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RESEARCH ARTICLE

Large-scale spatial pollution patterns around the North Sea indicated by coastal bird eggs within an EcoQO programme

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Abstract To categorize the marine environmental health status, the Oslo and Paris commissions have recently formulated Ecological Quality Objectives (EcoQOs) for many ecological features including the contamination of coastal bird eggs with mercury and organochlorines. In this study, we describe spatial and temporal patterns of egg contamination around the North Sea and compared them to the EcoQOs. Concentrations of mercury, polychlorinated biphenyl (ΣPCB) congeners, dichlorodiphenyltrichloroethane (ΣDDT) and derivatives, hexachlorobenzene (HCB) and hexachlorocyclohexane (ΣHCH) isomers were analysed in two tern species (*Sterna hirundo* and *Sterna paradisaea*) and Oystercatcher (*Haematopus ostralegus*) eggs collected between 2008 and 2010 in a total of 21 sites in seven countries surrounding the North Sea. Hg, ΣPCB and HCB were highest in the southern sites, while ΣDDT and ΣHCH concentrations were greatest in eggs from the western North Sea and the Elbe estuary. There were rarely any consistent decreases over time for any compounds. In the terns, Hg, HCB and ΣHCH increased at most sites, ΣPCB and ΣDDT in Sweden and Norway. In the Oystercatcher, HCB and ΣHCH increased at more than the half of the sites, ΣPCB, ΣDDT and Hg at several German sites. In the terns, Hg, ΣPCB and ΣDDT exceeded the EcoQO in all, HCB in most years and sites. At most sites, ΣHCH fulfilled the EcoQO in some study years. In the Oystercatcher, Hg, ΣPCB and ΣDDT exceeded the EcoQO in all or most years and sites. HCB and ΣHCH fulfilled the EcoQO in some or all years at most sites. The EcoQO was exceeded most frequently in estuaries. We conclude that EcoQOs are suitable for drawing contamination patterns of the coastal North Sea in an easily understandable

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manner, offering the opportunity to harmonize the EcoQOs with coordinated environmental monitoring programmes.

Keywords EcoQO · Mercury · Organochlorines · North Sea · Monitoring · POPs

Introduction

The Oslo and Paris commissions (OSPAR), uniting currently 15 governments of the western coasts and catchments of Europe, together with the European community, 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 and to conduct quality assessments of the marine environment (Stagg 1998; Hagger et al. 2006). Under the framework of the Joint Assessment and Monitoring Programme (JAMP) an upgrade of the Coordinated Environmental Monitoring Programme (CEMP) is currently under development and will incorporate various chemical parameters. Current environmental health is evaluated using Ecological Quality Objectives (EcoQOs) 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 approximate the expected status without or after a complete stop of any further input of anthropogenic pollutants. 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. In this sense, a good EcoQO 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 eggs of coastal birds are suitable indicators of the environmental pollution with anthropogenic contaminants such as heavy metals and organochlorines (e.g. Furness 1993; Becker 2003; Becker et al. 2003; Becker and Dittmann 2009). Bird eggs reunite four major advantages for the monitoring of environmental pollution: (1) Sampling is logistically simple and cost-effective. (2) Biomagnification effects allow to record substances even if their absolute concentrations in the environment are low. (3) A low variation of values per site and species results in a high power of statistics. (4) A comparatively well-known ecology of the species enables a reliable interpretation of results.

After toxic effects in historical times, causing population collapses in bird populations, pollution of coastal bird eggs with Hg and organochlorines has been monitored for decades in different countries surrounding the North Sea or in the Baltic Sea (Pereira et al. 2009; Becker and Dittmann 2009; Bignert et al. 2011), but these studies involved different bird species and often covered only single sites. This is partially due to the fact that several of the species studied have a restricted distribution in the North Sea area (BirdLife International 2004), hampering the creation of a large-scale overview of the contamination of the North Sea. In contrast, the recent EcoQO approach of OSPAR has emphasized the necessity to reveal the pollution patterns of the coastal North Sea with a hitherto unique density of sampling sites, covering most countries surrounding the North Sea which are situated within the areas of responsibility of OSPAR.

In 2005, following advice from the International Council of the Exploration of the Sea, OSPAR agreed on the threshold concentrations proposed as EcoQOs for Hg and organochlorines in the eggs of coastal birds. Common Tern (*Sterna hirundo*) or Arctic Tern (*Sterna paradisaea*) and Eurasian Oystercatcher (*Haematopus ostralegus*) were chosen as indicator species. Whereas the terns feed predominantly on small pelagic fishes and crustaceans (Becker and Ludwigs 2004), the diet of the oystercatcher consists mainly on mussels and worms (Cramp et al. 1983). Thus, the oystercatcher represents a slightly lower trophic level than the terns, but nevertheless, both species are considered as top predators in the marine food chain. They are widely distributed throughout the coastal zones of the North Sea and other regions covered by the OSPAR convention (BirdLife International 2004) and enable a spatially large-scale sampling. Contamination in Common Tern eggs has already been studied throughout the Wadden Sea (Becker and Dittmann 2009), and contamination levels in the eggs of Arctic terns are similar to that in Common Tern eggs in the same area; Arctic Tern is therefore considered to be a suitable alternative to the Common Tern in areas where the latter is rare (Dittmann et al. 2011). EcoQOs have been defined as substance-specific and, in the case of Hg, also species-specific threshold concentrations that may be considered "background" in eggs from relatively pristine sites; they therefore are a baseline against which current pollution levels in eggs at different sites can be compared (OSPAR 2007a, b; Dittmann et al. 2011).

The current study was a pilot to provide the first assessment of the extent to which the proposed coastal bird egg EcoQuos for Hg and four groups of chlorinated persistent organic pollutants (POPs), i.e. polychlorinated biphenyls (PCBs) (different congeners), dichlorodiphenyltrichloroethane (DDT) and metabolites, hexachlorobenzene (HCB) and hexachlorocyclohexane (HCH) isomers were met. These contaminants were measured in the eggs of Common

Tern or Arctic Tern and Oystercatcher collected at a total of 21 sites in seven states surrounding the North Sea. For the majority of sites, sampling took place annually in 2008, 2009 and 2010. The methods of sampling and analysing were performed according to internationally recognized standards (JAMP, OSPAR). The arithmetic mean concentrations of pollutants in the fresh egg content per site and year were compared with the substance specifically defined EcoQOs which were defined through threshold concentrations (Dittmann et al. 2011). In this sense, an EcoQO was achieved if the mean concentration of a substance group was equal or below the threshold value, otherwise the EcoQO had to be seen as exceeded and, in consequence, as not achieved: Hg concentrations in tern eggs were compared with an EcoQO of 160 ng/g, for those in Oystercatcher eggs, the EcoQO was 100 ng/g. For the organochlorines, the EcoQOs defined were equal for terns and the oystercatcher and were as follows: sum of 62 PCB congeners (ΣPCB), 20 ng/g; HCB, 2 ng/g; sum of DDT and its metabolites (∑DDT), 10 ng/g; sum of HCH isomers (ΣHCH), 2 ng/g.

The specific aims of the study were to:

1. Document spatial contamination patterns for the North Sea as revealed by pollution concentrations in the eggs of coastal birds;
2. Determine whether there was significant short-term/inter-year variation in contamination,
3. Assess the extent to which concentrations achieved the EcoQO, and
4. Recommend future monitoring strategies based on the findings of the current study.

Methods

Study species

Common and Arctic Terns are both considered to be income breeders as eggs are formed largely from nutrients incorporated by the female in the 2 weeks of courtship feeding by the male mate immediately before egg-laying (Wendeln and Becker 1996; Wendeln 1997). In the breeding season, Common and Arctic Terns forage mostly within 10 and 3 km, respectively, of their breeding colonies (Cramp 1985; Becker et al. 1993) and so both species are characterized as inshore feeders. The terns are long distance migrants: Common Terns winter in west/southwest Africa, Arctic Terns in the Antarctic (Cramp 1985; Becker and Ludwigs 2004). The Oystercatcher is a capital breeder, eggs being formed from substances stored in the body over longer time periods. The species is a resident breeder over large parts of the North Sea (Koffijberg et al. 2006). In contrast to the terns, it feeds on macrozoobenthic organisms, such as mussels and worms and may have a slightly smaller feeding range than the terns 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 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 particularly suitable monitors of contamination of the local marine environment.

Sampling sites

In total, 21 coastal sites from across seven countries surrounding the North Sea were chosen as monitoring sites (Fig. 1). According to the requirements of OSPAR (2007a), sites ranged from those remote from industrial development which could be regarded as reference sites to those located on the estuaries of large rivers that drain major industrial conurbations. It was not possible to sample all species at each site, and a summary of which species were sampled at each site is given in Table 1. The sites from Balgzand to Langli have been subject of the Trilateral Monitoring Programme for pollutants in the Wadden Sea conducted since 1998 (TMAP; Becker et al. 2001; Becker and Muñoz Cifuentes 2004; Becker and Dittmann 2009).

Collection of egg samples

Eggs were sampled according to the guidelines of the Joint Assessment and Monitoring Programme (JAMP; OSPAR 1997), the Trilateral Monitoring and Assessment Programme (TMAP; Becker et al. 2001) and Verein Deutscher Ingenieure (VDI) (Verein Deutscher Ingenieure 2009). Ten fresh eggs per species, site and year were taken under license. Since in general, intra-clutch variation is low compared to inter-clutch variation, one egg per clutch was chosen randomly (e.g. Becker et al. 1991). Because contaminant levels in eggs 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. The eggs were kept frozen at −18 °C until they were analysed. The egg's content without shell was homogenized using an Ultra-Turrax, filled into suitable polypropylene cups and frozen at −18 °C until chemical analysis.

Chemical analyses

All egg samples from continental Europe were analysed in one single laboratory, the ICBM-Terramare Wilhelmshaven according to the OSPAR guidelines (OSPAR 1997) with sample preparation following Heidmann (1986). The

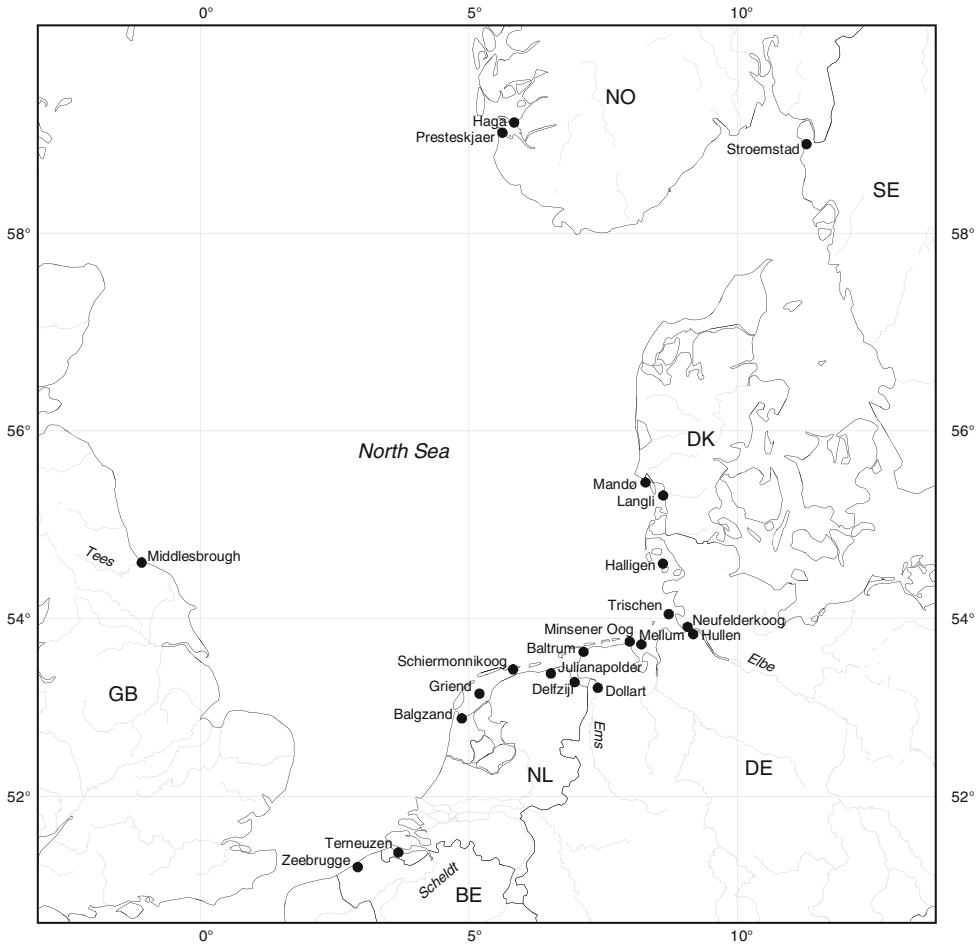

Fig. 1 Sampling sites. For an overview of sample sizes per species and year see Table 1

concentration of total Hg was determined by atomic absorption spectrometry, those of 62 PCB congeners and further organochlorine substances were determined by gas chromatography (Becker et al. 1991, 1998). The substances were analysed by gas chromatography–mass spectrometry detection (Agilent 6890, coupled to a quadruple Agilent 5973), using a 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, an 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 and Denker (1995). Most of the PCBs were baseline-separated during the gas chromatographic separation, but 21 PCBs co-elute in nine peaks. The selection of the 62 PCB congeners (abbreviated to ΣPCB 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 (dichlorodiphenyldichloroethane), and p,p′-DDE (dichlorodiphenyldichloroethylene; ∑DDT= sum of all metabolites), as well as the alpha, beta and gamma isomers of hexachlorocyclohexane (∑HCH). For further details of the chemical analysis, detection limits and quality assessment/quality control measures see Becker et al. (2001) and, respectively, Dittmann et al. (2011).

The egg samples from Middlesbrough were analysed at the Centre of Ecology and Hydrology (CEH), UK. The total Hg content was determined by inductively coupled plasma mass spectrometry, those of organochlorines were analysed by gas chromatography mass spectrometry (Agilent 6890, coupled to a quadruple Agilent 5973). The 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. For details of the chemical analyses see Pereira et al. (2009).

To compare the pollutant levels measured in the UK to those in continental Europe and to the EcoQOs defined, an inter-calibration of both methodological approaches was conducted. We exchanged ten eggs (five Oystercatcher and five Common Tern eggs) to compare the results obtained by the two laboratories. We derived calibration factors to convert the concentrations from the British eggs analysed by CEH to levels comparable with those from eggs from all other sites analysed by ICBM-Terramare (cf. Dittmann et al. (2011) for details). 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 significant differences were found

Table 1 Coordinates of the sampling sites with state, short characteristics, species and number of eggs sampled per year

State	Site	Coordinates	Characteristics	Sample size 2008/2009/2010		
				OC	AT	CT
UK	Middlesbrough	54.36 N, 01.13 W	Tees estuary			–/10/10
Belgium	Zeebrugge	51.22 N, 03.13 E	Mainland coast			10/–/10
The Netherlands	Terneuzen	51.20 N, 03.48 E	Scheldt estuary			10/–/–
	Balgzand[a]	52.54 N, 04.53 E	Mainland coast	10/10/10		10/10/10
	Griend[a]	53.15 N, 05.15 E	Island	10/10/10		10/10/9
	Julianapolder[a]	53.24 N, 06.20 E	Mainland coast	10/10/7		
	Schiermonnikoog[a]	53.24 N, 06.10 E	Island			10/10/9
	Delfzijl[a]	53.20 N, 06.58 E	Ems estuary, industrial area	10/10/10		10/10/6
Germany	Dollart[a]	53.16 N, 07.14 E	Ems estuary	10/10/10		
	Baltrum[a]	53.44 N, 07.22 E	Island			10/10/10
	Minsener Oog[a]	53.46 N, 08.00 E	Island			10/10/10
	Mellum[a]	53.43 N, 08.09 E	Island	10/10/10		
	Hullen[a]	53.51 N, 09.03 E	Elbe estuary	10/10/10		
	Neufelderkoog[a]	53.54 N, 0858 E	Elbe estuary			10/10/10
	Trischen[a]	54.04 N, 08.40 E	Island	10/10/3		10/10/10
	Hallig Hooge[a]	54.34 N, 08.32 E	Island	10/10/10		10/7/10
Denmark	Langli[a]	55.31 N, 08.19 E	Island	10/4/–	10/–/–	
	Mandø	55.16 N, 08.33 E	Island	–/–/4	–/10/–	
Sweden	Strömstad	58.53 N, 11.09 E	Mainland coast	10/2/9		10/10/10
Norway	Haga	58.93 N, 05.62 E	Island and mainland coast	10/10/10		
	Presteskjaer	58.57 N, 05.36 E	Islet		10/10/10	

Cf. Fig. 1 for location of sites

OC Oystercatcher, *CT* Common Tern, *AT* Arctic Tern

[a] Site covered by the TMAP monitoring programme (Becker and Dittmann 2009). Egg samples collected and analysed in 2008/2009/2010 (total, 720 eggs)

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 (Dittmann et al. 2011). The concentrations of chemicals measured are given in nanogrammes per gramme fresh weight of egg content.

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 (Common and Arctic Tern eggs pooled), site and year (2008–2010). Year effects at specific sites were tested for with ANOVA. For inter-year 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. We defined an increase in contamination over the study period 2008–2010 as the case that pollutant concentration was significantly higher in at least one later year compared to an earlier year. A decrease was complementarily defined as the case that a pollutant concentration was 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 detectable throughout the 3 years. The sample size of Oystercatcher eggs from the Danish Wadden Sea was below five in 2008 and 2010, so here no statistical comparison of years was possible to detect temporal changes.

Results

Influence of species, site and year on contaminant concentration

Species, site and year, as well as most of their interactions, were significant factors that explained variation the concentrations of the different contaminants. Site was the strongest source of variation for all contaminants except ΣHCH, followed by species for Hg, HCB and ΣDDT (Table 2). Comparing the arithmetic mean contamination of eggs sampled between 2008 and 2010, pollutant levels measured in tern eggs were 1.7-times (ΣPCB) to 2.6-times (HCB) higher than those in Oystercatcher eggs at the same or adjacent sites (Fig. 2).

Spatial and temporal contamination patterns

In Common and Arctic Tern eggs, concentrations of Hg, ΣPCB, HCB and ΣDDT were highest at the inner Elbe estuary. ΣHCH concentrations were clearly elevated in eggs from Middlesbrough (Fig. 2). The lowest mean concentrations for four out of the five contaminant groups were in eggs from Presteskjaer (Hg) or Stroemstad (ΣPCB, ΣDDT and ΣHCH) while the lowest mean HCB concentration was measured in eggs from Terneuzen (Fig. 2). Overall, there was a clear, continuous decrease in Hg, ΣPCB, HCB and ΣDDT concentrations as distance away from the Elbe estuary increased towards the north and an abrupt decrease towards the west (Fig. 2). Spatial patterns of contamination were more substance-specific in Oystercatcher eggs. For Hg and ΣDDT, highest arithmetic mean concentrations were measured in eggs from the Elbe estuary (Neufelderkoog) or its immediate surroundings (Trischen). Eggs from the Elbe estuary also had the second highest HCB concentration, but concentrations were markedly elevated at Delfzijl. Comparatively, high ΣPCB concentrations were found in eggs from the sites from Julianapolder, NL to Trischen, D (Fig. 2). Sites with the lowest contaminant concentrations in eggs were Presteskjaer (Hg), Hallig Hooge (ΣPCB and ΣDDT), the Danish Wadden Sea (HCB) and Stroemstad (ΣHCH). There was a decrease in Hg, ΣPCB and HCB concentrations with increasing distance from the river Elbe or the island of Trischen (Fig. 2), but this was less pronounced than in the Common Tern. The significance of the effect of year in explaining temporal variation in contaminant concentrations was greatest for ΣHCH (Table 2). For the sites where both the Oystercatcher and a tern species were sampled, mean contamination of the Oystercatcher per site and year was strongly and positively correlated with that of the terns in all substance groups (N=27; Hg: r_S=0.477, p=0.014; ΣPCB: r_S=0.703, p<0.001; HCB: r_S=0.812, p<0.001; ΣDDT: r_S=0.653, p<0.001; ΣHCH: r_S=0.598, p=0.001).

Table 2 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)

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

Partial Eta² shows the proportion of the variance explained by a factor. Error(df)=643 for all chemicals. In the analysis, the two adjacent Norwegian sites were treated as one site

In the tern species, Hg, HCB and ΣHCH contamination increased in the three study years at most sites. ΣPCB and ΣDDT concentrations in eggs increased at Stroemstad and Presteskjaer and at one or several German sites, whereas they decreased at several Dutch sites.

In the Oystercatcher, HCB and ΣHCH concentrations also increased at more than the half of the sites. As in the terns, ΣPCB concentrations in eggs increased at Hullen

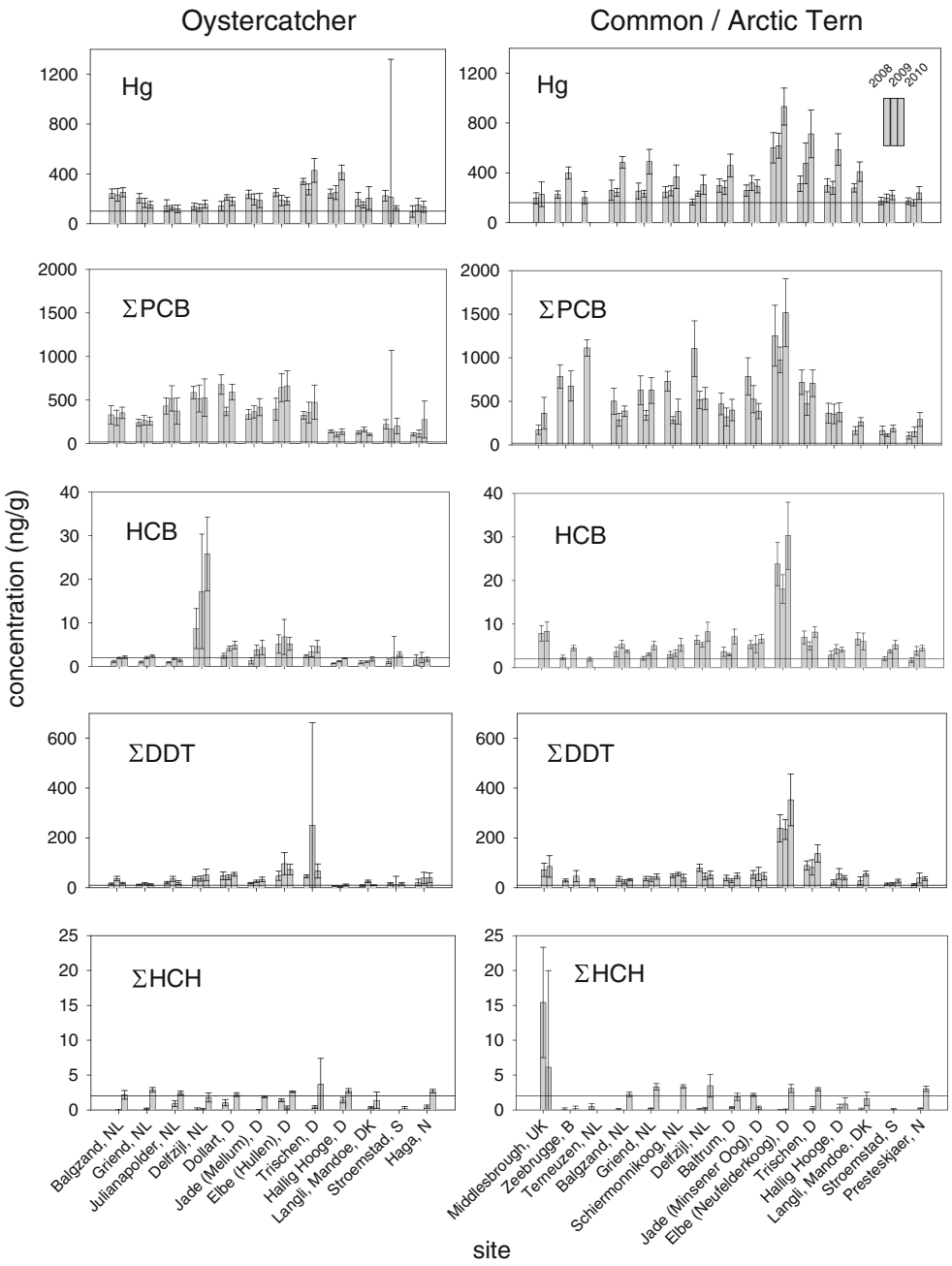

Fig. 2 Mean concentrations± 95 % confidence interval of Hg and organochlorines in eggs of Oystercatcher and terns during the study period 2008–2010 at different sampling sites around the North Sea. The left columns represent the values from 2008, the central columns those of 2009 and the right column those of 2010. The *horizontal lines* show the EcoQO, respectively. Sampling sites around the North Sea are anti-clockwise listed in the order of their position on the coast, beginning with the westernmost site. For an overview of sample sizes per year, see Table 1. If 95 % confidence intervals do not overlap concentrations differ significantly with $p<0.05$

(Elbe estuary) and ΣDDT increased at several German sites. In contrast, Hg concentrations increased over time only at two sites in Germany, whereas decreases were recorded at Griend, Hullen, Trischen and Stroemstad. For the other substance groups, decreases in concentration were only found at single sites. In summary (Table 3), decreases in contamination over time were patchy on sites. Increases were generally evident across sites for HCB and HCH in both the terns and the Oystercatcher and for Hg in the terns. For other contaminants, no such general pattern of change over time was detectable across all sites. Hence, there was little evidence for current declines overall in contaminants (Fig. 2; Table 3).

The interaction term between site and year was also significant for both terns and Oystercatcher in explaining variation in the concentrations of each contaminant.

Concentrations in eggs relative to the EcoQO values

Mercury, ΣPCB and ΣDDT exceeded the EcoQOs in tern eggs in all three study years at all sites. HCB remained below the 2 ng/g EcoQO only in 2008 at Zeebrugge (the only study year at that site) and in 1 year (2008) at Presteskjaer. At all sites except Middlesbrough and Elbe, ΣHCH fulfilled the EcoQO at least in some study years (Fig. 3).

Table 3 Overview of significant temporal changes in pollutant concentrations between 2008 and 2010 (cf. Table 2 and Fig. 2; results from post hoc Scheffé tests or, respectively, *t* tests if only data from 2 years available)

species	pollutant	Middlesbrough, UK	Zeebrugge, B[a]	Terneuzen, NL	Balgzand, NL	Griend, NL	Julianapolder, NL	Schiemonnikoog, NL	Delfzijl, NL	Dollart, D	Baltrum, D	Minsener Oog,, D	Mellum, D	Neufelderkoog, D	Hullen,, D	Trischen, D[b]	Hallig Hooge, D	Langli, Mandoe, DK[a]	Stroemstad, S[b]	Haga, N	Presteskjaer, N
Oystercatcher	Hg				=	-	=		=	+			=		-	-	+		=	=	
	∑PCB				=	=	=		=	±			=		+	+	-		=	=	
	HCB				+	+	±		+	+			+		=	=	+		=	=	
	∑DDT				±	=	±		=	=			+		+	=	+		=	=	
	∑HCH				+	+	+		+	±			+		±	+	+		+	+	
Common/Arctic Tern	Hg	=	=		+	+		+	+		+	=		+		+	+	=	=			+
	∑PCB	=	=		-	±		-	-		=	-		+		±	=	=	+			+
	HCB	=	=		+	+		+	+		+	=		+		+	+	=	+			+
	∑DDT	=	=		=	=		-	-		+	=		=		+	+	=	+			+
	∑HCH	=	=		+	+		+	+		+	-		+		+	=	+	=			+

For more details on the definitions see "Methods"; (+) increase, (−) decrease, (±) fluctuations, (=) no significant changes, grey: site not sampled or data insufficient to determine a temporal development

[a] In the terns, only data from 2 years are available

[b] In the oystercatcher, only data from 2 years are available

In the Oystercatcher, Hg exceeded the EcoQO of >100 ng/g at all study sites in all study years except at Presteskjaer in 2008. ΣPCB exceeded the target 20 ng/g concentration in all years at all sites. The EcoQO for HCB was met in some or even all of the three study years at most sites except at those sites situated in or near the estuaries of the Ems and Elbe; concentrations exceeded the EcoQO in all study years at these sites. ΣDDT concentrations in eggs were below the EcoQO in some years at Hallig Hooge and in the Danish Wadden Sea but exceeded the EcoQO at all other sites. ΣHCH concentrations met the EcoQO at all sites in some years but concentrations have increased at nine of 11 sites.

Discussion

Spatial and temporal patterns of contamination

The spatial pattern of contamination in eggs is consistent with the idea that large rivers draining into the North Sea are major input sources for environmental pollutants. The Elbe appears to be an important input source for Hg, the industrial chemicals ΣPCB and HCB, and remains, even in 2007–2010, a source for the metabolites of the insecticide DDT. There was a clear, continuous decrease in the egg concentrations of Hg, PCB, HCB and, in the case of terns, DDT as distance from the Elbe estuary increased towards the north, and concentrations decreased abruptly towards the west (Fig. 2). This suggests there is a dilution effect on contaminant concentrations, most likely caused by anti-clockwise flowing water currents along the German coast (e.g. Lozán et al. 1990). With regard to other rivers, eggs from German and Dutch sites at or near the river mouth of the Ems had relatively high concentrations of the industrial chemicals HCB and ΣPCB while tern eggs from the Tees site (Middlesbrough, GB) had particularly high concentrations of ΣHCH, and relatively high levels of HCB and ΣDDT; oystercatcher eggs were not sampled at that site. Overall, concentrations of the insecticides ΣDDT and ΣHCH, and also levels of HCB, were greatest in eggs collected from GB and in or near the Elbe estuary, whereas peak concentrations of the industrial chemicals Hg, ΣPCB and HCB particularly occurred in eggs from sites that were in or near estuarine areas on the mainland (cf. Fig. 1). Given the documented inter-site and inter-year differences within species, we cannot entirely exclude that these were potentially additionally affected by some variation in trophic variation of the prey species composition between sites and their trophic level (Cotin et al. 2011; Dänhardt and Becker 2011). However, given the fact that pollution levels of Oystercatcher and terns were strongly and positively correlated despite the differences in their feeding ecology, we consider that the spatial contamination patterns reliably reflected spatial patterns in marine environmental pollution.

In the Wadden Sea, the pattern of spatial contamination from The Netherlands to Denmark largely reflected that documented by Becker and Dittmann (2009). Assuming a mean water content of 75 % and 76 % for Oystercatcher and tern eggs, respectively (Mattig et al. 2000), Hg concentrations in tern eggs between 2008 and 2010 were broadly of

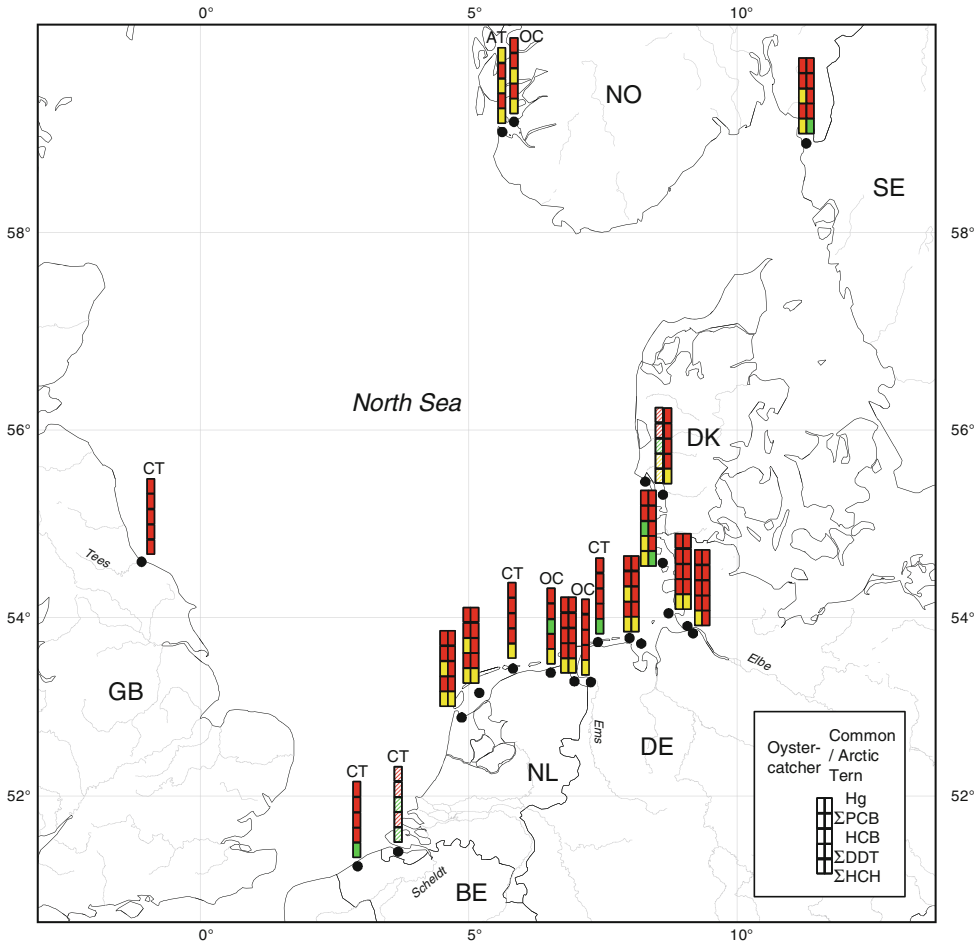

Fig. 3 Fulfillment of EcoQOs for five groups of environmental pollutants in 2008–2010. *Green*, EcoQO fulfilled in all study years; *yellow*, EcoQO fulfilled in some of the study years; *red*, EcoQO not fulfilled in any study year. *Dashed bar*, sample size only in 1 year larger than four. For a complete overview of sample sizes per year, see Table 1. Where Oystercatcher and Terns were sampled at the same or at adjacent sites, *paired stacked bars* are shown with the Oystercatcher being represented by the left and the terns by the right bar. For the further sites where only one species each was sampled, species are abbreviated as *OC* Oystercatcher, *CT* Common Tern and *AT* Arctic Tern

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. The similarities and differences between species may reflect their specific positions in the food web as fish (Gannet, tern) and benthos (Oystercatcher) feeders. Thus, higher pollution values have to be expected in fish feeders, covering a slightly higher trophic level which may result in stronger bioaccumulation effects. 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 only as low as that in Guillemot eggs at the least contaminated sites of the North Sea area. 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 and 1.3–20.4-fold, respectively, lower than that of Baltic Sea Guillemots from Sweden (Bignert et al. 2011) which indicates a stronger pollution of the studied site in the Baltic Sea with this pollutant. The concentration levels of the different contaminants studied remained below the levels considered as toxic for the single components (Becker et al. 2001; Becker 2003; Muñoz Cifuentes et al. 2003; Muñoz Cifuentes 2004) and contaminant concentrations have decreased in the Wadden Sea over the past 20–30 years (Becker and Dittmann 2009). In consequence, a direct relationship to the observed decline in Oystercatcher and tern populations in the Wadden Sea during the past two decades (Blew et al. 2007; CWSS 2010) seems to be rather unlikely. However, it must be stressed that the knowledge about cumulative effects of the chemicals analysed here on the avian organism is very poor and the same is true for those of further and/or new substances which were not subjects of the study.

There were no clear spatial patterns in terms of egg concentrations meeting EcoQO targets. EcoQO targets were fulfilled, at least in some years, at all (Oystercatcher) or almost all (terns) sites for ΣHCH and at seven out of 11 sites for HCB in the Oystercatcher (Fig. 3). In contrast, concentrations remained above the EcoQO for Hg, ΣPCB and ΣDDT at nearly all sites for both species. However, the overall spatial pattern suggests that, where contamination appears to be decreasing over time, the EcoQOs for most substances will probably be met first in the northern and northeastern part of the North Sea. 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).

In terms of spatially larger-scaled temporal changes, there were significant increases between 2008 and 2010 at the majority of sites for Hg and HCB in tern eggs and for HCB and ΣHCH in Oystercatcher eggs. No such general changes were detectable for the other contaminant groups. The increases recorded contrast with the significant mid- and long-term decreases prior to 2008 that were detected at the majority of sites (Becker and Dittmann 2009). The reasons for this apparent anomaly are unknown, and this 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. However, based on experience from long-term temporal trend monitoring (e.g. Bignert et al. 2011), unexplained differences between single years can be considerable without necessarily indicating a general significant change over a period of time. Hence, significant differences between years, reported here, should be interpreted with caution.

It should be remembered that at least in the Wadden Sea the concentrations of several substances analysed here were relatively high prior to 1990, decreased steeply during the 1990s, and then decreased only slowly or remained broadly constant albeit with some fluctuations thereafter (Becker et al. 2001; Bakker et al. 2009; Becker and Dittmann 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. If pollutant concentrations decrease significantly in the future, EcoQO targets are likely to be reached most quickly at sites distant from large rivers and/or in Oystercatcher eggs because of the slightly lower trophic level of this species compared to the terns (cf. Fig. 3).

Suitability of the species studied

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 (Cramp et al. 1983; Cramp 1985; Exo 1992; Becker et al. 1993; Koffijberg et al. 2006). 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. Both are well-known species, and their differing life strategies have pros and cons in terms of environmental monitoring.

As fish-eating top predators that forage mostly at sea, tern species bioaccumulate marine pollutants particularly effectively and are considered as income breeders (Wendeln and Becker 1996; Wendeln 1997). Thus, contaminants in tern eggs originate from 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 (Dittmann et al. 2011). Both species are colonial breeders, enabling the sampling of a sufficient number of eggs in a short time almost without impact to the colony as a whole. However, although tern egg pollution show clear breeding site-specific patterns which correspond well to patterns found in other biota or water (Becker et al. 2001), 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 (Dänhardt and Becker 2011), thereby magnifying the spatial range over which the pollutants measured in the eggs may have originated, increasing the variance and 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 of the year. This enhances the ability to identify local pollution events and hotspots, such as the historical heavy pollution of sediments with HCB in the harbour of Delfzijl (Eggens and Bakker 2001) which is still clearly evident from a strong HCB peak in the eggs of Oystercatchers from Delfzijl in this study (Fig. 2). 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 which might be the reason for the generally weaker spatial variation in this species (Fig. 2). 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 spatial or temporal trends.

Recommendations for methods and monitoring concept

An annual sampling scheme is recommended to be able to detect short-term changes in pollution, underlining the function of the monitoring as an early warning system. Additionally, the maintenance of an annual monitoring may have large logistical advantages, in particular with respect to the

continuity of labs, staff and methodical details. To ascertain additionally the reliability of the spatial and temporal patterns found in bird eggs with respect to potential spatial and temporal variation in the prey composition and their trophic level, a stable isotope analysis of C and N would be useful (cf. Roscales et al. 2010).

In addition to the substances studied here, a variety of new environmental pollutants have emerged that may 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 and Dittmann 2009), a pilot assessment of phthalates has been initiated in recent years (unpublished). Further experience exists for analyses of further POPs such as polybromates, the dioxins polychlorinated dibenzodioxins (PCDD) and polychlorinated dibenzofurans (PCDF), as well as for perfluorinated compounds (PFCs; Shore et al. 2006; Bignert et al. 2011; Leslie et al. 2011; Crosse et al. 2012). 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 a preliminary coordinated environmental monitoring program (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).

With respect to a potential organizational framework of a further environmental monitoring of the mentioned contaminants, it has to be mentioned that there are several requirements for monitoring of hazardous substances in various matrices of biota including coastal bird 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 respective monitoring programmes, are under development. There are opportunities to tune these activities across countries to address both the requirements under the various EU directives, the objectives of OSPAR and the Trilateral Wadden Sea Cooperation, and make explicit the link between the concepts of good environmental status (GES) and of EcoQOs.

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