Contaminants in Bird Eggs in the Wadden Sea

Spatial and Temporal Trends 1991 - 2000

WADDEN SEA ECOSYSTEM No. 11 - 2001

Contaminants in Bird Eggs in the Wadden Sea

Spatial and Temporal Trends 1991 - 2000

Colophon

Publishers
Common Wadden Sea Secretariat (CWSS), Wilhelmshaven, Germany
Trilateral Monitoring and Assessment Group (TMAG)

Authors
Peter H. Becker, Jacqueline Muñoz Cifuentes, Brigitte Behrends, Institut für Vogelforschung "Vogelwarte Helgoland", An der Vogelwarte 21, D-26386 Wilhelmshaven, Germany; Klaus R. Schmieder, ITI, Fachhochschule Wilhelmshaven, Fiedrich-Paffrath-Str., D-26386 Wilhelmshaven, Germany

This work was funded by
Niedersächsische Wattenmeerstiftung, Hannover, Germany; Institute of Avian Research "Vogelwarte Helgoland" (Institut für Vogelforschung"Vogelwarte Helgoland"), Wilhelmshaven, Germany; Institute of Technology and Research at the University of Applied Sciences (Institut für Technisch-Wissenschaftliche Innovation (ITI) at the Fachhochschule Wilhelmshaven), Wilhelmshaven, Germany, National Park Agency Lower Saxony (Nationalparkverwaltung Niedersächsisches Wattenmeer), Wilhelmshaven, Germany; Ministry of Transport, Public Works and Water Management (Ministerie van Verkeer en Waterstaat, Rijksinstituut voor Kust en Zee/RIKZ), Den Haag, The Netherlands, Ministry of Agriculture, Nature Management and Fisheries, (Ministerie van Landbouw, Natuurbeheer en Visserij), Den Haag, The Netherlands, Ministry of Environment and Energy (Miliø-og Energiministeriet) Copenhagen, Denmark, National Park Agency Schleswig-Holstein (Landesamt für den Nationalpark Schleswig-Holsteinisches Wattenmeer), Tönning, Germany, TERRAMARE Research Centre (Forschungszentrum TERRAMARE), Wilhelmshaven, Germany,

The preparation of the report was financed by
Ministry of Transport, Public Works and Water Management, Den Haag, The Netherlands,
National Park Agency Lower Saxony, Wilhelmshaven, Germany,
Ministry of Environment and Energy, Copenhagen, Denmark.

Cover photo
J.-D. Ludwigs

Language support
Marijke Polanski

Lay-out
CWSS

Print
Druckerei Plakativ, Kirchhatten, +49(0)4482-97440

Paper
Cyclus – 100% Recycling Paper

Number of copies
1,200

Published
2001

ISSN 0946-896X

This publication should be cited as:
Becker, P.H., J. Muñoz Cifuentes, B. Behrends, K.R. Schmieder, 2001. Contaminants in Bird Eggs in the Wadden Sea. Temporal and spatial trends 1991 - 2000. Wadden Sea Ecosystem No. 11. Common Wadden Sea Secretariat, Trilateral Monitoring and Assessment Group, Wilhelmshaven, Germany.

WADDEN SEA ECOSYSTEM No. 11

Contaminants in Bird Eggs in the Wadden Sea

Spatial and Temporal Trends 1991 - 2000

Peter H. Becker
Jacqueline Muñoz Cifuentes
Brigitte Behrends
Klaus R. Schmieder

2001
Common Wadden Sea Secretariat
Trilateral Monitoring and Assessment Group

Editorial Foreword

This report documents the second assessment of the contaminant level of coastal bird eggs in the Wadden Sea carried out in the framework of the Trilateral Monitoring and Assessment Program (TMAP). The first report entailed the results of a trilateral pilot project carried out in 1996 – 1997 which had the aim to analyze the feasibility of this monitoring parameter for the TMAP and which focused mainly on the German Wadden Sea (Wadden Sea Ecosystem No. 8, 1998). After inclusion of this parameter into the TMAP in 1998, it is the first time that the whole Wadden Sea from Balgzand (Western Dutch Wadden Sea) to Langli (Ho Bugt, Denmark) is covered in a trilateral report on contaminants in bird eggs.

The experiences from the last years have shown that monitoring contaminant levels in bird eggs is an excellent instrument to indicate the environmental state of the Wadden Sea. The available time series, which partly goes back to 1981, is also valuable for future analysis and this value will increase with the continuation of the program.

The report has revealed, in general, strong decreasing levels of contaminants in bird eggs during the last two decades. However, some "hot spots" still deserve attention and "new" toxic substances may be a problem in the future. Chapter 6 of the report summarizes these issues and gives recommendations for future management, monitoring and research.

The preparation of the report has been financially supported by the National Institute for Coastal and Marine Management (Den Haag), the National Park Administration Lower Saxony (Wilhelmshaven) and the Forest and Nature Agency (Copenhagen). Because of this support and the "one-lab approach" it was possible to publish the results only a few months after sampling and chemical analysis. It would be desirable that other monitoring results could also be assessed and published trilaterally within such a relatively short period of time.

We would like to thank all those who contributed to the surveys and to the preparation of the report, amongst which the field workers and organizations involved and, in particular, Peter H. Becker and his team. Only their continuous effort and great commitment made this report possible.

Harald Marencic
Common Wadden Sea Secretariat
September 2001

Acknowledgements

During the 1990s, this monitoring program was supported by the Niedersächsische Wattenmeerstiftung, Hannover, the Institute of Avian Research "Vogelwarte Helgoland", Wilhelmshaven, the Institute of Technology and Research at the University of Applied Sciences (ITI), Wilhelmshaven, Germany, and the TERRAMARE Research Centre, Wilhelmshaven, as well as by the governmental organizations of the Wadden Sea states, the Nationalparkverwaltung "Niedersächsisches Wattenmeer", Wilhelmshaven, the National Institute for Coastal and Marine Management/RIKZ, Den Haag, The Netherlands, the Ministry of Environment and Energy, Copenhagen, Denmark, and the Landesamt für den Nationalpark Schleswig-Holsteinisches Wattenmeer, Tönning, Germany, in the framework of the Trilateral Cooperation on the Protection of the Wadden Sea represented by the Common Wadden Sea Secretariat, Wilhelmshaven.

The collection of eggs was supported repeatedly by A. Brenninkmeijer, T. Grünkorn, E. Hartwig, V. Hennig, J. Jungman, B. Koks, H. Krethe, J. Ludwig, U. Schneider, L.M. Rasmussen, P. Todt, P. Tydeman, M. Schulze-Dieckhoff, E. Stienen, M. Wagener, MELLUMRAT e.V., Verein JORDSAND and NABU, and by many employees doing their civil service and by nature conservation wardens.

The laboratory work was done or supported by U. Sommer, U. Pijanowska, S. Kahle, and S. Schuhmann. G. Liebezeit supported establishing the chemical analyses in TERRAMARE. A. Büthe and E. Denker offered valuable information on PCB-analyses. T. Dittmann, S. Kahle, S. Mickstein, and S. Thyen helped with statistical analyses and graphics, G. Haude and H.-J. Rogall with administration. The development of the parameter within the TMAP was encouraged by B. Reineking, H. Farke and H. Marencic. M. Polanski checked the English. F. Bairlein, R. Czeck, K. Essink, H. Farke, H. Marencic, B. Reineking, P. Südbeck, and S. Thyen presented helpful comments on the manuscript. We thank J.-D. Ludwigs, R. Nagel and P. Tydeman for photos and K. Essink and A.G. Ragborg for the translations of the summaries.

We would like to thank all persons and institutions concerned.

Peter H. Becker
Jacqueline Muñoz Cifuentes
Brigitte Behrends
Klaus R. Schmieder

September 2001

Contents

Summary

After the pilot study in 1996 and 1997, the parameter "Contaminants in Bird Eggs" was fully and successfully implemented within the Trilateral Monitoring and Assessment Program (TMAP) in 1998. Since 1999, the entire Wadden Sea from Balgzand in the western Dutch Wadden Sea to Langli in the Danish northern Wadden Sea has been covered by 13 sampling sites to monitor spatial and temporal trends in contamination of coastal birds. Residues of the heavy metal mercury and of the organochlorines PCBs, DDT and metabolites, HCB, HCH isomers and chlordanes (trans-chlordan, cis-chlordan, trans-nonachlor, and cis-nonachlor) were analyzed in Common Tern *Sterna hirundo* and Oystercatcher *Haematopus ostralegus* eggs. Sampling and analyses were carried out according to standardized methods and guidelines (JAMP, OSPAR) tested successfully in previous years. The parameter profits from similar former studies in the German Wadden Sea back to 1981, now allowing the analysis of time trends over two decades (1981 – 2000). Main focus of the report is the spatial pattern of the recent contamination in 2000 covering the entire Wadden Sea and the temporal trend during the last decade.

Interspecific variation

Common Tern eggs were higher contaminated by the analyzed chemicals than Oystercatcher eggs, with the exception of chlordanes. The differences between the species in accumulation of these substances depend on the environmental contaminants' load, and are explained by different feeding strategies, breeding and migration behavior.

Geographical trends

Discriminant analyses clearly separate the breeding sites by the given concentrations of all chemicals in the mixture within the eggs. Spatial trends were more distinct in the Common Tern. In general, eggs from breeding sites at the inner German Bight (Elbe estuary and Trischen) were contaminated on much higher levels (about 3 – 20 fold in the Common Tern) than those collected at western and northern breeding sites of the Wadden Sea indicating the lasting high importance of the Elbe as input source of environmental chemicals. The geographical differences in PCBs' levels were linked also with changes in the proportions of PCB congeners of high and low degrees of chlorination, as well as in the most toxic non-, mono- and di-ortho PCBs. In the Oystercatcher, however,

highest PCB, HCB and chlordane levels in eggs were found in the western part of the Wadden Sea, at the Julianapolder and Dollard, indicating recent discharges or high local loads originating from former years.

Temporal trends

At most study sites, the temporal trends from 1991 – 2000 revealed decreasing residues of mercury and organochlorines, though some increases have been recorded (e.g. Elbe estuary). The proportion of low chlorinated congeners within the PCB-mixture decreased during the last decade indicating an advancing metabolization. When considering both last decades (since 1981) the strongest drops in the eggs' contamination occurred during the late 1980s and early 1990s.

Target assessment

The results are discussed and assessed with respect to the Targets of the Wadden Sea Plan. Most results and developments are favorable in view of the state of Wadden Sea pollution, which was clearly reduced during the last two decades. So far critical levels are known, an impairment of breeding success by the recent levels of the toxicants seems no longer likely. However, there are some recent local problems of discharges or of persistence of contaminants prohibited long time ago, which need further efforts of environmental protection, monitoring and investigations.

Bird eggs as indicators of contamination

The results emphasize the advantages that eggs of coastal birds have in monitoring the pollution state of the Wadden Sea. Consequently, the endorsement to install the parameter has been justified by the experiences during the first years of the monitoring after its implementation. "Contaminants in Coastal Bird Eggs" has proved to be a very valuable, reliable, feasible and logistically favorable instrument to monitor the contamination of the Wadden Sea. As top predators the two bird species integrate the chemical pollution of different trophic levels.

Recommendations and gaps in monitoring

Considering the fact of still high local contamination, the policies to reduce the application of xenobiotic hazardous substances in the framework of OSPAR, the North Sea Conferences and the EU should be intensified. With respect to the monitoring we recommend:

1. to meet the requirement to distinguish short-term fluctuations from time trends, a long-term disposition of the parameter is advised;
2. some "new" chemicals to date not studied in birds should be considered (e.g. TBT, polybrominated biphenyls, bromocyclen or musk xylol);
3. an additional sampling site at the delta of the Rhine influencing the Wadden Sea by its contaminants' loads should be included;
4. to implement the parameter "Breeding Success of Coastal Birds", using birds as sensitive indicators of environmental change including chemical pollution. This instrument should be established as a valuable supplement to "Contaminants in Bird Eggs" and to the three other TMAP parameters utilizing birds to monitor the ecological state of the Wadden Sea.

Sammenfatning

Efter pilotprojektet i 1996 og 1997 er der efterfølgende med succes gennemført en overvågning af parameteren "Forurenende stoffer i fugleæg" inden for det Trilaterale Undersøgelses- og Vurderingsprogram. Siden 1999 er der i hele Vadehavet fra Balgzand i det vesthollandske Vadehav til Langli i det danske nordlige Vadehav oprettet 13 prøvetagningsstationer, hvor der sker en indsamling med henblik på overvågning af udbredelse og tidsmæssige tendenser i forureningsgraden hos kystfugle. Indhold af tungmetallet kviksølv og de organiske klorforbindelser PCBere, DDT og metabolitter, HCB, HCH-isomere og klordaner (transklordan, cisklordan, transnonaklor og cisnonaklor) er blevet analyseret i æg fra fjordterne *Sterna hirundo* og strandskade *Haematopus ostralegus.*

Prøvetagning og analyser er i overensstemmelse med retningslinier fra JAMP, OSPAR udført efter standardmetoder, som i tidligere år er blevet anvendt med positive resultater. Parametrene er analyseret ved lignende undersøgelser i det tyske Vadehav siden 1981, hvilket giver mulighed for analyse af den tidsrelaterede udvikling over en horisont på to årtier (1981-2000). Rapportens hovedemne er udbredelsesmønstret af forureningen år 2000, dækkende hele Vadehavet samt de tidsmæssige tendenser gennem det seneste årti.

Interspecifik variation

Indholdet af de analyserede kemiske stoffer er højere i æg fra fjordterne end i æg fra strandskade, med undtagelse af klordaner. Forskellene mellem de to arter mht. ophobningen af disse stoffer afhænger af baggrundsniveauerne i omgivelserne og kan forklares ved forskelle i fødestrategier, samt yngle- og trækadfærd.

Geografisk udvikling

På baggrund af resultaterne over koncentrationer af samtlige kemiske stoffer i materialet fra æggene kan man via diskriminantanalyserne klart adskille ynglestederne fra hinanden. Den geografiske tendens var tydeligst for fjordterne. Generelt var forureningen af æg fra ynglesteder ved Elbens udmunding og Trischen langt højere (ca. 3-20 gange mere hos den fjordterne) end forureningen af æg, der var indsamlet på de vestlige og nordlige ynglesteder i Vadehavet. Dette understreger Elbens stedse store betydning som input-kilde for miljøskadelige stoffer. De geografiske forskelle i PCB-niveauer blev tillige sammenkædet med forandringer i fordelingen af PSB-grupper med forskellige antal af klorgrupper, samt med de mest toksiske non-, mono- and di-ortho PCBere. Hos strandskaden blev de højeste PCB, HCB og chlordan-niveauer i æg imidlertid fundet i den vestlige del af Vadehavet, ved Julianapolder og

Dollard hvilket peger på, at der pågår store lokale udledninger af belastende stoffer eller at baggrundsniveauerne er høje på grund af udledninger fra tidligere år.

Tidsrelateret udvikling

I de fleste områder viser den tidsrelaterede udvikling fra 1991 til 2000 et faldende indhold af kviksølv og organiske klorforbindelser, men der er også registreret stigninger, fx i Elbens udmunding. Andelen af lavklorinerede grupper i PCB-blandingen er blevet lavere i løbet af det seneste årti, hvilket indikerer stigende metabolisk omsætning. Når man ser på de seneste årtier (siden 1981), fandt det største fald i forureningen af æg sted sidst i 80ere og i begyndelsen af 90ere.

Målvurdering

Resultaterne er analyseret og vurderet i relation til Vadehavsplanens målsætninger. Størstedelen af resultaterne og udviklingen viser en positiv udvikling for så vidt angår belastningen med forurenende stoffer i Vadehavet, og der er påvist et klart fald gennem de seneste to årtier. På baggrund af kendskab til kritiske niveauer, synes det ikke længere sandsynligt, at de registrerede niveauer for giftstoffer vil kunne virke hæmmende på fuglenes ynglesucces. Der er imidlertid fortsat lokale problemer med udledning eller tilstedeværelse af forurenende stoffer som er forbudte i dag, hvilket kræver yderligere tiltag inden for miljøbeskyttelse, monitering og undersøgelser.

Fugleæg som forureningsindikatorer

Resultaterne af de gennemførte undersøgelser understreger de fordele, som en monitering af æg fra kystfugle har i forbindelse med overvågningen af Vadehavets forureningstilstand. De erfaringerne der er indhentet gennem de første år med implementering af overvågningen af "Forurenende stoffer i æg fra kystfugle" bekræfter hensigten med at inkorporere denne parameter i TMAP-programmet. Undersøgelserne af "Forurenende stoffer i æg fra kystfugle" har således vist sig at være et meget værdifuldt, pålidelig, praktisk og logistisk gunstigt redskab i overvågningen af forureningen af Vadehavet. Som øverste led i fødekæden integrerer de to fuglearter den kemiske forurening på forskellige trofiske niveauer.

Anbefalinger og mangler i moniteringen

I betragtning af, at der fortsat lokalt er en høj forurening, bør man intensivere tiltag med henblik på at mindske brugen af farlige, miljøfremmede stoffer inden for rammerne af OSPAR, Nordsøkonferencen og EU. Hvad angår monitering, anbefales:

1. at behovet for, at der skelne mellem kortsigtede variationer og tidsrelaterede tendenser, imødekommes ved, at parameteren disponeres ud fra en langsigtet synsvinkel;
2. at "nye" kemiske stoffer, som til dato ikke er undersøgt i fugle, bør inddrages, fx TBT, polybrombifenyler, bromcyclen eller musk xylol;
3. at der bør tilføjes yderligere et prøvetagningssted i Rhindeltaet, med henblik identificiering af påvirkerne i Vadehavet fra Rhinen;
4. at der gennemføres en overvågning af parameteren "Kystfugles yngleevne" ("Breeding Success of Coastal Birds") med anvendelse af fugle som følsomme indikatorer for miljøforandringer, herunder kemisk forurening. Denne parameter bør etableres som et værdifuldt supplement til "Forurening i Fugleæg" ("Contaminants in Bird Eggs") og de øvrige tre andre TMAP parametre, som anvender fugle i monitoreringen af Vadehavets økologiske tilstand.

Zusammenfassung

Nach der Pilotstudie aus den Jahren 1996 und 1997 wurde der Parameter „Schadstoffe in Vogeleiern" gemäß des Vorschlags der TMAG im Jahre 1998 länderübergreifend und erfolgreich im TMAP eingeführt. Das gesamte Wattenmeer von Balgzand in den Niederlanden bis nach Langli in Dänemark ist seit 1999 durch 13 Untersuchungsgebiete abgedeckt, um räumliche und zeitliche Trends in der Kontamination von Küstenvögeln aufzuzeigen. Rückstände des Schwermetalls Quecksilber sowie der Organochlorverbindungen PCB, HCB, DDT und Metaboliten, HCH-Isomere und Chlordane wurden in Eiern von Flußseeschwalben *Sterna hirundo* und Austernfischern *Haematopus ostralegus* untersucht. Probennahme und chemische Analytik entsprachen standardisierten Methoden und Richtlinien, die in früheren Jahren erarbeitet und erfolgreich getestet worden waren (JAMP, OSPAR). Der Parameter baut auf ähnlichen Studien im deutschen Wattenmeer auf, die bis zum Jahre 1981 zurück reichen und jetzt Zeittrendanalysen über zwei Dekaden ermöglichen (1981-2000). Im Mittelpunkt dieses Reports stehen das räumliche Muster der Kontamination von Küstenvögeln im Jahre 2000, als das gesamte Wattenmeer beprobt wurde, und die zeitliche Variation während der vergangenen Dekade.

Zwischenartliche Variation

Flußseeschwalbeneier waren stärker mit den untersuchten Chemikalien kontaminiert als Austernfischereier, ausgenommen die Chlordane. Die Artunterschiede in der Akkumulation der Kontaminanten stiegen mit zunehmender Umweltbelastung des Entnahmegebiets an und finden ihre Erklärung in unterschiedlichen Ernährungsstrategien, Brut- und Zugverhalten der Vogelarten.

Geographische Trends

Auf der Basis der Konzentrationen aller Chemikalien im Ei lassen sich die untersuchten Brutgebiete durch Diskriminanzanalysen klar voneinander trennen. Geographische Unterschiede fielen bei der Flußseeschwalbe deutlicher aus als beim Austernfischer. Im Allgemeinen waren Eier von der Inneren Deutschen Bucht (Elbeästuar und Trischen) viel höher kontaminiert (3-20-fach bei der Flußseeschwalbe) als Eier, die im westlichen und nördlichen Wattenmeer gesammelt wurden. Dies zeigt den nach wie vor großen Einfluß der Elbe als Eintragsquelle von Umweltchemikalien in das Wattenmeer an. Im Zusammenhang mit den geographischen Unterschieden in den PCB-Gehalten standen Veränderungen in den Anteilen von hoch- und niedrigchlorierten PCBs und den toxischen

PCB-Kongeneren (Non-, Mono- und Di-Ortho-PCB). Beim Austernfischer wurden höchste PCB-, HCB- und Chlordan-Konzentrationen in Eiern von Julianapolder und Dollart aus dem westlichen Wattenmeer gefunden, was jüngste Immissionen oder hohe lokale Belastungen aus früheren Jahren anzeigt.

Zeittrends

An den meisten Standorten waren die Zeittrends von 1991-2000 durch Abnahmen der Rückstände an Quecksilber und Organohalogenen im Ei gekennzeichnet, obwohl auch einige Zunahmen festgestellt wurden (z.B. Elbeästuar). Der Anteil der niedrigchlorierten Kongenere innerhalb des PCB-Gemischs nahm während der letzten Dekade ab und zeigte eine zunehmende Metabolisierung der PCB an. Unter Berücksichtigung der 80er Jahre ergaben sich die deutlichsten Schadstoffrückgänge in den Eiern um die Zeit des Dekadenwechsels in den späten 80er und frühen 90er Jahren.

Qualitätsziele

Die Ergebnisse werden diskutiert und bewertet in Hinsicht auf die ökologischen Qualitätsziele, die in der trilateralen Kooperation für den Schutz des Ökosystems Wattenmeer formuliert wurden. Die meisten Ergebnisse und Entwicklungen sind als günstig hinsichtlich der Kontamination des Wattenmeeres zu bewerten, die eindeutig während der vergangenen beiden Dekaden zurück gegangen ist. Soweit kritische Werte für den Bruterfolg der Vögel bekannt sind, ist dessen Beeinträchtigung auf Grund der gegenwärtigen Kontamination mit Umweltgiften unwahrscheinlich. Dennoch mahnen jüngste lokale Belastungsprobleme sowie die Persistenz einiger Umweltchemikalien, die viele Jahre zuvor bereits verboten wurden und nach wie vor in hohen Konzentrationen feststellbar sind, zu weiteren Anstrengungen im Umweltschutz, Fortführung des Monitorings und zu speziellen Untersuchungen.

Vorteile von Vogeleiern im Schadstoffmonitoring

Die Ergebnisse unterstreichen die Vorteile, die Küstenvogeleier beim Monitoring der Belastung des Wattenmeeres mit Umweltchemikalien bieten und zeigen, dass die angestrebten Ziele vom Parameter „Schadstoffe in Vogeleiern" im Rahmen des TMAP erreicht wurden. Auf Grund der guten Erfahrungen während der ersten Jahre des Monitorings erwies sich somit die Entscheidung, diesen Parameter aufzugreifen, als gerechtfertigt. „Schadstoffe in Vogeleiern" hat sich als sehr wertvolles, zuverlässiges, gut handhabbares und logistisch günstiges Instrument für das Monitoring der Schadstoffbelastung des Wattenmeeres erwiesen. Als Top-Prädatoren integrieren beide Zielarten die Kontamination verschiedener trophischer Stufen des Wattenmeeres.

Empfehlungen und Lücken im Monitoring

Aufgrund der lokal nach wie vor hohen Kontamination müssen die politischen Aktivitäten und Maßnahmen im Rahmen von OSPAR, der Nordseeschutz - Konferenzen und der EU intensiviert werden, um die Anwendung und Einleitung von Umweltchemikalien weiter einzuschränken. Wir empfehlen, einige noch existierende Lücken im Monitoring zu schließen:

1. Um kurzzeitige Schwankungen der Eikonzentrationen der Schadstoffe von Zeittrends unterscheiden zu können, ist eine langzeitliche Fortführung des Monitorings mit diesem Parameter angebracht;
2. in der Analytik sind wegen ihrer Toxizität relevante, bislang aber unbearbeitete Schadstoffe zu berücksichtigen (z.B. TBT, Flammschutzmittel, Bromocyclen oder Moschusxylol);
3. ein Untersuchungsgebiet nahe des Rheindeltas sollte hinzu kommen, da der Rhein nach wie vor mit seiner Schadstofffracht das Wattenmeer beeinflußt;
4. außerdem sollte dringend der Parameter „Bruterfolg von Küstenvögeln" implementiert werden, der Vögel als sensitive Indikatoren für Umweltänderungen einschließlich der Schadstoffbelastung nutzt. Dieses Instrument ist eine wertvolle Ergänzung zu „Schadstoffen in Vogeleiern" sowie zu den drei anderen TMAP-Parametern, die Vögel für das Monitoring des ökologischen Zustandes des Wattenmeeres einsetzen.

Samenvatting

Na een oriënterende studie in 1996 en 1997, werd de parameter "Verontreinigingen in vogeleieren" in 1998 volledig en met succes opgenomen in het Trilaterale Monitoring en Beoordelings Programma (TMAP). Sinds 1999 worden in de gehele Waddenzee, van het Balgzand (westelijk in de Nederlandse Waddenzee) tot Langi (noordelijk in de Deense Waddenzee), 13 locaties bemonsterd voor het monitoren van ruimtelijke en temporele trends in het voorkomen van verontreinigingen in kustvogels. Gehaltes van het zware metaal kwik, en van de gechloreerde koolwaterstoffen PCB's, DDT en metabolieten, HCB, HCH isomeren, en chloordanen (trans-chloordaan, cis-chloordaan, transnonachloor en cis-nonachloor) werden bepaald in eieren van de Visdief (*Sterna hirundo*) en de Scholekester (*Haematopus ostralegus*). Bemonstering en chemische analyses werden uitgevoerd volgens gestandaardiseerde methoden en richtlijnen (JAMP, OSPAR), die in de voorafgaande jaren met succes waren beproefd. Hierbij is geprofiteerd van eerdere vergelijkbare studies in de Duitse Waddenzee die teruggaan tot 1981, waardoor nu de analyse mogelijk is van temporele trends over twee decennia (1981-2000). Het rapport besteedt vooral aandacht aan het ruimtelijk patroon van verontreiniging in 2000 in de gehele Waddenzee, en aan de temporele trend gedurende de laatste tien jaar.

Verschillen tussen de vogelsoorten

De eieren van de Visdief waren sterker verontreinigd door de geanalyseerde chemicaliën, met uitzondering van de chloordanen, dan de eieren van de Scholekster. De verschillen tussen de vogelsoorten in ophoping van deze stoffen hangen af van de hoeveelheid verontreiniging die in het milieu voorkomt, en worden verklaard door verschillende voedselstrategieën en verschillen in broed- en trekgedrag.

Ruimtelijke trends

Toepassing van zg. 'discriminant analyses' laat zien dat vooral de broedlocaties van de onderzochte vogels verschillen in concentraties van alle verontreinigingen in het aangetroffen mengsel van verontreinigingen in de eieren. De ruimtelijke trends waren meer uitgesproken bij de Visdief. In het algemeen waren de eieren van broedlocaties in de binnenste Duitse Bocht (Elbe estuarium en Trischen) veel sterker verontreinigd (in eieren van de Visdief ongeveer 3 tot 20 maal) dan eieren verzameld op de westelijk en noordelijk gelegen broedlocaties in de Waddenzee. Dit geeft aan dat de Elbe nog steeds een belangrijke bron van in het milieu aangetroffen chemicaliën is. De ruimtelijke verschillen in PCB-gehalten hadden zowel te maken met verschillen in het aandeel van de hoog en laag gechloreerde PCB congeneren, als met verschillen in het aandeel van de meest toxische non-, mono- en di-ortho PCBs. In scholekstereieren, daarentegen, werden de hoogste gehaltes van PCBs, HCB en chloordanen aangetroffen in het westelijk deel van de Waddenzee (Julianapolder en Dollard). Dit geeft aan dat hier sprake is van recente lozingen of van een hoge belasting als gevolg van lozingen uit vroegere jaren.

Temporele trends

Op de meeste van de onderzochte locaties is er van 1991 tot 2000 sprake van een trend van afnemende gehaltes van kwik en organochloorverbindingen, hoewel ook enkele toenames werden waargenomen (bijv. in het Elbe estuarium). Het aandeel van laag gechloreerde congeneren in het PCB-mengsel nam gedurende de laatste tien jaar af, hetgeen wijst op een voortschrijdende omzetting en afbraak. In de laatste twee decenia (vanaf 1981) vond de sterkste daling van de verontreiningen in de eieren plaats aan het eind van de tachtiger, en het begin van de negentiger jaren.

Beoordeling van de doelstelling

De verkregen resultaten zijn bediscussieerd en gebruikt ter beoordeling van de doelstellingen van het Waddenzee Plan. Het merendeel van de resultaten geeft aan dat de verontreinigingstoestand van de Waddenzee zich in de laatste twee decennia in gunstige zin heeft ontwikkeld. Voor zover bekend lijkt er gezien de recent gemeten gehalten van verontreinigende stoffen in de vogeleieren niet langer sprake te zijn van een nadelige beïnvloeding van het broedsucces. Er zijn echter nog wel sprake van enkele recente lokale lozingen of van aanwezigheid van reeds lang geleden verboden verontreinigende stoffen, die om verdere inspanning vragen op het gebied van milieubescherming, monitoring en onderzoek.

Vogeleieren als indicator van verontreiniging

De gepresenteerde resultaten benadrukken de waarde van het gebruik van kustvogeleieren ten behoeve van het monitoren van de verontreinigingstoestand van de Waddenzee. De ervaringen gedurende de eerste jaren van monitoring geven dan ook een rechtvaardiging voor het opnemen van deze parameter in het monitoringprogramma. De parameter "Verontreinigingen in vogeleieren" heeft bewezen een waardevol, betrouwbaar, en goed uitvoerbaar instrument te zijn voor het monitoren van verontreiniging van de Waddenzee. Als toppredatoren integreren de twee gebruikte vogelsoorten de chemische verontreiniging van verschillende niveaus in de voedselketens.

Aanbevelingen en tekortkomingen

Aangezien er lokaal nog steeds sprake is van een hoge verontreiniging, dient het beleid voor het terugdringen van de toepassing van gevaarlijke xenobiotische stoffen in het kader van OSPAR, de Noordzee Conferenties en de Europese Unie te worden aangescherpt. Met betrekking tot monitoring wordt aanbevolen:

1. de parameter langdurig in het monitoringprogramma op te nemen om trends van korte termijn fluctuaties te kunnen onderscheiden;
2. thans nog niet in vogels onderzochte 'nieuwe' chemische stoffen in overweging te nemen (bijv. TBT, polybroom biphenylen, cyclische broomverbindingen en musk xylol);
3. een bemonsteringslocatie in de Rijndelta toe te voegen omdat vanuit de Rijndelta de Waddenzee wordt beïnvloed door verontreinigende stoffen;
4. de parameter "broedsucces van kustvogels" in het TMAP te implementeren. Hierbij worden vogels gebruikt als gevoelige indicatoren van milieuverandering inclusief chemische verontreiniging. Dit instrument dient als waardevolle aanvulling op de parameter "Verontreinigingen in vogeleieren" en op de drie andere TMAP parameters waarin vogels gebruikt worden om de ecologische toestand van de Waddenzee te monitoren.

1. Introduction

The general aim of the Trilateral Monitoring and Assessment Program (TMAP) is to provide a scientific assessment of the status of the ecosystem Wadden Sea, and to assess the implementation status of the Targets of the Wadden Sea Plan (Stade Declaration 1997, Marencic 1997). Among the concert of biological, chemical and other parameters selected to verify this monitoring program, "Contaminants in Bird Eggs" was classified as a parameter of high priority and as an adequate instrument to supervise the contamination of biota in the Wadden Sea ecosystem (Bakker et al. 1997).

Birds are an important part of this ecosystem: Several millions of individuals of many species are concentrated here each year during migration, wintering or the breeding season (e.g. Exo 1994, Fleet et al. 1994, Meltofte et al. 1994, Rasmussen et al. 2000), and the Wadden Sea plays a dominant role in the life cycles of these species. Coastal birds such as seabirds and waders occupy high trophic levels, tend to accumulate persistent chemicals and are vulnerable to their effects. On the other hand, we can make use of these characteristics: Coastal birds are of value as accumulative and sensitive indicators of even this marine pollution with chemicals, caused by man.

Because of the prominent role of birds as indicators of various aspects of environmental pollution with chemicals, biomonitoring of contaminants with birds has been introduced in various countries since the 1960s. The most frequently used matrix to monitor avian contamination is the bird egg. The main objectives of programs utilizing birds as accumulative indicators – such as the TMAP in the Wadden Sea - are as follows (e.g. Newton et al. 1993, Bignert et al. 1995, 1998, Bignert 2001, TMAP 1997, Becker et al. 1998):

- to provide long-term residue data on several representative species and sites;
- to assess long-term temporal trends in chemical residues and to estimate the rate of changes (time trend monitoring);
- to assess spatial variation in contaminants' levels, on small and large geographical scales (spatial trend monitoring);
- to explore site-specific and time-specific patterns in the composition of compound mixtures like PCBs, DDTs, HCHs, chlordanes, toxaphenes and organotin;

Such information can be used:

- to detect incidents of local contamination discharges;
- to detect renewed application or discharge of banned contaminants;
- to assess the effectiveness of measures to reduce contamination, against the background of successive governmental restrictions on organochlorine and mercurial pesticide use;
- to assess population changes of affected species: Hence, the response of species to changes in pollution levels may affect the physiology, reproduction or abundance of species leading to structural changes in the ecosystem (TMAP 1997). To achieve this aim, chemical monitoring has to be combined with bird population monitoring (see 5.5).

Table 1: History of the parameter "contaminant in bird eggs" during the 1990s organised by a co-operation of Institute of Avian Research Wilhelmshaven, Germany, Institute of Technology and Research at the University of Applied Sciences, Wilhelmshaven, Germany, and TERRAMARE Research Centre, Wilhelmshaven, Germany.
Abbreviations: DK, Denmark; NERI, Ministry of Environment and Energy, Copenhagen, Denmark; NL, The Netherlands; NLPV, National Park Agency Lower Saxony, Wilhelmshaven, Germany; NPA, National Park Agency Schleswig-Holstein, Tönning, Germany; Nds, Lower Saxony; RIKZ, Ministry of Transport, Public Works and Water Management, Den Haag, The Netherlands; SH, Schleswig-Holstein, Germany.

Period	Sampling sites in (states):	TMAP status	Funded by:
1991-1995	SH, Nds (NL)	-	Niedersächsische Wattenmeerstiftung
1996-1997	SH, Nds, NL	Pilot study	Niedersächsische Wattenmeerstiftung, RIKZ
1998	SH, Nds, NL	Implementation by Nds, NL	Niedersächsische Wattenmeerstiftung, RIKZ
1999-2000	DK, SH, Nds, NL	Full Implementation	RIKZ, NERI, NLPV, NPA

The Institute of Avian Research "Vogelwarte Helgoland" started to use seabirds as accumulative indicators of contaminants in the North Sea in 1981, cooperating with the Chemical Institute of School of Veterinary Medicine, Hannover, where the chemical analyses were performed. After repeating the egg sampling in 1986, the two institutes initiated and performed a four-year program "Seabirds as monitors of environmental chemicals" which was supported by the Environmental Agency, Berlin, and elaborated the basis for an international monitoring program with seabirds in the North Sea (Becker 1989, 1991, Becker et al. 1991, 1992).

From 1991-1995, the Institute of Avian Research "Vogelwarte Helgoland" continued to collect and store further samples from the sampling sites chosen in the German Wadden Sea (see Table 1). From 1995-1998, these samples could be analyzed by a cooperation with the ITI at the University of Applied Sciences, Wilhelmshaven, supported by the Niedersächsische Wattenmeer-Stiftung. This program was also the core of the pilot study "Monitoring pollutants in coastal bird eggs" in the Wadden Sea" from 1996-1997 within the TMAP (Becker et al. 1998). The positive results of this study became the basis of the implementation of the parameter in 1998 within the framework of the TMAP, when the Netherlands joined the program to measure this parameter, using birds' eggs on the basis of generally agreed guidelines for monitoring design sampling procedures and chemical analytical procedures (Bakker et al. 1997, OSPAR 1997, TMAP 1997, Becker et al. 1998). In 1999, the program was fully implemented by the four states Denmark, Schleswig-Holstein (Germany), Lower Saxony (Germany), and the Netherlands (Table 1). Several sampling sites along the Wadden Sea coast permit the evaluation of geographical trends and hot spots of contamination (Fig. 1). Since 1999, the cooperating institutions, Institute of Avian Research "Vogelwarte Helgoland" and University of Applied Sciences Wilhelmshaven, use the chemical laboratory of the TERRAMARE Research Center in Wilhelmshaven to perform the chemical analyses. All eggs collected in the Wadden Sea are now analyzed for the selected environmental chemicals in the TERRAMARE labs. Organization and analysis in "one hand" offer many advantages for monitoring (see 5.5).

In 1998, on the basis of projects carried out in the 1980s, and of a pilot study within the TMAP framework (Table 1), monitoring chemicals in the Wadden Sea area was started using birds' eggs on the basis of generally agreed guidelines for monitoring design, sampling procedures and chemical analytical procedures.

This report includes the monitoring results of three years after full implementation of the TMAP parameter in 1998 and proceeds to evaluate spatial and temporal trends of bird contamination in the Wadden Sea since the report of the pilot study (Becker et al. 1998). For the first time we present data of the insecticide Chlordane and from two new Danish sampling sites, filling the gap of the northern part of the Wadden Sea in monitoring of coastal bird's pollution. We focus on:

- spatial trends including 13 sampling sites along the Wadden Sea coast;
- temporal trends from 1991 – 2000;
- analyses and assessment of contamination trends over a period of 20 years using data sets available from the early 1980s;
- the assessment of the recent bird egg contamination with respect to possible negative effects on the bird populations, to the ecological targets and to the ecological state of the Wadden Sea;
- recommendations for management;
- the experiences made with the implementation of this new parameter within the TMAP;
- the discussion of the question, whether the objectives of this parameter requested by the TMAP are fulfilled, and of the value of the parameter within the future TMAP.

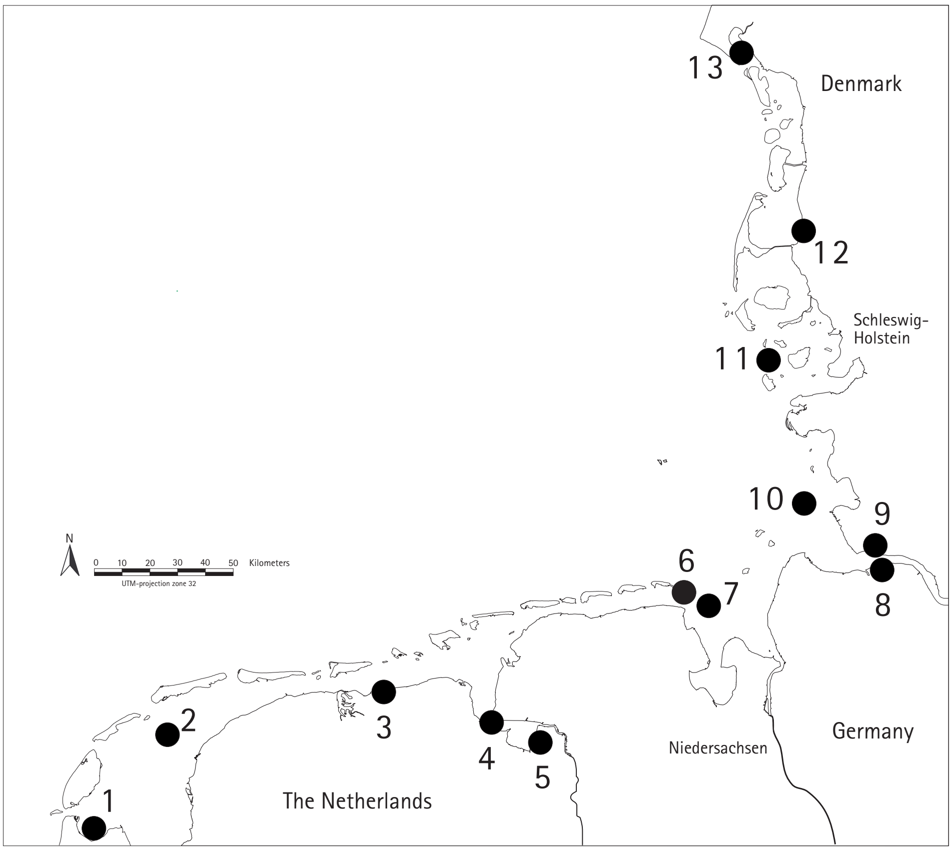

Figure 1: Sampling sites of Common Tern and Oystercatcher eggs in the Wadden Sea in 2000. The Netherlands: 1 Balgzand, 2 Griend, 3 Julianapolder, 4 Delfzijl; Germany, Lower Saxony: 5 Dollart, 6 Minsener Oog, 7 Mellum (6 and 7 = Jade), 8 Hullen, 9 Neufelderkoog (8 and 9 = Elbe estuary); Germany, Schleswig Holstein: 10 Trischen, 11 Norderoog; Denmark: 12 Margrethekoog, 13 Langli. At sites 5, 7, 8,11 and 13 only Oystercatcher eggs, at sites 6, 9 and 12 only Common Tern eggs have been taken; at all other sites eggs of both species have been collected.

2. Bird Species, Sampling Sites, Chemicals

Common Tern
(Photo J.-D. Ludwigs)

2.1 Species

Common Tern (*Sterna hirundo*) and Oystercatcher (*Haematopus ostralegus*) are among the most common species using the Wadden Sea as breeding area. The numbers of breeding pairs of Common Terns and Oystercatchers represent about 12 and 20%, respectively, of the total of the population that breeds in northwestern Europe (Rasmussen et al. 2000).

The Common Tern is a long-distance migratory species that arrives in the Wadden Sea in spring and forms large breeding colonies. Oystercatchers, however, are resident birds in the Wadden Sea area. Both species display different habits of foraging and eat different diets: Common Terns feed mainly on fish which are taken by plunge-diving, and are considered a top-predator of the Wadden Sea food-chain, while Oystercatchers mainly feed on macrozoobenthic organisms like mussels and worms (Smit & Wolff 1980, Cramp & Simmons 1985).

The good knowledge of the biology and ecology of these species, their large populations and abundance in the Wadden Sea, the high position they occupy within the marine food-chains, and the capacity to accumulate persistent contaminants make them especially suitable as monitors of the contamination of the environment.

Oystercatcher
(Photo J.-D. Ludwigs)

Table 2: Sampling sites (c.f. Fig. 1), species, and years of the monitoring project during the decade 1991-2000. CT= Common Tern, OC = Oystercatcher, NL = the Netherlands, NS = Lower Saxony, Germany, SH = Schleswig Holstein, Germany and DK = Denmark. Jade = Minsener Oog and Mellum, Elbe Estuary = Hullen and Neufelderkoog. 10 eggs per species, site and year were collected (exceptions: Griend: 1996=2 OC; Julianapolder: 1998= 2 CT, 2 OC, 1999=9 CT; Dollard: 1995=6 OC, 1996= 4 OC, 1997=9 OC, 1999=5 OC; Trischen: 1999=1 CT; Norderoog: 2000=9 OC). [1] Eggs not yet analyzed.

	1991	1992	1993	1994	1995	1996	1997	1998	1999	2000
1. Balgzand, NL	-	-	-	-	-	-	-	CT/OC	CT/OC	CT/OC
2. Griend, NL	-	-	CT	-	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC
3. Julianapolder, NL	-	-	-	-	-	-	CT/OC	CT/OC	CT/OC	CT/OC
4. Delfzijl, NL	-	-	-	-	-	-	-	CT/OC	CT/OC	CT/OC
5. Dollard, Nds	OC	OC	OC	-	OC	OC	OC	OC	OC	OC
6. Minsener Oog, Nds	CT	CT	CT	CT	CT	CT	CT	CT	CT	CT
7. Mellum, Nds	OC	OC	OC	OC	OC	OC	OC	OC	OC	OC
8. Hullen, Nds	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	OC	OC	OC	OC	OC
9. Neufelder Koog, Nds	-					CT	CT	CT	CT	CT
10. Trischen, SH	CT/OC	CT/OC	CT/OC	-	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC
11. Norderoog, SH	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	CT/OC	OC[1]	OC
12. Margrethekoog, DK	-	-	-	-	-	-	-	-	CT	CT
13. Langli, DK	-	-	-	-	-	-	-	-	OC	OC

2.2 Sampling Sites

In order to study spatial patterns in bird pollution, sampling sites from the western to the northern parts of the Wadden Sea have been chosen (Fig. 1, Table 2) including different habitats of the Wadden Sea coast. Furthermore, the choice of breeding colonies attempted to cover those locations which represent potentially high loads of environmental contaminants, such as the estuaries of Ems, Jade/Weser and Elbe as potential pollutant sources. The sampling areas in the Netherlands were, from west to east, Balgzand (salt marsh), Griend (island), Julianapolder (salt marsh) and Delfzijl (dike foot vegetation). In Germany, the Dollart (salt marsh, Ems estuary), Minsener Oog and Mellum (islands, Jade Bay), Hullen and Neufelderkoog (Elbe estuary), Trischen (island, inner German Bight) and Norderoog (island) have been selected. In Denmark, Margrethekoog (salt marsh) and Langli (island, adjacent to the Vardeå estuary) were chosen.

Balgzand, Western Dutch Wadden Sea (Photo P. Tydeman)

Delfzijl, Eastern Dutch Wadden Sea, (Photo R. Nagel)

Minsener Oog, Jade, FRG (Photo R. Nagel)

Trischen, Dithmarschen, FRG (Photo P. Becker)

Norderoog, FRG
(Photo R. Nagel)

2.3 The Environmental Chemicals Under Study: Characteristics, Production, Application and Regulations by Law

Environmental chemicals are hazardous compounds emitted by man into the environment and of toxicological relevance for organisms. Among the chemicals studied are the heavy metal and natural micropollutant mercury. The other analyzed chemicals are xenobiotics, i.e., they are exclusively introduced into the environment by human activities (Koch 1991). In this study, we classify the analyzed substances in industrial chemicals (mercury, PCBs and HCB) and pesticides (ΣDDT, ΣHCH, and chlordanes).

Mercury

Mercury occurs naturally in the environment ("background values"; Koch 1991, Haarich 1994, Schlüter 2000). It is used by man in many products (thermometers, barometers, batteries, etc.; in former decades, it was also used as biocide in seed disinfection) and as catalyst in many industrial processes (paper manufacturing, production of vinyl chloride, urethane foam, etc). Mercury that is released into the environment will remain there indefinitely. Mercury is transformed into a very toxic form (methylmercury) by bacteria and chemical processes. In its organic form, this heavy metal is highly accumulated in the food-chain. For this reason, relatively low levels of mercury in aquatic ecosystems can lead to toxic contamination in organisms high within the food-chain (e.g. predators). In man, an oral intake of 4 $ng \cdot g^{-1} \cdot day^{-1}$ is assumed to be toxic. Birds eliminate body mercury into growing feathers and eggs (e.g. Furness 1993, Gochfeld 1997).

PCBs

Polychlorinated biphenyls (PCBs) are industrial products or by-products formed in industrial processes (Holoubek et al. 1994; Koèan et al. 1994, 1996), and are composed of 209 individual congeners with varying levels of toxicity. Owing to appropriate physical-chemical properties (inert and lipophilic), PCBs were widely applied in industry. The excellent properties of PCBs for industrial use make them also hazardous to the environment. 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: The smaller their number is, the greater is the toxicity of the congener (e.g. Parkinson & Safe 1987). Therefore, the coplanar congeners "non-ortho PCBs" (without substitution in ortho-position of the phenyl ring) have a higher toxicity than mono-ortho PCBs (one chlorine atom in ortho-position) or di-ortho-PCBs (two chlorine atoms in ortho-position), because of a completely flat (planar) conformation which is close to that of dioxins. The coplanar congeners strongly induce activity of Cytochrom-P-448 and have similar effects as the 2,3,7,8-TCDD (tetrachloro-dibenzo-p-dioxin). A propeller-like conformation of ortho-chlorines prevents the planar conformation to a variable degree, and therefore ortho-congeners are less dioxin-like than non-ortho-congeners. Chlorines in ortho-positions seem to be hardly 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 by-product in the production of chlorine gas and chlorinated compounds, including several pesticides. 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 three Wadden Sea countries during the 1980s. HCB enters into the environment for example as contaminant of other chemical products, from the reduction of PCBs, as metabolite of Lindane or with the production of pentachlorophenol (Becker et al. 1991).

DDT

The insecticide p,p'-DDT is the pesticide most widely known, owing to its toxic effects on other biota (Koch 1991). ΣDDT 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, whose presence at high levels is associated with the thinning of egg shell in birds (see reviews, e.g. Moriarty et al. 1986, Furness 1993). This pesticide and its metabolites are considerably 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 West Europe during the 1970s but was still used in several countries of East Europe during the 1980s.

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. Due to the long-lasting systematic application, the hexachlorocyclohexanes are spread in the environment and will remain in soils for some decades.

Chlordanes

Chlordanes are chlorinated hydrocarbons and belong to the cyclodienes, originally registered as pesticides in 1948. Chlordane was banned in Germany in 1988. Technical chlordane is a mixture of at least 50 compounds; the major constituents are cis- and trans-chlordane, cis- and trans-nonachlor and Heptachlor (Agency for Toxic Substances and Disease Registry United States Public Health Service, 1994). In the aquatic environment, chlordanes are very persistent in the adsorbed state. Trans-nonachlor is bioaccumulated the most.

Oystercatcher
(Photo J.-D. Ludwigs)

3. Methods

The sampling, preparation and chemical analyses follow the standardized methodological guideline for JAMP Biota Monitoring (OSPAR 1997), which is part of the TMAP guidelines.

3.1 Collection of Egg Samples

Ten fresh eggs per species, site and year were taken under license (Table 2). Since in general intraclutch versus interclutch variation is low, except for mercury, one egg within a clutch was chosen randomly (for details and discussion see Becker et al. 1991). Because egg levels reflect the contamination of the egg-laying female (e.g. Becker et al. 1989, Lewis et al. 1993), the ten eggs collected per area and species indicate the current contamination of ten females breeding in the respective area and year. The eggs were frozen at –18°C until they were analyzed.

3.2 Physical Egg Parameters

Total egg weight (to the nearest 0.1 g), length, and width (0.01 mm) were recorded. In fertilized eggs, the embryonic age was determined by measuring the diameter of the eye (1 mm). The eggshells were air-dried and weighed (0.01 g), 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.

3.3 Chemicals Analyses

To avoid expensive intercalibration between several laboratories and to guarantee correct and exact results, since 1991 all egg samples from the international Wadden Sea have been analyzed in one lab, the ITI of the University of Applied Sciences Wilhelmshaven. The ITI participated in an intercalibration with two other labs, and in 1996, 1997 and 2000 in an international quality assurance (QUASIMEME project), whose results were ranked as satisfactory in most analyses.

3.3.1 Spectrum of examined chemicals

Besides the heavy metal mercury (Hg), 62 polychlorinated biphenyls (PCBs, Appendix 1) and 10 other organochlorine substances (see below) have been determined since 1991 (Becker et al. 1991, 1998). Most of the PCBs are baseline separated during the gaschromatographic separation, but 21 PCBs coelute in nine peaks (Appendix 1). 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 coplanar PCBs contribute significantly to the toxicity of the PCB mixtures (order of decreasing toxicity: non-, mono-, di-ortho-PCBs; see Parkinson & Safe 1987; Appendix 1).

The other organochlorine substances analyzed are hexachlorobenzene (HCB) and the insecticide p,p'-DDT (dichlorodiphenyltrichloethane), the isomer o,p'-DDD, and metabolites p,p'-DDD (dichlorodiphenyldichlorethane), o,p'-DDE, and p,p'-DDE (dichlorodiphenyldichlor-ethene; ΣDDT = sum of all isomers and metabolites), and the alpha-, beta- and gamma-isomers (Lindane) of hexachlorocyclohexane (ΣHCH). The o,p'-isomers of DDT, DDD and DDE were excluded from 1999 onwards, as they were no more detected. Since 1998, chlordanes have been included in the monitoring program. The levels of chlordane are presented as Σchlordane and Σnonachlor, corresponding to the sum of cis- and trans-chlordane, and cis- and trans-nonachlor, respectively).

Sample preparation (Photo R. Nagel)

3.3.2 Sample preparation for organochlorines: determination and measurement

The methods are in agreement with the OSPAR guidelines (OSPAR 1997). Sample preparation coincides with the method used by the Chemical Institute of School of Veterinary Medicine, Hannover (Heidmann 1986). All standards were obtained from Ehrenstorfer, Augsburg, Germany, and the solvents (for organic trace analysis) were obtained from Merck, Darmstadt, Germany.

Approximately 2 g of the fresh egg homogenate was blended with 100 µl internal standard (tetrabromobenzene and hexabromobenzene, 2 and 4 ppm, respectively) and dried by stirring with sodium sulphate. The mixture was given onto a column filled with two deactivated silica gel phases (10 % water and 40 % sulphuric acid) and eluted with n-hexane:dichloromethane (8:2), evaporated, and taken up in 250 µl toluene.

For the determination of the organochlorines, a mixture containing the following compounds was prepared: o,p'-DDT, o,p'-DDD, o,p'-DDE (until 1998 only), p,p'-DDT, p,p'-DDD, p,p'-DDE, a-, b- and g-HCH, HCB, trans- and cis-chlordane, trans- and cis-nonachlor (since 1998), and the PCB congeners 28, 52, 66, 92, 101, 118, 126, 138, 153, 155, 180, 189 and 194. Calibration of the GC-MS was carried out with seven standard solutions with a concentration range from 5 - 5000 ppb. The limit of determination varied between 0.3 – 0.4 ppb for the single organochlorines, but were 0.9 ppb for p,p'-DDT and 0.6 ppb for chlordanes and nonachlors, respectively.

A gaschromatograph HP5890, series II coupled with a mass selective detector HP 5971 (electron impact ionization, single ion monitoring) was used for the determination. All compounds could be separated in one run (30 min) with a HT-5 column (SGE, 25 m, 0.22 mm I.D., 0.1 µm layer) with helium as carrier gas. The identification took place by retention time and by the abundance of two representative masses of the ions (Büthe & Denker 1995). Quantification of the compounds was carried out by integration of a single mass (target ion). Due to their retention times, groups (timeframes) of ions were defined, to keep the number of ions to be measured at a time as low as possible, because of lowering the detection limit. Those PCB congeners, which were not included in the standard solution, were quantified by a PCB with the same degree of chlorination in the same group (Büthe & Denker 1995).

Gaschromatograph coupled with a mass selective detector (Photo R. Nagel)

3.3.3 Sample preparation for mercury: determination and measurement

The preparation coincides with the instructions of Kruse (1979). For digestion, approx. 100 mg egg content are blended in a test tube with 3 ml digestion acid, a mixture of nitric acid, chloric acid, and perchloric acid. The tube is closed with a capillary stopper and heated in a dry-block to 145 °C for two hours. After cooling off the sample, the graduated test tubes are filled up with bidest. water up to the desired volume.

Dependent on the expected amount of mercury, the standard solutions are prepared within a range from 0.5 $ng \cdot g^{-1}$ to 10 $ng \cdot g^{-1}$ egg content. For mercury determination, a carrier and a reductant solution are needed. The carrier solution is hydrochloric acid 1% and the reductant solution is stannous(II)-chloride-dihydrate 1.25% in hydrochloric acid 1%. A flow Injection Mercury System (FIMS 400, Perkin Elmer) with an integrated flow injection module of the FIAS series is used for the measurement. The determination limit is 0.1 $ng \cdot g^{-1}$.

3.4 Statistical Methods

The concentration of chemicals are given always in $ng \cdot g^{-1}$ fresh weight of egg content. Contaminant values were log-transformed (log n + 1) to achieve homogeneity of variances and normal distribution. Interspecific variation was analyzed by t-tests, intersite variation by ANOVA with post-hoc Scheffé tests. In some cases, non-parametric tests (Kruskal-Wallis) had to be used. To separate sampling sites by the patterns of chemical's concentrations in eggs, we also performed discriminant analyses. Spearman's rank correlation coefficient were calculated to reveal temporal trends on the basis of the raw data (n = number of the eggs; only in = four years of monitoring per site). Results were considered as significant at p-values < 0.05 (*), < 0.01 (**, highly significant), and < 0.001 (***, very highly significant). All tests were conducted two-tailed. All statistics were performed by SPSS 8.0 for Windows.

4.1 Interspecific Variability of Contaminants' Accumulation

The general pattern of contaminants' levels in eggs of the two species is shown in the samples of the year 2000 (Table 3). Among the chemicals analyzed, the summed PCB congeners had highest levels in the eggs of both species and all sites. Also mercury and ΣDDT were found in higher concentrations than the remaining contaminants.

In general, Common Tern eggs were significantly higher contaminated at most sampling sites than Oystercatcher eggs (Table 3). However, some exceptions were found: chlordane and nonachlor levels were higher in Oystercatcher eggs or similar between the two species. In the Ems estuary (Delfzijl, Dollart), Oystercatcher eggs had higher HCB and DDT levels than Common Tern eggs. On the other hand, eggs of Common Tern had higher proportions of PCB congeners of low degrees of chlorination (PCB_{3Cl} to PCB_{5Cl}), whereas Oystercatcher eggs were more contaminated with congeners of high chlorination degree (PCB_{6Cl} to PCB_{8Cl}; Table 4).

Common Tern	Hg	Σ PCB	Σ DDT	HCB	Σ HCH	Chlordane	Nonachlor
Balgzand	483.6 ± 221.1 N ***	1356,4 ± 220,8 N,M	95,2 ± 64,3 N	7,1 ±1,2 N,T	4,2 ± 0,9 N,T	0,2 ± 0,1	0,4 ± 0,2 **
Griend	304.9 ± 97.4 N	862,9 ± 244,4 N,T *	47,5 ± 13,5 N,T ***	4,8 ±1,0 J,D,O,N,T ***	3,3 ± 0,8 O,N,T **	0,1 ± 0,2 T **	0,5 ± 0,6 T **
Julianapolder	364.9 ± 60.0 N ***	1017,9 ± 197,6 N,M ***	89,9 ± 24,9 N	10,0 ±3,3 G,N,T,M	3,7 ± 0,6 N,T ***	0,2 ± 0,1 ***	0,5 ± 0,3 ***
Delfzijl	437.2 ± 107.8 N ***	1122,3 ± 334,5 N,M **	76,2 ± 43,5 N,T *	9,1 ±3,4 G,N,T **	3,1 ± 0,9 O,N,T	0,4 ± 0,1 M	1,0 ± 0,5 M **
Minsener Oog (Jade)	297.4 ± 81.6 N *	895,1 ± 296,2 N,T	93,3 ± 52,3 N ***	9,5 ±2,3 G,N,T,M ***	5,9 ± 1,9 G,D,N,T,M **	0,5 ± 0,2 M	0,9 ± 0,3 M **
Neufelderkoog (Elbe)	961.7 ± 348.1 B,J,G,D,O,T,M ***	2532,7 ± 591,1 B,J,G,D,O,M ***	390,4 ± 91,3 B,J,G,D,O,T,M ***	89,9 ±32,3 B,J,G,D,O,T,M ***	22,7 ± 6,3 B,J,G,D,O,M *	0,3 ± 0,1 T	0,4 ± 0,2 T ***
Trischen	422.7 ± 75.8 N ***	1805,7 ± 1025,9 G,O,M	157,7 ± 82,6 G,D,N,M	17,0 ±6,9 B,J,G,D,O,N,M **	27,7 ± 10,5 B,J,G,D,O,M **	0,6 ± 0,7 G,N,M	1,4 ± 1,5 G,N,M
Margrethekoog	297.7 ± 51.5 N	603,2 ± 163,7 B,J,D,N,T	59,6 ± 22,6 N,T	5,5 ±1,5 J,O,N,T	3,3 ± 0,8 O,N,T	0,0 ± 0,1 D,O,T	0,1 ± 0,1 D,O,T
Oystercatcher	**Hg**	**Σ PCB**	**Σ DDT**	**HCB**	**Σ HCH**	**Chlordane**	**Nonachlor**
Balgzand	208,5 ± 53,0 H ***	1112,9 ± 314,8 G,N,L	74,3 ± 40,5 G,H,L	8,2 ±5,3 D	11,4 ± 11,4	0,3 ± 0,2 J,Do	1,1 ± 0,6 J,Do **
Griend	275,2 ± 109,7 J,Do	621,7 ± 192,9 B,J,Do,H,L *	22,5 ± 6,6 B,J,D,H,T ***	2,5 ±1,0 D,Do,H,T ***	4,8 ± 1,1 H,T **	0,4 ± 0,1 J,Do **	1,1 ± 0,5 J,Do **
Julianapolder	140,6 ± 41,1 G,H,T ***	1895,3 ± 700,9 G,D,M,T,N,L ***	81,8 ± 27,2 G,M,L	7,9 ±3,6 D,L	12,4 ± 7,4 D,Do,M,N,L ***	1,1 ± 0,5 B,G,M,H,T,L ***	3,7 ± 2,2 B,G,M,H,T,L ***
Delfzijl	216,6 ± 83,5 H ***	755,8 ± 193,3 J,Do,L **	46,9 ± 16,8 G,Do,H,T *	43,0 ±40,7 B,J,G,M,T,N,L **	3,0 ± 0,7 J,H,T	0,5 ± 0,2	1,9 ± 0,6 **
Dollart	125,0 ± 15,4 G,M,H,T,L	1428,2 ± 230,3 G,D,M,N,L	97,9 ± 17,2 G,D,M,N,L	11,7 ±4,4 G,M,N,L	4,7 ± 1,1 J,H,T	1,2 ± 0,3 B,G,H,T,L	3,2 ± 0,9 B,G,H,T,N,L
Mellum (Jade)	216,2 ± 59,5 Do,H *	738,0 ± 199,1 J,Do,L	41,5 ± 11,5 J,Do,H,T ***	3,6 ±0,6 D,Do,H ***	3,7 ± 0,4 J,H,T **	0,5 ± 0,1 J	1,6 ± 0,6 J **
Hullen (Elbe)	358,2 ± 64,6 J,D,Do,M ***	1127,4 ± 222,1 G,N,L ***	131,9 ± 34,9 B,G,D,M,N,L ***	17,3 ±6,5 G,M,N,L ***	15,4 ± 3,3 G,D,Do,M,N,L *	0,3 ± 0,0 J,Do	1,1 ± 0,2 J,Do ***
Trischen	263,2 ± 53,2 J,Do ***	991,5 ± 207,7 J,N,L	119,6 ± 25,6 G,D,M,N,L	10,6 ±3,1 G,D,N,L **	15,2 ± 5,6 G,D,Do,M,N,L **	0,4 ± 0,1 J	1,2 ± 0,5 J,Do
Norderoog	203,9 ± 39,3 H	538,0 ± 199,3 B,J,Do,H,T,L	45,2 ± 17,7 G,Do,H,T	3,1 ±1,4 D,Do,H,T	4,2 ± 1,1 J,H,T	0,6 ± 0,3	2,0 ± 0,9
Langli	223,5 ± 35,2 Do	302,2 ± 99,2 B,G,J,D,Do,M,H,T,N	27,3 ± 5,2 B,J,Do,H,T	2,4 ±0,9 J,D,Do,H,T	4,2 ± 1,0 J,H,T	0,4 ± 0,1 J,Do	1,1 ± 0,4 J,Do

Table 3: Concentrations of all contaminants analyzed in Common Tern and Oystercatcher eggs collected at different breeding sites along the Wadden Sea coast in 2000. Mean concentrations ($ng \cdot g^{-1}$ fresh weight of egg content) and standard deviation are presented. The first letter of the sites' name below the values on the cell's left indicate significant differences between sites (ANOVA, Scheffé tests). Sites used to test interspecific differences (* p<0.05, ** p<0.01, *** p<0.001, t-test; on the cell's right) are shaded. See Table A2 and A3 in Appendix for values of all single compounds.

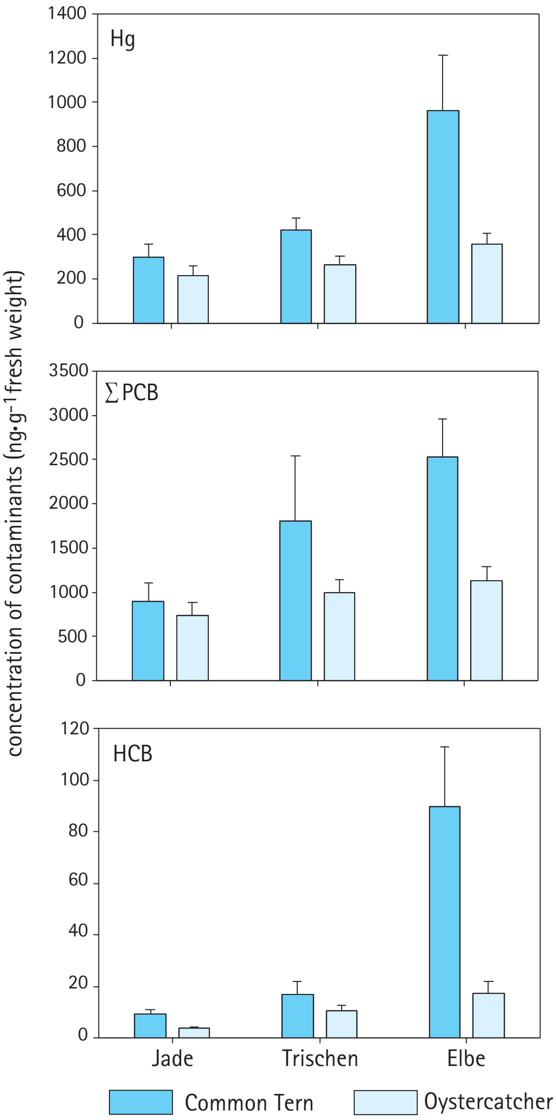

Figure 2: Interspecific variation in the concentrations of mercury, HCB and ΣPCB from selected breeding sites in eggs sampled in 2000. Mean concentration ($ng \cdot g^{-1}$ fresh weight of egg content) and 95% confidence intervals are presented.

The interspecific variation in accumulation is exemplary shown in Fig. 2. The interspecific difference in egg pollution is increasing with the level of contamination at the respective site: At those sites where the general concentration of the contaminants was high (Elbe, Trischen), the analyzed eggs show noticeable and higher differences in the levels of contamination between both species than at sites of lower pollution (Jade).

In 2000, at the Elbe for example (Fig. 2, Table 3), contaminant levels of Common Tern eggs were higher than those of Oystercatcher eggs, for mercury 2.7 fold, HCB 5.2 fold and PCBs 2.2 fold. Eleven years before (1989), when contamination was on much higher levels (Figs. 8, 9), these factors were much higher in mercury and PCBs (mercury 15.7, HCB 4.9 and PCBs 3.4; Becker et al. 1991).

4.2 Spatial Trends in Contamination in the Year 2000

Intersite variation of chemicals' levels in coastal bird eggs in the Wadden Sea is exemplary presented for the most recent year of this study (2000), when all sites were covered by sampling (Fig. 3, see Appendix 2, 3 for egg levels of all compounds analyzed).

Common Tern

The spatial pattern of Common Tern pollution is very pronounced and distinctly shows a higher egg pollution at the Elbe estuary and the inner German Bight (Fig. 3, Table 3). The contaminants' values in eggs of the Dutch and Danish parts of the Wadden Sea were on a similar and lower level. At Balgzand, slightly higher mercury, ΣPCB and ΣDDT levels were determined than at the other Dutch sites of the western part of the Wadden Sea. The concentrations of mercury, ΣDDT and HCB in eggs sampled at the Elbe were significantly higher than in eggs from the other sites, and were usually followed by the eggs collected on Trischen (Fig. 3). Also the PCB and ΣHCH levels in bird eggs found at the Elbe and on Trischen were found to be considerably higher in comparison to samples found on other sites. At all sampling sites, the contamination of bird eggs by DDT was mainly based on the metabolite p,p'-DDE. The samples of the Elbe displayed the highest values of this metabolite (Appendix 2).

Oystercatcher

In contrast to the Common Tern, the spatial pattern of Oystercatcher egg pollution was more divers and not as distinct (Fig. 3, Table 3). The highest rank in contamination among the sites was also taken at the Elbe estuary (and/or on Trischen) in case of mercury, ΣDDT and ΣHCH, but changed to other sites with respect to ΣPCB (Julianapolder and Dollart where remarkable high levels of PCBs were found) and with respect to HCB (Delfzijl). ΣPCB and HCB concentrations were considerably high at Delfzijl, but not significantly different from the samples obtained at the Elbe and Dollard. This result holds also true for the former years 1998 and 1999. Eggs from Balgzand and Julianapolder also contained high levels of ΣDDT and ΣHCH, besides eggs from the Elbe. The relatively high contamination of Oystercatcher eggs from Julianapolder in 2000 with ΣPCB, ΣDDT, ΣHCH, chlordane and nonachlor was not as pronounced in 1998 and 1999. However, also in these last mentioned years, at Julianapolder, higher egg residues of these contaminants than in eggs from the more western site Griend were found. Furthermore, the Oystercatcher eggs from Julianapolder in 2000 were higher contaminated with ΣHCH than eggs from Balgzand, and also higher with chlordanes than eggs from Delfzijl.

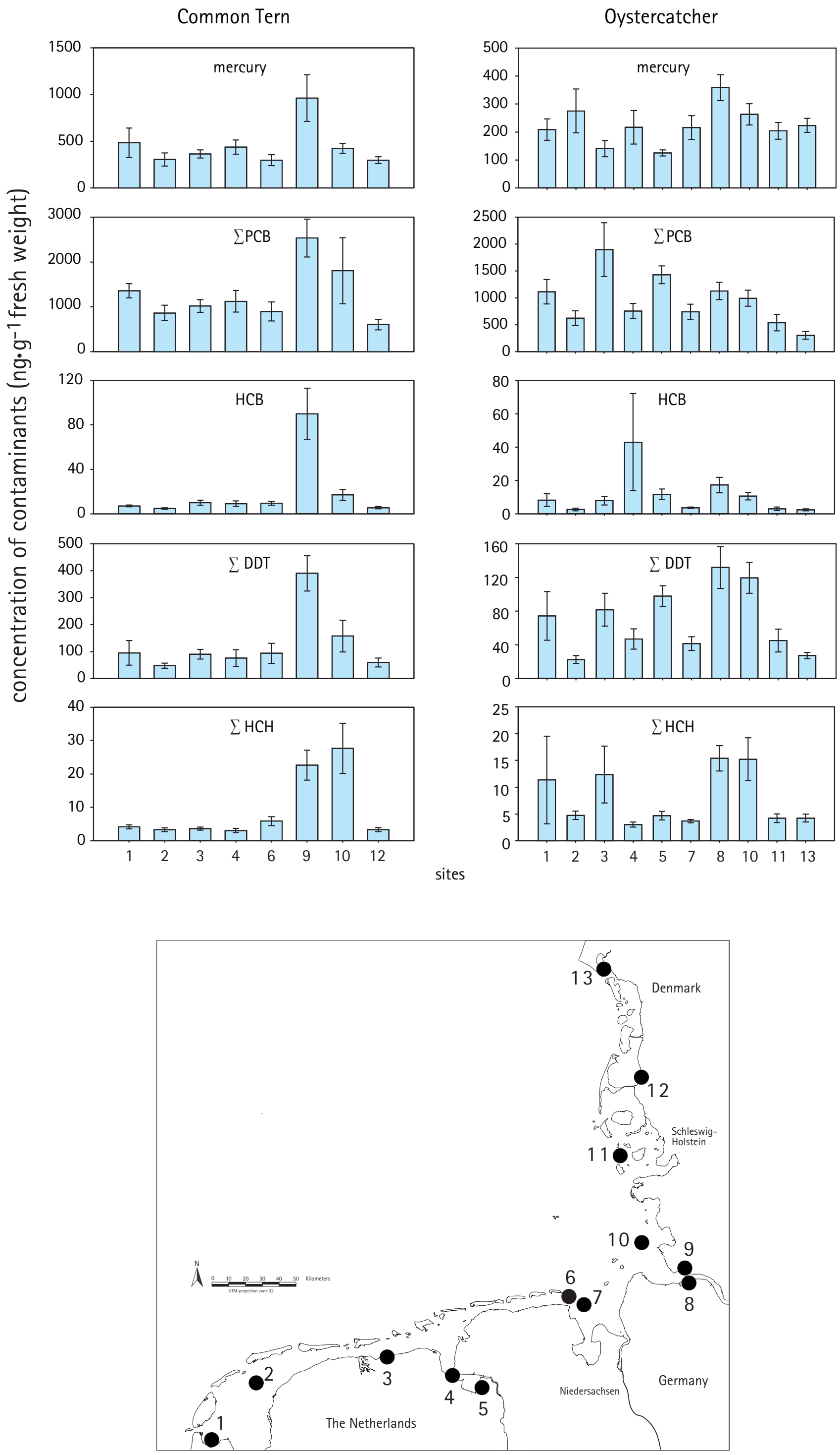

Figure 3: Geographic variation of the most important contaminants in Common Tern and Oystercatcher eggs in 2000. Mean concentration (ng·g^{-1} fresh weight of egg content) and 95% confidence intervals are presented. N=10 eggs per site and species were analyzed (Oystercatcher, Norderoog: n=9).

Table 4:
Spatial variation in the PCB composition in eggs of Common Tern and Oystercatcher in 2000. The average proportion (%) of PCBs of different degree of chlorination in the PCB mixture of 62 congeners. Highly significant intersite differences were found within each chlorination group and in both species (p<0.001, respectively, K-W-tests). Most important higher values are indicated in bold.

Common Tern	PCBs (%)					
	3 Cl	4 Cl	5 Cl	6 Cl	7 Cl	8 Cl
Balgzand	**0,4**	**3,4**	**16,1**	52,7	**24,3**	**3,1**
Griend	**0,4**	2,9	**16,4**	53,4	24,0	2,9
Julianapolder	**0,5**	**3,6**	**18,0**	53,1	22,0	2,9
Delfzijl	**0,4**	**3,0**	**16,8**	53,3	23,6	2,9
Minsener Oog (Jade)	**0,4**	**3,7**	**17,9**	52,3	22,8	2,9
Neufelder Koog (Elbe)	0,3	2,3	13,0	**55,8**	**25,8**	2,8
Trischen	0,2	2,0	14,2	**56,9**	**24,1**	2,5
Margrethekoog	0,2	2,1	13,9	**56,4**	**24,3**	**3,0**
Oystercatcher	PCBs (%)					
	3 Cl	4 Cl	5 Cl	6 Cl	7 Cl	8 Cl
Balgzand	0,3	**2,5**	**15,1**	55,6	23,7	2,8
Griend	0,4	**2,1**	**13,8**	**56,5**	23,9	3,4
Julianapolder	0,4	2,3	**13,7**	55,9	24,6	3,1
Delfzijl	0,4	1,9	12,0	53,6	**28,3**	**3,7**
Dollart	0,4	**2,3**	12,9	55,2	26,0	3,3
Mellum (Jade)	0,3	2,1	**13,5**	**57,1**	24,0	3,0
Hullen (Elbe)	0,1	1,0	8,6	54,8	**31,5**	**4,0**
Trischen	0,2	1,5	10,8	**58,0**	**26,2**	3,2
Norderoog	0,3	1,5	11,0	**58,6**	24,9	**3,6**
Langli	0,4	2,4	**13,5**	56,1	24,3	3,4

PCB-Mixtures

The composition of the PCB mixture (Table 4) showed some intersite variation which can be generalized in both species as follows: In the western part of the Wadden Sea, the PCB congeners of low chlorination degree (PCB_{3Cl} to PCB_{5Cl}) show higher proportions than in the eastern Wadden Sea, where congeners of high degree of chlorination (PCB_{6Cl} to PCB_{8Cl}) are found in higher proportions. In the Oystercatcher eggs from the Elbe, the PCB-mixture differed from that of the other sites. At Delfzijl, the composition was more similar to that found at the sites in the inner German Bight.

In accordance with the general spatial PCB-pollution pattern, highest levels of the more toxic PCB congeners, non-, mono- and di-ortho-PCBs, were observed in Common Tern eggs from Neufelder Koog (Elbe estuary, Table 5). These values were significantly different from those measured in samples from Trischen, and Balgzand, which in general also displayed high concentrations of these toxic congeners. Oystercatcher eggs from Julianapolder exhibited higher values of these congeners, usually followed by the samples from the Dollart.

Table 5:
Intersite variation in the levels of coplanar PCBs (non-, mono-, and di-ortho congeners, see Table A1 in Appendix 1 for single congeners) in eggs of Common Tern and Oystercatcher. Mean concentrations ($ng \cdot g^{-1}$ fresh weight) and standard deviations. Highly significant intersite differences were found within each coplanar PCB group (p<0.001, respectiveley; K-W-testes).

Common Tern	Non-ortho-PCBs	Mono-ortho-PCBs	Di-ortho-PCBs
Balgzand	0.5 ± 0.1	105.5 ± 20.5	216.4 ± 34.2
Griend	0.4 ± 0.1	66.7 ± 16.9	131.9 ± 36.9
Julianapolder	0.6 ± 0.1	92.2 ± 19.4	149.5 ± 39.2
Delfzijl	0.5 ± 0.1	90.7 ± 31.9	170.7 ± 55.8
Minsener Oog	0.5 ± 0.1	74.3 ± 23.4	134.7 ± 43.7
Neufelder Koog	0.9 ± 0.2	150.3 ± 36.3	420.7 ± 101.3
Trischen	0.7 ± 0.3	126.6 ± 70.1	296.6 ± 169.7
Margrethekoog	0.3 ± 0.1	49.9 ± 13.9	105.4 ± 29.4
Oystercatcher	**Non-ortho-PCBs**	**Mono-ortho-PCBs**	**Di-ortho-PCBs**
Balgzand	0.6 ± 0.2	87.9 ± 22.9	173.3 ± 49.4
Griend	0.2 ± 0.1	62.7 ± 20.7	93.4 ± 29.6
Julianapolder	0.6 ± 0.2	175.1 ± 59.9	305.9 ± 110.7
Delfzijl	0.6 ± 0.4	66.7 ± 26.1	118.7 ± 38.2
Dollart	0.6 ± 0.2	119.6 ± 19.1	225.5 ± 38.3
Mellum	0.4 ± 0.1	75.2 ± 19.7	114.1 ± 30.9
Hullen	0.4 ± 0.1	70.1 ± 13.8	175.9 ± 39.5
Trischen	0.4 ± 0.1	76.3 ± 14.5	155.8 ± 32.4
Norderoog	0.3 ± 0.1	45.5 ± 15.8	92.0 ± 35.6
Langli	0.2 ± 0.1	31.1 ± 11.2	48.5 ± 13.7

Chlordanes

In Common Tern eggs, the concentrations of chlordanes reached the highest levels in samples from Trischen, Oldeoog (Jade Bay) and Delfzijl. In samples from Margrethekoog, residues of chlordane were very low (almost not detectable). Oystercatcher eggs, however, showed a different spatial pattern, and eggs from Julianapolder and Dollart had the highest levels of chlordane and nonachlor.

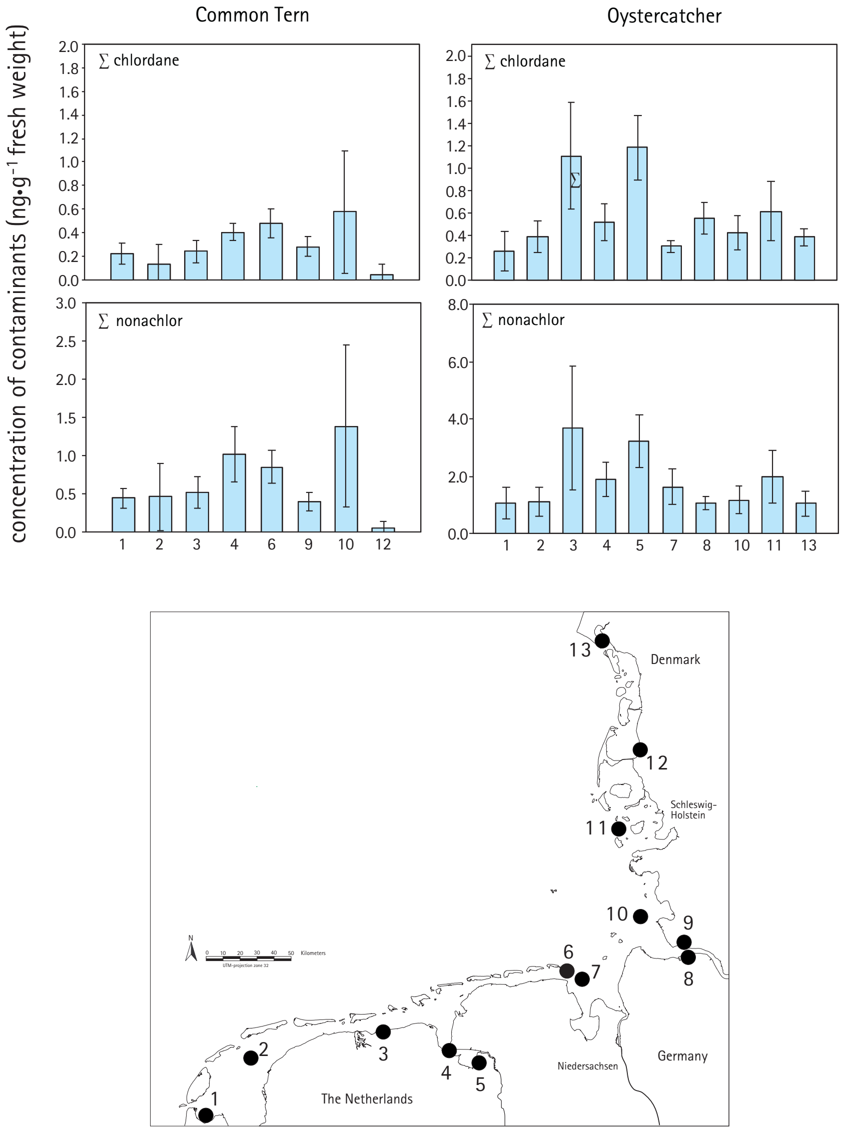

Figure 4: Spatial variation in the concentrations of Σchlordane and Σnonachlor in Oystercatcher and Common Tern eggs in 2000. Mean concentration (ng·g^{-1} fresh weight of egg content) and 95% confidence intervals are presented; n = 10 eggs each (Norderoog: n=9).

Site-specific contaminants' mixtures

To reveal intersite variation in the composition and pattern of the pollutant mixture in the eggs sampled, we applied discriminant analyses for each species on the basis of all studied single contaminants. In case of the Common Tern, each sampling site could be very well separated from the others (Fig. 5). The contaminants that displayed the highest discriminant value characterizing the sites were PCB congeners with high chlorination degree (given in order of decreasing importance: PCB171, PCB160, PCB175, PCB118 and PCB172; see Appendix 2). The breeding sites located at the Elbe, Julianapolder, Trischen (Common Tern), and Dollart (Oystercatcher) could be better separated from the other sites on the basis of the contaminants' pattern. Also in this species the concentrations of specific PCB congeners with high degrees of chlorination were the discriminant variables of importance (PCB177, PCB175, PCB153, PCB160, and PCB156; see Appendix 3). Consequently, the eggs laid at one site are clearly characterized by the contaminants' site-specific mixture. Theoretically, by analysis of an egg from unknown origin its provenance can be predicted.

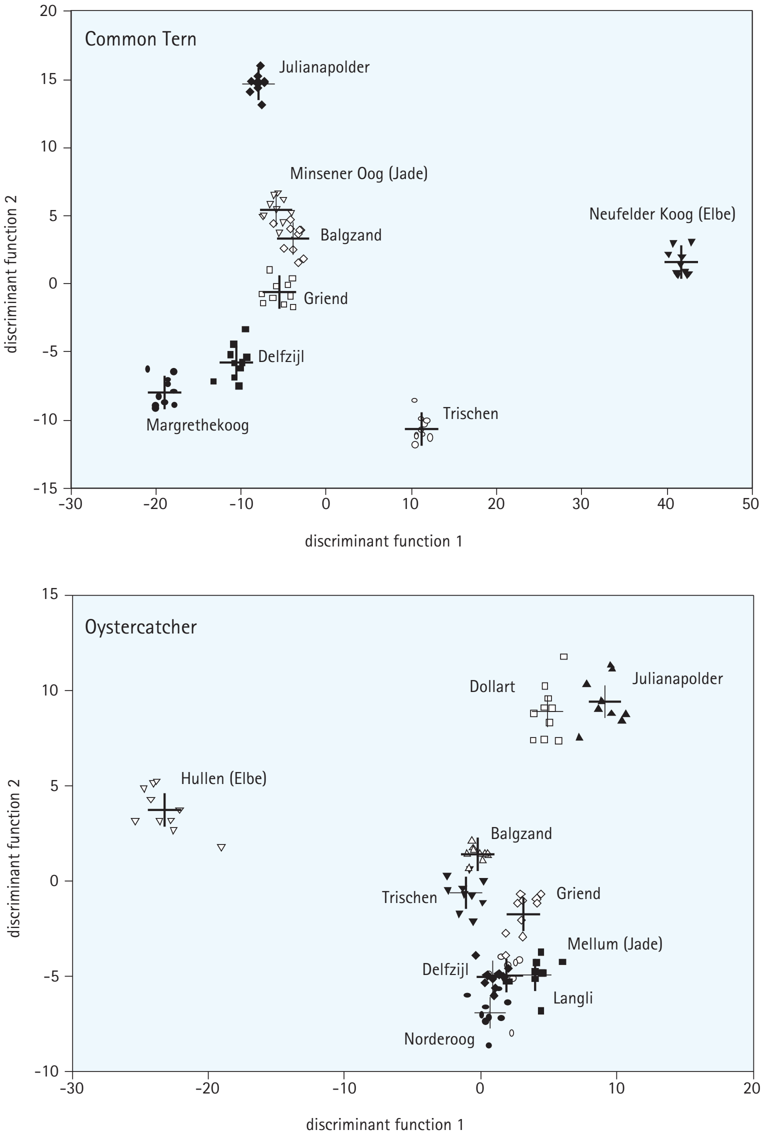

Figure 5: Diagram of the results of discriminant analyses for separation of breeding sites (different symbols) on the basis of concentrations of all contaminants measured (Hg, HCB, 62 PCB-congeners, DDT and metabolites, HCH-isomers, Chlordane (cis- and trans-chlordane) and Nonachlor (cis and trans-nonachlor) in eggs of Common Tern and Oystercatcher collected in 2000. Main discriminant variables in order of decreasing importance; Common Tern: PCB171, PCB160, PCB175, PCB118 and PCB172; Oystercatcher: PCB177, PCB175, PCB153, PCB160, and PCB156.

4.3 Temporal Trends

4.3.1 Temporal trends 1991 – 2000

In general, with some exceptions that will be mentioned below, in both species the levels of contaminants decreased during the last decade (Figs. 6, 7; Table 6). Some positive trends, however, have been detected, too (Table 6): Slight increases have been observed in the levels of HCB, ΣDDT, and ΣHCH in the Common Tern samples from the Elbe, as well as in the mercury and PCBs concentrations at the Dutch locality Delfzijl during the three years under study. Increases in Oystercatcher egg contamination were detected at Balgzand, Julianapolder (from 1997-2000, respectively) and Hullen: At Balgzand, a significant increase was only observed in the HCB concentration, whereas in Julianapolder, with exception of mercury, all the contaminants were found to have increased significantly. At Hullen (Elbe), the mercury level increased significantly during the last decade, too.

During the decade 1991-2000, the composition of the PCB mixture did change (Table 7): The proportion of PCB congeners of lower degree of chlorination showed reductions with the years [Common Tern: Elbe ($p<0.01$, Spearman rank correlations), Trischen ($p<0.05$); Oystercatcher: Trischen and Jade ($p<0.001$, respectively), Griend ($p<0.01$), and Norderoog ($p<0.05$)].

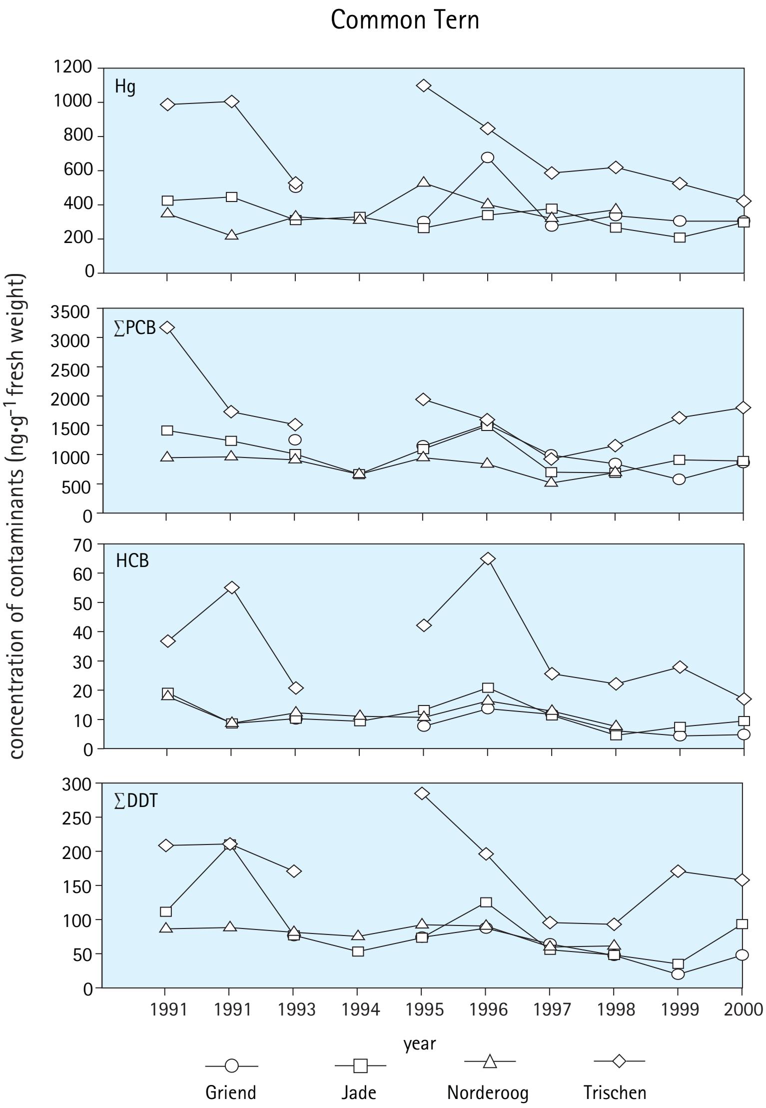

Figure 6: Temporal trends of mercury, sum of PCB congeners (ΣPCB), HCB and Sum of DDT and metabolites (ΣDDT) concentrations in eggs of Common Tern from selected sampling sites 1991–2000. Arithmetic means ($ng \cdot g^{-1}$ fresh weight of egg content) are presented.

Figure 7: Temporal trends of mercury, sum of PCB congeners (ΣPCB), HCB and Sum of DDT and metabolites (ΣDDT) concentrations in eggs of Oystercatcher from selected sampling sites 1991-2000. Arithmetic means (ng·g[-1] fresh weight of egg content) are presented.

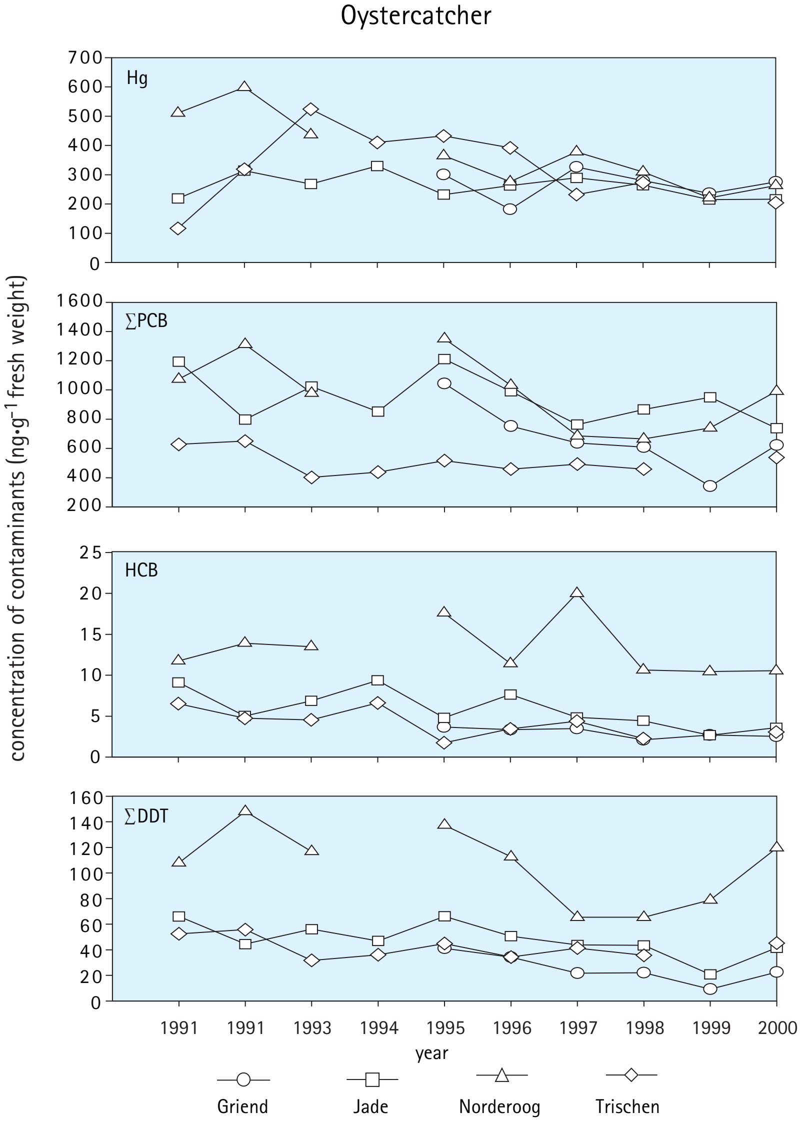

Table 6: Temporal trends in pollutant levels in Common Tern (CT) and Oystercatcher (OC) eggs from 1991 until 2000. Numbers of years studied at each site in brackets. Only trends from sites studied for more than three years are included. For significant trends, Spearman rank coefficients (r_s) calculated on the basis of n eggs and p-values are presented. n.s: not significant, *<0.05, **<0.01, ***<0.001. Elbe; Common Tern = Hullen 1991-1995 + Neufelderkoog 1996-2000; Oystercatcher = Hullen. Positive trends are given in bold.

Common Tern	Hg	HCB	ΣPCB	ΣDDT	ΣHCH
Balgzand (4)	n.s.	n.s.	n.s.	n.s.	n.s.
Griend (7)	-0.40 **	-0.64 ***	-0.49 ***	-0.55 ***	-0.70 ***
Julianapolder (4)	n.s.	n.s.	n.s.	n.s.	n.s.
Minsener Oog (Jade, 10)	-0.39 ***	-0.33 **	-0.30 **	-0.37 ***	n.s.
Elbe (10)	-0.55 ***	**0.21 ***	n.s.	**0.26 ****	**0.28 ****
Trischen (9)	-0.52 ***	-0.27 *	-0.35 **	-0.32 **	n.s.
Norderoog (8)	n.s.	n.s.	-0.39 ***	-0.30 **	-0.34 **
Oystercatcher	**Hg**	**HCB**	**ΣPCB**	**ΣDDT**	**ΣHCH**
Griend (6)	n.s.	-0.46 **	-0.57 ***	-0.59 ***	-0.54 ***
Julianapolder (4)	-0.77 ***	**0.56 *****	**0.47 ****	**0.34 ***	**0.57 *****
Dollart (9)	n.s.	-0.43 ***	n.s.	n.s.	-0.30 *
Mellum (Jade, 10)	-0.22 *	-0.59 ***	-0.21 *	-0.41 ***	-0.67 ***
Elbe (10)	**0.43 *****	n.s.	-0.34 **	-0.31 **	n.s.
Trischen (9)	-0.69 ***	n.s.	-0.42 ***	-0.31 **	-0.58 ***
Norderoog (9)	n.s.	-0.46 ***	n.s.	n.s.	-0.57 ***

Table 7: Temporal trend in the PCBs' composition in eggs of Common Tern and Oystercatcher. The intersite range (see Table 2 for the sites studied) of the average proportion (%) of PCBs with low chlorination degree (PCB_{3Cl}, PCB_{4Cl}, PCB_{5Cl}) and with high chlorination degree (PCB_{6Cl}, PCB_{7Cl}, PCB_{8Cl}) in the PCB mixture of 62 congeners is presented.

	Common Tern		Oystercatcher	
year	$\%PCB_{3Cl-5Cl}$	$\%PCB_{6Cl-8Cl}$	$\%PCB_{3Cl-5Cl}$	$\%PCB_{6Cl-8Cl}$
1991	17.1-31.1	68.9-82.9	20.3-23.4	76.6-79.7
1994	25.1-28.4	71.6-74.9	16.8-21.8	78.2-83.2
1997	21.8-30.6	69.4-78.2	16.6-25.5	74.5-83.4
2000	15.5-22.2	77.8-84.5	9.7-17.6	82.4-90.3

4.3.2 Temporal trends 1981 – 2000

The comparison of the temporal trends from the 1990s – the period in focus of this report – with the temporal development of coastal bird pollution during the 1980s in the German Wadden Sea reveals that the burden of Common Tern and Oystercatcher eggs with environmental chemicals had decreased strongly since the beginning of the 1990s (Figs. 8, 9). Especially the contamination with mercury, PCBs and HCB (Fig. 11) from 1990 onwards, and also the strong interannual fluctuations of the concentrations in eggs from the Elbe estuary, Trischen and Jade were constantly reduced. This process of pollutant reduction was most pronounced in samples from the Elbe estuary, followed by Trischen and Jade. Concentrations in the 1990s were roughly more than half of those from the decade before (Figs. 8, 9), but since the mid 1990s, the decrease of concentrations seems to stagnate at levels above the target concentrations.

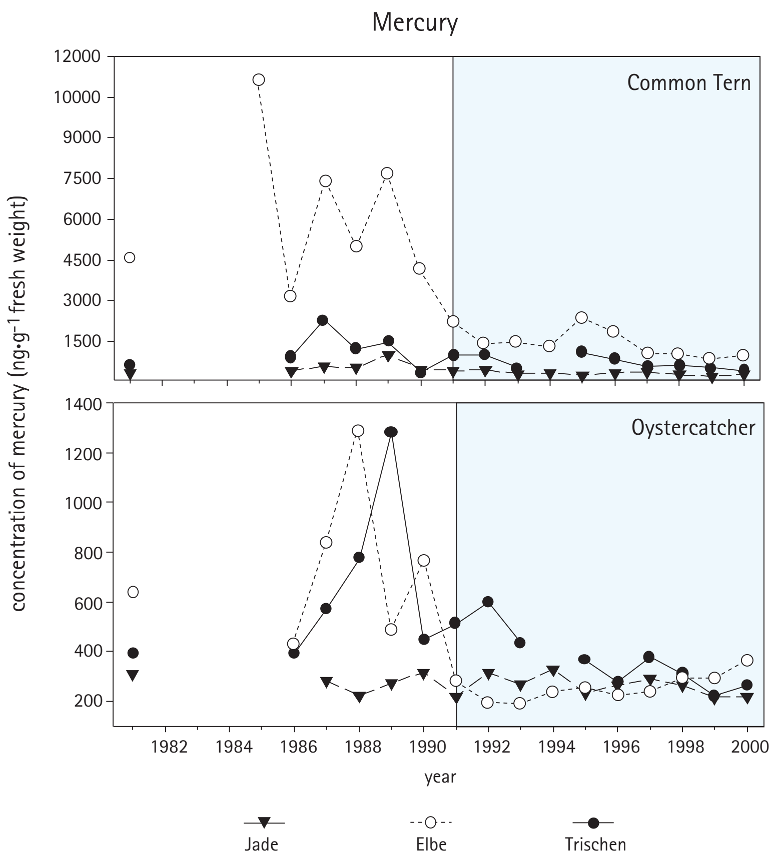

Figure 8: Temporal trends of mercury concentrations ($ng \cdot g^{-1}$ fresh weight, arithmetic means) in Common Tern and Oystercatcher eggs from selected breeding sites from the German Wadden Sea 1981–2000. The period covered by this report is colored. Data from 1981–1990 after Becker et al. 1991, 1992.

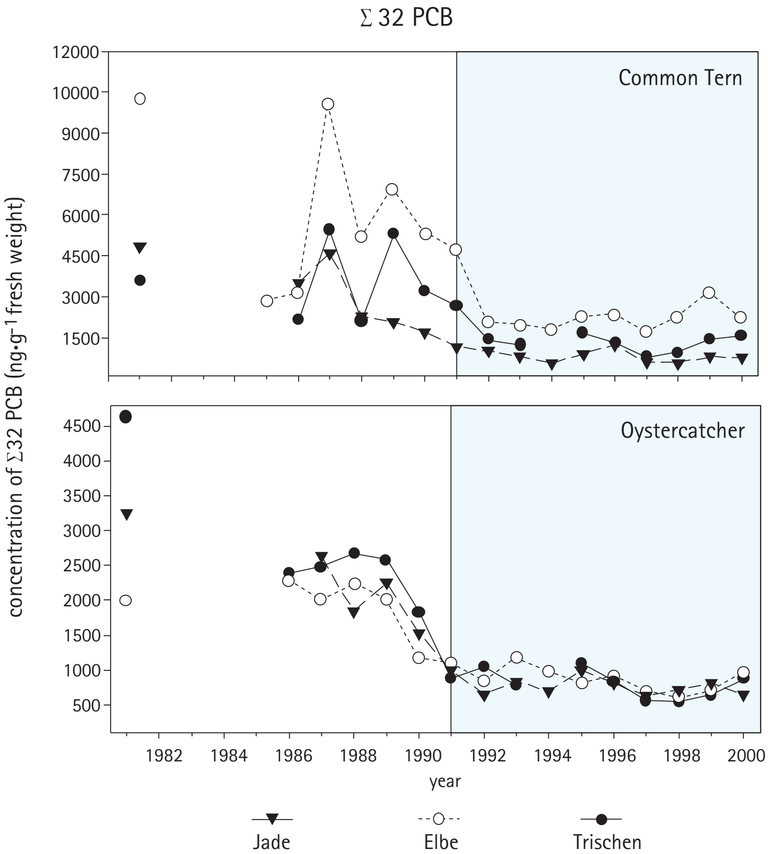

Figure 9: Temporal trends of Σ32PCB (sum of 32 congeners) concentrations ($ng \cdot g^{-1}$ fresh weight, arithmetic means) in Common Tern and Oystercatcher eggs from selected breeding sites from the German Wadden Sea 1981–2000. The period covered by this report is colored. Data from 1981–1990 after Becker et al. 1991, 1992.

5. Discussion

The results presented in this report substantiate the success of the implementation of the parameter "Contaminants in Bird Eggs" within the TMAP during the late 1990s and confirms its usefulness to assess the current ecological state of the Wadden Sea ecosystem with respect to contamination. In the discussion we focus on the following aspects:

- The suitability and meaning of coastal bird eggs contamination as parameter to monitor chemical pollution in the Wadden Sea,
- the variability of the contamination of coastal birds in the Wadden Sea, dependent on species, site and year, with the aim to understand the patterns found and to recognize causes of spatial and temporal trends,
- the evaluation of the recent pollution levels with respect to possible effects on birds,
- the assessment of the bird contamination state regarding the Wadden Sea ecotargets, and
- the significance of the parameter within the TMAP and recommendations to enhance its meaning.

5.1 Suitability of Coastal Bird Eggs Contamination as Parameter to Monitor Chemical Pollution in the Wadden Sea

Birds are good indicators of those environmental chemicals, which tend to biomagnify through food-chains and which are accumulated in lipid-rich tissues, e.g. organochlorines and methylmercury. The relevance of contaminants' levels in organisms in high trophic positions like many birds (Fig. 10) as warnings of human health risks may be greater than their concentrations in water, sediment or soil (ICES 1999). In particular, the ability to "integrate pollutant signals over time and space" by bioaccumulating contaminants in tissues means that to obtain a given level of accuracy measurements, a smaller number of animal samples is required than of physical samples (Furness et al. 1993) thus increasing the power of trend analyses. This is especially true in birds: together with the biomagnification effect (Fig. 10), the lower within-sample variance, especially in the egg as matrix (Gilbertson et al. 1987, ICES 1999), means that spatial or temporal differences in con-

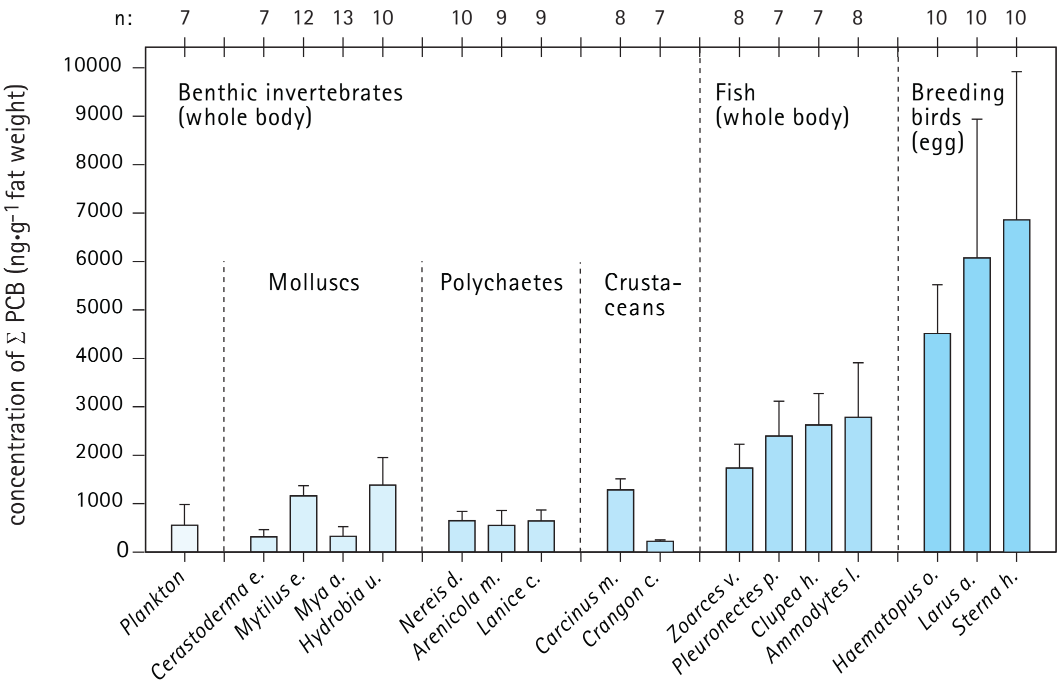

Figure 10: Bioaccumulation of PCBs in the food web of the Wadden Sea. Sum of concentration of 8 PCB-congeners (ng·g^{-1}, means ± 1 standard deviation on fat weight basis) is presented for plankton, 9 benthic invertebrates (*Cerastoderma edule, Mytilus edulis, Mya arenaria, Hydrobia ulvae, Nereis diversicolor, Arenicola marina, Lanice conchilega, Carcinus maenas, Crangon crangon*), 4 fishes (juvenile stages: *Zoarces viviparus, Pleuronectes platessa, Clupea harengus, Ammodytes lancea*) and 3 coastal bird species (*Haematopus ostralegus, Larus argentatus, Sterna hirundo*). After Mattig et al. (1996).

tamination are more conspicuous than by physical samples or by samples of the birds' prey.

Both, laboratory experiments and oral dosing of birds in the field with mercury and other chemicals, have shown that concentrations in tissues like liver, kidney, feathers and eggs are dose-dependent (e.g. mercury: Teijning 1967, Lewis & Furness 1991). For this reason, birds indicate the current environmental burden with a chemical, and also react relatively fast to its change (e.g. mercury can be detected in the egg within a few days after dosing; Bäckström 1969). This is clearly shown by the long-term data of Common Terns on the Elbe river (Fig. 11), of Guillemots *Uria aalge* in the Baltic (Bignert et al. 1998), as well as by small-scale spatial patterns in bird contamination (see below).

Birds' contamination reflects that of the food they eat (e.g. Nisbet & Reynolds 1984, Dirksen et al. 1995). In coastal birds of the Wadden Sea, the fish-feeding species like terns and the Herring Gull (*Larus argentatus*) were found to be more contaminated than benthic feeders such as Oystercatchers (Fig. 10, Becker 1991, Becker et al. 1991, Mattig et al. 2000; Thyen & Becker 2000).

The bird egg has been proven to be a favorable matrix and has been used as sample unit in numerous studies and monitoring projects. The removal of eggs is less damaging than that of adults, having only a minor impact on the hatching success of the studied population. Intraclutch variation between egg chemical levels is low, so one egg taken from a clutch provides all the information needed. Several studies have shown seabird eggs to be good indicators of local pollutant contamination, even in migrating species like terns, since concentrations in eggs tend to reflect pollutant uptake by the female foraging close to the colony in the few days prior to egg laying. Advantages and disadvantages of eggs as a matrix are discussed, e.g., by Gilbertson et al. (1987), Becker (1989), Furness (1993), Walker (1994), Becker et al. (1991, 1998), and ICES (1999):

Advantages of the matrix egg: High lipid content and accumulation of lipophilic persistent compounds; consistent composition; originate

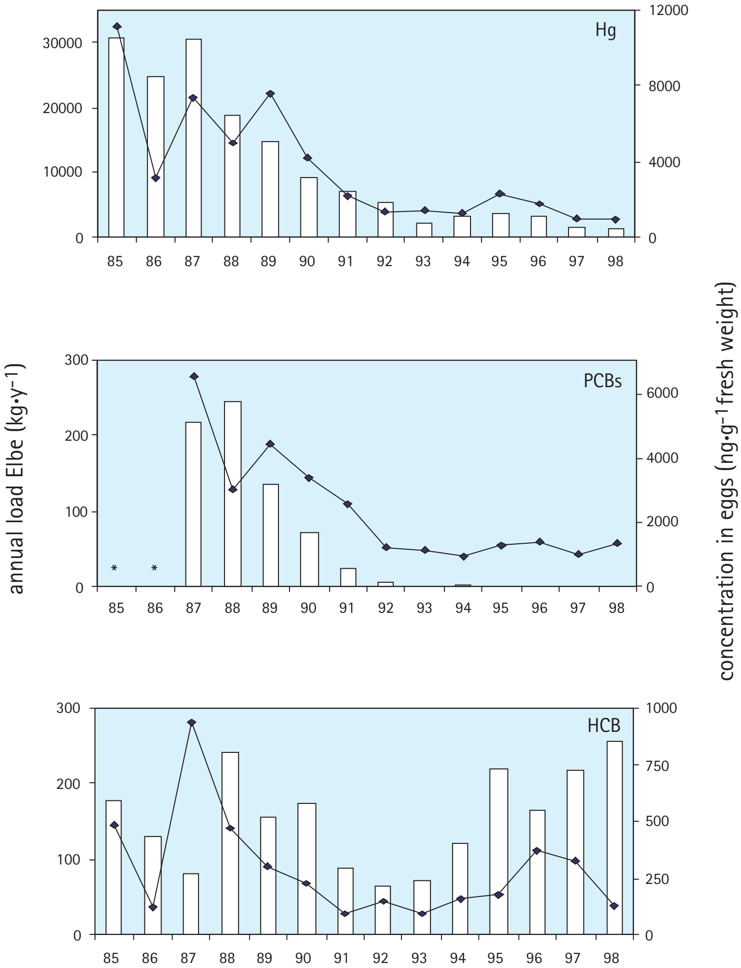

Figure 11: Concentrations of chemicals in Common Tern eggs (lines) reflect the annual loads of the river Elbe (columns). Mercury, PCBs and HCB are chemicals discharged in vast quantities by the Elbe. Annual loads are derived from the sum of weekly discharges from June of the previous year until May of the reference year when the eggs have been sampled. PCBs: sum of the congeners CB101, 138, 153 and 180. *: no data available; from 1993–1998, weekly water concentrations of PCBs were often below the detection limit; therefore, no loads have been calculated. Data from annual reports of the Wassergütestelle Elbe and from Becker et al. (1998).

from a defined area and year; reflection of the contamination of breeding females (healthy and reproductive part of the population); being restricted to the breeding season, reduced seasonal variability in chemicals' levels due to limitations of the breeding period; ease and speed of sampling; ease of handling and storing samples; sensitive reaction of birds during egg development and early chick stage to toxic chemicals and relevance of egg residues to embryotoxic effects; feasibility of studies bearing on the relationships between contaminants, eggshell quality and hatching success.

Disadvantages: Relevance to only a part of the population (reproductive females) and of the year (breeding season); variation of pollutant levels with the laying sequence; failure of some heavy metals to accumulate in the egg (e.g. cadmium and lead).

The advantages of the matrix egg were clearly confirmed by its use during the first years within the TMAP. This matrix is the basis for the quick and "in time" analyses of the present bird contamination in the Wadden Sea, as well as for the reliable, representative and repeatable monitoring results we were able to present by this report. Also the JAMP guidelines of this parameter (OSPAR 1997), supplemented by the mostly fulfilled recommendations of the pilot phase (Becker et al. 1998), have been a good basis to run the parameter in a meaningful way for monitoring the ecological state of the Wadden Sea.

Trischen, FRG
(Photo P. Becker)

5.2 The Variability of the Contamination of Coastal Birds in the Wadden Sea

5.2.1 Interspecific variation

The observed differences in the contamination between Common Tern and Oystercatcher eggs can be explained to a great extent by their different biology, especially in diet choice, reproductive ecology and migration (Becker et al. 1998, Thyen & Becker 2000a). The Common Tern is a long distance migrant arriving in the Wadden Sea two to three weeks before egg laying. During the period of courtship, the females have to feed great quantities of fish as the main diet taken exclusively from the breeding area to be able to produce the clutch (Nisbet 1973, Becker et al. 1987, Wendeln & Becker 1996). Oystercatchers mostly stay at the Wadden Sea during winter and feed on benthic invertebrates (Hulscher 1985, Goss-Custard 1996). When feeding inland they take edaphic invertebrates (living in soil).

Common Tern eggs contained higher residues of most persistent contaminants than Oystercatcher eggs, because the terns feed on fish occupying higher trophic levels than benthic organisms taken by Oystercatchers (Fig. 10). Through biomagnification, fish is contaminated higher by most of the environmental chemicals examined than benthic invertebrates (Fig. 10, Mattig et al. 1997). This fact explains the higher tern contamination although Oystercatchers are exposed to the chemical load of the Wadden Sea for longer periods of the year than the terns. Thus, Common Tern and Oystercatcher reflect the contaminants' load of their prey occupying specific trophic levels.

However, the degree of the interspecific difference is linked with the level of chemical contamination in the environment: The higher the environmental contamination with a compound, the greater is the difference between Common Tern and Oystercatcher egg levels. Consequently, the difference between the two species in egg contamination is reduced if the total chemical burden of the ecosystem declines spatially (Fig. 2) or with regard to time (see 5.3). That means that the Common Tern is characterized by higher accumulation rates than the Oystercatcher (ICES 1999), caused by the trophic differences discussed above, and possibly by the relatively higher amounts of food a tern female has to assimilate in a short period to produce the clutch. The mass of an average clutch of three eggs in the Common Tern is about 50% of a females body mass, in the Oystercatcher only about 26% (data from Bezzel 1985).

There are a few exceptions showing higher Oystercatcher than Common Tern egg contamination (for HCB at Delfzijl see 6.2.2). Generally, Oystercatcher eggs had higher levels of nonachlor, and at the lower contaminated sites, also higher ΣHCH residues than Common Tern eggs. With respect to HCH, we can explain this result by different fat-solubility of the substances and by different fat content of prey: Polychaetes, which belong to the preferred Oystercatcher diet, are more often highly contaminated by HCH isomers than fish (Mattig et al. 1997), resulting in the higher Oystercatcher pollution with these compounds. If this could be also the explanation for higher nonachlor levels in Oystercatcher eggs is unclear.

The exposure to environmental chemicals stimulate the activity of catabolizing enzymes in the liver of the birds. This could be another source of interspecific variation in egg residues, especially of PCBs (Elliott et al.1990, Schwarz & Stalling 1991). In birds which are confronted continuously with higher amounts of chemicals (e.g. Oystercatcher staging in the Wadden Sea throughout the year), such enzymes are more induced. Therefore, these birds are able to degrade the contaminants more efficiently than birds which are exposed only temporarily (e.g. Common Tern). This hypothesis was confirmed by Beyerbach et al. (1993) and by the results of this study with respect to the PCB-mixtures. The high levels of PCBs and the reduced levels of PCBs with low degree of chlorination ($PCB_{3Cl-5Cl}$) found in Oystercatcher eggs from Julianapolder allow us to assume recent inputs, since the high activity of enzymes catabolize such unstable compounds and lead to its elimination (Beyerbach et al. 1993, Denker et al. 1994).

5.2.2 Intersite variation

Bird studies can highlight significant differences in egg contamination not only between regions but also on smaller geographical scales. In the Wadden Sea, discriminant analyses clearly separated eggs from various breeding sites by the overall patterns of chemicals' concentrations (Fig. 5). In Herring Gull eggs from the Great Lakes, PCBs and HCB had the greatest power to separate the bird samples from the lakes' breeding sites (Weseloh et al. 1990). Eggs from breeding sites at the inner German Bight (Elbe estuary, Trischen island) were contaminated on higher levels than those from western and northern breeding sites of the Wadden Sea, indicating the discharge of the industrial chemicals (mercury, HCB, and PCBs) and of pesticides by the river Elbe into the North Sea. This characteristic spatial pattern was obvious during the two decades under study (Becker et al. 1985a,b, 1991, 1998 with detailed data on spatial trends 1996 and 1997) and points to pollutant sources of high significance for the Wadden Sea contamination, also in the late 1990s.

On sites west and north of the Elbe estuary, however, egg contamination was considerably lower, corresponding to the results of our earlier studies. In the Danish Wadden Sea, covered for the first time by the parameter "Contaminants in Bird Eggs", lowest egg residue levels were recorded (Figs. 3, 4). Because of the direction of North Sea currents, the pollutant loads originating from the Elbe are distributed in northeastern and northern directions (e.g. Lee 1980). Accordingly, bird contamination decreased from the estuary via Trischen to Norderoog, Margrethekoog and Langli in the Danish Wadden Sea, obviously in parallel with an increasing dilution of chemicals in water, sediment and food web (Fig. 3).

The breeding sites west of the Elbe estuary are influenced to a lower extent by the pollutant loads brought in by this river. Nevertheless, elevated concentrations of PCBs were measured also at Dollart (as in 1997, Becker et al. 1998) and Delfzijl. At these sites, Oystercatcher eggs were additionally characterized by very high HCB levels, indicating a local source of contamination. HCB is ingested by individuals feeding on benthic organisms from the Sea Harbor Channel in Delfzijl. Due to industrial activities carried out in the period 1969-1986, large amounts of HCB were drained into this channel. Consequently the measured concentrations exceed the maximum permissible risk levels of HCB accepted in the Netherlands (five times higher in sediments and ten times higher in biota, Nereis and Oystercatcher eggs; Eggens & Bakker 2001).

But also at some Dutch sites distant from estuaries, at Balgzand and Julianapolder, considerable contamination by PCBs in Oystercatcher eggs was ascertained. At the most western site Balgzand, Common Tern eggs had elevated levels of PCB and mercury as well (Fig. 3). These data suggest that the Dutch Wadden Sea ecosystem is still influenced by contaminants' loads from the river Rhine and IJsselmeer (De Jonge 1990, De Jonge & Essink 1991, Bester & Faller 1994), where the Balgzand birds may forage in addition to the Wadden Sea. High PCBs and Lindane levels in eggs of Common Terns (Muñoz Cifuentes & Becker 1998, Becker & Sommer 1998) or Oystercatchers breeding at the lower Rhine, as well as high levels in further bird species from this area (Stronkhorst et al. 1993, Bosveld et al. 1995, Dirksen et al. 1995, Exo et al. 1998), and other environmental samples (Bester & Faller 1994) identify the Rhine as an important source of these chemicals.

The causes of the considerable contamination of Oystercatcher eggs by various chemicals at Julianapolder in 2000 are not known, but some speculation is presented here. Inputs from inland sources of substances, such as chemicals used in agriculture (e.g. in the culture of seed potatoes), could reach the Wadden Sea via the Lauwersmeer (K. Essink pers. comm.). Besides some of the eggs collected in 2000 were laid later than usually (in early June), and this may explain the high contamination, since it is known that late eggs are higher contaminated than early clutches (Becker et al. 1992). Unlike the Oystercatcher eggs, Common Tern eggs from this site showed no elevated levels of contamination. This may indicate: (i) a contamination of Oystercatchers in this area mainly outside the breeding season (when Common Terns are on migration); or (ii) some foraging of the Oystercatchers in the Lauwersmeer itself, or inland in contaminated habitats. Other TMAP matrices investigated for chemicals in this central Dutch Wadden Sea area in 2000 should be consulted to elucidate this contamination problem. Furthermore, the verification of the higher Oystercatcher egg contamination of Julianapolder remains to be seen during the next years.

Thus, the results of spatial monitoring imply that by the careful selection of sampling sites, the hot spots of pollution and input sources of chemicals can be identified on the basis of the birds' samples. By reference to the degree of spatial differences, distinct local discharges of contaminants can be distinguished, as in mercury or HCB transported by the Elbe into the North Sea, or in the highest concentrations of HCB in Oystercatcher eggs measured at the Ems estuary (Delfzijl), indicating inputs from inland industrial areas. The less distinct spatial patterns of some chemicals such as the PCBs may be due to atmospheric deposition besides the riverine discharge.

Further hints to pollution sources and events can be derived from the detailed analyses of the chemicals' mixtures in the birds' samples. Within-mixture patterns of the isomers of HCH, the metabolites of DDT or the PCB-congeners are interesting in this respect. A high ratio of p,p'-DDT may indicate a more recent application of this insecticide, and the composition of the HCH-mixtures can point to the use of Lindane or to loads of the technical mixture and beta–HCH from industrial sources, as in the river Elbe (Becker et al. 1998). Significantly higher ΣDDT concentrations were measured in Common Tern eggs during the 1990s at the Elbe, and the higher proportion of p,p'-DDT suggests a recent input of this pesticide, legally no more in use in eastern Europe since the late 1980s (2.3). Oystercatcher eggs with relatively high concentrations of unmetabolised p,p'-DDT were collected at the Ems estuary (Delfzijl). Also in former years, higher DDT- residues in eggs from breeding sites at the Dollard gave rise to assume an illegal use of DDT in this area (Becker et al. 1991, 1998).

The composition of PCB-mixtures in birds depends on the degree of metabolism, which differs between species and sites (Beyerbach et al. 1993, Denker et al. 1994, Dietrich et al. 1997). In coastal birds of the Wadden Sea, the degree of metabolism and the percentage of highly chlorinated congeners within the mixture have been found to be higher in areas of stronger PCB-pollution, indicating an increased induction of metabolizing enzymes (Beyerbach et al. 1993, Becker et al. 1998; see also 6.2.1). Since PCBs with a low chlorination degree are biodegraded at higher rates than PCBs with more chlorine atoms (Beyerbach et al. 1993), the higher levels of PCB_{3Cl} to PCB_{5Cl} found in eggs from Julianapolder and Balgzand suggest the occurrence of more recent events of contamination.

When analyzing the intersite variation of the most toxic PCB congeners (non-, mono-, and di-ortho-PCBs), Common Tern eggs from the Elbe showed not only higher levels of ΣPCB than samples from the other sites, but also of its most toxic congeners. Consequently, at the Elbe river, the PCB-mixture may be the most hazardous to the bird populations among the Wadden Sea breeding sites. This may probably explain the negative effects of PCBs on hatchability in Common Terns breeding at the Elbe estuary found in the late 1980s (Becker et al. 1993b).

Levels of chlordanes in Common Tern eggs again point to the Elbe river as contamination source. Chlordanes' concentrations in Oystercatcher eggs identify inputs of these compounds at the Julianapolder and Ems estuary originating from possible inland sources. The high local concentration of nonachlors in Oystercatcher eggs in 2000 suggests that more recent inputs of chlordanes have occurred at these areas.

The monitoring results show steep contamination decreases at the beginning of the 1990s and further reductions during the last decade which is corroborated by the analyses of abiotic and other biotic samples in the Wadden Sea (Bakker et al. 1999; see 6.2.3). Yet despite the lowered inputs of chemicals into the North Sea through rivers and by atmosphere, the contamination of the seabird eggs clearly indicates distinct geographical trends today, with the hot spot Elbe estuary still persisting to date. During the 1980s, however, the spatial variation in bird egg contamination in the German Wadden Sea was more distinct, and intersite differences were higher, owing to the formerly huge contaminants' inputs by the river Elbe into the North Sea (see Becker et al. 1985a,b, 1991 for spatial patterns from the 1980s, and Bakker et al.1999). Nevertheless, the Wadden Sea adjacent to the inner German Bight was and will further be the hot spot where birds are most likely at risk of hazardous environmental chemicals, which should be kept under careful observation. Also a possible contamination source at the Julianapolder and Delfzijl, mainly reflected by Oystercatcher eggs, needs attention and further investigation in order to avoid or reduce anthropogenic inputs of chemicals into these areas of the Wadden Sea.

5.2.3 Temporal variation

In the past, seabirds have often revealed temporal changes in the chemical pollution of marine areas. In Britain, Northern Gannet (*Sula bassana*, Newton et al. 1990) and European Shag (*Phalacrocorax aristotelis*, Coulson et al. 1972) eggs, and in Canada, the eggs of several seabird species have revealed decreasing temporal trends (Chapdelaine et al. 1987, Elliott et al. 1994, Braune at al. 2001). Common Guillemot *Uria aalge* (Bignert et al. 1998) and Little Tern *Sterna albifrons* eggs (Thyen et al. 2000a) have shown the declines of organochlorine pollution in the Baltic Sea. The long-term studies of the contamination of seabirds' eggs in the Wadden Sea revealed decreases in the levels of most organochlorines from 1981-2000 (Figs. 8, 9), but the predominant changes occurred during the late 1980s/early 1990s (Becker et al. 1991, 1992, 1998).

Before we discuss some aspects of the observed trends and their causes some remarks are necessary on the data basis available. In contrast to the long-term studies on eggs from the German Wadden Sea sites, eggs from the Dutch coast were not sampled before 1997 (except Griend, Table 2) and from the Danish sites have only been sampled since 1999. Therefore, the preliminary time trend analyses for the Dutch sites Balgzand and Julianapolder have to be treated with caution, and lack of significance at the Danish sites. Furthermore, when analyzing the temporal variation of contaminants other factors affect contaminant levels in biota and in the birds, masking the year effect in question. Floods of the rivers or storms during springtime may have caused strong variation of egg contamination during the 1980s at the Elbe and inner German Bight (Figs. 8, 9). Floods cause resuspension and therefore remobilization of deposited organically bound contaminants (Becker et al. 1991, Haarich 1996, Bartholdy & Aagaard 2001). Other factors causing remobilization of contaminants may be deepening of river beds and harbor basins (Farke 1994). The resulting interyear fluctuations of contaminant levels are high at some sites (Figs. 6, 7), therefore long time periods (e.g. on a decadal scale) of annual sampling are required to identify temporal trends as is possible by the valuable data series of two decades now (Figs. 8, 9).

All bird studies mentioned above have clearly shown a continuous decrease of concentrations of most environmental chemicals in the industrial countries since the 1970s. Obviously, many measures aimed at protecting the environment have had positive effects of reduced pollution of the environment and the birds as well. For a long time the Elbe estuary was the hot spot of contamination of the North Sea, especially by mercury: Common Terns from the Elbe were considered to be among the most heavily contaminated birds in the world with mercury (Becker et al. 1993a,b; Furness et al. 1995). Also PCB levels were high and endangering the reproductive success of the Common Tern colonies at the Elbe estuary (Becker et al. 1993b). The main drop of the contamination with mercury and organochlorines at the Elbe, and consequently at the Wadden Sea, happened after the German reunification in 1989 (Figs. 8, 9, 11; Bakker et al. 1999). The reasons were the closure of many chemical industries without purification plants, subsequent effective measures of environmental protection in the eastern part of Germany, reductions and purification of industrial effluents, as well as the final ban of PCB and DDT production and use in eastern European states. So, in the Wadden Sea, the policy of protection applied by the governments concerned appears to have been successful since the burden of environmental chemicals could be strongly reduced.

On the other hand, the contamination of seabird eggs of the Wadden Sea indicate ongoing significant inputs of PCBs through the big rivers Rhine and Elbe, and of mercury, HCB and DDT and metabolites through the Elbe, even though production and use of these chemicals have been forbidden in central Europe, the catchment area of these rivers, since the 1980s. Concentrations of some chemicals are still unacceptably high, especially of the toxic PCBs (Table 5). In Sweden, for example, the rate of decrease was slower in PCBs than in other compounds studied, indicating an ongoing pollution from sources like refuse tips (Bignert et al. 1998). In Common Tern eggs from the Elbe no significant decrease in PCB contents was found during the last decade, and the concentrations seem to stagnate at levels above the target concentrations. In this context, however, the change in the proportion of toxic PCB compounds in the mixture between 1995 and 2000 (Table 5) is worth to be mentioned: the proportion of non-ortho and mono-ortho-PCBs decreased from 0.6% to 0.2%, and from 52% to 26%, respectively (Munoz Cifuentes & Becker 1998; concentrations of non-, mono- and di-ortho-PCBs measured in 1995: 4.61 ± 0.4, 378.8 ± 46.1, and 347.9 ± 100.7, respectively; cf. Table 5). A lower proportion of non-ortho-PCBs indicates reductions in recent PCB uptake by the birds (Fiedler & Lau 1998). In addition, also the proportions of the PCB congeners with lower degree of chlorination - PCB_{3Cl} to PCB_{5Cl} – showed clear reductions during the last decade (Table 7, cf. Tables 7 and 8 in Becker et al. 1998). These results hold true for the Elbe area, but also for the Wadden Sea sites in both species, and is a further hint to advancing metabolization of the PCB burden of the birds and of ceasing new contamination with PCBs.

The concentrations of HCB measured at the Elbe are still at a high level and have increased slightly during the last decade in the Common Tern, while trends of HCB egg concentrations in both species were declining at most sites. In Oystercatchers at the Elbe, the mercury egg levels have increased but on a very low level (Table 3 for the year 2000 value). Also on the Dutch coast of the Wadden Sea, some positive trends have been observed (Table 7). However these trends are based on a few years of monitoring and do not allow for safe conclusions.

To conclude, our findings in general reveal ongoing reductions of pollution of Wadden Sea biota including birds. This is a positive development for the Wadden Sea improving the environmental quality towards the ecotargets (5.4). However, at some sites, stagnating or even increasing levels of some chemical compounds were obvious, which demand further measurements of environmental protection, and the control of the success of those activities by the continuation of relevant TMAP parameters including top predators, such as birds, on the required long-term and annual basis of sampling.

Oystercatcher clutch
(Photo J.-D. Ludwigs)

5.3 Effects of the Recent Contamination on Coastal Birds

All analyzed chemical compounds are toxic and persistent, and biomagnify in lipid-rich tissues of top-predators as coastal birds (e.g. Moriarty 1990, Furness 1993, DesGranges et al. 1998). They can exert negative impacts on the birds' health, survival and reproduction, for example causing hormonal disruption, behavioral changes, thinning of egg shells, embryonic death and reproductive failure (see reviews of Ohlendorf et al. 1978, Moriarty et al. 1986, Moriarty 1990, Scheuhammer 1987, Furness 1993, Nisbet 1994, Walker 1994, Grasman et al. 1998). Critical levels of contaminants, however, are difficult to derive, especially in wild birds. They vary among chemical compounds, species and other environmental factors, but also owing to different methods applied, which has taken into account if results are assessed. Numerous studies including this monitoring project have investigated the variation of contaminant levels in coastal birds occurring at the Wadden Sea, but only a few addressed toxic effects of contaminants or threshold levels (e.g. Koeman et al. 1967, Becker et al. 1993b, Murk et al. 1994, Bosveld et al. 1995, Dirksen et al. 1995). As mentioned above, threshold levels taken from the literature must be considered carefully. Further difficulties originate from possible interactions and synergistic effects of single toxicants (e.g. Wachs 1994, Busch 1996).

The critical range for mercury in eggs varies widely among species and studies, from about 500 – 6,000 ng·g^{-1} (Ohlendorf et al. 1978, Ohlendorf 1993, Scheuhammer 1987). Becker et al. (1993) found no influences of high egg mercury levels (6,200 ng·g^{-1}) on hatchability of Common Terns. Consequently, since the highest mercury concentrations in Common Tern (960 ng·g^{-1}) and in Oystercatcher eggs (350 ng·g^{-1}) measured in 2000 were far below the critical range known, no adverse effects would be expected by the recent mercury contamination of coastal birds in the Wadden Sea.

In case of PCBs, too, critical levels for birds reported in the literature range widely, from 1,000 to 8,000 ng·g^{-1}. According to Lorenz & Neumeier (1982), concentrations of more than 3,000 to 5,000 ng·g^{-1} can influence the reproductive success of birds. This critical range has been confirmed by several authors: Blus & Prouty (1979) found no negative effects on the breeding success of Least Tern (*Sterna antillarum*) at concentrations of 1,200 – 3,700 ng·g^{-1}. Becker et al. (1993) found significant differences in the PCB concentrations between unhatched Common Tern eggs and eggs sampled at random (7,600 ng·g^{-1} vs. 5,100 ng·g^{-1}, respectively). But in Forster's terns (*Sterna forsteri*) breeding on Green Bay (U.S. Lake Michigan), concentrations of PCBs in eggs as high as 7,000 ng·g^{-1} and oral intake rates of 40 ng·g^{-1}·day^{-1} were found to cause no observable adverse effects on the reproductive success (Harris et al. 1993). Eggs of Caspian Tern (*Sterna caspia*) from the Great Lakes had even higher PCB concentrations of 18,500 – 39,300 ng·g^{-1}, but again no adverse reproductive effects were noted (Struger & Weseloh 1995). Considering negative effects of the PCBs' congeners of higher toxicity, Bosveld et al (1995) showed that high concentrations of non- and mono-ortho-PCBs in yolk sacs of hatchlings of Common Tern from the Rhine estuary (22 and 7,947 ng·g^{-1}, respectively) were linked with an increase of incubation time and an enhanced activity of cytochrome-P450 in the liver of chicks. The maximum values of non- and mono-ortho-PCBs measured in 2000 (Table 5) were very low compared to those egg levels, making it unlikely that the hatching success is impaired by the recent PCB contamination in the Wadden Sea.

It is widely known that high levels of DDT and metabolites are associated with significant egg shell thinning, embryotoxicity and reduced reproductive output in a variety of species including gulls, terns, raptors and cormorants (see reviews, e.g. Furness 1993, Moriarty et al. 1986). The threshold range of DDE egg levels associated with impairment of reproduction varies between 500 to more than 6,000 ng·g^{-1}. Switzer et al. (1973) and Fox (1976) supposed that 4,000 ng DDE ·g^{-1} egg cause a reduction of hatching success in terns, whereas Pearce et al. (1979) report a reduction of hatchability in Common Terns at concentrations of about 25,000 ng·g^{-1}. No association between DDE levels and hatching success was found in Caspian and Elegant Terns (*Sterna elegans,* Ohlendorf et al. 1985) at DDE egg levels of 9,300 and 3,800 ng·g^{-1}, respectively. In the Wadden Sea, DDE levels were much lower than these threshold levels during the 1990s (Table 3, Fig. 3; Tables A2 and A3 in Appendix; about 90% of the ΣDDT level is represented by DDE).

Chlordane is widely distributed in the environment, but usually found at ppb levels in biota (Eisler 1990). Several factors determine the transfer of these compounds from the environment to biota. Studies carried out with insectivorous birds showed that the poisoning by chlordane was initially low because the compounds were highly effective killing insects which could no more be consumed by the birds. Years later, however, insects developed resistance that made the food-chain transfer of lethal amounts of chlordane from soil to birds possible (Stansley et al. 2001). There is a lack of studies, however, reporting levels of chlordane in bird eggs associated with reproductive impairment. Concentrations of chlordane in unhatched Bald Eagle (*Haliaetus leucocephalus*) eggs from Lake Erie could be used as reference of possible toxic effects: 640 ng·g^{-1} and 160 ng·g^{-1} (for the years 1974-1980 and 1989-1994, respectively), and 40 ng·g^{-1} from Lake of the Woods, Canada (Donaldson et al. 1999). Other studies report on chlordane concentrations with no detected negative effects on reproduction: Osprey (*Pandion haliaetus*) eggs collected at the Fraser River (U.S.) contained concentrations of cis- and trans-chlordane and of cis- and trans-nonachlor over a range of 0.01–2.0 and 0.1–10 ng·g^{-1}, respectively (Elliot et al. 2000). In eggs of Black-crowned Night Heron (*Nycticorax nycticorax*) from Baltimore Harbor, a highly contaminated site in the U.S., higher trans-nonachlor levels of 66 ng·g^{-1} were found, although without measurable effects on reproduction (Rattner et al. 2001). The measured concentrations of chlordanes at the end of the 1990s in the Wadden Sea (Table 3, Fig. 4) were very low in comparison with these mentioned levels.

As the contaminant levels of mercury and organochlorines found in eggs of Wadden Sea birds during the late 1990s were by far lower than the critical levels reviewed, the recent contamination of coastal birds appeared not to impair reproduction, as it was the case in the 1980s. However, possible sublethal effects of the contaminants, interactions between single compounds and synergistic effects could be harmful. Such negative impacts cannot be excluded and should be taken into consideration, especially at the "hot spots" of contamination in the Wadden Sea, where the birds' body burden with contaminants is still high.

5.4 Assessment of the Bird Contamination Levels Regarding the Ecotargets

In the sense of obtaining extensive natural conditions as the basic element of the Trilateral Wadden Sea Cooperation, the absolute quantity of anthropogenic contaminants influencing the ecosystem should be as small as possible. That means that the concentrations of naturally occurring substances should be at natural levels, and the discharges of non-natural substances should be zero, as these man-made chemicals have only been produced during the past century (Bakker et al. 1997). Recently, ICES (2001) proposed Ecological Quality Objectives for seabirds in the North Sea, including mercury and organochlorines. For all OSPAR substances the ultimate goal is achieving concentrations in the marine environment near background values for naturally occurring substances and close to zero for man-made synthetic substances (OSPAR 1997).

As the long-term trends and recent results gained by the monitoring of contaminants in bird eggs from the Wadden Sea reveal, the anthropogenic discharges of micropollutants converge slowly towards the ecotargets, although riverborne inputs are still considerable and short-term pollution still occurs (5.2.2, 5.2.3; see also Bakker et al. 1999). There is still a latent risk of renewed contamination of the ecosystem's biota triggered by events discussed in 5.2.2 and 5.2.3. Presumably the target levels for man-made chemicals of zero values in biota, including bird eggs, could not be achieved in the near future as these persistent chemicals have long half-lives. Despite the immense strides in preventing chemical pollution of the Wadden Sea during the last two decades by legal regulations, more effort is needed to reduce the chemicals' inputs into the environment.

As with most organochlorines, decreasing mercury egg contamination could be proved during the last two decades. However, natural mercury concentrations can be presumed not to have been achieved until now, although suitable comparative reference levels for eggs are not available. In all Wadden Sea areas, during the mid 1990s, mercury levels in sediment have been 3 - 10 times higher and mercury levels in mussels about 2 – 4 times higher than the agreed background range (Bakker et al. 1999). Herring Gull feather mercury levels from the German North Sea, indeed decreasing since the 1960s, have not yet arrived at reference levels (ICES 2001) of the last century (Thompson et al. 1993, Furness et al 1995, Mattig et al. 1996).

However, as a first step to develop assessment criteria of an ecotarget for mercury in bird eggs within the TMAP and OSPAR (Bakker et al.1997), we are cautiously approaching a reference level using the recent egg values in the low polluted areas. The lowest mercury levels in Oystercatcher eggs from the Wadden Sea were found to be about 100 $ng \cdot g^{-1}$ fresh egg mass during the last decade (Table 3, Fig. 3). At the lower polluted sites, Common Tern mercury egg levels were found to be comparable with those of the Oystercatcher (see Fig.2; minimum levels in 2000 about 300 $ng \cdot g^{-1}$, Table 3; see also Mattig et al. 2000). Mercury egg levels of both species were the highest values among a group of eight coastal bird species from the Wadden Sea island Spiekeroog (Mattig et al. 2000). Consequently the reference and target levels for mercury should roughly be around 100 $ng \cdot g^{-1}$ or below. Mercury levels detected in coastal bird eggs in 2000 were up to 10-fold this level (Table 3).

The decline in chemical contamination of the Wadden Sea during the last decade means an improvement of the environmental conditions for biota in this ecosystem, and may be a contribution to stable or increasing breeding populations of typical Wadden Sea bird species during the 1990s with only a few exceptions (Melter et al. 1997, Rasmussen et al. 2000, Hälterlein et al. 2000). The Oystercatcher population increased, but the Common Tern population showed slight decreases during the mid 1990s (for discussion of possible causes other than chemicals see Becker & Sudmann 1998, ICES 2000). The other tern species had stable or increasing population sizes, the gull populations increased, too, with the exception of Herring Gulls. Also Cormorant *Phalacrocorax carbo* and Shelduck *Tadorna tadorna* increased in numbers. Whether reproduction is impaired by the recent chemical contamination could be revealed by the proposed parameter "Breeding Success", which is however not yet implemented in the TMAP (see 5.5).

5.5 The Significance and Future of the Parameter "Contaminants in Bird Eggs" within the TMAP and Recommendations

The results presented and discussed in this report show that monitoring contaminants in coastal bird eggs is qualified as a valuable and significant contribution with respect to inputs of contaminants and its corresponding effects on natural processes and species in the Wadden Sea. So far, the objectives of monitoring this parameter have been achieved (TMAP 1997, see chapter 1.), and the endorsement of the Trilateral Governmental Conference in Stade in October 1997 to install the parameter has been justified by the experiences during the first years of monitoring (Enemark 1997, Marencic & Lüerßen 1997).

The successful implementation of the parameter in 1998 was possible on the basis of the harmonized methods proven during the pilot studies and based on the successful projects from the 1980s (Table 1). The methods applied follow the guidelines (OSPAR 1997, TMAP 1997, Becker et al. 1998) and have been found to be practicable and adequate to achieve the monitoring aims (see chapter 1) by a reasonable effort.

Most of the requirements and recommendations indicated in the pilot study (Becker et al. 1998) have been fulfilled by the later implementation of the parameter:

- Through a selection of additional sampling sites in the western and northern part of the Wadden Sea (Fig. 1) a good coverage of the islands, coast and estuaries was achieved. This enabled an adequate spatial scale in order to discover spatial trends and local pollution sources, and to recognize the influence of big rivers and exchange of water with adjacent Wadden Sea areas.
- The use of one established laboratory with the relevant expertise was the right step making expensive, permanent and time consuming intercalibrations unnecessary.
- Also the project organization, data evaluation and assessment by an institution experienced in ornithological research, the Institute of Avian Research "Vogelwarte Helgoland" in Wilhelmshaven, has been advantageous for monitoring the parameter. In each year since 1998, respectively, until the end of the year of sampling raw data could be provided to the TMAP data units, and the results supplemented by a preliminary evaluation have been presented.
- We adapted the spectrum of the organochlorines analyzed (3.3) and included the insecticides chlordanes and nonachlors into our analysis.

Because of the parameters' significance for the assessment of the state of the Wadden Sea, and of the value of one of the longest time series to show temporal trends of pollution in the Wadden Sea (cf. Bakker et al. 1999), the TMAG is well advised to continue to apply this significant instrument further. Both Common Tern and Oystercatcher as typical Wadden Sea bird species should be used to indicate contamination of different trophic levels. As mentioned above (5.2.3), monitoring contaminants should be performed under a long-term perspective in order to be able to recognize significant changes over the years, also with respect to the ecotargets. Nevertheless, there are some requirements open in order to increase the value of the parameter "Contaminants in Bird Eggs", and we repeat the following recommendations made already before (Becker et al. 1998).

The TMAG should focus attention on some new environmental chemicals with persistence and toxicological relevance, such as organotin compounds and their degradation products, which are known to accumulate in birds (Stäb et al. 1996, Guruge et al. 1997); flame retardants, such as polybrominated biphenyls (de Boer et al. 2000); the antiparasitic agent bromocyclen used in veterinary medicine; or musk xylol utilized in the production of cosmetics or detergents. Both last mentioned compounds have been found in eggs of birds using the Wadden Sea (Mattig et al. 2000). For a preliminary screening of concentrations in Wadden Sea bird eggs, the analytics have to be developed or adapted for such substances by pilot studies, before a decision on their inclusion into the TMAP can be made.

As revealed by the results of this report, the Rhine still appears to play an important role as a source of environmental chemical, especially of PCBs, transported by the currents into the Wadden Sea. Therefore, a sampling site near the Rhine delta itself would help to investigate the chemicals' input by this river into the North Sea in a more direct way than by the site Balgzand in the westernmost part of the Wadden Sea. We suggest, e.g., the localities Slijkplaat and/or Westplaat as possible sampling sites, which are situated in the sedimentation area of the Rhine and Meuse Rivers (see Bosveld et al. 1995)

Besides the value of birds as accumulative indicators of chemicals' levels, the capacity of avian top predators to act as sensitive indicators should be utilized to monitor for possible biological effects of chemicals in the Wadden Sea. This approach uses birds (i) as an early warning of new chemicals not covered by the regular monitoring program (ii) to spot dangerous interactions of chemicals, (iii) to derive critical levels for toxic chemicals. In this way in Sweden, for example, the reproductive success of White-tailed Sea Eagles (Helander et al. 1998, Olsson et al. 1998, Helander 1999) is a parameter of the national marine monitoring program. In Canada, monitoring contamination of aquatic birds at the Great Lakes is supplemented by studies on reproductive performance, congenital anomalies, mutagenicity and on biochemical and physiological effects (Mineau et al. 1984, Ryckman et al. 1998, Grasman et al. 1998, CWS 2001) in order to receive an early warning of possible toxic effects of environmental chemicals. This approach was also recommended (Becker 1991, 1992, Exo et al. 1996), successfully tested and adopted by the TMAP in the Wadden Sea (Thyen et al. 1998, 2000b) and includes the species Common Tern and Oystercatcher, but has not yet been applied.

Programs monitoring chemicals should be combined with studies of bird demography, in particular of possible effects on reproduction, in order to use birds acting as an early warning system. Such studies should especially cover the hot spots of environmental pollution relevant also in the future: rivers, estuaries, industrial areas, where pollution sources and environmental chemicals become concentrated, and where the first signs of negative effects on nature are apt to become manifest – such as the Elbe estuary. But sites with low pollution levels also have to be covered, in order to compare natality between sites of different degrees of contamination and to be able to derive critical levels and species' sensitivity towards chemicals.

In consequence, we recommend strongly to install the parameter "Breeding Success" as a necessary supplement of the TMAP, a requirement also observed by Rasmussen et al. (2000), who remind its installation in the package of the parameters monitoring birds, "Numbers and Breeding Bird Distribution", "Monitoring of Migratory Birds", "Beached Bird Survey" and "Contaminants in Bird Eggs". The parameter "Breeding Success" should be measured preferably at the hot spots of pollution and at some other logistically favorable sites, in coincidence with "Contaminants in Bird Eggs" and using the same indicator species to be linked with this parameter. Within a selected colony, egg contamination can be linked with reproductive success (hatching success, fledging success) on a colony-basis or even on a clutch-basis: chemical data of one sampled egg can be related to the reproductive outcome of the other eggs in the same clutch (e.g. Becker et al. 1993). In this way, coastal birds may act as a real early warning system against deleterious effects of pollution on the ecosystem Wadden Sea as well as on man using its resources.

6. Conclusions and Recommendations

It is concluded that:

- in general, the levels of chemicals in bird eggs have decreased strongly during the last two decades;
- the policy of protection applied by the governments concerned appears to have been successful in the Wadden Sea, since the burden of environmental contamination was reduced. Many measures aimed to protect the environment have had positive effects of decreased contamination of the ecosystem and the birds as well;
- some increases of organochlorine levels in eggs occurred, however, and that geographical pattern of bird egg contamination points to pollutant sources of high significance also during the 1990s;
- the Elbe estuary and the inner German Bight still were the "hot spot" of chemical contamination;
- other local sources of contamination were evident at Delfzijl and Julianapolder;
- the chemical inputs by the river Rhine still affect the Wadden Sea ecosystem;
- the positive development with respect to contamination improved the environmental quality in the Wadden Sea towards the ecotargets;
- negative effects on bird reproduction and populations by the recent levels of chemicals seem unlikely so far critical levels are known;
- the parameter Contaminants in Bird Eggs using birds as accumulative indicators has great advantages in monitoring the contamination state of the Wadden Sea;
- one organization doing the sampling and reporting of the data, and one laboratory doing the chemical analyses as in this parameter reveal logistical advantages within the TMAP.

It is recommended that:

- point sources of contamination with environmental chemicals (e.g. Julianapolder, Delfzijl) need attention in order to elucidate their causes and to avoid or reduce anthropogenic inputs of contaminants;
- the Elbe estuary and the Wadden Sea adjacent to the inner German Bight should be kept under careful observation, being further the hot spot where birds and ecosystem are most likely at risk of hazardous environmental chemicals;
- considering the existence of still high local contamination, the policies for the reduction of the application of xenobiotic hazardous substances in the framework of OSPAR, the North Sea Conferences and the EU should be intensified;
- measures to minimize further inputs of substances into the ecosphere by the rivers Elbe, Weser, Ems and Rhine, and by the atmosphere, should be reinforced;
- relevant parameters of the TMAP, including birds as top predators and accumulative indicators, have to be measured in a long-term perspective, in order to distinguish short-term fluctuations from time-trends;
- spatial and temporal trend monitoring in different TMAP parameters should be interrelated to discriminate whether an increase in contamination is caused by new inputs or by remobilization of chemicals released a long time ago and being a latent risk;
- xenobiotics which have been forbidden many years ago in Europe as some pesticides and PCBs, but still causing recent local chemical contamination, should be continued to be monitored;
- the spectrum of chemicals studied should be adapted, and some "new" toxic substances should be proved if it makes sense to include it into the monitoring by bird eggs (e.g. TBT, polybrominated biphenyls, bromocyclen or musk xylol);
- additional sampling sites at the Rhine delta should be included to investigate bird contamination at this area, in order to determine the impact of contaminants originating from this river and transported to the Wadden Sea;
- the proposed TMAP-parameter "Breeding Success" should be implemented to also utilize birds as sensitive indicators of chemical contamination and as an early warning system, in addition to the other parameters within the TMAP making use of birds to monitor the ecological state of the Wadden Sea.

7. References

Agency for Toxic Substances and Disease Registry United States Public Health Service (1994): Toxicological Profile for Chlordane. In www.eco-usa.net/toxics/chlordane.html.

Bakker, J.F., W. Bartelds, P.H. Becker, K. Bester, D. Dijkshuizen, B. Fredericks & B. Reineking (1999): 4. Marine Chemistry. In: F. de Jong, J.F. Bakker, C.J.M. van Berkel, N. M. J.A. Dankers, K. Dahl, C. Gätje, H. Marencic & P. Potel (eds.): 1999 Wadden Sea Quality Status Report. Wadden Sea Ecosystem No. 9, Common Wadden Sea Secretariat, Trilateral Monitoring Group, Quality Status Group. Wilhelmshaven, Germany: 85-117.

Bakker, J., N. Dankers, F. de Jong, C. Gätje, T. Pedersen, P. Potel & K. v. Berkel (1997): Assessment of the Wadden Sea ecosystem No.7. CWSS, QSR Group & TMAG, Wilhelmshaven.

Bartholdy, J. & T. Aagaard (2001): Storm surge effects on a back-barrier tidal flat of the Danish Wadden Sea. Geo-Marine Letters. 20: 133-141.

Bäckstrom, J. (1969): Distribution studies of mercury pesticides in quail and some fresh-water fishes. Acta Pharmacol. Toxicol. 27: 1-103.

Becker, P.H. (1989): Seabirds as monitor organisms of contaminants along the German North Sea coast. Helgoländer Meeresunter. 43: 395-403.

Becker, P.H. (1991): Population and contamination studies in coastal birds: the Common Tern *Sterna hirundo*. In: C.M. Perrins, J.D. Lebreton & G.J.M. Hirons (eds.): Bird population studies: relevance to conservation and management: 433-460.

Becker, P.H. (1992): Seevogelmonitoring: Brutbestände, Reproduktion, Schadstoffe. Vogelwelt 113: 262-272.

Becker, P.H., A. Büthe & W. Heidmann (1985a): Schadstoffe in Gelegen von Brutvögeln der deutschen Nordseeküste. I. Chlororganische Verbindungen. J. Ornithol. 126: 29-51.

Becker, P.H., D. Frank & U. Walter (1987): Geographische und jährliche Variation der Ernährung der Flußseeschwalbe (*Sterna hirundo*) an der Nordseeküste. J. Ornithol. 128: 457-475.

Becker, P.H., R.W. Furness & D. Henning (1993a): The value of chicks feathers to assess spatial and interspecific variation in the mercury contamination of seabirds. Environ. Monitoring Assessment 28: 255-262.

Becker, P.H., W.A. Heidmann, A. Büthe, D. Frank & C. Koepff (1992): Umweltchemikalien in Eiern von Brutvögeln der deutschen Nordseeküste: Trends 1981-1990. J. Ornithol. 133: 109-124.

Becker, P.H., C. Koepff, W.A. Heidmann & A. Büthe (1991): Schadstoffmonitoring mit Seevögeln. Forschungsbericht UBA-FB 91-081, TEXTE 2/92, Umweltbundesamt, Berlin.

Becker, P.H., S. Schuhmann & C. Koepff (1993b): Hatching Failure in Common Terns (*Sterna hirundo*) in relation to environmental chemicals. Environ. Pollut. 79:209-213.

Becker, P.H. & U. Sommer (1998): Die derzeitige Belastung der Flußseeschwalbe *Sterna Hirundo* mit Umweltchemikalien in Mitteleuropa. Vogelwelt 119:243-249.

Becker, P. H. & S. R. Sudmann (1998): Quo vadis *Sterna hirundo*? Schlußfolgerungen für den Schutz der Flußseeschwalbe in Deutschland. Vogelwelt 119: 293-304.

Becker, P.H., W. Ternes & H.A. Rüssel (1985b): Schadstoffen in Gelegen von Brutvögeln der deutschen Nordseeküste. II. Quecksilber. J. Ornithol. 126: 253-262.

Becker, P.H., S. Thyen, S. Mickstein, U. Sommer & K.R. Schmieder (1998): Monitoring pollutants in coastal bird eggs in the Wadden Sea. In: Wadden Sea Ecosystem No. 8, final report of the pilot study 1996-1997: 59-98.

Bester, K. & J.T. Faller (1994): Persistente synthetische organische Schadstoffe im Wattenmeer. In: J.L. Lozán, E. Rachor, K. Reise, H. v. Westernhagen & W. Lenz (eds.): Warnsignale aus dem Wattenmeer: 37-42. Blackwell Wissenschaftsverlag , Berlin.

Beyerbach, M., P.H. Becker, A. Büthe, E. Denker, W.A. Heidmann & G. Staats de Yanés (1993): Variation von PCB-Gemischen in Eiern und Vögeln des Wattenmeeres. J. Ornithol. 134: 325-334.

Bezzel, E. (1985): Kompendium der Vögel Mitteleuropas. Nonpasseriformes. Aula, Wiesbaden.

Bignert, A. (2001): Comments concerning the National Swedish Contaminant Monitoring Programme in Marine Biota. Report Contaminant Research Group Swedish Museum Natural History: 1-114.

Bignert , A., K. Litzen, T. Odsjö, M. Olsson, W. Persson & L. Reutergardh (1995): Time related factors influence the concentrations of, PCBs and shell parameters in eggs of Baltic Guillemot (*Uria aalgae*), 1961-1989. Environm. Pollut. 89: 27-36.

Bignert , A., M. Olsson, W. Persson, S. Jensen, S. Zakrisson, K. Litzen, U. Eriksson, L. Häggberg & T. Alsberg (1998): Temporal trends of organochlorines in Northern Europe, 1967-1995. Relation to global fractionation, leakage from sediments and international measures. Environm. Pollut. 99: 177-198.

Bishop, C.A., P. Ng, R.J. Norstrom, R.J. Brooks & K.E. Pettit (1996): Temporal and geographic variation of organochlorine residues in eggs of the Common Snapping Turtle (*Chelydra serpentina serpentina*) (1981-1991) and comparisons to trends in the Herring Gull (*Larus argentatus*) in the Greats Lakes in Ontario, Canada. Arch. Environ. Contam. Toxicol. 31:512-524.

Blus, L. J. & R. M. Prouty (1979): Organochlorine pollutants and population status of Least Terns in South Carolina. Wilson Bull. 91: 62-71.

BMU (1988): Verordnung über Höchstmengen an Schadstoffen in Lebensmitteln (Schadstoff- Höchstmengenverordnung – SMmV) Bundesgesetzblatt 1988: 422-424.

Boer, J. de, K. de Boer & J.P. Boon (2000): Polybrominated Biphenyls and Diphenylethers. In: The Handbook of Environmental Chemistry. Vol. 3 Sreies: Anthropogenic Compound, Chapter 4: 61-95.

Bosveld, A.T.C., J. Gradener, A.J. Murk, A. Brower, M. van Kampen, E.H.G. Evers & M. van den Berg (1995): Effects of PCDDs, PCDFs and PBCs in Common Tern (*Sterna hirundo*) breeding in estuarine and coastal colonies in the Netherlands and Belgium. Environm. Toxicol. Chem. 14: 99-116.

Busch, D. (1996): Die Belastung der Biozönosen durch Schadstoffe. In: J.L. Lozán & H. Kausch (eds.): Warnsignale aus Flüssen und Ästuaren: 259-265.

Büthe, A. & E. Denker (1995): Qualitative and quantitative determination of PCB congeners by using a HAT-5 GC column and an efficient quadrupole MS. Chemosphere 30: 753-771.

Braune, B.M., G.M. Donaldson & K.A. Hobson (2001): Contaminant residues in seabird eggs from the Canadian Artic. Part I. Temporal trends 1975-1998. Environmental Pollution 114: 39-54.

Chapdelaine, G., P. Laporte & D.N. Nettleship (1987): Population, productivity, and DDT contamination trends of Northern Gannets (*Sula bassanus*) at Bonaventure Island, Quebec, 1997-1984. Can. J. Zool. 65: 2992-2996.

Coulson, J. C., I. R. Deans, G. R. Potts, J. Robinson & A. N. Crabtree (1972): Changes in organochlorine contamination of the marine environment of eastern Britain monitored by Shag eggs. Nature 236: 454-456.

Cramp, S. & K. E. L. Simmons (eds.) (1985): Handbook of the birds of Europe, the Middle East and North Africa. Oxford University Press, Oxford.

CWS (2001): Wildlife Toxicology Division Canadian Wildlife Service, Internet Homepage: www.cws-scf.ec.gc.ca/nwrc/wildtox.ntm.

De Jonge, V.N. (1990): Response of the Dutch Wadden Sea ecosystem to phosphorus discharges from the river Rhine. Hydrobiologia 195: 49-62.

De Jonge, V.N. & K. Essink (1991): Long-term changes in nutrients loads and primary and secondary production in the Dutch Wadden Sea. In: M. Elliott & J.P. Ducrotoy (eds.): Estuaries and coasts: spatial and temporal intercomparisons: 307-316.

Denker, E., P.H. Becker, M. Beyerbach, A. Büthe, W.A. Heidmann & G. Staats de Yanés (1994): Concentration and metabolism of PCBs in eggs of waterbirds on the German North Sea Coast. Bull. Environ. Contam. Toxicol. 52: 220-225.

DesGranges, J.-L., J. Rodrigue, B. Tardif & M. Laperle (1998): Mercury accumulation and biomagnification in Ospreys (*Pandion haliaetus*) in the James Bay and Hudson Bay regions of Québec. Arch. Environ. Contam. Toxicol. 35: 330-341.

Dietrich, S., A. Büthe, E. Denker & H. Hötker (1997): Organochlorines in eggs and food organisms of Avocets (*Recurvirostra avosetta*). Bull. Environ. Contam. Toxicol. 58: 219-226.

Dirksen, S.T.J., Boudewijn, L.K. Slager, R.G. Mes, M.J.M. Schaick & P. de Voogt (1995): Reduced breeding success of Cormorants (*Phalacrocorax carbo sinensis*) in relation to persistent organochlorine pollution of aquatic habitats in the Netherlands. Environ. Pollut. 88: 119-132.

Donaldson, G.M., J.L. Shutt & P. Hunter (1999): Organochlorine Contamination in Bald Eagle Eggs and Nestlings from the Canadian Great Lakes. Arch. Environ. Contam. 36: 70-80.

Eggens, M.L. & J.F. Bakker (2001): Toxicity of dredged material polluted with Hexachlorobenzene (HCB) – Is there a risk for organisms living in the Ems-Dollard estuary? Wadden Sea Newsletter 2001- 2: 13 – 15.

Enemark, J.A. (1997): The 8th Trilateral Governmental Conference on the Protection of the Wadden Sea in Stade, Germany, October 22, 1997. WSNL 97-2: 32-36.

Eisler, R. (1990): Chlordane hazards to fish, wildlife and invertebrates: a synoptic review. Biological Report 85 (1.21), US Fish and Wildlife Service, Washington, DC.

Elliot, J.E., S.W. Kenngy, D.B. Peakall & H. Won (1990): Polychlorinated biphenyl (PCB) effects on hepatic mixed function oxidases and porphyria in birds. I. Japanese quail. Comp. Biochem. Physiol. 96: 205-210.

Elliot, J.E., D.G. Noble, R.J. Norstrom, P.E. Whitehead, M. Simon & D.B. Peakall (1994): Patterns and trends of organic contaminants in Canadian seabirds eggs, 1968-90. In: C.H. Walker & D.R. Livingstone (eds.): Persistent pollutants in marine ecosystems. Pergamon Press, Oxford, New York, Seoul: 181-194.

Elliot, J.E., M.M. Machmer, L.K. Wilson & C.J. Henny (2000): Contaminants in Ospreys from the Pacific Northwest: II. Organochlorine pesticides, Polychlorinated Biphenyls, and mercury. Arch. Environ. Contam. Toxicol. 38:93-106.

Exo, K.M. (1994): Bedeutung des Wattenmeeres für Vögel. In: Lozán, J.L., E. Rachor, K. Reise, H.v. Westernhagen & W. Lenz (eds.): Warnsignale aus dem Wattenmeer: 261-270.

Exo, M., P.H. Becker, B. Hälterlein, H. Scheufler, H. Hötker, A. Stiefel, M. Stock, P. Südbeck & P. Thorup (1996): Bruterfolgsmonitoring bei Küstenvögeln. Vogelwelt 117: 287-293.

Exo, M., P.H. Becker & U. Sommer (1998): Umweltchemikalien in Eiern von Binnenland- und Wattenmeerbrütern des Austernfischers (*Haematopus ostralegus*). J. Ornithol. 139: 401-405.

Farke, H. (1994): Eingriffe durch Baggerarbeiten. In: Lozán, J.L., Rachor, E., Reise, K., Von Westernhagen, H. & W. Lenz (eds.): Warnsignale aus dem Wattenmeer: 60-64.

Fiedler, H. & C. Lau (1998): Environmental fate of chlorinated organics. In: G. Schürmann & B. Markert (eds.): Ecotoxicology: Ecological Fundamentals, Chemical Exposure, and Biological effects: 317-370.

Fleet, D.M., J. Frikke, P. Südbeck & R.L. Vogel (1994): Breeding Birds in the Wadden Sea 1991. Wadden Sea Ecosystem No. 1. Common Wadden Sea Secretariat, Wilhelmshaven. 108 pp.

Fox, G.A. (1976): Eggshell quality: its ecological and physiological significance in a DDE-contaminated Common Tern population. Wilson Bull. 88: 459-477.

Furness, R.W. (1993): Birds as a monitors of pollutants. In: R.W. Furness, & J.J.D. Greenwood (eds.): Birds as monitors of environmental change. Chapman & Hall, London: 86-143.

Furness, R.W., D.R. Thompson & P.H. Becker (1995): Spatial and temporal variation in mercury contamination of seabirds in the North Sea. Helgoländer Meeresunters. 17: 108-125.

Gilbertson, M., J.E. Elliot & D.B. Peakall (1987): Seabirds as indicators of marine pollution. In: A.W. Diamond & F.L. Fileon (eds.): The value of birds. ICBP Technical Public. Vol. 6: 231-248.

Gochfeld, M. (1997): Spatial patterns in a bioindicator: heavy metal and selenium concentration in eggs of Herring Gulls (*Larus argentatus*) in the New York Bight. Arch. Environ. Contam. Toxicol. 33: 63-70.

Goss-Custard, J.D. (1996): The Oystercatcher. Oxford, New York, Tokyo.

Grassman, K.A., P.F. Seanlon & G.A. Fox (1998): Reproductive and physiological effects of environmental contaminants in fish-eating birds of the Great Lakes: a review of historical trends. Environ. Monit. Assessment 53: 117-145.

Guruge, K.S., H. Iwata, H. Tanaka & S. Tanabe (1997) Butylin accumulation in the liver and kidney of seabirds. Mar. Environm. Res. 44: 191-199.

Haarich, M. (1994): Schwermetalle in Wasser und Sediment. In: Lozán, J.L., Rachor, E., Reise, K., Von Westernhagen, H. & W. Lenz (eds.): Warnsignale aus dem Wattenmeer: 30-34.

Haarich, M. (1996): Schadstoff-Frachten durch die Flüsse. In: Lozán & H. Kausch (eds.): Warnsignale aus Flüssen und Ästuaren: 144-148.

Hälterlein, B., P. Südbeck, W. Knief & U. Köppen (2000): Population trends of coastal breeding birds of the German North and Baltic sea coast. Vogelwelt 121: 241-267.

Harris, J.H., T.C. Erdman, G.T. Ankley & K.B. Lodge (1993): Measures of reproductive success and polychlorinated biphenyl residues in eggs and chicks of Forster's Terns on Green Bay, Lake Michigan, Wisconsin –1988. Arch. Environ. Contam. Toxicol. 25: 304-314.

Heidmann, W.A. (1986): Isomer specific determination of polychlorinated biphenyls in animal tissues by gas chromatography mass spectrometry. Chromatographia, 22: 363-369.

Helander, B., A. Olsson, A. Bignert, K. Litzén, L. Asplund & Å. Bergman (1998): Abnormal dehydration in eggs of the white-tailed sea eagle (*Haliaeetus albicilla*) in Sweden – a persisting effect from high exposure to organochlorines? Organohalogen Compounds 39: 423-426.

Holoubek I., J. Čáslavský, J. Helešic, R. Vančura, J. Kohoutek, A. Kočan, J. Petrik & J. Chovancová (1994): Project TOCOEN - The fate of selected organic pollutants in the environment. Part XXI. The contents of PAHs, PCBs, PCDDs/Fs in sediments from Danube river catchment area. Toxicol. Environ. Chem. 43: 203-215.

Hothem, R.L. & A.N. Powell (2000): Contaminants in eggs of Western Snowy Plovers and California Least Terns: Is there a link to population decline? Bull. Environ. Contam. Toxicol. 65:42-50.

Hulscher, J.B. (1985): Growth and abrasion of the Oystercatcher bill in relation to dietary switches. Neth. J. Zool. 35: 124-154.

ICES (1999): Report of the Working Group on Seabird Ecology. ICES CM 1999/C:5: 1-58. Copenhagen.

ICES (2001): Report of the Working Group on Seabird Ecology. ICES CM2001/C:5, Copenhagen.

Koch, R. (1991): Umweltchemikalien. VCH Verlagsgessellschaft, Germany. 426 pp.

Kočan A., J. Petrik, B. Drobná & J. Chovancová (1994): Levels of PCBs and some organochlorine pesticides in the human population of selected areas of the Slovak Republic. I. Blood. Chemopshere 29: 2315-2325.

Kočan A., M. Ursinyová, E. Reichertová, V. Hladíková, J. Petrik, J. Chovancová, H. Uhrinová, L. Randová, B. Drobná & L. Rosíval (1996): Occurence of selected toxic and carcinogenic organic and inorganic compounds in ambient air in selected locations of the Slovak Republic. Report. Institute of Preventive and Clinical Medicine, Bratislava, Slovakia, PHARE EC/93/AIR/22 project. June 1996, 61 pp.

Koeman, J.H., A.A.G. Oskamp, J. Veen, E. Brouwer, J. Rooth & P. Zwart (1967): Insecticides as a factor in the mortality of the Sandwich Tern (*Sterna sandvicensis*). Medelingen Rijksfaculteit Landbourwetenschappen Gent 27: 841-854.

Kruse, R. (1979): Ein verlustfreier offener Aufschluß mit $HNO_3/HClO_3/HClO_4$ für die Bestimmung von Gesamt-Quecksilber in Fischen. Z. Lebensm. Unters. Forsch. 169: 259-262.

Lewis, S.A., P.H. Becker & R.W. Furness (1993): Mercury levels in eggs, tissues and feathers of Herring gulls *Larus argentatus* from the German Wadden Sea coast. Env. Pollut. 80: 293-299.

Lewis, S.A. & R.W. Furness (1991): Mercury accumulation and excretion in laboratory reared Black-headed Gull *Larus ridibundus* chicks. Arch. Environ. Contam. Toxicol. 21: 316-320.

Lee, A.J. (1980): North Sea: Physical oceanography. From the north west European shelf seas. The sea bed and the sea in motion. II. Elsevier Oceanogr. Ser. 24B.

Lorenz, H. & G. Neumeier (1972): Polychlorierte Biphenyle - ein gem. Bericht des BGA und UBA. MMV-Verlag, München.

Marencic, H. (1997): The Trilateral Monitoring and Assessment Program (TMAP) of the Wadden Sea. Nat. Landsch. 72: 507-512.

Marencic, H. & G. Lüerßen (1997): Common package of TMAP parameters adopted at the Stade Conference. Wadden Sea Newsletter 1997-2: 43-45.

Mattig, F.R., P.H. Becker, H. Bietz & K. Gießing (1996): Schadstoffanreicherung im Nahrungsnetz des Wattenmeeres. Forschungsbericht 10802085/21. Umweltbundesamt, Berlin.

Mattig, F.R., H.U. Rösner, K. Gießing & P.H. Becker (2000): Umweltchemikalien in Eiern des Alpenstrandläufers (*Calidris alpina*) aus Nordnorwegen im Vergleich zu Eiern von Brutvogelarten des Wattenmeeres. J. Ornithol. 141:361-369.

Melter, J., P. Südbeck, D.M. Fleet, L.M. Rasmussen & R.L. Vogel (1997): Changes in breeding bird numbers on census areas in the Wadden Sea 1990 until 1994. Wadden Sea Ecosystem No. 4. Common Wadden Sea Secretariat, Wilhelmshaven: 7-93.

Meltofte, H., J. Blew, J. Frikke, H.U. Rösner & C.J. Smit (1994): Numbers and distribution of waterbirds in the Wadden Sea. IWRB Publication 34, Wader Study Group Bull. 74 special issue, 192 pp.

Mineau, P., G.A. Fox, R.J. Norstrom, D.V. Weseloh, D.J. Hallen & J.A. Ellenton (1984): Using the Herring Gull to monitor levels and effects of organochlorine contamination in the Canadian Great Lakes. In: J.O. Nriagu & M.S. Simmons (eds.): Toxic contaminants in the Great Lakes. J. Willey and Sons, Inc.: 426-452.

Moriarty, F. (1990): Ecotoxicology – the study of pollutants in ecosystems. Academic Press, London.

Moriarty, F., A.A. Bell & H. Hanson (1986): Does p,p'-DDE thin eggshells? Environ. Pollut. 40: 257-286.

Muñoz Cifuentes, J. & P.H. Becker (1998): Eier der Flußseeschwalbe (*Sterna hirundo*) als Indikator für die aktuelle Belastung von Rhein, Weser und Elbe mit Umweltchemikalien. UWSF-Z. Umweltchem. Ökotox. 10: 15-21.

Murk, A.J., A.T.C. Bosveld, M. van den Berg & A. Brouwer (1994): Effects of polyhalogenated aromatic hydrocarbons (PHAHs) on biochemical parameters in chicks of the Common Tern. Aquatic Toxicol. 30: 91-115.

Newton, I., M.B. Haas & P. Freestone (1990): Trends in organochlorine and mercury levels in gannet eggs. Environ. Pollut. 63: 1-12.

Newton, I., I. Wyllie & A. Asher (1993): Long-term trends in organochlorine and mercury residues in some predatory birds in Britain. Environ. Pollut. 79: 143-151.

Nisbet, I.C.T. (1973): Courtship feeding, egg size, and breeding success in Common Tern. Nature 241: 141-142.

Nisbet, I.C.T. (1994): Effects of pollution on marine birds. Bird Life Conserv. Series: 8-25.

Nisbet, I.C.T. & L.M. Reynolds (1984): Organochlorine residues in Common Tern and associated estuarine organisms, Massachusetts, USA, 1971-81. Marine Environ. Research 11: 33-66.

Ohlendorf, H.M. (1993): Marine birds and trace elements in the temperate North Pacific. In: K. Vermeer, K.T. Briggs, K.H. Morgan & D. Siegel-Causey (eds.): The Status Ecology and Conservation of Marine Birds of the North Pacific. Canadian Wildlife service, Ottawa: 232-240.

Ohlendorf, H.M., R.W. Riesebrough & K. Vermeer (1978): Exposure of marine birds to environmental pollutants. Wildlife Research Report 9. Washington, D.C.

Ohlendorf, H. M., F. C. Schaffner, T. h. W. Custer & C. J. Stafford (1985): Reproduction and organochlorine contaminants in terns at San Diego Bay. Colonial Waterbirds 8: 42-53.

Olsson, A., B. Helander, A. Bignert, K. Litzen, L. Asplund & A. Bergman (1998): Is PCB responsible for embryo toxicity in White-tailed Sea-Eagle (*Haliaeetus albicilla*) from the Swedish Baltic coast? Organohalogen Compounds 39: 17-20.

OSPAR (1997): Oslo and Paris Conventions for the Prevention of Marine Pollution. Joint meeting of the Oslo and Paris Commissions, Brussels 2-5 September 1997. Summary record OSPAR 97/15/1.

Parkinson, A. & S. Safe (1987): Mammalian biologic and toxic effects of PCB. In: S. Safe (ed.): Polychlorinated biphenyls: Mammalian and Environmental Toxicolgy.

Pearce, P.A., D.B. Peakall & L.M. Reynolds (1979): Shell thinning and residue of organochlorines and mercury in seabird eggs, eastern Canada, 1970-76. Pestic. Monitor. J. 13: 61-68.

Rasmussen, L.M., D.M. Fleet, B. Hälterlein, B.J. Koks, P. Potel & P. Südbeck (2000): Breeding birds in the Wadden Sea in 1996. Wadden Sea Ecosystem No. 10. Common Wadden Sea Secretariat, Wilhelmshaven. 122 pp.

Rattner, B.A., P.C. McGowan, J.S. Hatfield, C.-S. Hong & S.G. Chu (2001): Organochlorine contaminant exposure and reproductive success of Black-crowned Night Herons (*Nycticorax nycticorax*) nesting in Baltimore Harbor, Maryland. Arch. Environ. Contam. Toxicol. 41: 73-82.

Ryckman, D.P., D.V. Weseloh, P.Hamr, G.A. Fox, B. Collins, P.J. Ewins & R.J. Norstrom (1998): Spatial and temporal trends in organochlorine contamination and bill deformities in Double-crested Cormorants (*Phalacrocorax auritus*) from the Canadian Great Lakes. Environ. Monit. Asses. 53: 169-195.

Scheuhammer, A.M. (1987): The chronic toxicity of Aluminium, Cadmium, Mercury, and Lead in birds: A review. Environ. Pollut. 46: 263-295.

Schlüter, K. (2000): Review: evaporation of mercury from soils. An integration and synthesis of current knowledge. Environmental Geology 39: 249-271.

Schwarz, T.R. & D.L. Stalling (1991): Chemometric comparison of Polychlorinated biphenyl residues and toxicologically active biphenyl congeners in the eggs of Forster Terns (*Sterna forsteri*). Arch. Environ. Cotam. Toxicol. 20: 183-199.

Smit, C. J. & W. J. Wolff (1980): Birds of the Wadden Sea. Report 6, Wadden Sea Working Group Leiden.

Stade Declaration (1997): Ministerial Declaration of the Eighth Trilateral Governmental Conference on the protection of the Wadden Sea. Stade, 1997 (English version). Common Wadden Sea Secretariat. Wilhelmshaven, 100 pp.

Stansley, W., D.E. Roscoe, E. Hawthorne & R. Meyer (2001): Food chain aspects of chlordane poisoning in birds and bats. Arch. Environ. Contam. Toxicol. 40: 285-291.

Stäb, J.A., T.P. Traas, G. Stroomberg, J.v. Kesteren, P. Leonard, B.v. Hattum, U.A.T. Brikman & W.P. Cofino (1996): Determination of organotin compounds in the foodweb of a shallow freshwater lake in the Netherlands. Arch. Environ. Contam. Toxicol. 31: 319-328.

Stronkhorst, J., T.J. Ysebaert, F. Smedes, P.L. Meinigers, S. Dirksen & T.J. Boudewijn (1993): Contaminants in eggs of some waterbirds species from the Scheldt estuary, SW Netherlands. Mar. Poll. Bulletin 26: 572-578.

Struger, J. & D. V. Weseloh (1985): Great Lakes Caspian Terns: Egg contaminants and biological implications. Colonial Waterbirds 8: 142-149.

Switzer, B., V. Lewin & F.H. Wolfe (1973): DDE and reproductive success in some Alberta Common Terns. Can. J. Zool. 51: 1081-1086.

Teijning, S. (1967): Biological effects of methyl mercury dicyandiamide-treated grain in the domestic fowl *Gallus gallus* L. Oikos Suppl. 8: 1-116.

Thompson, D.R., P.H. Becker & R.W. Furness (1993): Long-term changes in mercury concentrations in Herring Gulls *Larus argentatus* and Common Tern *Sterna hirundo* from the German North Sea coast. J. Appl. Ecol. 30: 316-320.

Thyen, S., & P.H. Becker (2000): Aktuelle Ergebnisse des Schadstoffmonitorings mit Küstenvögeln im Wattenmeer. Vogelwelt 121:281-291.

Thyen, S., P.H. Becker & H. Behmann (2000a): Organochlorine and mercury contamination of Little Terns (*Sterna albifrons*) breeding at the western Baltic Sea, 1978-1996. Environ. Pollut. 108: 225-238.

Thyen, S., P.H. Becker, K.-M. Exo, B. Hälterlein, H. Hötker & P. Südbeck (1998): Monitoring breeding success of coastal birds. Wadden Sea Ecosystem No. 8. Common Wadden Sea Secretariat, Wilhelmshaven: 9-55.

Thyen, S., P.H. Becker, K.-M. Exo, B. Hälterlein, H. Hötker & P. Südbeck (2000b): Bruterfolgsmonitoring bei Küstenvögeln im Wattenmeer 1996 und 1997. Vogelwelt 121: 269-280.

TMAP (1997): Trilateral Monitoring and Assessment Program Manual. Common Wadden Sea Secretariat, Wilhelmshaven.

Walker, C.H. (1994): The ecotoxicology of persistent pollutants in marine fish-eating birds. In: C.H. Walker & D.R. Livingstone (eds.): Persistent pollutants in marine ecosystems. Pergamon Press, Oxford: 211-232.

Walsh, P.M. (1990): The use of seabirds as monitors of heavy metals in the marine environment. In: Furness, R.W., P.S. Rainbow (eds.) Heavy metals in the marine environment. CRC Press, Boca Raton, FL, 183pp.

Wendeln, H. & P.H. Becker (1996): Body mass change in breeding Common Terns (*Sterna hirundo*). Bird Study 43: 85-95.

Weseloh, D.V., P. Mineau & J. Struger (1990): Geographical distribution of contaminants and productivity measures of Herring Gulls in the Great Lakes: Lake Erie and connecting channels. Sci. Total Environ. 91: 141-159.

Appendix 1

List of all examined 62 PCB (Σ PCB) congeners analyzed since 1991. The shaded cells represent the 34 congeners analyzed since 1987 (Becker et al. 1991); bold letters represent the 6 congeners according to the "Schadstoff-Höchstmengenverordnung" of the German food law (BMU, 1988).

Congener	Degree of chlorination	Structure
PCB 28	3	
PCB 47/48	4	
PCB 52	4	
PCB 64	4	
PCB 66	4	
PCB 70	4	
PCB 74	4	
PCB 84/92	5	
PCB 85	5	
PCB 87/115	5	
PCB 95	5	
PCB 99	5	
PCB 101/90	5	
PCB 105	5	mono-ortho
PCB 107	5	
PCB 110	5	
PCB 114	5	mono-ortho
PCB 118	5	mono-ortho
PCB 123	5	mono-ortho
PCB 126	5	non-ortho
PCB 128	6	di-ortho
PCB 130	6	
PCB 132/146	6	
PCB 138	6	di-ortho
PCB 141	6	
PCB 149	6	
PCB 153	6	
PCB 155	6	
PCB 156	7	
PCB 157	6	mono-ortho
PCB 158/129	6	di-ortho/-
PCB 160/3/4	6	
PCB 166	6	di-ortho
PCB 167	6	mono-ortho
PCB 169	6	non-ortho
PCB 170	7	di-ortho
PCB 171	7	
PCB 172	6	mono-ortho
PCB 174	7	
PCB 175/187	7	
PCB 177	7	
PCB 178	7	
PCB 180/193	7	
PCB 183	7	
PCB 189	7	mono-ortho
PCB 190	8	
PCB 194	8	
PCB 195	8	
PCB 196/203	7	
PCB 199	8	
PCB 202	8	

Appendix 2

Concentrations of all single chemicals analyzed in Common Tern eggs sampled at different breeding sites along the Wadden Sea coast in 2000. Mean concentration ($ng \cdot g^{-1}$ fresh weight) and standard deviations are presented.

	Balgzand	Griend	Julianapolder	Delfzijl	Minsener Oog (Jade)	Neufelder Koog (Elbe)	Trischen	Margrethekoog
Hg	483,6 ± 221,1	304,9 ± 97,4	364,9 ± 60,0	437,2 ± 107,8	297,4 ± 81,6	961,7 ± 348,1	422,7 ± 75,8	297,7 ± 51,5
alpha-HCH	0,2 ± 0,1	0,1 ± 0,0	0,5 ± 0,1	0,2 ± 0,1	0,3 ± 0,1	0,4 ± 0,2	0,2 ± 0,1	0,2 ± 0,1
beta-HCH	2,6 ± 0,7	1,7 ± 0,6	1,8 ± 0,3	1,3 ± 0,3	3,8 ± 1,6	18,3 ± 5,0	25,9 ± 10,0	2,5 ± 0,8
gamma-HCH	1,4 ± 0,3	1,5 ± 0,3	1,4 ± 0,5	1,6 ± 0,7	1,8 ± 0,5	4,0 ± 2,4	1,5 ± 1,0	0,7 ± 0,3
HCB	7,1 ± 1,2	4,8 ± 1,0	10,0 ± 3,3	9,1 ± 3,4	9,5 ± 2,3	89,9 ± 32,3	17,0 ± 6,9	5,5 ± 1,5
p,p'-DDT	0,2 ± 0,2	0,3 ± 0,2	0,2 ± 0,1	0,4 ± 0,1	0,5 ± 0,2	4,9 ± 2,5	1,5 ± 1,6	0,2 ± 0,2
p,p'-DDD	1,0 ± 0,6	1,2 ± 0,8	0,6 ± 0,5	1,9 ± 1,5	2,3 ± 1,5	40,8 ± 29,7	11,4 ± 8,0	1,0 ± 0,6
p,p'-DDE	94,0 ± 63,9	46,0 ± 13,5	89,1 ± 24,9	73,9 ± 43,0	90,5 ± 52,4	344,7 ± 83,3	144,7 ± 79,1	58,4 ± 22,5
PCB28	5,6 ± 1,4	3,1 ± 0,8	5,5 ± 1,5	4,3 ± 1,6	4,0 ± 1,5	8,8 ± 3,0	3,6 ± 1,6	1,5 ± 0,3
PCB52	0,9 ± 0,5	1,0 ± 0,8	1,1 ± 0,8	1,0 ± 0,6	1,0 ± 0,4	5,8 ± 2,8	2,2 ± 2,1	0,5 ± 0,1
PCB47/48	8,8 ± 2,2	4,5 ± 0,9	7,5 ± 1,8	5,7 ± 2,2	5,9 ± 2,4	12,0 ± 3,8	6,2 ± 3,0	2,4 ± 0,4
PCB64	0,3 ± 0,1	0,4 ± 0,3	0,5 ± 0,5	0,5 ± 0,3	0,4 ± 0,2	1,1 ± 0,4	0,6 ± 0,5	0,2 ± 0,0
PCB95	0,9 ± 0,4	1,0 ± 0,7	1,1 ± 0,4	1,0 ± 0,5	0,8 ± 0,3	4,4 ± 1,9	2,2 ± 2,2	0,7 ± 0,2
PCB74	9,6 ± 1,9	4,6 ± 1,0	8,4 ± 1,6	7,1 ± 2,2	6,7 ± 2,5	10,3 ± 3,1	6,9 ± 3,7	2,8 ± 0,8
PCB70	6,1 ± 2,1	4,4 ± 1,1	2,9 ± 2,3	5,9 ± 2,2	5,3 ± 1,9	7,0 ± 1,6	5,0 ± 2,1	1,9 ± 0,4
PCB66	20,5 ± 3,9	10,0 ± 2,5	16,0 ± 3,5	13,4 ± 4,9	13,5 ± 5,1	21,3 ± 6,5	14,8 ± 7,3	5,2 ± 1,5
PCB92/84	1,5 ± 0,7	1,5 ± 1,2	1,6 ± 0,7	2,1 ± 1,3	1,4 ± 0,5	6,4 ± 3,6	4,3 ± 4,8	0,7 ± 0,2
PCB101/90	33,5 ± 12,0	25,8 ± 9,2	26,9 ± 10,1	32,7 ± 12,9	29,0 ± 11,5	71,3 ± 21,9	47,9 ± 25,6	11,5 ± 3,2
PCB99	63,2 ± 12,0	34,4 ± 8,7	50,5 ± 12,2	44,8 ± 15,2	38,1 ± 14,7	72,8 ± 20,8	58,1 ± 34,7	20,0 ± 6,6
PCB87/115	3,1 ± 1,2	2,5 ± 0,8	2,5 ± 0,8	3,1 ± 1,3	2,9 ± 1,0	8,9 ± 2,8	4,5 ± 2,1	1,2 ± 0,3
PCB85	4,1 ± 0,8	2,2 ± 0,6	3,6 ± 0,8	3,0 ± 1,3	2,7 ± 1,1	5,6 ± 1,6	4,0 ± 2,4	1,6 ± 0,5
PCB110	25,4 ± 8,6	19,6 ± 6,6	19,8 ± 7,6	26,3 ± 7,1	22,6 ± 9,3	48,0 ± 13,3	35,4 ± 19,4	8,0 ± 3,0
PCB107	4,0 ± 1,1	2,9 ± 0,9	2,4 ± 1,2	4,8 ± 1,5	3,6 ± 1,4	5,4 ± 1,4	5,5 ± 3,2	2,3 ± 0,6
PCB123	0,7 ± 0,1	0,4 ± 0,1	0,7 ± 0,1	0,5 ± 0,2	0,5 ± 0,1	1,5 ± 0,4	1,0 ± 0,5	0,3 ± 0,1
PCB118	69,6 ± 14,2	43,1 ± 10,8	61,3 ± 13,2	58,0 ± 20,8	48,3 ± 16,0	87,4 ± 22,5	77,7 ± 43,0	30,7 ± 9,0
PCB114	0,9 ± 0,2	0,8 ± 0,3	1,3 ± 0,4	0,9 ± 0,2	1,0 ± 0,3	1,9 ± 0,5	1,6 ± 1,0	0,5 ± 0,2
PCB105	11,8 ± 2,7	7,8 ± 2,0	11,5 ± 2,8	11,3 ± 4,1	9,7 ± 3,3	16,6 ± 4,6	15,2 ± 8,4	6,4 ± 1,9
PCB126	0,4 ± 0,1	0,3 ± 0,1	0,4 ± 0,1	0,4 ± 0,1	0,4 ± 0,1	0,8 ± 0,2	0,6 ± 0,3	0,3 ± 0,1
PCB155	1,4 ± 0,2	0,7 ± 0,2	1,0 ± 0,2	0,9 ± 0,3	3,8 ± 9,7	6,4 ± 1,6	2,3 ± 1,3	0,5 ± 0,2
PCB149	27,8 ± 8,2	25,1 ± 11,8	21,7 ± 11,3	33,2 ± 11,8	25,3 ± 11,1	69,1 ± 18,2	60,4 ± 31,5	11,1 ± 4,0
PCB132/146	60,3 ± 11,9	45,1 ± 19,3	49,4 ± 11,7	60,0 ± 14,2	43,7 ± 14,7	121,8 ± 33,1	98,5 ± 53,1	26,4 ± 9,3

	Balgzand	Griend	Julianapolder	Delfzijl	Minsener Oog (Jade)	Neufelder Koog (Elbe)	Trischen	Margrethekoog
PCB153	354,2 ± 54,7	215,3 ± 55,0	264,3 ± 59,7	273,2 ± 89,2	215,0 ± 70,9	682,9 ± 165,2	476,7 ± 286,9	164,2 ±47,0
PCB141	5,1 ± 1,9	3,4 ± 1,1	4,0 ± 1,7	4,5 ± 1,3	3,5 ± 1,5	18,4 ± 6,5	7,7 ± 4,1	2,4 ±1,4
PCB130	4,8 ± 1,4	4,5 ± 2,9	4,6 ± 1,3	5,9 ± 1,6	4,6 ± 1,6	10,2 ± 3,0	10,1 ± 5,6	2,6 ±0,9
PCB178	2,0 ± 0,5	2,1 ± 0,8	2,2 ± 0,6	2,4 ± 0,8	2,0 ± 0,7	4,6 ± 2,0	5,9 ± 3,0	1,4 ±0,5
PCB160/3/4	61,5 ± 12,2	42,5 ± 12,1	49,6 ± 13,6	58,3 ± 17,4	44,4 ± 14,1	112,4 ± 27,1	88,1 ± 55,5	31,4 ±9,5
PCB138	164,4 ± 26,3	102,8 ± 28,7	120,6 ± 33,2	134,5 ± 43,5	106,9 ± 33,7	319,3 ± 79,9	236,7 ± 134,7	85,5 ±25,0
PCB158/129	8,8 ± 2,6	4,1 ± 1,2	4,9 ± 1,3	5,5 ± 2,0	4,1 ± 1,4	20,9 ± 5,4	10,1 ± 6,1	3,1 ±0,8
PCB175/187	70,4 ± 13,4	55,7 ± 20,8	61,7 ± 15,5	75,3 ± 20,1	56,5 ± 18,0	144,1 ± 32,1	126,7 ± 75,9	40,0 ±13,4
PCB166	0,4 ± 0,1	0,3 ± 0,2	0,5 ± 0,2	0,3 ± 0,1	0,3 ± 0,1	0,9 ± 0,3	0,7 ± 0,4	0,0 ±0,0
PCB183	31,6 ± 8,1	16,0 ± 4,7	17,6 ± 4,5	20,0 ± 6,6	14,7 ± 5,8	65,4 ± 14,8	33,8 ± 20,4	11,4 ±3,0
PCB202	1,2 ± 0,3	1,2 ± 0,4	1,5 ± 0,3	1,9 ± 1,8	1,3 ± 0,3	2,1 ± 0,7	2,9 ± 1,4	0,8 ±0,2
PCB174	19,5 ± 4,1	13,2 ± 3,6	15,7 ± 3,2	17,4 ± 5,7	13,7 ± 4,0	34,6 ± 9,0	29,0 ± 15,9	11,2 ±3,2
PCB128	5,2 ± 1,8	3,4 ± 1,7	3,3 ± 1,5	4,0 ± 1,4	3,1 ± 1,8	12,1 ± 3,5	7,9 ± 4,4	2,0 ±1,9
PCB177	17,4 ± 3,4	13,0 ± 5,1	16,1 ± 2,8	17,6 ± 5,1	14,1 ± 4,9	25,4 ± 5,3	28,8 ± 16,4	9,6 ±2,6
PCB167	8,2 ± 1,5	5,3 ± 1,3	6,0 ± 1,2	7,1 ± 2,5	5,1 ± 1,4	16,8 ± 3,4	11,1 ± 6,5	4,3 ±1,1
PCB171	10,8 ± 2,5	5,9 ± 1,8	6,9 ± 1,4	7,5 ± 2,3	5,9 ± 2,0	19,3 ± 4,4	13,1 ± 7,5	4,6 ±1,3
PCB172	6,1 ± 1,5	3,9 ± 1,4	4,0 ± 0,9	4,7 ± 1,4	3,5 ± 1,2	14,4 ± 3,3	7,9 ± 4,7	2,6 ±0,9
PCB156	10,4 ± 1,9	6,7 ± 1,8	8,0 ± 1,9	9,1 ± 3,3	6,7 ± 1,9	19,1 ± 4,3	13,7 ± 7,9	5,5 ±1,4
PCB157	2,0 ± 0,5	1,5 ± 0,5	1,9 ± 0,4	2,0 ± 0,6	1,8 ± 0,8	3,0 ± 1,3	3,7 ± 1,9	1,2 ±0,4
PCB180/193	124,2 ± 28,5	69,8 ± 18,2	70,4 ± 18,2	85,7 ± 29,2	65,8 ± 24,9	254,8 ± 50,5	135,3 ± 82,6	45,6 ±12,3
PCB199	10,4 ± 3,4	6,7 ± 2,4	7,5 ± 2,1	8,1 ± 2,3	6,3 ± 2,2	17,1 ± 2,9	11,8 ± 6,4	4,3 ±1,3
PCB170	37,6 ± 8,1	21,3 ± 5,6	20,2 ± 9,1	26,4 ± 9,7	20,4 ± 7,6	67,4 ± 13,5	41,2 ± 24,9	14,8 ±4,3
PCB196/203	15,5 ± 5,7	8,3 ± 2,1	10,3 ± 2,9	10,7 ± 3,2	8,9 ± 3,6	27,1 ± 4,3	15,4 ± 8,6	6,5 ±1,7
PCB190	8,3 ± 2,0	5,2 ± 1,3	7,6 ± 5,1	6,9 ± 2,3	5,8 ± 1,8	18,2 ± 3,8	11,7 ± 7,2	4,1 ±1,0
PCB169	0,1 ± 0,0	0,1 ± 0,0	0,2 ± 0,0	0,1 ± 0,1	0,1 ± 0,0	0,1 ± 0,1	0,1 ± 0,0	0,0 ±0,0
PCB195	3,8 ± 1,3	2,3 ± 0,5	2,7 ± 0,8	3,1 ± 0,9	2,6 ± 0,9	6,1 ± 0,8	4,3 ± 2,4	1,8 ±0,5
PCB189	1,9 ± 0,4	1,3 ± 0,3	1,5 ± 0,4	1,7 ± 0,6	1,4 ± 0,4	4,1 ± 0,6	2,6 ± 1,6	1,0 ±0,4
PCB194	10,9 ± 3,9	6,7 ± 1,6	7,4 ± 1,7	8,4 ± 2,4	6,7 ± 3,0	18,8 ± 2,4	11,2 ± 6,3	4,9 ±1,4
c-Chlordane	0,0 ± 0,0	0,0 ± 0,0	0,0 ± 0,0	0,1 ± 0,0	0,1 ± 0,0	0,1 ± 0,1	0,1 ± 0,3	0,0 ±0,0
t-Chlordane	0,2 ± 0,1	0,1 ± 0,2	0,2 ± 0,1	0,3 ± 0,1	0,4 ± 0,2	0,2 ± 0,1	0,5 ± 0,5	0,0 ±0,1
c-Nonachlor	0,2 ± 0,1	0,2 ± 0,2	0,3 ± 0,1	0,5 ± 0,1	0,4 ± 0,2	0,2 ± 0,1	0,5 ± 0,4	0,0 ±0,0
t-Nonachlor	0,2 ± 0,1	0,3 ± 0,4	0,3 ± 0,2	0,6 ± 0,4	0,5 ± 0,1	0,3 ± 0,1	0,9 ± 1,1	0,1 ±0,1

Appendix 3

Concentrations of all single chemicals analyzed in Oystercatcher eggs sampled at different breeding sites along the Wadden Sea coast in 2000. Mean concentration (ng·g[-1] fresh weight) and standard deviations are presented.

	Balgzand	Griend	Julianapolder	Delfzijl	Dollart	Mellum (Jade)	Hullen (Elbe)	Trischen	Norderoog	Langli
Hg	208,5 ± 53,0	275,2 ± 109,7	140,6 ± 41,1	216,6 ± 83,5	125,0 ± 15,4	216,2 ± 59,5	358,2 ± 64,6	263,2 ± 53,2	203,9 ± 39,3	223,5 ± 35,2
alpha-HCH	0,2 ± 0,1	0,1 ± 0,0	0,2 ± 0,1	0,2 ± 0,1	0,3 ± 0,2	0,4 ± 0,3	0,2 ± 0,1	0,3 ± 0,1	0,2 ± 0,1	0,2 ± 0,1
gamma-HCH	1,1 ± 0,4	1,6 ± 0,6	2,4 ± 0,6	1,6 ± 0,4	1,8 ± 0,5	0,7 ± 0,2	0,9 ± 0,2	0,6 ± 0,2	0,5 ± 0,1	0,8 ± 0,2
HCB	8,2 ± 5,3	2,5 ± 1,0	7,9 ± 3,6	43,0 ± 40,7	11,7 ± 4,4	3,6 ± 0,6	17,3 ± 6,5	10,6 ± 3,1	3,1 ± 1,4	2,4 ± 0,9
p,p'-DDT	0,8 ± 0,7	0,3 ± 0,2	1,2 ± 0,4	1,6 ± 0,8	1,3 ± 0,4	0,6 ± 0,2	3,8 ± 1,2	1,5 ± 0,5	0,5 ± 0,3	0,8 ± 0,3
p,p'-DDD	5,3 ± 8,8	0,5 ± 0,1	1,3 ± 0,3	0,7 ± 0,3	1,8 ± 0,5	0,6 ± 0,1	1,9 ± 0,6	4,0 ± 1,6	1,1 ± 0,3	0,5 ± 0,2
p,p'-DDE	68,2 ± 34,4	21,7 ± 6,4	79,2 ± 26,6	44,6 ± 15,8	94,9 ± 16,6	40,3 ± 11,3	126,2 ± 34,8	114,1 ± 24,3	43,6 ± 17,1	26,0 ± 5,0
PCB28	3,8 ± 1,8	2,3 ± 0,8	7,3 ± 2,5	3,0 ± 1,3	5,9 ± 1,3	2,4 ± 0,6	1,4 ± 0,2	2,2 ± 0,6	1,5 ± 0,4	1,1 ± 0,4
PCB52	0,9 ± 0,7	0,4 ± 0,1	0,7 ± 0,1	0,5 ± 0,1	0,9 ± 0,2	0,4 ± 0,1	0,4 ± 0,1	0,6 ± 0,2	0,3 ± 0,0	0,4 ± 0,1
PCB47/48	5,1 ± 3,1	1,7 ± 0,4	5,3 ± 1,5	2,2 ± 0,7	4,3 ± 0,9	1,9 ± 0,3	1,7 ± 0,4	2,3 ± 0,6	1,2 ± 0,2	1,1 ± 0,2
PCB64	0,4 ± 0,2	0,2 ± 0,0	0,6 ± 0,1	0,2 ± 0,1	0,5 ± 0,1	0,3 ± 0,0	0,2 ± 0,1	0,3 ± 0,1	0,2 ± 0,0	0,2 ± 0,0
PCB95	1,3 ± 0,7	0,9 ± 0,2	1,8 ± 0,4	1,0 ± 0,3	2,8 ± 0,9	0,8 ± 0,2	1,1 ± 0,4	1,3 ± 0,3	0,7 ± 0,1	0,7 ± 0,1
PCB74	5,9 ± 2,5	3,1 ± 1,1	10,8 ± 3,8	3,6 ± 1,4	8,6 ± 1,8	3,8 ± 0,9	2,2 ± 0,3	3,3 ± 0,9	1,9 ± 0,6	1,7 ± 0,8
PCB70	3,3 ± 2,3	0,9 ± 0,2	2,1 ± 0,5	0,8 ± 0,2	1,8 ± 0,4	0,8 ± 0,1	0,4 ± 0,1	0,9 ± 0,2	0,6 ± 0,2	0,5 ± 0,1
PCB66	12,3 ± 5,5	6,9 ± 2,4	24,5 ± 8,3	7,4 ± 2,5	16,3 ± 3,9	8,7 ± 2,3	6,1 ± 1,3	7,9 ± 2,0	4,0 ± 1,5	3,3 ± 1,7
PCB92/84	3,0 ± 1,9	2,9 ± 0,7	8,5 ± 2,6	3,6 ± 1,8	7,8 ± 1,7	3,0 ± 0,9	3,7 ± 0,8	4,7 ± 1,4	2,0 ± 0,8	1,1 ± 0,1
PCB101/90	23,9 ± 19,2	3,3 ± 0,8	9,7 ± 2,5	3,4 ± 0,8	5,8 ± 1,4	2,9 ± 0,7	3,3 ± 0,6	4,4 ± 1,3	2,3 ± 0,7	1,8 ± 0,3
PCB99	45,1 ± 15,5	21,8 ± 7,6	79,7 ± 26,9	26,0 ± 8,2	58,4 ± 10,2	26,5 ± 7,4	31,5 ± 7,8	30,4 ± 6,4	14,9 ± 5,7	9,4 ± 3,9
PCB87/115	2,4 ± 1,7	0,7 ± 0,2	2,1 ± 0,5	1,1 ± 0,4	1,8 ± 0,3	0,8 ± 0,2	0,8 ± 0,1	1,1 ± 0,3	0,7 ± 0,2	0,5 ± 0,1
PCB85	2,4 ± 1,5	0,7 ± 0,2	2,2 ± 0,7	0,8 ± 0,3	1,6 ± 0,3	0,8 ± 0,2	0,8 ± 0,2	1,0 ± 0,3	0,5 ± 0,2	0,3 ± 0,1
PCB110	17,8 ± 15,2	2,0 ± 0,6	5,2 ± 1,4	1,7 ± 0,4	3,9 ± 0,7	1,7 ± 0,5	2,1 ± 0,2	2,5 ± 0,5	1,4 ± 0,3	1,1 ± 0,2
PCB107	3,7 ± 1,1	2,5 ± 0,8	7,7 ± 2,7	2,4 ± 0,9	5,8 ± 1,2	4,6 ± 5,1	2,4 ± 0,4	2,9 ± 0,8	1,8 ± 0,7	1,3 ± 0,6
PCB123	0,5 ± 0,1	0,6 ± 0,4	1,5 ± 0,6	0,6 ± 0,2	1,1 ± 0,2	3,3 ± 8,3	0,8 ± 0,2	0,7 ± 0,2	0,3 ± 0,1	0,3 ± 0,1
PCB118	57,0 ± 15,2	43,1 ± 14,8	118,6 ± 39,5	40,8 ± 16,4	78,2 ± 13,2	48,6 ± 12,3	40,9 ± 8,1	48,1 ± 8,8	29,0 ± 9,9	20,0 ± 7,7
PCB114	1,0 ± 0,3	0,7 ± 0,2	1,8 ± 0,7	0,9 ± 0,2	2,0 ± 0,4	1,1 ± 0,3	1,4 ± 0,3	1,3 ± 0,3	0,5 ± 0,2	0,4 ± 0,2
PCB105	10,1 ± 2,9	7,0 ± 2,3	21,7 ± 7,3	9,1 ± 4,4	15,5 ± 2,5	8,7 ± 2,3	8,2 ± 1,6	8,7 ± 1,7	5,3 ± 1,9	4,1 ± 1,5
PCB126	0,3 ± 0,1	0,2 ± 0,1	0,5 ± 0,2	0,3 ± 0,1	0,4 ± 0,1	0,3 ± 0,1	0,4 ± 0,1	0,3 ± 0,1	0,2 ± 0,1	0,2 ± 0,1
PCB155	1,1 ± 0,5	0,4 ± 0,1	1,3 ± 0,5	0,4 ± 0,1	0,9 ± 0,3	0,4 ± 0,1	1,6 ± 0,5	1,2 ± 0,4	0,3 ± 0,1	0,1 ± 0,1
PCB149	30,0 ± 20,6	7,1 ± 1,5	26,8 ± 9,8	10,0 ± 2,9	20,5 ± 4,8	7,0 ± 2,1	18,4 ± 3,4	10,7 ± 3,4	5,4 ± 1,8	2,7 ± 0,4
PCB132/146	60,3 ± 18,5	38,1 ± 11,4	110,0 ± 39,9	44,9 ± 9,1	82,8 ± 15,9	43,8 ± 13,5	69,8 ± 13,0	59,1 ± 13,4	28,7 ± 11,6	16,5 ± 7,9
PCB153	299,6 ± 71,3	186,2 ± 61,3	543,4 ± 205,6	198,2 ± 48,9	403,7 ± 71,2	224,1 ± 62,8	303,2 ± 64,4	308,0 ± 66,3	168,1 ± 59,2	86,2 ± 26,4
PCB141	3,5 ± 2,6	0,7 ± 0,3	1,8 ± 0,6	1,3 ± 0,5	1,7 ± 0,4	0,6 ± 0,1	2,1 ± 0,5	1,0 ± 0,4	0,6 ± 0,5	0,5 ± 0,1

	Balgzand	Griend	Julianapolder	Delfzijl	Dollart	Mellum (Jade)	Hullen (Elbe)	Trischen	Norderoog	Langli
PCB130	5,4 ± 2,0	3,2 ± 1,0	10,5 ± 3,6	4,7 ± 1,5	7,6 ± 2,0	3,6 ± 1,2	7,0 ± 1,8	5,5 ± 1,5	2,5 ± 1,2	2,3 ± 2,5
PCB178	6,2 ± 4,2	6,9 ± 2,3	18,3 ± 8,8	12,1 ± 2,2	17,5 ± 6,3	8,8 ± 2,9	18,5 ± 3,4	13,6 ± 3,6	6,1 ± 2,6	3,9 ± 2,5
PCB160/3/4	52,5 ± 20,7	24,2 ± 7,1	70,1 ± 23,4	29,6 ± 7,8	55,6 ± 10,1	29,1 ± 8,6	49,7 ± 9,5	38,0 ± 8,2	20,5 ± 8,7	12,5 ± 4,3
PCB138	139,5 ± 38,2	77,9 ± 24,8	255,3 ± 91,8	96,6 ± 31,2	186,2 ± 31,9	96,8 ± 27,5	141,1 ± 31,5	130,9 ± 27,4	77,4 ± 29,9	41,4 ± 11,8
PCB158/129	5,0 ± 2,8	1,9 ± 0,7	7,9 ± 2,7	3,1 ± 1,7	5,2 ± 0,8	1,9 ± 0,5	4,9 ± 1,2	3,2 ± 0,8	1,6 ± 0,6	1,0 ± 0,3
PCB175/187	76,4 ± 21,6	47,4 ± 14,7	141,3 ± 57,6	63,7 ± 12,4	113,8 ± 21,0	58,8 ± 17,8	121,4 ± 19,2	82,0 ± 19,7	43,7 ± 18,6	24,8 ± 10,2
PCB166	0,5 ± 0,2	0,2 ± 0,1	0,6 ± 0,2	0,4 ± 0,3	0,5 ± 0,1	0,3 ± 0,1	0,5 ± 0,1	0,4 ± 0,1	0,3 ± 0,3	0,2 ± 0,1
PCB183	19,4 ± 5,2	11,0 ± 3,5	39,0 ± 16,6	13,8 ± 3,6	28,2 ± 4,5	13,1 ± 4,1	23,1 ± 4,2	19,7 ± 4,5	10,0 ± 3,8	5,0 ± 1,8
PCB202	2,8 ± 1,4	3,2 ± 0,9	7,2 ± 3,4	5,5 ± 1,0	7,4 ± 2,7	4,5 ± 1,8	8,0 ± 1,6	5,8 ± 1,3	3,0 ± 1,1	1,8 ± 1,1
PCB174	18,2 ± 4,3	10,5 ± 3,2	34,0 ± 12,3	14,9 ± 4,9	24,7 ± 6,3	13,1 ± 3,9	18,1 ± 3,6	16,8 ± 3,5	10,3 ± 3,6	5,7 ± 2,0
PCB128	3,5 ± 2,6	0,8 ± 0,3	2,2 ± 0,7	1,3 ± 0,4	1,9 ± 0,4	1,0 ± 1,4	2,5 ± 0,6	1,0 ± 0,3	0,6 ± 0,3	0,4 ± 0,1
PCB177	17,5 ± 5,6	10,5 ± 3,1	31,0 ± 12,7	17,0 ± 3,7	27,9 ± 8,2	13,0 ± 4,4	27,0 ± 6,2	19,8 ± 5,3	10,0 ± 4,6	6,3 ± 2,9
PCB167	6,6 ± 1,6	4,7 ± 1,5	12,1 ± 4,8	5,8 ± 1,7	8,9 ± 1,4	5,4 ± 1,6	7,6 ± 1,5	7,4 ± 1,6	4,1 ± 1,4	2,3 ± 0,8
PCB171	7,1 ± 2,0	4,4 ± 1,3	16,0 ± 6,5	5,5 ± 1,4	11,6 ± 1,8	5,4 ± 1,6	8,5 ± 1,6	7,7 ± 1,7	4,3 ± 1,6	2,2 ± 0,8
PCB172	4,1 ± 1,5	1,7 ± 0,5	4,9 ± 1,7	2,7 ± 0,9	4,4 ± 0,7	1,9 ± 0,5	4,9 ± 1,2	3,1 ± 0,7	1,8 ± 0,9	0,9 ± 0,3
PCB156	8,4 ± 2,2	4,7 ± 1,5	14,1 ± 5,4	6,8 ± 2,7	10,0 ± 1,6	5,8 ± 1,6	7,9 ± 1,6	7,1 ± 1,5	4,3 ± 1,7	2,7 ± 0,8
PCB157	2,8 ± 1,1	1,2 ± 0,3	2,9 ± 1,0	1,8 ± 0,8	2,2 ± 0,3	1,4 ± 0,4	1,7 ± 0,3	1,7 ± 0,3	1,0 ± 0,4	0,7 ± 0,2
PCB180/193	81,5 ± 22,8	38,3 ± 12,3	127,5 ± 54,8	60,1 ± 15,2	99,3 ± 14,6	42,4 ± 12,5	97,2 ± 22,1	68,9 ± 16,0	30,3 ± 12,4	16,5 ± 6,0
PCB199	7,9 ± 2,3	4,3 ± 1,4	12,2 ± 4,7	7,2 ± 1,5	11,5 ± 1,9	4,4 ± 0,9	12,6 ± 3,0	7,5 ± 1,9	4,3 ± 2,2	2,3 ± 1,0
PCB170	24,7 ± 6,8	12,6 ± 4,3	40,0 ± 15,9	17,4 ± 5,3	31,7 ± 5,8	14,1 ± 4,2	26,9 ± 6,7	20,3 ± 4,6	12,1 ± 4,9	5,6 ± 2,0
PCB196/203	10,1 ± 2,1	7,1 ± 2,3	22,0 ± 9,8	7,8 ± 1,8	16,0 ± 2,7	7,2 ± 2,2	12,3 ± 2,9	10,2 ± 2,2	6,4 ± 2,3	3,1 ± 1,2
PCB190	6,8 ± 1,8	4,4 ± 1,4	12,8 ± 5,5	5,7 ± 2,0	10,0 ± 1,8	5,5 ± 2,2	8,0 ± 2,1	7,1 ± 1,6	4,9 ± 2,1	2,1 ± 0,7
PCB169	0,3 ± 0,2	0,0 ± 0,0	0,0 ± 0,0	0,3 ± 0,3	0,1 ± 0,0	0,1 ± 0,0	0,1 ± 0,0	0,1 ± 0,0	0,1 ± 0,0	0,1 ± 0,0
PCB195	2,9 ± 0,7	2,2 ± 0,7	5,7 ± 2,3	2,2 ± 0,7	4,3 ± 0,7	2,1 ± 0,7	3,8 ± 0,9	2,9 ± 0,6	2,1 ± 0,6	1,0 ± 0,4
PCB189	1,3 ± 0,5	0,9 ± 0,3	2,3 ± 1,0	0,9 ± 0,3	1,6 ± 0,3	1,1 ± 0,3	1,6 ± 0,3	1,3 ± 0,3	0,9 ± 0,3	0,5 ± 0,2
PCB194	7,2 ± 2,0	4,2 ± 1,3	11,1 ± 4,5	5,5 ± 1,7	8,1 ± 1,6	3,9 ± 1,1	8,8 ± 2,5	5,4 ± 1,4	3,8 ± 1,5	2,0 ± 0,8
c-Chlordane	0,0 ± 0,1	0,1 ± 0,0	0,2 ± 0,1	0,1 ± 0,1	0,4 ± 0,1	0,1 ± 0,0	0,0 ± 0,0	0,1 ± 0,0	0,1 ± 0,1	0,1 ± 0,0
t-Chlordane	0,2 ± 0,1	0,3 ± 0,1	0,9 ± 0,4	0,4 ± 0,1	0,8 ± 0,2	0,4 ± 0,1	0,3 ± 0,1	0,3 ± 0,1	0,5 ± 0,2	0,3 ± 0,1
c-Nonachlor	0,4 ± 0,2	0,3 ± 0,1	0,9 ± 0,5	0,6 ± 0,2	0,8 ± 0,3	0,4 ± 0,2	0,3 ± 0,1	0,3 ± 0,1	0,5 ± 0,3	0,3 ± 0,1
t-Nonachlor	0,7 ± 0,5	0,8 ± 0,4	2,8 ± 1,6	1,3 ± 0,4	2,4 ± 0,6	1,2 ± 0,5	0,8 ± 0,2	0,8 ± 0,4	1,5 ± 0,7	0,8 ± 0,3

Issues of the Publication Series „Wadden Sea Ecosystem“