surrounding the North Sea which were analyzed synoptically. Of the 21 sites studied here, 15 sites (or corresponding sites in the very vicinity) situated in the Dutch, German and Danish Wadden Sea have been sampled for decades by the Institute of Avian Research within the TMAP program (e. g. Becker & Dittmann 2009), whereas six new sites have been additionally sampled in the three years of the pilot study of the EcoQO from 2008 to 2010. These have been one site each situated in Great Britain, Belgium, western Netherlands, Sweden and two sites in Norway. In particular the three latter sites were situated in non or slightly industrialized areas, providing values of high importance for a comparison of pollution levels with those from sites in the estuaries of large rivers draining highly industrialized areas of Middle and Western Europe including concomitant atmospheric deposition in the river shed area. Based on the new minimum concentration measured for Hg at Presteskjaer in Norway, the EcoQO for this heavy metal has now been set to 160 ng/g fresh egg mass for tern eggs and 100 ng/g for Oystercatcher eggs. An overview over the actual threshold values defined as EcoQOs is given in Table 4.

4.1 Spatial contamination patterns

The spatial contamination pattern is obviously a result of several large rivers draining into the North Sea as main important input sources for environmental pollutants. So the river Elbe has proved to be an important input source for Hg, the industrial chemicals ∑PCB and HCB, but also still for derivates of the insecticide DDT in Oystercatcher and Common Tern still during the end of the first decade of the 2000s. For the concentrations of Hg, PCB, HCB, in case of the terns also for DDT, a clear, continuous decrease was found with increasing distance from the Elbe estuary towards the north whereas concentrations decreased abruptly towards the west (Fig. 2, Fig. 4.1-4.8), indicating effects of dilution through the northwards streaming water currents (Lozán et al. 1990). Besides the river Elbe, the German and Dutch sites at or near the river mouth of the Ems exhibit concentration peaks of HCB and ∑PCB. At the river Tees (Middlesbrough, GB), high levels of ∑HCH were measured in the Common Tern, further, lower peaks were recorded for HCB and ∑DDT concentrations (the Oystercatcher was not sampled at that site). Summarizing, the concentrations of the derivates of the insecticides, ∑DDT, ∑HCH and HCB were greatest in GB and in or near the Elbe estuary, whereas peak concentrations of the industrial chemicals Hg, ∑PCB and HCB particularly occurred in eggs from sites that were in or near estuarine areas on the mainland.

In the Wadden Sea from The Netherlands to Denmark, the pattern of spatial contamination largely reflects those documented by Becker & Dittmann (2009). Assuming a mean water content of 75% and, respectively, 76% for Oystercatcher and tern eggs respectively (Mattig et al. 2000), Hg concentrations in tern eggs between 2008 and 2010 were broadly of the same order of magnitude as those in Gannet *Morus bassanus* eggs from two Scottish colonies in the beginning of the 2000s (Pereira et al. 2009); concentrations in Oystercatcher eggs were 1.5-3.0 fold lower. This may reflect the specific positions of Gannets, terns and Oystercatchers in the food web as fish and benthos feeders. However, the range of Hg contamination in the Oystercatcher eggs strongly overlapped that in Guillemot *Uria aalge* eggs from the Swedish Baltic Sea (Bignert et al. 2011), whereas Hg contamination in tern eggs was as low as that in Guillemot eggs only at the least contaminated sites. Assuming a fat content of 8.30 % and, 7.95 % for Oystercatcher and tern eggs respectively (Mattig et al. 2000), the HCB content of North Sea Oystercatcher and tern eggs was by factors of 1.6-60.2

fold lower and, respectively, 1.3-20.4 fold lower than that of Baltic Sea Guillemots from Sweden (Bignert et al. 2011).

There were no clearly detectable spatial patterns in terms of meeting EcoQO targets. EcoQO targets were fulfilled, at least in some years, at all (Oystercatcher) or almost all (terns) sites for ∑HCH and at 7 out of eleven sites for HCB in the Oystercatcher (Fig. 5). In contrast, concentrations remained above the EcoQO in Hg, ∑PCB and ∑DDT values at nearly all sites for both species. However, the overall spatial pattern suggests that, where contamination appears to be decreasing over time, EcoQO targets for most substances will probably first be met in the northern and northeastern part of the study area. How likely or rapid any such decrease may be is unclear given that pollutants, including those measured in this study, may accumulate even in remote areas of the world through long-term transportation by atmospheric and water currents (e. g. Bakker et al. 2009; see above).

4.2 Temporal changes

In terms of North Sea wide temporal changes, significant increases between 2008 and 2010 at the majority of sites were detected for Hg and HCB in tern eggs and for HCB and ∑HCH in Oystercatcher eggs. The reasons for these spatially large-scaled developments are unknown. No such general changes were detectable for the other contaminant groups. This contrasts with the mid- and long-term significant decreases prior to 2008 at the majority of sites (Dittmann & Becker 2009). This apparent anomaly emphasizes the need for long-term environmental monitoring based on annual measurements to evaluate whether recent increases of some substances indicate the beginning of a spatially large-scale trend. Pollutant concentrations reached during the increases between 2008 and 2010 still ranged within the contaminant fluctuations recorded during the 20 years before 2008. It should be remembered that the concentrations of several substances analyzed here were relatively high prior to1990, decreased steeply during the 1990s, and then only decreased slowly, or remained broadly constant albeit with some fluctuations, thereafter (Becker et al. 2001, Bakker et al. 2009, Dittmann & Becker 2009). Thus, without specific measures to further reduce the pollutant input into the marine environment, EcoQO targets based on minimum concentrations measured in recent times are unlikely to be achieved rapidly. When there are future significant concentration decreases, EcoQO targets are likely to be reached first at sites distant from large rivers and/or in Oystercatcher eggs because of the lower trophic level of this species (Fig. 5).

4.3 Suitability of the species studied

A key question is whether it is necessary to monitor both the Oystercatcher and a tern species to obtain a reliable picture of the contamination of the marine environment within the EcoQO framework of the concept. Both are well known species and their differing life strategies have pros and cons in terms of environmental monitoring.

Tern species, as top predators foraging mostly at sea, are effective bioaccumulators of marine pollutants and are income breeders. Thus, contaminants in tern eggs accumulation that has mainly occurred over a short and well defined period on a specific site. The Arctic Tern proved a suitable species to replace the Common Tern as study species in areas where the latter is rare. Both species are colonial breeders, enabling the sampling of a sufficient number of eggs in short time. However, both tern species are long-distance migrants and, theoretically, some pollutants transferred into eggs could have been ingested at unknown sites during migration. Furthermore, the fish prey eaten by terns may also migrate to some

extent, thereby magnifying the spatial range over which the pollutants measured in the eggs may have originated, and also potentially reducing measurable heterogeneity in contaminant levels that may occur spatially.

The Oystercatcher, as a largely sedentary capital breeder feeding on sedentary prey, integrates the signal of pollution of a given site over a relatively long time period. The enhances ability to identify local pollution events and hotspots, such as the historical heavy pollution of sediments with HCB in the harbour of Delfzijl (Eggens & Bakker 2001) which is still clearly evident from a strong HCB peak in the eggs of Oystercatchers from Delfzijl in this study (Fig. 3). On the other hand, the capital breeding strategy of Oystercatchers, coupled with limited winter movements (particularly prevalent in birds from the northern sites and during cold winters), mean that some contaminants in eggs may originate from locations distant from the breeding site. Furthermore, Oystercatchers may feed to a higher degree than terns in non-marine habitats, such as nearby inland meadows, and contaminants in eggs may partially reflect terrestrial pollution.

Given this, it can be argued that monitoring of both the Oystercatcher and a tern species should be continued, as the relative advantages and disadvantages of both species as sentinels complement each other. Such combined monitoring enhances the reliability of detection of any patterns and the ability to explain such trends. Furthermore, if the EcoQO is reached both in the Oystercatcher and the tern at a given site, the environmental health status of that site is confirmed at both a lower and higher trophic level.

4.4 Recommendations for monitoring concept and methods

Common Tern, Arctic Tern and Oystercatcher eggs have proved suitable matrices for the monitoring of the pollution of the marine environment with Hg and organochlorines. Due to the high position of the study species in the marine food web and the high level of bioaccumulation of lipophilic contaminants, eggs of these species can be used to detect very small concentrations of chemicals in the marine environment with relatively low logistical sampling effort. Intra-site variation in contaminant concentrations is also comparatively low, providing high statistical power for analyses of spatial and temporal trends, and for defining EcoQO thresholds. This is a clear advantage of the bird egg matrix compared with, for example, sediments, water, or organisms with a lower position in the food web such as invertebrates or herring *Clupea harengus* (cf. Becker et al. 2001, Bignert et al. 2007, OSPAR 2007b). The good understanding of the biology and the ecology of the species also make the outputs from monitoring quite easy to interpret and explain to a broad public.

We recommend maintaining an annual sampling scheme, as described here, to guarantee sufficient statistical power to detect middle-term changes of the pollutant levels in the environment. Such annual analyses furthermore facilitate a variety of logistics, including analytics being performed for longer periods in the same labs with constant methods, which is essential for a comparability of datasets. Furthermore, we strongly recommend to continue treating each egg as an individual sample and not to merge the content of the eggs of each site into a pooled sample. We argue that the statistical benefits of maintaining measures of individual variation outweigh any benefits of pooling samples that are gained from reduced analytical cost.

To avoid costly intercalibrations between laboratories, we recommend that laboratories specialize in their analysis and all samples are analysed for one or more contaminant groups in the same laboratory. Thus, a number of laboratories could be involved in monitoring,

each specializing on different contaminant groups. This would require the egg samples to be sub-divided and sent to the different laboratories involved. The tern and oystercatcher eggs are sufficiently large for this to be practical (terns about 19 g, Oystercatcher about 40 g fresh mass). Each laboratory would need to have in place adequate quality assurance and control measures in order to be able to demonstrate appropriate accuracy and repeatability of analysis.

We propose the inclusion of further regions into the EcoQO concept. Among the countries surrounding the North Sea, expansion is particularly needed on further sites in UK which has been represented up to now only by the one site Middlesbrough. The UK has a high degree of industrialization with large rivers such as the Thames, Humber and Forth, which might be expected to contribute significant amounts of pollutants into the North Sea. Sites with low levels of pollution are also likely to exist in Northern Scotland, for example the Moray Firth and these could represent suitable reference areas and have already been recommended by OSPAR (2007a, b, 2009c). In addition, a site at the Rhine delta should be included into the monitoring scheme to consider this important central European river.

On a larger spatial scale within the framework of the Oslo-Paris convention, the inclusion of further European regions such as the French Atlantic Coast is recommended. Furthermore, it is desirable to include the Baltic Sea, which is particularly threatened through pollution events by its specific hydrology, into the EcoQO program, and also the Northwest Atlantic. The latter would be facilitated by the occurrence of the same (Common and Arctic Tern) or closely related and ecologically corresponding (American Oystercatcher *Haematopus palliatus*) coastal bird species, thus representing comparable study species as bioindicators.

In addition to new areas, a variety of new environmental pollutants have emerged that may additionally require monitoring or may replace the monitoring of older legacy compounds that fall permanently below the EcoQO threshold. In the Wadden Sea, chlordanes and nonachlor-compounds have been monitored for decades (Becker & Dittmann 2009), a pilot assessment of phthalates has been initiated in recent years (unpublished). Polybromates (e. g. BDE 47, 99, 100, 183, 209, HBCD and TBBP-A), persistent organic pollutants (POPs) such as the dioxins polychlorinated dibenzodioxins (PCDD) and polychlorinated dibenzofurans (PCDF), and perfluorinated compounds (PFCs) have begun to be included in monitoring schemes for predatory birds in GB (Leslie et al. 2011, Shore et al. 2006, Crosse et al. in press) and Guillemots in Sweden (Bignert et al. 2011). According to Agenda Item 11 following the Meeting of the ASMO in June 2010 (OSPAR 2010), the monitoring of PCDD, PCDF and PFCs in bird eggs has been determined as part of the pre-CEMP and tools for quality assurance procedures and assessment criteria are under development. The inclusion of these comparatively new substances into the coordinated monitoring is dependent on resolving the status of the EcoQO (OSPAR 2010). The same is true for TBTs, bromocyclen and musk xylol for which the development of an EcoQO has also been recommended (OSPAR 2007b).

5 Existing monitoring programs on coastal bird egg contamination in the OSPAR region

The monitoring of Hg, organochlorines and other pollutants in coastal bird eggs has been well established for decades at many sites in the North Sea and adjacent areas. An overview of the currently running monitoring programs among the contracting parties gives Table 2. Since 1981, Common Tern and Oystercatcher eggs have been sampled throughout the Wadden Sea in several research projects. Contaminants in eggs of these species from The Netherlands, Germany and Denmark have been monitored since 1998 in the Trilateral Monitoring and Assessment Program (TMAP) according to the OSPAR JAMP monitoring guidelines (OSPAR 1997). In the North Sea area, this monitoring program covers 13 stations and a broad range of regularly analyzed chemicals.

The COMBINE monitoring program of HELCOM quantifies the impacts of nutrients and hazardous substances in the marine environment. In Sweden and Germany, long-term sampling of bird eggs – and a variety of other environmental parameters - is carried out by the national Environment Specimen Banks (ESB) of both countries. Both conduct monitoring for contaminants in the national environment. In Sweden, samples have been collected since the late 1960s according to international standards. Samples include Guillemot eggs that are analysed for Hg, Pb, Cd, Ni, Cr, Cu and Zn and organochlorines. Further time series data have been generated for piperidinocyclohexanecarbonitrile (PCC; Wideqvist et al. 1993), dioxins/dibenzofurans and polybrominated compounds (Sellström 1996, Sellström et al. 2003), bis(4-chlorophenyl) sulfone, methylsulfonyl-DDE and -PCBs (Jörundsdóttir et al. 2006) and PFOS/PFOA (Holmström et al. 2005). The German ESB, established in 1985, has been sampling Herring Gull eggs annually at two sites in the Wadden Sea (Jade Bay and Trischen) and at one site in the Baltic Sea (Bodden National Park). Compounds that have been monitored continuously are Hg, As, Se, Tl, Cu, Pb, 7 PCBs, DDTs, HCHs, HCB, OCS, Dieldrin and PeCBZ..

The Predatory Bird Monitoring Scheme (PBMS; http://pbms.ceh.ac.uk/) has been conducting a long-term monitoring of pollutants in the livers and eggs of mainly terrestrial predatory birds since the late 1960s in UK. The PBMS currently contributes to the North Sea monitoring by the measurement of contaminants in Gannet eggs from Bass Rock in the Firth of Forth, Scotland. Gannet eggs are analysed for Hg, HCB, DDT, DDE, TDE, α-HCH, γ-HCH, HEOD and 35 PCB congeners (Pereira et al. 2009).

Further monitoring programs on chemicals in coastal birds are conducted elsewhere, for example in the high Arctic (e.g. Ólafsdóttir et al. 2005, Braune 2007, Braune et al. 2007).

6 Synergies between EcoQOs and other monitoring and reporting requirements

There are several requirements for monitoring of hazardous substances in various matrices of biota including coastal birds eggs. Drivers for this are inter-related and come from the OSPAR EcoQO approach, the relevant EU Directives (Water Framework Directive, Birds and Habitats Directive, Marine Strategy Framework Directive) and the Trilateral Wadden Sea Cooperation and there is a need for harmonization and coherence in approach and activities. Presently, the definition of specific objectives and reference values, as well as monitoring programs are under development. Therefore, there are opportunities to tune these activities across the countries in order to address both the requirements under the various EU Directives, the objectives of OSPAR and the Trilateral Wadden Sea Cooperation. Aims and scopes of the various directives and agreements and the potential chances to integrate the EcoQOs are given in the following:

EU Water Framework Directive (WFD)

The EU Water Framework Directive (WFD) (Directive 2000/61/EC), which came into force in December 2000, establishes a new integrated framework for the protection, improvement and sustainable use of all waters, including transitional and coastal waters up to 1 nautical mile from territorial sea baselines. The key objective (Article 1) of the Directive that is most relevant to marine ecosystems is: "to prevent further deterioration and protect and enhance the status of aquatic ecosystems and associated wetlands." A key aim (Article 4) is that Member States will be required to achieve a "good surface water status" and also to prevent deterioration in the quality of waters that are already "good" by 2015. The guidance on monitoring with regard to the selection of matrices (water, sediment and biota - WFD monitoring guidance for surface waters) underlines that analysis of sediment and biota (especially with regard to hydrophobic and lipophilic substances) is resource effective for trend monitoring. The use of sediment and biota is also advised to assess compliance with the "non-deterioration" objective, to assess long-term changes resulting from anthropogenic activity and to monitor the progressive reduction in the contamination of priority substances (PS) and phasing out of priority hazardous substances (PHS). For marine areas, the guidance refer to the OSPAR guidelines for sampling and analysis of contaminants and metals in fish, shellfish and coastal bird eggs (OSPAR 1997).The EcoQOs for coastal bird eggs can be usefully integrated into the WFD objectives to define the "good status" and to monitor the "non-deterioration" target.

Marine Strategy Framework Directive (MSFD)

The Marine Strategy Framework Directive" – MSFD (2008/56/EC) represents the environmental pillar of the new EU Maritime Integrated Policy. The main objective of the Directive is to achieve a Good Environmental Status (GES) of the marine environment by 2020. According to the directive, this aim should be realized by a series of organizational steps which are the initial assessment of the environmental status, the definition of GES by July 2012, the implementation of a monitoring program by July 2014 and, in consequence, of a program of measures by July 2016 to achieve or maintain a GES by 2020. The definition of GES includes the establishment of a series of environmental targets and indicators.

As a general descriptor for the GES with respect to contaminants has been formulated that their concentrations are at levels not giving rise to pollution effects (EU 2010). Heavy metals, industrial chemicals and pesticides are listed in Tables 1 and 2 of the Annex III of the MSFD (EU 2008). The MSFD (EU 2008) also states that, when devising targets and indicators, Member States shall take into account the continuing application of relevant existing environmental targets laid down at national, Communitiy or international level in respect of the same waters, ensuring that these targets are mutually compatible and that relevant transboundary impacts and features are also taken into account, to the extent possible.

In a large part of the North Sea, the international Wadden Sea, a monitoring of pollutants in bird eggs has been running for decades (TMAP; Becker & Dittmann 2009) and this monitoring has been a substantial base for the EcoQOs developed and presented in this report. Therefore there is potential to standardize GES with respect to the already established EcoQO, thus enabling effective cooperation between both programs. Spatially, a combination of these programs could also provide a cost-effective way to include further sites into the EcoQO program, as, for example the Baltic Sea by a cooperation with the MSFD Baltic Sea program (see above) and/or in case of tying the HELCOM monitoring of indicators more closely to the MSFD requirements, e. g. through the current HELCOM CORESET project.

EU Birds and Habitats Directive

The EU Habitat Directive (Directive 92/43/EEC) aims to maintain or restore, at favourable conservation status, natural habitats and species of wild fauna and flora of Community interest (Article 2). The EU Birds Directive (Directive 79/409/EEC) aims at the conservation of all species of naturally occurring birds in the wild state in the European territory of the Member States. It covers the protection, management and control of these species and elaborate rules for their exploitation (Article 1). In order to assess a favorable conservation status, bird monitoring will be carried out with regard to abundance, distribution, population trend, breeding success, habitat and food requirement of birds. Monitoring of contaminants and application of EcoQOs supports this assessment in combination with other biological parameters.

Trilateral Wadden Sea Cooperation

The objective of the trilateral management is to guarantee the natural functioning of the ecosystem through proper regulation of human activities. The best guarantee for a natural ecosystem is to achieve the full scale of habitat types which belong to a natural and dynamic Wadden Sea. Each of these habitats needs a certain quality, which can be reached by proper management of the area. This quality can be described by certain characteristic structures, processes, the presence of certain organisms, and the absence of disturbance and toxic effects by the chemical condition of the habitat.

The trilateral policy and management regarding pollution issues is closely related to the developments in the framework of the North Sea Conferences, OSPAR, IMO and the European Union. The aim of the Trilateral Monitoring and Assessment Program (TMAP) is to provide a scientific assessment of the status of the Wadden Sea ecosystem and to assess the implementation of the Targets of the Wadden Sea Plan (Stade Declaration 1997, Wadden Sea Plan 2010). One of these Targets is to achieve concentrations of contaminants in the marine environment as resulting from zero discharges (Becker et al. 2001). This aims at values in the environment and in indicator species which are near background values for substances which do also occur naturally such as heavy metals (e.g. Hg) and close to zero

for man-made synthetic substances (xenobiotic compounds) (http://www.waddensea-secretariat.org/TMAP/Monitoring.html). The parameter "Breeding Success" has been included recently into the TMAP monitoring, first datasets have been collected in 2010. Therefore, in future, the contamination levels can be linked to breeding success to identify possible effects of chemical egg contamination on reproduction. The EcoQOs for coastal bird eggs can already be integrated into the TMAP in order to assess the achievement of these targets in the Trilateral Wadden Sea and to decide on further management measures.

7 Estimated Costs of the EcoQO monitoring

This pilot study had no own budget and was financially based mainly on the TMAP (Germany, The Netherlands and Denmark), The Norwegian Directorate for Nature Management (Norway), the CEH (UK), the Swedish Environmental Protection Agency (Sweden) and the CWSS (Germany). A continuation and further development of this EcoQO, however, requires additional financial sources and a reliable budget.

Here we present an estimation of financial support needed to run this EcoQO in future. We base our calculation on the EcoQO proposal by OSPAR for the North Sea which we followed with our pilot study, comprising two species, Oystercatcher and Common/Arctic Tern, 10 eggs per species (OC, CT/AT), site and year as well as on the substances Hg, ΣPCB, HCB, ΣDDT and ΣHCH.

The annual costs per egg sampled would be €50 for sampling and transport, €350 for chemical analysis and reporting, and €80 (20%) overheads, in total €480 per egg, i. e. €4,800 per species and site (Table 6). In the Wadden Sea, these costs are covered by the TMAP – parameter "Contaminants in Bird Eggs". For the other sites, an additional €86,400 (sites x eggs x costs) would have to be funded. The analysis of additional contaminants would further increase the price per sample and the amount of funding needed, according to the additional effort necessary for the analysis. The organization of the sampling, the coordination of the work between participating labs, data compilation and analysis as well as reporting would require a half-time position for a scientific assistant (biologist or chemist) with costs of €31,200/year. Further costs may originate from meetings of the cooperating partners.

Table 6: Estimated annual total costs of the EcoQO monitoring around the North Sea as proposed by OSPAR (2007), for sites sampled within the TMAP program (Becker & Dittmann 2009), sites sampled specifically in this pilot project and further sites proposed for future monitoring. OC = Oystercatcher, CT = Common Tern, AT = Arctic Tern.

Position	State	Site	Species	Costs (€)		
				TMAP	This pilot project	Pro-posed
Sampling, transport, chemical analysis	UK	Moray Firth	OC, CT			9,600
		Forth estuary	OC, CT			9,600
		Tees estuary (Middlesbrough)	OC, CT		4,800	4,800
		Humber estuary	OC, CT			9,600
		Thames estuary	OC, CT			9,600
	Belgium	Zeebrugge	CT		4,800	
	Netherlands	Scheldt estuary (Terneuzen)	CT		4,800	
		Rhine delta	CT			4,800
		Balgzand	OC, CT	9,600		
		Griend	OC, CT	9,600		
		Julianapolder	OC	4,800		
		Schiermonnikoog	CT	4,800		
		Delfzijl	OC, CT	9,600		
	Germany	Dollart	OC	4,800		
		Baltrum	CT	4,800		
		Jade (Minsener Oog)	CT	4,800		
		Jade (Mellum)	OC	4,800		
		Elbe estuary (Hullen)	OC	4,800		
		Elbe estuary (Neufelderkoog)	CT	4,800		
		Trischen	OC, CT	9,600		
		Hallig Hooge	OC, CT	9,600		
	Denmark	Langli	OC	4,800		
		Mandø	AT		4,800	
	Sweden	Strömstad	OC, AT		9,600	
	Norway	Haga	OC		4,800	
		Presteskjaer	AT		4,800	
Project coordination, data analysis, reporting						31,200
Annual meeting of cooperation partners						3,500
Subtotal				91,200	38,400	82,700
Total			212,300			

Appendix 1: Analytical Methods

A.1.1 Laboratory ICBM-Terramare, Wilhelmshaven, Germany

For the chemical analytics the egg contents were homogenized and frozen at -18 °C until analysis.

For the determination of the organochlorines (11 organochlorine pesticides and 62 PCB congeners) 2 g egg-homogenate were spiked with an internal standard solution, dried with sodiumsulfate extracted in n-hexane, dichlormethane (8:2) by a silica gel column, evaporated and re-dissolved in 250 µl isooctane. For the detection of the substances, an Agilent 6890 was used, coupled to a mass-selective detector Agilent 5973, cold injection system (KAS, Gerstel), with helium as the carrier gas. Measurements were performed in the SIM mode using an electron impact ionization. For separation a HT-5-column with a length of 25 m was used. The qualification and the quantification of the pesticides and PCBs were performed according to Büthe & Denker (1995). 41 of 62 PCB congeners could be determined separately. The remaining 21 congeners were detected together (nine peaks with two PCBs and one peak with three PCBs). The detection limits were within the range 0.1 ppb to 0.9 ppb. Concentrations are given in ng/g fresh mass.

To determine mercury concentrations in 2008 and 2009, 100 mg egg homogenate were prepared for mercury determination with a mixture of nitric acid, chloric and perchloric acid in a partly closed test tube due to Kruse (1979). An atomic absorption spectrometer (FIMS-400, Perkin Elmer) with an integrated flow injection module of the FIAS series was used for the measurement. The determination limit was 0.1 ng/g. Since 2010, the mercury measurement has been accomplished with a DMA80 Direct Mercury Analyzer by MLS GmbH. Compared to the former device, this mercury analyzer had the advantage to analyze samples in a shorter time without sample preparation and no waste disposal.

Approximately 30mg egg-homogenate were weighed and introduced into the sample boat. The sample was initially dried and burned at 750 °C in an oxygen current. The high temperature decomposed mercury compounds and elementary mercury was set free. Mercury vapors were collected on a gold amalgamation trap and subsequently desorbed for quantification. Mercury content was determined using atomic absorption spectrometry at 254 nm. The automatical process to analyze 40 samples took about 4 hours. All samples were analyzed in duplicate.

The system was based on two Measuring methods: EPA 7473 and ASTM D-6722-01 (MLS 1998, 2006), giving the concentration in ng/g fresh mass with a detection limit of 0.003 ng total mercury. To certify results, parallel reference materials DORM-2 and DORM-3 (NRC-CNRC Canada) were measured. We intercalibrated both methods of mercury determination with 10 eggs measured by both methods. Mean values were almost identical (151 ± 23 vs. 150 ± 17 ng/g; means ± se; paired t-test) and values were highly positively correlated ($r = 0.979$, $p < 0.001$).

A.1.2 Laboratory Centre of Ecology and Hydrology, Lancaster, UK

To determine the concentrations of organochlorines, a sub- sample of each egg (~1 g) was thawed, weighed accurately, ground with sand and dried with anhydrous sodium sulphate. Each sample was spiked with labelled recovery standards (^{13}C OCs and ^{13}C PCBs) and soxhlet extracted in DCM for 16 h. A small portion of the extract (5 ml) was evaporated to zero volume and the lipid content was determined gravimetrically. The remaining of the extract was cleaned using automated size exclusion chromatography followed by deactivated alumina column. The extract was then spiked with labelled internal standards (^{13}C OCs and ^{13}C PCBs) and 20 µl of the sample was injected into a GC-MS with programmable temperature vaporization (PTV) inlet. The PTV injector was kept at 20 °C for 0.6 min, and heated to 400 °C at a rate of 700 °C/min and kept at 400 °C for 5 min. Then the temperature was reduced to 350 °C/min at a rate of 10 °C/min. The GC-MS had a 50 m HT8 column (0.22 mm internal diameter and 0.25 µm film thickness, SGE Milton Keynes, UK) and the carrier gas was helium (2.0 ml/min). The temperature programme was: isothermal at 50 °C for 2 min, 45 °C/min to 200 °C, 1.5 °C/min to 240 °C and was held at 240 °C for 12.5 min, 2 °C/min to 285 °C, 50 °C/min to 325 °C and isothermal at 325 °C for 10 min. Measurements were performed in the SIM mode using electron impact ionization mode. Residues were quantified using internal standard method and also calibration curves of the standard PCBs (Greyhound Ltd, Birkenhead, UK) and were recovery corrected. The method limits of detection (LoDs) were calculated as the instrumental LoD multiplied by the dilution factor and divided by the sample weight used. LODs ranged between 0.5-0.9 ng/g wet weight and 0.5-2.2 ng/g wet weight for PCBs and OCs, respectively.

To measure mercury concentration, a 1g wet weight sample of the egg was digested in 10 ml of 70 % ultrapure nitric acid (Baker, Ultrex II) in a microwave digestion system at 200 ºC for 15 minutes. The digested samples were then made up to an initial digest volume of 25 ml using ultrapure water (Millipore, MilliQ). Samples were further diluted ten-fold using ultrapure water immediately prior to analysis by inductively couple plasma mass spectrometry (ICPMS) using a Perkin Elmer DRCII ICPMS. Hg was measured using the ICPMS operating under standard conditions after the addition of Au as a stabiliser (as chloride at 5 mg/l). The moisture content of the sample was determined by drying a 0.5 g sub-sample at 70 ºC for a minimum of 24 hours. Dry weight concentrations were calculated based upon the wet weight of the analysed sample and the moisture content of the sub-sample. Instrumental limits of detection (LoDs) for each element were calculated as 4.03 times the standard deviation of six replicate blank determinations. The method LoDs were calculated as the instrumental LoD multiplied by the dilution factor and digest volume divided by the minimum sample weight used. Certified reference materials (CRMs) were run alongside the egg samples. Certified reference materials TORT-2 and DOLT-4 (NRCC-CNRC, Canada) were used to verify the Hg microwave digestion and analysis; a recovery of 121 % and 100 % respectively was achieved when compared with reported certified values.

A.1.3 Intercalibration between the two laboratories

We exchanged 10 eggs (5 Oystercatcher eggs, Mellum, and 5 Common Tern eggs, Elbe estuary, from 2007) to intercalibrate the different methods used by the German and British team. The results were satisfactory and are presented in Table A.4.

We calculated calibration factors to convert the concentrations from the British eggs analysed by the Centre of Ecology and Hydrology to levels comparable with those from eggs from all other sites, analysed by ICBM-Terramare. These calibration factors were for ΣPCB (based on 23 congeners) 1.207, for HCB 0.696 and for ΣDDT 0.789. For Hg and for γ-HCH, no differences were found between Lancaster and Wilhelmshaven, so a calibration factor of 1.0 was applied.

The sum of the 26 PCB-congeners which were analysed by both labs accounted for 77.6 % of the sum of the 62 PCB-congeners analysed by ICBM-Terramare (n = 371 Common and Arctic Tern eggs, 2008-2010). To be able to compare the summarized PCB-levels of British eggs with those on the mainland and with the EcoQO determined for the continent, the summarized PCB-concentrations of British eggs were multiplied by a factor of 1.29.

Apendix 2: Comparison of Common Tern and Arctic Tern eggs

To answer the question whether Common Tern and Arctic Tern egg contamination is comparable, we investigated the contaminants in eggs of both species in 2006 at the same site, at Hallig Hooge, Schleswig-Holstein Wadden Sea. Both tern species were contaminated with environmental chemicals in the same order of magnitude (Table A.6). No significant differences were found in the industrial chemicals, the heavy metal mercury, ΣPCB and HCB.

In the insecticides, however, except the Σchlordanes, which are not of further interest in this report, higher levels of ΣDDT and ΣHCH were found in Arctic Tern eggs. These elevated concentrations of insecticides might be related to the dietary differences between the two species (e.g. Frick & Becker 1995), or to different areas used on their migration routes. On the other hand, the interspecific differences were relatively low and the ΣHCH levels close to the determination limit, resulting in a higher inaccuracy of measurements.

In consequence, both tern species can be used mutually as indicators of environmental contamination using the egg concentrations of chemicals.

Tables Appendix

Table A.1: Coordinates of the sampling sites with state, short characteristics and species sampled. OC = Oystercatcher, CT = Common Tern, AT = Arctic Tern. Cf. Fig. 1 for location of sites. *: Site covered by the TMAP monitoring program (Becker & Dittmann 2009).

State	Site	Coordinates	Characteristics	Species sampled
UK	Middlesbrough	54.36N, 01.13W	Tees estuary	CT
Belgium	Zeebrugge	51.22N, 03.13E	mainland coast	CT
The Netherlands	Terneuzen	51.20N, 03.48E	Scheldt estuary	CT
	Balgzand*	52.54N ,04.53E	mainland coast	OC, CT
	Griend*	53.15N, 05.15E	island	OC, CT
	Julianapolder*	53.24N, 06.20E	mainland coast	OC
	Schiermonnikoog*	53.24N, 06.10E	island	CT
	Delfzijl*	53.20N, 06.58E	Ems estuary, industrial area	OC, CT
Germany	Dollart*	53.16N, 07.14E	Ems estuary	OC
	Baltrum*	53.44N, 07.22E	island	CT
	Minsener Oog*	53.46N, 08.00E	island	CT
	Mellum*	53.43N, 08.09E	island	OC
	Hullen*	53.51N, 09.03E	Elbe estuary	OC
	Neufelderkoog*	53.54N, 08.58 E	Elbe estuary	CT
	Trischen*	54.04N, 08.40E	island	OC, CT
	Hallig Hooge*	54.34N, 08.32E	island	OC, CT, AT
Denmark	Langli*	55.31N, 08.19E	island	OC, AT
	Mandø*	55.16N, 08.33E	island	OC, AT
Sweden	Strömstad	58.53N, 11.09E	mainland coast	OC, AT
Norway	Haga	58.93N, 05.62E	island and mainland coast	OC
	Presteskjaer	58.57N, 05.36E	islet	AT

Table A.2: Egg samples collected and analyzed in 2008/2009/2010 (total: 720 eggs). Eggs from Middlesbrough, UK, were analyzed at the Centre of Ecology and Hydrology, Lancaster, all other eggs were analyzed at ICBM-Terramare,Wilhelmshaven, D. *: Site covered by the TMAP monitoring program (Becker & Dittmann 2009).

Site	Common Tern	Arctic Tern	Oystercatcher
Middlesbrough, UK	- / 10 / 10		
Zeebrugge, B	10 / - / 10		
Terneuzen, NL	10 / - / -		
Balgzand, NL*	10 / 10 / 10		10 / 10 / 10
Griend, NL*	10 / 10 / 9		10 / 10 / 10
Julianapolder, NL*			10 / 10 / 7
Schiemonnikoog, NL*	10 / 10 / 9		
Delfzijl, NL*	10 / 10 / 6		10 / 10 / 10
Dollart, D*			10 / 10 / 10
Baltrum, D*	10 / 10 / 10		
Jade (Minsener Oog, Mellum), D*	10 / 10 / 10		10 / 10 / 10
Elbe (Hullen, Neufelderkoog), D*	10 / 10 / 10		10 / 10 / 10
Trischen, D*	10 / 10 / 10		10 / 10 / 3
Hallig Hooge, D*	10 / 7 / 10		10 / 10 / 10
Langli, Mandø, DK*		10 / 10 / -	10 / 4 / 4
Stroemstad, S	10 / 10 / 10		10 / 2 / 9
Haga, N			10 / 10 / 10
Presteskjaer, N		10 / 10 / 10	

Table A.3: Methods for the determination of mercury and organochlorine concentrations at the laboratories Terramare at Wilhelmshaven, D, and the Centre of Ecology and Hydrology (CEH) at Lancaster, GB.

Substance group	Analytical step	Wilhelmshaven	Lancaster
Hg	Digestion	Nitric/chloric/ perchloric acid	70% nitric acid
	Analysis	Atomic absorption spectrometer (FIMS-400)	ICPMS
	Reference Materials	CRMs	CRMs
	Estimations	double	single
	Determination limits (ww)	0.1 ng/g	0.5-2.2 ng/g
Organochlorines	Extraction	dried, column extracted	soxhlet extracted
	Clean-up	Silica gel column, eluted	size exclusion chromatography and alumina
	Internal standards	yes	yes
	Detection	GC-MS with CIS, HT5 column	GC-MS with PTV, HT8 column
	Estimations	double	single
	Correcting calibration curves	yes	yes
	Determination limits	0.1 – 0.9 ng/g	0.5-2.2 ng/g

Table A.4: Mean concentrations of contaminants ± SD in 10 Common Tern eggs analyzed both by the Centre of Ecology and Hydrology, Lancaster, UK, and ICBM-Terramare, Wilhelmshaven, Germany for intercalibration purposes. P values are given according to t-tests for related samples; r = Pearson correlation coefficients (all $p < 0.001$). [1)] ΣPCB based on 23 congeners. The calibration factor was assumed to be 1.0 for Hg and γ-HCH whose concentrations did not differ significantly between Lancaster and Wilhelmshaven.

Contaminant	Lancaster	Wilhelmshaven	p	r	Calibration factor
Hg	492 ± 117	494 ± 113	n.s.	0.99	1.000
ΣPCB[1]	878 ± 216	1060 ± 271	0.037	0.99	1.207
HCB	30.3 ± 10.1	21.1 ± 6.7	0.013	0.98	0.696
ΣDDT	490 ± 161	387 ± 131	0.009	0.99	0.789
γ-HCH	2.8 ± 2.0	0.0 ± 0.0	n.s.	-	1.000

Table A.5: PCB congeners analyzed in Lancaster, GB, and Wilhelmshaven, D. Congeners analyzed by both labs are marked grey. Most toxic congeners for which toxic equivalence factors (TEFs) have been developed are written in red.

Contaminant	Lancaster	Wilhelmshaven	Contaminant	Lancaster	Wilhelmshaven
PCB 8	X		PCB 153	X	X
PCB 18	X		PCB 155		X
PCB 28	X	X	PCB 156	X	X
PCB 29	X		PCB 157	X	X
PCB 31	X		PCB 158/129		X
PCB 47/48		X	PCB 160/3/4	X	X
PCB 52	X	X	PCB 166		X
PCB 64		X	PCB 167	X	X
PCB 66		X	PCB 169	X	X
PCB 70		X	PCB 170	X	X
PCB 74		X	PCB 171	X	X
PCB 77	X		PCB 172		X
PCB 81	X		PCB 174	X	X
PCB 84/92		X	PCB 175/187		X
PCB 85		X	PCB 177		X
PCB 87/115		X	PCB 178		X
PCB 95		X	PCB 180/193	X	X
PCB 99		X	PCB 183	X	X
PCB 101/90	X	X	PCB 187	X	
PCB 105	X	X	PCB 189	X	X
PCB 107		X	PCB 190		X
PCB 110		X	PCB 194	X	X
PCB 114	X	X	PCB 195		X
PCB 118	X	X	PCB 196/203		X
PCB 123	X	X	PCB 199	X	X
PCB 126	X	X	PCB 201	X	
PCB 128	X	X	PCB 202		X
PCB130		X	PCB 205	X	
PCB 132/146		X	PCB 206	X	
PCB 138	X	X	PCB 209	X	
PCB 141	X	X	Total	37	62
PCB 149	X	X			

Table A.6: Comparison of contaminants (ng/g) in Common Tern and Arctic Tern eggs (n=10 per species), sampled in 2006 at Hallig Hooge, Schleswig-Holstein Wadden Sea. T-tests on logarithmic values.

Chemicals	Common Tern	Arctic Tern	p
Hg	312.5 ± 53.7	290.4 ± 56.6	n.s.
ΣPCB	394.9 ± 118.9	379.8 ± 62.1	n.s.
HCB	8.5 ± 26.0	8.4 ± 1.4	n.s
ΣDDT	40.3 ± 10.9	51.7 ± 10.5	<0.05
ΣHCH	0.8 ± 0.5	3.4 ± 0.4	<0.001

Table A.7: Effects of species, site and year and their interactions on the concentration of environmental pollutants measured in Oystercatcher and tern eggs in 2008-2010 (GLM, based on logarithmic values). Partial Eta² shows the proportion of the variance explained by a factor. Error(df) = 643 for all chemicals.

Chemical	Factor	df	F	p	Eta²
Hg	Species	1	170.9	0.000	0.21
	Site	19	45.1	0.000	0.57
	Year	2	34.6	0.000	0.10
	Species*Site	8	4.4	0.000	0.05
	Species*Year	2	23.4	0.000	0.07
	Site*Year	31	3.4	0.000	0.14
	Species*Site*Year	12	3.9	0.000	0.07
ΣPCB	Species	1	106.8	0.000	0.14
	Site	19	83.6	0.000	0.71
	Year	2	23.9	0.000	0.07
	Species*Site	8	11.9	0.000	0.13
	Species*Year	2	2.2	0.115	0.01
	Site*Year	31	4.8	0.000	0.19
	Species*Site*Year	12	2.2	0.011	0.04
HCB	Species	1	249.4	0.000	0.28
	Site	19	96.2	0.000	0.74
	Year	2	116.1	0.000	0.27
	Species*Site	8	30.4	0.000	0.27
	Species*Year	2	2.1	0.129	0.01
	Site*Year	31	3.8	0.000	0.16
	Species*Site*Year	12	3.9	0.000	0.07
ΣDDT	Species	1	186.1	0.000	0.22
	Site	19	76.3	0.000	0.69
	Year	2	22.3	0.000	0.07
	Species*Site	8	19.3	0.000	0.19
	Species*Year	2	1.9	0.148	0.01
	Site*Year	31	3.4	0.000	0.14
	Species*Site*Year	12	6.8	0.000	0.11
ΣHCH	Species	1	59.6	0.000	0.09
	Site	19	42.8	0.000	0.56
	Year	2	460.8	0.000	0.59
	Species*Site	8	6.5	0.000	0.08
	Species*Year	2	18.7	0.000	0.06
	Site*Year	31	23.7	0.000	0.53
	Species*Site*Year	12	7.1	0.000	0.12

Figures Appendix

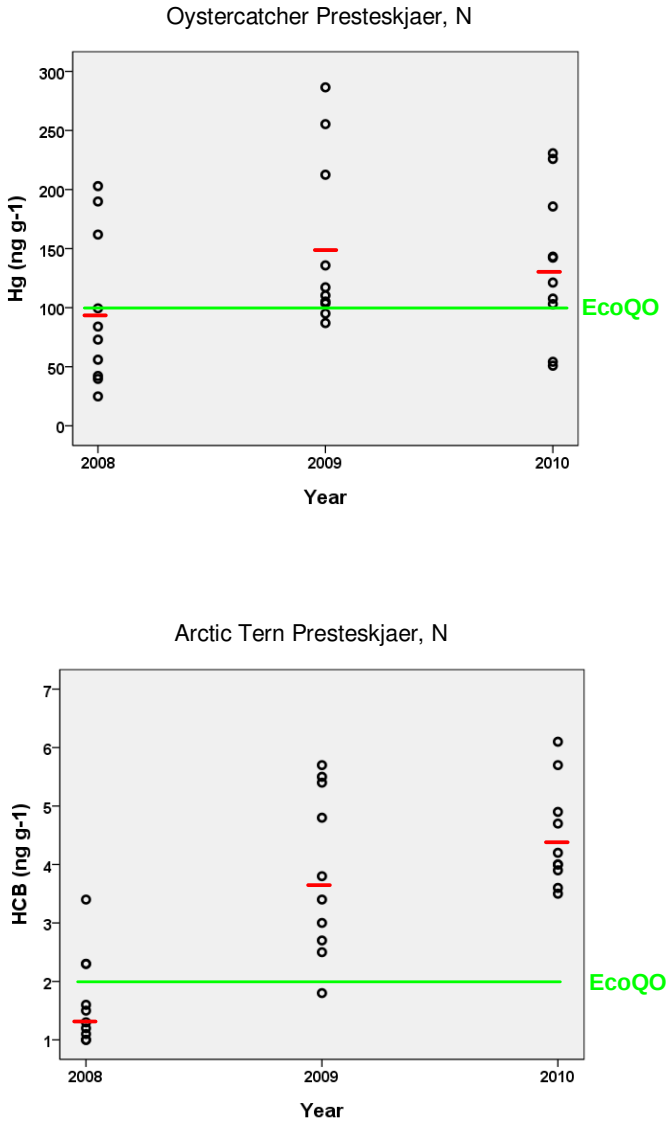

Fig. A.1: Two examples of the fluctuation of chemicals' levels around the EcoQO in three years: Mercury in the Oystercatcher (top) and HCB in the Arctic Tern from Presteskjaer, Norway (bottom). The individual egg levels each representing one female are shown by circles, mean values by red lines and the EcoQO by the green line (for Hg in the Oystercatcher 100 ng/g, for HCB 2 ng/g). In both examples the EcoQO has been met (mean below the EcoQO-level) in the first year 2008, whereas in 2009 and 2010 the mean was above the level.

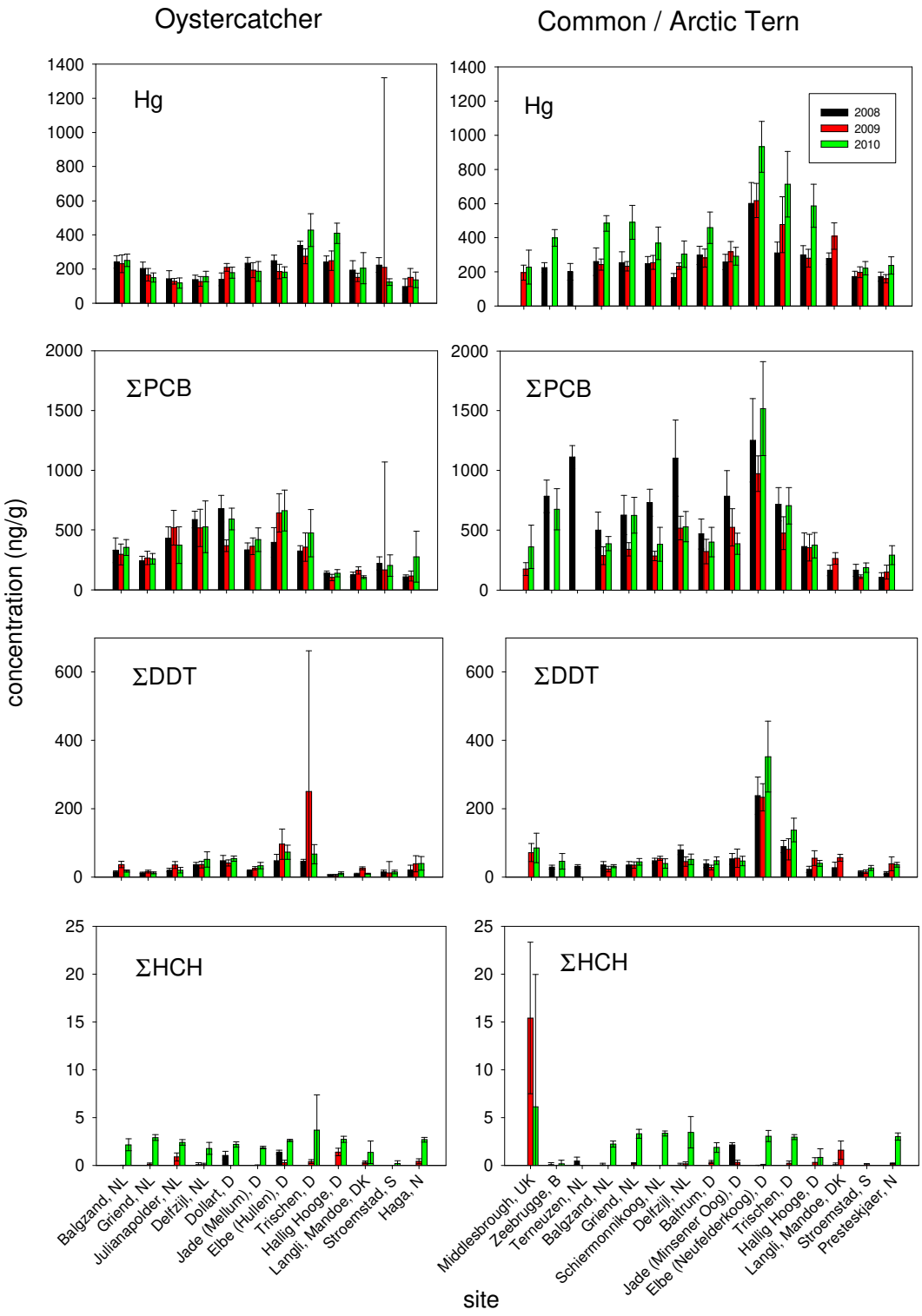

Fig. A.2: Mean contamination ± 95%-confidence interval of Oystercatcher and Common Tern with Hg, ΣPCB (26 congeners), ΣDDT and ΣHCH (α-, β- and γ-HCH, only α- and γ-HCH at Middlesbrough) in the study years 2008-2010 at different sampling sites around the North Sea. For sample sizes per year, see Table A.2. For HCB see Fig. 2

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