



Halogenated organic compounds in the eggs of aquatic birds from the Gulf of Gdansk and Wloclawek Dam (Poland)



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HIGHLIGHTS

- Deca-BDE was found only in gull eggs from an inland reservoir.
- Highly chlorinated furans have the lowest concentration in birds' eggs.
- Chlorinated compounds dominated in tern eggs, brominated in gull eggs.

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ABSTRACT

Diet is the major route of exposure to environmental contaminants in biota and, after penetration into an organism, xenobiotics continue to accumulate in the body. In birds the egg-laying process acts as a transfer mechanism for the accumulated pollutants and results in the burdening of the next generation at an early stage of development. This transfer has a strong influence on developmental disorders and even breeding success. With this in mind polybrominated diphenyl ethers (PBDE), polychlorinated dibenzo-p-dioxins and polychlorinated dibenzo furans (PCDD/Fs), as well as polychlorinated biphenyls (PCBs), were analyzed in the eggs of aquatic birds from different habitats and with different diet preferences. The highest concentration of PBDE was noted in gull eggs ($198.31 \text{ ng g}^{-1} \text{ dw.}$) and the lowest in tern eggs (sandwich tern: $76.96 \text{ ng g}^{-1} \text{ dw.}$; common tern: $113.73 \text{ ng g}^{-1} \text{ dw.}$). Deca-BDE was detected only in herring gull eggs from the Wloclawek reservoir. PCDD/Fs were found in the eggs of terns from the southern coast of the Baltic Sea and gulls from an inland reservoir (dam) on the River Vistula close to the town of Wloclawek. The highest toxicity (birds Toxic Equivalent Factor) was found in the eggs of terns (sandwich tern – $93.97 \text{ pg g}^{-1} \text{ dw.}$, common tern – $68.35 \text{ pg g}^{-1} \text{ dw.}$), and this was found to be several times higher than in herring gull eggs ($18.80 \text{ pg g}^{-1} \text{ dw.}$). Non-dioxin like PCBs were ten times higher than other analyzed PCB congeners, but the congener pattern was similar to other studies.

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1. Introduction

Coastal areas, lagoons and reservoirs are places where water birds are inadvertently drawn into the biogeochemical cycle of certain halogenated persistent organic pollutants (POPs). The combination of shallow waters and anthropogenic pressure causes them to be exposed to a wide range of pollutants as a matter of course, and they can also be greatly affected by accidental ones. Halogenated persistent organic pollutants including polybrominated diphenyl ethers (PBDE) and brominated, chlorinated

and mixed brominated/chlorinated derivatives of biphenyl, dioxin and dibenzofurans are among the primary environmental contaminants deemed to be of high risk (Van den Berg et al., 1998).

PBDEs are chemicals used in plastics, textiles, electronic circuitry and other materials to prevent fires. There are currently 209 recognized PBDE congeners and technical products contain mainly penta-BDEs, octa-BDEs and deca-BDE (De Wit, 2002; Geyer et al., 2000; Hanari et al., 2006; La Guardia et al., 2006). Polychlorinated biphenyls (PCBs) were produced commercially between the 1930s and 1970s in most industrialized countries and were widely used in industry and commercial products (Rice et al., 2003). Unlike PBDEs and PCBs, polychlorinated dibenzo-p-dioxins and furans (PCDD/Fs) were never produced intentionally. They are instead by-products of industrial processes (e.g. chlorinated phenol production), formed during combustion, and are detected in

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ash from industrial heating facilities and particles from combustion of municipal and chemical waste (Hites, 2011).

These organic pollutants are toxic, persist in the environment even with long distance atmospheric transportation and are resistant to photo degradation, and chemical or biological decomposition. Most of them cause disruption to the endocrine system and may therefore be held responsible for abnormalities in development and functional disorders (Baker and Kjellerup, 2016; Fernie et al., 2009; Suzuki et al., 2010; Wohlfahrt-Veje et al., 2014). Recent studies show that poor breeding success in birds often results from a high body burden of these compounds (Desforges et al., 2018; Falandysz et al., 2019; Marteinson et al., 2010; Toschik et al., 2005).

POPs are present in aquatic biota worldwide and are regarded as an important pollutant on account of their abilities to accumulate and magnify in the trophic chain (eg. Aznar-Alemany et al., 2019; Eljarrat et al., 2019; Morales et al., 2016; Quinn et al., 2013; Polder et al., 2016; Zicus and Rave, 2004). In the Baltic Sea region, chlorinated and brominated organic compounds have been found both in marine sediment and the water (eg. Dabrowska et al., 2017; Falandysz and Strandberg, 2004; Szlinder-Richert et al., 2012), as well as in Baltic fish (eg. Falandysz et al., 2019; Karl and Ruoff, 2007; Szlinder-Richert et al., 2009; Waszak et al., 2012) and fish products (eg. Falandysz, 1994; Falandysz et al., 2019; Usydus et al., 2009). Recent studies on piscivorous birds and other top predators from the Baltic Sea region demonstrate their contamination by several organic pollutants (eg. Falandysz, 1984, 1986; Falandysz and Szefer, 1984; Falandysz et al., 1994; 2000; Falkowska et al., 2016; Flidner et al., 2012; Kannan et al., 2003; Lundstedt-Enkel et al., 2006; Thyen et al., 2000). This points to a highly significant dependence between habitat pollution and potential risk of contamination in biota.

Food, especially seafood, is considered to be the major source of human and animal exposure to several organic xenobiotics, including chlorinated and brominated ones (Falandysz et al., 2019; Fernandes et al., 2018; Knutsen et al., 2008; Tard et al., 2007; Wan et al., 2005). Birds provide an early warning in terms of exposure to environmental hazards within the trophic chain. As pollution may cause unexpected changes in their numbers, health or breeding success, these animals are commonly viewed as sentinel organisms (Grove et al., 2009) and their eggs are often used as a noninvasive tool for environmental pollution monitoring (eg. Bignert et al., 2016; Miller et al., 2014; Rigét et al., 2016).

Concentrations and congener patterns of halogenated organic compounds in aquatic birds from different ecosystems may be strongly dependent on their food sources. To confirm the strong connection between organism and environment we determined levels of PBDEs, PCDD/Fs and PCBs in the eggs of three species of aquatic birds using different food sources: Sandwich terns (*Sterna sandvicensis*) and Common terns (*Sterna hirundo*) from the Gulf of Gdansk (coastal sea area) represent marine food consumers, while Herring gulls (*Larus argentatus*) from a breeding colony on the Wloclawek Dam (inland reservoir) represent omnivores and scavengers. Even the results shown data from 8 to 9 years ago and continuously declining trends for these pollutants are indicated they are still present in birds system. In this case the results of this study might allow to focus on further research on the biochemical processes, such as maternal transfer, in birds during breeding, as important source of pollutants transfer to the next generation and birds' body burden since the first life stage.

2. Materials and methods

Research was carried out on herring gull (*Larus argentatus*) eggs (n = 25) collected in 2011 from a breeding colony located at the River Vistula dam in Wloclawek (52°39'N, 19°08'E), and on

sandwich tern (*Sterna sandvicensis*) (n = 35) and common tern (*Sterna hirundo*) eggs (n = 10) collected in 2010 from Mewia Lacha Bird Sanctuary (54°21'N, 18°56'E) which were combined into three composite samples respectively. The herring gull breeding colony consisted of 138 pairs and was a mixed colony consisting mainly of herring gulls but also featuring individual caspian gulls, and other hybrid specimens. The size of the tern breeding colony was estimated at 276 pairs of sandwich terns and 100 pairs of common terns. The sandwich tern eggs were collected in 2010 after flooding resulted in laid eggs being washed out of their nests. All of the analyzed eggs had probably been abandoned by the parents. A detailed description of the sample collection is given in a previous paper by Grajewska et al. (2015) - for the present study only unhatched eggs were used. All samples were collected with the permission of national authorities.

The eggs were transported to the laboratory, washed and weighed. The materials were frozen and whole eggs were separated from shell and membrane. The obtained materials were freeze dried with sodium sulfate and kept in amber glass vessels until analysis.

The main 17 congeners of PCDDFs listed by the WHO as having strong toxicity potential, dioxin-like compounds (non-ortho PCBs: 77, 81, 126, 169; mono ortho PCB: 105, 114, 118, 123, 156, 157, 167, 189) and non-dioxin like PCBs (PCB: 28, 52, 101, 138, 153, 180), as well as brominated diphenylethers (BDE: 17, 28, 47, 49, 66, 71, 77, 85, 99, 100, 119, 126, 138, 153, 154, 156, 183, 184, 191, 196, 197, 206, 207, 209), were analyzed by Eurofins GfA Lab Service GmbH (Hamburg) - a laboratory with an applied quality control system based on ISO 17025.

To determine POPs in biological material PBDE, PCDD/F and PCB standards were used. The material, supplied by AccuStandard, was over 99% pure (GC area $\geq 99.8\%$). Extraction solvents for use in chromatographic analysis: n-hexane, acetone, n-tetradecane with a minimum purity of 99% (GC area $\geq 99\%$), were supplied by MERCK. The procedure also involved the use of: sulfuric acid (min. 95%), ammonium acetate (pure per analysis), SPE columns with Thermo Scientific HyperSep type octadecyl-C18 cartridges, and rounded cellulose extraction thimbles from Whatman. Technical gases from Linde were also used: 99.995% pure helium, and 99.994% pure nitrogen.

The fundamental steps of analysis were as follows: addition of internal $^{13}\text{C}_{12}$ marked PCDD/F, PCB and PBDE standard substances, soxhlet extraction and extract clean-up by column chromatography, and finally quantification of the native PCDD/Fs, PCBs and PBDEs via the internal $^{13}\text{C}_{12}$ marked standards. PCDDFs were determined by means of high resolution mass spectrometry (HRGC/HRMS) and GC/MS-Triple Quad. PCBs and PBDEs were determined by means of high resolution mass spectrometry with high-resolution mass spectrometry (HRGC/LRMS) as has been previously described elsewhere (eg. Król et al., 2012; Toms et al., 2009; US EPA, 1994). LOQ for: PCDD/Fs was 0.1 pg WHO-TEQ g^{-1} lw, non-ortho PCBs 0.19–0.25 pg/g lw., mono-ortho PCBs 0.32–47 pg/g lw., non-dioxin like PCBs 0.15–0.21 pg/g lw. and for PBDEs 0.002–0.04 ng/g lw.

The toxic equivalency (TEQ) was determined using the toxic equivalency factor (TEF) defined by the WHO in 2005, and additionally using the factor (WHO TEF for birds) described by Van den Berg et al. (1998). In that latter model, the TEF factor for birds has the highest toxic equivalence for the following isomers: 2,3,7,8-TCDD; 1,2,3,7,8-PCDD; 2,3,7,8-TCDF and 2,3,4,7,8-PCDF.

3. Results

The average PBDE concentration found in herring gull eggs (198.31 $\mu\text{g/kg dw}$) was about two times higher than in tern eggs

Table 1

Average concentration (ng/g dw.) of polybrominated diphenyl ether (PBDE) congeners in aquatic birds' eggs.

PBDE congeners		Herring gull	Sandwich tern	Common tern
2,4,4'-Tri BDE	BDE-28	0.593	0.912	0.715
2,2',4'-Tri BDE	BDE-17	<0.0296	0.0135	<0.0102
2,2',4,4'-Tetra BDE	BDE-47	91.90	37.40	57.3
2,2',4,5'-Tetra BDE	BDE-49	1.11	2.10	1.70
2,3',4,4'-Tetra BDE	BDE-66	0.555	0.745	0.595
2,3',4',6'-Tetra BDE	BDE-71	<0.0592	<0.0102	<0.0102
3,3',4,4'-Tetra BDE	BDE-77	0.0739	0.147	0.097
2,2',3,4,4'-Penta BDE	BDE-85	0.288	<0.0128	<0.0128
2,2',4,4',5'-Penta BDE	BDE-99	27.90	8.01	17.20
2,2',4,4',6'-Penta BDE	BDE-100	26.30	14.70	18.90
2,3',4,4',6'-Penta BDE	BDE-119	0.866	1.71	1.34
3,3',4,4',5'-Penta Bde	BDE-126	0.163	0.0722	0.0945
2,2',3,4,4',5'-Hexa BDE	BDE-138	0.65	<0.0191	0.24
2,2',4,4',5,5'-Hexa BDE	BDE-153	14.80	4.19	8.20
2,2',4,4',5,6'-Hexa BDE	BDE-154	7.89	6.67	7.25
2,3,3',4,4',5'-Hexa BDE	BDE-156	<0.178	<0.0191	<0.0191
2,2',3',4,4',5',6'-Hepta BDE	BDE-183	5.44	0.291	0.102
2,2',3,4,4',6,6'-Hepta BDE	BDE-184	<0.296	<0.0319	<0.0319
2,3,3',4,4',5',6'-Hepta BDE	BDE-191	<0.296	<0.0319	<0.0319
2,2',3,4,4',5,5',6'-Octa BDE	BDE-196	0.87	<0.0638	<0.0638
2,2',3,3',4,4',6,6'-Octa BDE	BDE-197	3.21	<0.0638	<0.0638
2,2',3,3',4,4',5,5',6'-Nona BDE	BDE-206	<1.18	<0.128	<0.128
2,2',3,3',4,4',5,6,6'-Nona BDE	BDE-207	3.90	<0.128	<0.128
Deca BDE	BDE-209	11.80	<0.319	<0.319
Σ PBDE		198.31	76.96	113.73

(Table 1). In all eggs tetra-BDE, penta-BDE and hexa-BDE were dominant (Fig. 1). Higher brominated BDEs were only found in herring gull eggs.

All congeners of chlorinated dioxins and furans were found in the analyzed aquatic bird eggs (Table 2). The highest total concentrations of polychlorinated dibenzo-p-dioxins (PCDDs) were found in herring gull eggs (105.08 pg/g dw), which were also characterized by the lowest total concentration of pentachlorodibenzo-furan (PCDF) congeners (28.0 pg/g dw). The highest concentrations of PCDFs were found in sandwich tern eggs (95.16 pg/g dw), which had the highest PCDD/F concentration (144.11 pg/g dw) and were at the same time characterized by the highest toxicity determined using bird TEFs. PCDFs were found to constitute the greatest proportion of PCDD/Fs in the tern eggs, whilst in the herring gull eggs, highly chlorinated congeners (octachlorodibenzo-dioxin and heptachlorodibenzo-furan) were dominant (Fig. 2).

Dioxin-like PCBs were found in all of the bird eggs (Table 3) and mono-ortho congeners had the highest concentration. Of the non-dioxin-like PCBs, the higher chlorinated congeners were dominant in all of the analyzed eggs (Table 4).

4. Discussion

There is no comparative data on the presence of PBDEs in aquatic bird eggs as extensive as is presented in this work, but several studies have analyzed PBDEs in the eggs of Baltic birds. BDE-47, -99 and -203 were detected in guillemot eggs from Stora Karlsö analyzed between 1969 and 2003 (Jörundsdóttir et al., 2009; Lundstedt-Enkel et al., 2006; Sellström et al., 2003). From 1988 to 2008 the sum of 35 PBDE congeners in herring gull eggs from Heuwiese decreased markedly with the exception of BDE-209. Penta-BDE was found to be a major contaminant (Fliedner et al., 2012). This study indicated the highest abundances of Tri-BDE and Penta-BDE, whilst Deca-BDE was detected only in herring gull eggs from the Wloclawek reservoir.

The main PBDE congeners can be assigned to three technical products: PENTA-BDE where BDE-47, -99, -100, and -153 dominate; OCTA-BDE which contains mainly BDE-183 and varying amounts of up to ten other congeners including BDE-196, -197, -203, -207 and -209; technical DECA-BDE which consists of over 97% of BDE-209 (Fliedner et al., 2012). The results demonstrated that Penta-BDE has the highest concentration in all

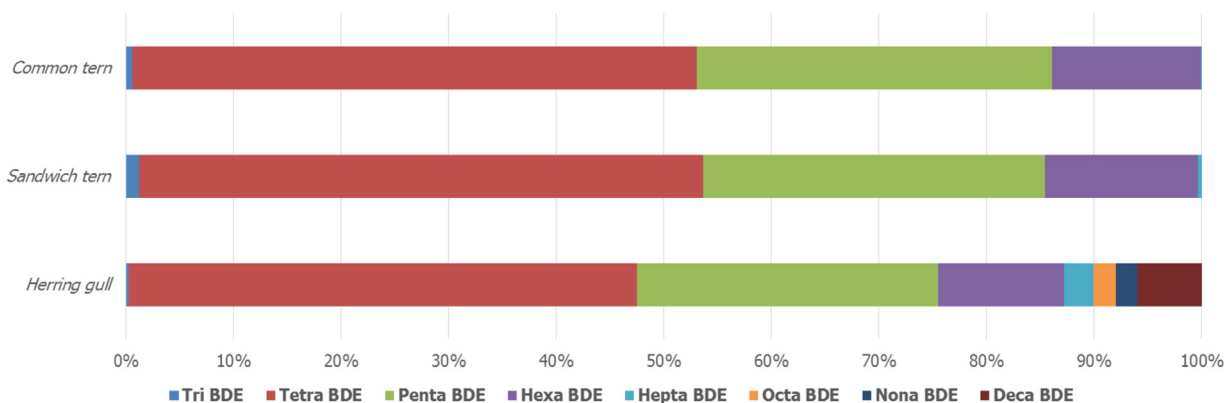


Fig. 1. PBDE contribution in the sum of analyzed PBDEs in aquatic birds' eggs.

Table 2
Average concentrations (pg/g dw.) of the main congeners of polychlorinated dibenzo-p-dioxin (PCDD) and polychlorinated dibenzo-furans (PCDFs) in aquatic birds' eggs.

Compounds	Herring gull	Sandwich tern	Common tern
Dry mass (%)	71%	67%	64%
Lipid content in dry mass (%)	10.3%	9.4%	8.9%
2,3,7,8-TCDD	4.89	6.21	5.24
1,2,3,7,8-PCDD	4.83	13.70	10.54
1,2,3,4,7,8-HxCDD	0.92	1.81	1.12
1,2,3,6,7,8-HxCDD	1.70	9.98	5.98
1,2,3,7,8,9-HxCDD	1.84	1.60	1.41
1,2,3,4,6,7,8-HpCDD	29.10	3.15	4.25
OctaCDD	49.80	12.50	15.80
ΣPCDD	105.08	48.95	44.34
2,3,7,8-TCDF	0.83	10.20	8.14
1,2,3,7,8-PCDF	<0.8	4.47	3.74
2,3,4,7,8-PCDF	6.17	61.80	42.84
1,2,3,4,7,8-HxCDF	5.64	1.81	1.47
1,2,3,6,7,8-HxCDF	7.36	9.98	8.10
1,2,3,7,8,9-HxCDF	<0.70	<1.3	<1.50
2,3,4,6,7,8-HxCDF	3.32	5.06	3.45
1,2,3,4,6,7,8-HpCDF	4.68	1.84	2.16
1,2,3,4,7,8,9-HpCDF	<0.69	<1.2	<2.50
OctaCDF	<5.80	<10.00	<7.80
ΣPCDF	28.00	95.16	69.90
ΣPCDDFs	133.08	144.11	114.24
TEQ WHO 2005	15.24	42.53	31.65
TEQ birds TEF	18.80	93.97	68.35

analyzed eggs, while BDE-209 which is the main compound of Deca-BDE was found only in eggs from the Włocławek reservoir. As contamination in a bird's eggs reflects the environmental condition of its habitat, it may suggest that the inland reservoir and surrounding area contribute to the higher deca-BDE observed in the birds while the coastal area with Penta-BDE. It might also indicate the differences between birds' food preferences and in the same way PBDE concentration in the prey.

Aquatic birds provide an early warning of pollution in their habitat. They are exposed to pollutants mainly through food but also via inhalation and others means of ingestion. It has been stated that pollutants that bio-accumulated in a bird's organs are selectively sequestered in the liver (Braune and Norstrom, 1989; Kubota et al., 2013). The lipids in bird egg yolk are primarily in the form of lipoproteins, which are synthesized in the liver and deposited in the ovarian follicles. Braune and Norstrom (1989) found that the transfer of PCDD/Fs to herring gull eggs decreased with increased chlorine content, thereby suggesting selective liver retention of those compounds.

The analyzed compounds were found in aquatic bird eggs

Table 3
Polychlorinated biphenyl (PCB) congeners – dioxin like compounds, average concentration (pg/g dw.) in aquatic birds' eggs.

Compounds	Herring gull	Sandwich tern	Common tern
non-ortho PCB			
PCB-77	439	<1200	657
PCB-81	<71	<260	<260
PCB-126	897	1870	951
PCB-169	<21	<780	<780
mono-ortho PCB			
PCB-105	68000	58100	41200
PCB-114	4290	2260	1970
PCB-118	242000	203000	178000
PCB-123	3590	3860	3010
PCB-156	60000	47900	57500
PCB-157	10600	9930	12050
PCB-167	25500	33900	21600
PCB-189	71440	9680	11570
TEQ (PCB birds TEF)	129.36	201.32	141.36

worldwide (Table 5), and the levels of chlorinated compounds in eggs from the southern part of the Baltic Sea and an inland reservoir (dam) on the river Vistula in region of the town Włocławek were similar to concentrations found in birds' eggs from Hong Kong (China) and the Ebro Delta (Spain). These represent areas of the highest anthropogenic activity, similar to the Baltic, confirming the importance of habitat pollution on the accumulation of POPs in wildlife. The situation was similar with contamination levels of PBDEs in Baltic bird eggs. However, higher occurrences were noted in the common eider than in the analyzed birds, which belong to a lower trophic level, demonstrating the significance of trophic position on bird egg contamination.

Nordl f et al. (2012) found 23478-PeCDF to be dominant in white-tailed sea eagle eggs from the South Bothnian Sea and Baltic Proper, and this congener is known to form in combustion processes (Rappe et al., 1987). This study showed the same congener to have the highest abundance in tern eggs, while in gull eggs more chlorinated dioxins (OCDD and HpCDD) were dominant. Such differentiation in dioxin and furan concentrations between tern and gull eggs, in spite of the physiological differences between the analyzed species, was most likely due to the environmental conditions of their habitats and the food consumed by females during the formation of eggs. Stable isotope studies ($\delta^{15}\text{N}$, $\delta^{13}\text{C}$) carried out on the same eggs by Grajewska et al. (2015) showed that the Herring gull occupied the lowest trophic position and this was associated with terrestrial food sources. The Common tern, which takes its food from a vast area and feeds both at sea and on land, occupied the highest trophic level. The Sandwich tern had a limited feeding

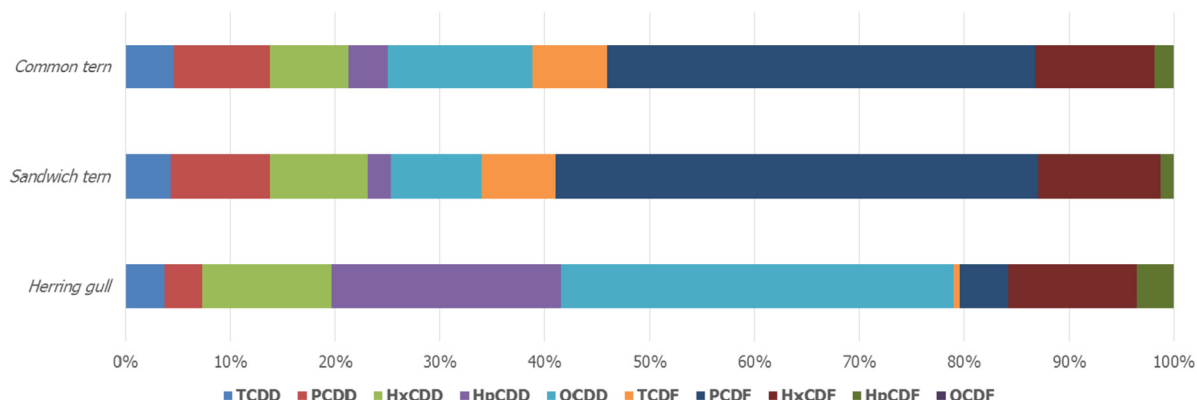


Fig. 2. Contribution of PCDDF congeners in the sum of analyzed PCDDFs in aquatic birds' eggs.

Table 4

Non-dioxin like PCB congeners average concentration (pg/g dw.) in aquatic birds' eggs.

Birds species egg	Non-dioxin like PCB congeners					
	28	52	101	138	153	180
Herring gull	13600	20500	37700	639000	997000	584000
Sandwich tern	<270	<270	85400	653000	1140000	621000
Common tern	10100	17800	69200	382000	785000	447000

area primarily associated with the marine environment.

Morales et al. (2012) found a completely different pattern of congeners in gulls from the Ebro Delta Natural Park (South

Catalonia) which is an area of intense agricultural activity. In eggs belonging to *Larus michahellis* and *Larus audouinii* from this region, the dominant congener was OCDD. This confirms the importance of bird eggs as a tool with which to assess pollution in the immediate environment and indicates the differences between marine and terrestrial habitat pollution. In the Baltic Sea region PCDD/Fs were found in guillemot eggs from Stora Karlsö and herring gull eggs from Heuwiese. The concentrations of PCDD/Fs in guillemot and herring gull eggs follow a decreasing trend. In gull eggs 23478-PeCDF was found to be the dominant congener, possibly indicating the dominance of seafood in the diet of females (Flüedner et al., 2012; Lundstedt-Enkel et al., 2006; Miller et al., 2014; Schröter-Kermani et al., 2005).

Table 5

PCDDFs, PCBs and PBDEs mean concentration in aquatic birds eggs worldwide.

Birds species	Location	Σ PCDDFs	Σ non-ortho PCB	Σ DL-PCBs	Σ PBDE	lipids [%]	Reference
<i>Larus argentatus</i> (n = 1)	Wloclawek Dam (Poland)	133.08 pg/g dw	1.34 ng/g dw	2533.8 ng/g dw	198.31 ng/g dw	10.3	this study
<i>Sterna hirundo</i> (n = 1)	Southern Baltic (Poland)	114.24 pg/g dw	1.61 ng/g dw	2763 ng/g dw	113.73 ng/g dw	8.9	this study
<i>Sterna sandvicensis</i> (n = 1)	Southern Baltic (Poland)	144.11 pg/g dw	1.87 ng/g dw	3471 ng/g dw	76.96 ng/g dw	9.4	this study
<i>Anhinga rufa</i> (n = 14)	South Africa (Parys)	—	—	—	17 ng/g lw	5.9	Polder et al., 2008
<i>Bubulcus ibis</i> (n = 9)	South Africa (Parys)	—	—	—	3.7 ng/g lw	5.8	Polder et al., 2008
<i>Bubulcus ibis</i> (n = 11)	South Africa (Barberspan)	—	—	—	2.3 ng/g lw	7.0	Polder et al., 2008
<i>Charadrius marginatus</i> (n = 1)	South Africa (Velddrif)	—	—	—	7.1 ng/g lw	11.0	Polder et al., 2008
<i>Egretta garzetta</i>	Hong Kong (China)	600 pg/g lw	5410 pg/g lw	—	—	—	Wang et al. (2012)
<i>Fulmarus glacialis</i> (n = 10)	Vestmannaeyjar Island (Iceland)	—	—	—	110 ng/g lw ^a	13	Jörundsdóttir et al. (2013)
<i>Larus argentatus</i>	Great Lakes (USA)	81.7 pg/g ww	—	—	—	10.1	Kannan et al. (2001)
<i>Larus argentatus</i>	Southern Baltic Sea (Germany)	—	—	9190 ng/g lw ^b	2021 ng/g lw ^b	—	Flüedner et al. (2012)
<i>Larus argentatus</i>	Southern Baltic Sea (Germany)	141 pg/g lw ^b	4.6 ng/g lw ^b	—	—	—	Schröter-Kermani et al. (2005)
<i>Larus argentatus</i> (n = 10)	Strömstad archipelago (Sweden)	—	—	—	8.3 ng/g ww	6	Carlsson et al. (2011)
<i>Larus audouinii</i> (n = 18)	Alboran Sea (North Africa)	2.1 pg/g ww	202 pg/g ww	—	9720 pg/g ww	8.30	Roscales et al., 2016
<i>Larus audouinii</i> (n = 3)	Elbro Delta and Medes Island (Spain)	141 pg/g dw	4.3 ng/g dw	—	—	—	Pastor et al. (1995)
<i>Larus audouinii</i>	Elbro Delta (Spain)	25.2 pg/g dw	1.1 ng/g dw	—	—	6.9	Morales et al. (2012)
<i>Larus audouinii</i> (n = 4)	Elbro Delta (Spain)	223 pg/g lw	4.8 ng/g lw	—	—	7.6	Morales et al. (2016)
<i>Larus crassirostris</i> (n = 10)	Hokkaido (Japan)	52.9 pg/g lw	4.91 ng/g lw	—	—	—	Choi et al. (2001)
<i>Larus dominicanus</i> (n = 1)	South Africa (Velddrif)	—	—	—	9.4 ng/g lw	10.0	Polder et al., 2008
<i>Larus fuscus</i> (n = 8)	Sandgerði (Iceland)	—	—	—	1200 ng/g lw	8.8	Jörundsdóttir et al. (2013)
<i>Larus marinus</i> (n = 9)	Sandgerði (Iceland)	—	—	—	830 ng/g lw	8.1	Jörundsdóttir et al. (2013)
<i>Larus michahellis</i> (n = 19)	Alboran Sea (North Africa)	2.47 pg/g ww	100 pg/g ww	—	6520 pg/g ww	7.78	Roscales et al., 2016
<i>Larus michahellis</i> (n = 3)	Elbro Delta and Medes Island (Spain)	79.1 pg/g dw	0.5 ng/g dw	—	—	—	Pastor et al. (1995)
<i>Larus michahellis</i>	Elbro Delta (Spain)	55.8 pg/g dw	1.9 ng/g dw	—	—	8.2	Morales et al. (2012)
<i>Larus michahellis</i> (n = 2)	Cabrera Archipelago (Spain)	49.1 pg/g lw	1.37 ng/g lw	—	—	7.4	Morales et al. (2016)
<i>Larus michahellis</i> (n = 2)	Chafarinas Island (Spain)	65.0 pg/g lw	1.04 ng/g lw	—	—	8.3	Morales et al. (2016)
<i>Larus michahellis</i> (n = 4)	Cies Island (Spain)	144.7 pg/g lw	0.62 ng/g lw	—	—	10.6	Morales et al. (2016)
<i>Larus michahellis</i> (n = 4)	Medes Island (Spain)	214.7 pg/g lw	1.09 ng/g lw	—	—	7.7	Morales et al. (2016)
<i>Larus michahellis</i> (n = 4)	Ebro Delta (Spain)	178 pg/g lw	2.8 ng/g lw	—	—	8.0	Morales et al. (2016)
<i>Nycticorax nycticorax</i>	Hong Kong (China)	1190 pg/g lw	2780 pg/g lw	—	—	—	Wang et al. (2012)
<i>Pagophila eburnea</i>	Seymour Island (Canada)	183 pg/g lw	4.9 ng/g lw	—	—	7.97	Braune et al. (2001)
<i>Pandion haliaetus</i>	Menorca (Spain)	7.36 pg/g ww	719 pg/g ww	—	—	—	Jiménez et al. (2007)
<i>Phalacrocorax gaimardi</i> (n = 3)	South Africa (Parys)	—	—	—	14 ng/g lw	4.3	Polder et al., 2008
<i>Somateria mollissima</i> (n = 10)	Strömstad archipelago (Sweden)	—	—	—	0.6 ng/g ww	18	Carlsson et al. (2011)
<i>Somateria mollissima</i> (n = 10)	Sandgerði (Iceland)	—	—	—	44 ng/g lw	18	Jörundsdóttir et al. (2013)
<i>Stercorarius skua</i> (n = 10)	Kvisker (Iceland)	—	—	—	2400 ng/g lw	8.8	Jörundsdóttir et al. (2013)
<i>Sterna paradisaea</i> (n = 6)	Sandgerði (Iceland)	—	—	—	110 ng/g lw	9.9	Jörundsdóttir et al. (2013)
<i>Sterna albifrons</i> (n = 17)	Southern Baltic Sea (Germany)	—	1.9 ng/g lw	1803.4 ng/g lw	—	—	Thyen et al. (2000)
<i>Tachybaptus ruficollis</i> (n = 1)	South Africa (Koppies)	—	—	—	19 ng/g lw	4.8	Polder et al., 2008
<i>Threskiornis aethiopicus</i> (n = 1)	South Africa (Parys)	—	—	—	396 ng/g lw ^b	5.0	Polder et al., 2008
<i>Uria aalge</i> (n = 30)	Northern Baltic Sea (Sweden)	—	—	8430 ng/g lw	77.4 ng/g lw	—	Lundstedt-Enkel et al. (2006)
<i>Uria aalge</i> (n = 10)	Vestmannaeyjar Island (Iceland)	—	—	—	61 ng/g lw ^a	13	Jörundsdóttir et al. (2013)
<i>Vanellus coronatus</i> (n = 1)	South Africa (Koppies)	—	—	—	120 ng/g lw	9.9	Polder et al., 2008

^a Sum of BDE-47; BDE-99; BDE-100; BDE-153; BDE-154.

^b Maximum values are given.

PCBs have been detected in bird eggs worldwide (Bustnes et al., 2015; Champoux and Boily, 2017; Peck et al., 2016) especially aquatic ones (Table 5). In the Baltic Sea region PCBs were found in the tissue of birds from the south coast including white-tailed sea eagles, cormorants and other birds wintering in Gdansk Bay (Falandysz and Szefer, 1984; Falandysz et al., 1994; 2000, 2002.; Kannan et al., 2003). Besides the burden in the birds' tissue, non-ortho PCBs (IUPAC No 77, 126, 169) were found in their eggs. White-tailed sea eagles from the Baltic Proper had a high concentration of PCB-126 (Nordl f et al., 2012), which stands in comparison to the results of the present study. *Uria aalge* eggs from Stora Karls  analyzed between 2000 and 2002 also indicated PCB burden (Lundstedt-Enkel et al., 2006). Little tern (*Sterna albifrons*) eggs from the east coast of Schleswig-Holstein analyzed between 1995 and 1996 (Thyen et al., 2000) and *Larus argentatus* eggs from Heuwise (Fliedner et al., 2012) were polluted by PCBs, and a decreasing trend was confirmed in both. The present study, however, indicates concentrations of PCBs in *Larus argentatus* eggs from the Wloclawek Dam which are almost three times higher than in the gulls from Heuwise.

The congener patterns of non-dioxin-like PCBs (herring gull: PCB-28<PCB-52<PCB-101<PCB-180<PCB-138<PCB-153; sandwich tern: PCB-101<PCB-180<PCB-138<PCB-153 and common tern: PCB-28<PCB-52<PCB-101< PCB-138<PCB-180<PCB-153) were similar to other study results and confirm the accumulation of higher chlorinated PCBs in bird eggs.

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References

- Aznar-Alemany,  . Sala, B., Pl n, S., Bouwman, H., Barcel , D., Eljarrat, E., 2019. Halogenated and organophosphorus flame retardants in cetaceans from the southwestern Indian Ocean. *Chemosphere* 226, 791–799.
- Baker, J.E., Kjellerup, B.V., 2016. Toxicological effects of polychlorinated biphenyls (PCBs) on freshwater turtles in the United States. *Chemosphere* 154, 148–154.
- Bignert, A., Danielsson, S., Faxneld, S., Nyberg, E., 2016. Comments Concerning the National Swedish Contaminant Monitoring Programme in Marine Biota, 2016.
- Braune, B.M., Norstrom, R.J., 1989. Dynamics of organochlorine compounds in herring gulls: III. Tissue distribution and bioaccumulation in Lake Ontario gulls. *Environ. Toxicol. Chem.* 8 (10), 957–968.
- Braune, B.M., Donaldson, G.M., Hobson, K.A., 2001. Contaminant residues in seabird eggs from the Canadian Arctic. Part I. Temporal trends 1975–1998. *Environ. Pollut.* 114 (1), 39–54.
- Bustnes, J.O., Bangjord, G., Yoccoz, N.G., 2015. Variation in concentrations of organochlorines and brominated flame retardants among eggs in abandoned clutches of a terrestrial raptor. *Chemosphere* 118, 357–360.
- Carlsson, P., Herzke, D., Wedborg, M., Gabrielsen, G.W., 2011. Environmental pollutants in the Swedish marine ecosystem, with special emphasis on polybrominated diphenyl ethers (PBDE). *Chemosphere* 82 (9), 1286–1292.
- Champoux, L., Boily, M., 2017. Temporal trends of mercury and organohalogen contaminants in great blue heron eggs from the St. Lawrence River, Qu bec, Canada, 1991–2011, and relationships with tracers of feeding ecology. *Sci. Total Environ.* 609, 1270–1285.
- Choi, J.W., Matsuda, M., Kawano, M., Wakimoto, T., Iseki, N., Masunaga, S., Hayama, S., Watanuki, Y., 2001. Chlorinated persistent organic pollutants in black-tailed gulls (*Larus crassirostris*) from Hokkaido, Japan. *Chemosphere* 44 (6), 1375–1382.
- Dabrowska, H., Kopko, O., Lehtonen, K.K., Lang, T., Waszak, I., Balode, M., Strode, E., 2017. An integrated assessment of pollution and biological effects in flounder, mussels and sediment in the southern Baltic Sea coastal area. *Environ. Sci. Pollut. Control Ser.* 24 (4), 3626–3639.
- De Wit, C.A., 2002. An overview of brominated flame retardants in the environment. *Chemosphere* 46 (5), 583–624.
- Desfor ges, J.P., Hall, A., McConnell, B., Ros ng-Asvid, A., Barber, J.L., Brownlow, A., et al., 2018. Predicting global killer whale population collapse from PCB pollution. *Science* 361 (6409), 1373–1376.
- Eljarrat, E., Aznar-Alemany,  ., Sala, B., Fr as,  ., Blanco, G., 2019. Decreasing but still high levels of halogenated flame retardants in wetland birds in central Spain. *Chemosphere* 228, 83–92.
- Falandysz, J., 1984. Metals and organochlorines in a female white-tailed eagle from Uznam Island, southwestern Baltic Sea. *Environ. Conserv.* 11 (3), 262–263.
- Falandysz, J., 1986. Metals and organochlorines in adult and immature males of white-tailed eagle. *Environ. Conserv.* 13 (1), 69–70.
- Falandysz, J., 1994. Polychlorinated biphenyl concentrations in cod-liver oil: evidence of a steady-state condition of these compounds in the Baltic area oils and levels noted in Atlantic oils. *Arch. Environ. Contam. Toxicol.* 27 (2), 266–271.
- Falandysz, J., Strandberg, B., 2004. Persistent organochlorine compounds in sludge and sediments from the gda nsk region, Baltic Sea. *Pol. J. Environ. Stud.* 13 (2).
- Falandysz, J., Szefer, P., 1984. Chlorinated hydrocarbons in fish-eating birds wintering in the Gda nsk Bay, 1981–82 and 1982–83. *Mar. Pollut. Bull.* 15 (8), 298–301.
- Falandysz, J., Yamashita, N., Tanabe, S., Tatsukawa, R., Ruci nska, L., Mizera, T., Jakuczun, B., 1994. Congener-specific analysis of polychlorinated biphenyls in white-tailed sea eagles *Haliaeetus albicilla* collected in Poland. *Arch. Environ. Contam. Toxicol.* 26 (1), 13–22.
- Falandysz, J., Strandberg, L., Mizera, T., Kalisi nska, E., 2000. The contamination of white-tailed sea eagles with organochlorines in Poland. *Rocz. Panstw. Zakl. Hig.* 51 (1), 7–13.
- Falandysz, J., Wyrzykowska, B., Puzyr, T., Strandberg, L., Rappe, C., 2002. Polychlorinated biphenyls (PCBs) and their congener-specific accumulation in edible fish from the Gulf of Gda nsk, Baltic Sea. *Food Addit. Contam.* 19 (8), 779–795.
- Falandysz, J., Smith, F., Panton, S., Fernandes, A.R., 2019. A retrospective investigation into the occurrence and human exposure to polychlorinated naphthalenes (PCNs), dibenzo-p-dioxins and furans (PCDD/fs) and PCBs through cod liver products (1972–2017). *Chemosphere* 231, 240–248.
- Falkowska, L., Reindl, A.R., Grajewska, A., Lewandowska, A.U., 2016. Organochlorine contaminants in the muscle, liver and brain of seabirds (*Larus*) from the coastal area of the Southern Baltic. *Ecotoxicol. Environ. Saf.* 133, 63–72.
- Fernandes, A.R., Mortimer, D., Holmes, M., Rose, M., Zhihua, L., Huang, X., et al., 2018. Occurrence and spatial distribution of chemical contaminants in edible fish species collected from UK and proximate marine waters. *Environ. Int.* 114, 219–230.
- Fernie, K.J., Shutt, J.L., Letcher, R.J., Ritchie, I.J., Bird, D.M., 2009. Environmentally relevant concentrations of DE-71 and HBCD alter eggshell thickness and reproductive success of American kestrels. *Environ. Sci. Technol.* 43 (6), 2124–2130.
- Fliedner, A., R del, H., J rling, H., M ller, J., Neugebauer, F., Schr ter-Kermani, C., 2012. Levels and trends of industrial chemicals (PCBs, PFCs, PBDEs) in archived herring gull eggs from German coastal regions. *Environ. Sci. Eur.* 24 (1), 7.
- Geyer, H.J., Rimkus, G.G., Scheunert, I., Kaune, A., Schramm, K.W., Kettrup, A., et al., 2000. Bioaccumulation and occurrence of endocrine-disrupting chemicals (EDCs), persistent organic pollutants (POPs), and other organic compounds in fish and other organisms including humans. In: *Bioaccumulation—New Aspects and Developments*. Springer, Berlin, Heidelberg, pp. 1–166.
- Grajewska, A., Falkowska, L., Szumilo-Pilarska, E., Hajdrych, J., Szubska, M., Fr czek, T., Brauze, T., 2015. Mercury in the eggs of aquatic birds from the Gulf of Gdansk and Wloclawek dam (Poland). *Environ. Sci. Pollut. Res.* 22 (13), 9889–9898.
- Grove, R.A., Henny, C.J., Kaiser, J.L., 2009. Osprey: worldwide sentinel species for assessing and monitoring environmental contamination in rivers, lakes, reservoirs, and estuaries. *J. Toxicol. Environ. Health Part B* 12 (1), 25–44.
- Hanari, N., Kannan, K., Miyake, Y., Okazawa, T., Kodavanti, P.R.S., Aldous, K.M., Yamashita, N., 2006. Occurrence of polybrominated biphenyls, polybrominated dibenzo-p-dioxins, and polybrominated dibenzofurans as impurities in commercial polybrominated diphenyl ether mixtures. *Environ. Sci. Technol.* 40 (14), 4400–4405.
- Hites, R.A., 2011. Dioxins: an overview and history. *Environ. Sci. Technol.* 45, 16–20.
- Jim nez, B., Merino, R., Abad, E., Rivera, J., Olie, K., 2007. Evaluation of organochlorine compounds (PCDDs, PCDFs, PCBs and DDTs) in two raptor species inhabiting a Mediterranean island in Spain. *Environ. Sci. Pollut. Res.* 14 (1), 61–68.
- J runds ttir, H., Bignert, A., Svavarsson, J., Nyg rd, T., Weihe, P., Bergman,  ., 2009. Assessment of emerging and traditional halogenated contaminants in guillemot (*uria aalge*) egg from north-western europe and the Baltic Sea. *Sci. Total Environ.* 407 (13), 4174–4183.
- J runds ttir, H., L fstrand, K., Svavarsson, J., Bignert, A., Bergman,  ., 2013. Polybrominated diphenyl ethers (PBDEs) and hexabromocyclododecane (HBCD) in seven different marine bird species from Iceland. *Chemosphere* 93 (8), 1526–1532.
- Kannan, K., Hilscherova, K., Imagawa, T., Yamashita, N., Williams, L.L., Giesy, J.P., 2001. Polychlorinated naphthalenes, -biphenyls, -dibenzo-p-dioxins, and -dibenzofurans in double-crested cormorants and herring gulls from Michigan waters of the Great Lakes. *Environ. Sci. Technol.* 35 (3), 441–447.
- Kannan, K., Kumar, K.S., Nakata, H., Falandysz, J., Oehme, G., Masunaga, S., 2003. Polychlorinated biphenyls, dibenzo-p-dioxins, dibenzofurans, and p, p'-DDE in livers of white-tailed sea eagles from eastern Germany, 1979–1998. *Environ. Sci. Technol.* 37 (7), 1249–1255.
- Karl, H., Ruoff, U., 2007. Dioxins, dioxin-like PCBs and chloroorganic contaminants in herring, *Clupea harengus*, from different fishing grounds of the Baltic Sea.

- Chemosphere* 67 (9), S90–S95.
- Knutsen, H.K., Kvalem, H.E., Thomsen, C., Frøshaug, M., Haugen, M., Becher, G., et al., 2008. Dietary exposure to brominated flame retardants correlates with male blood levels in a selected group of Norwegians with a wide range of seafood consumption. *Mol. Nutr. Food Res.* 52 (2), 217–227.
- Król, S., Zabiegała, B., Namieśnik, J., 2012. PBDEs in environmental samples: sampling and analysis. *Talanta* 93, 1–17.
- Kubota, A., Yoneda, K., Tanabe, S., Iwata, H., 2013. Sex differences in the accumulation of chlorinated dioxins in the cormorant (*Phalacrocorax carbo*): implication of hepatic sequestration in the maternal transfer. *Environ. Pollut.* 178, 300–305.
- La Guardia, M.J., Hale, R.C., Harvey, E., 2006. Detailed polybrominated diphenyl ether (PBDE) congener composition of the widely used penta-, octa-, and deca-PBDE technical flame-retardant mixtures. *Environ. Sci. Technol.* 40 (20), 6247–6254.
- Lundstedt-Enkel, K., Asplund, L., Nylund, K., Bignert, A., Tysklind, M., Olsson, M., Öberg, J., 2006. Multivariate data analysis of organochlorines and brominated flame retardants in Baltic Sea guillemot (*Uria aalge*) egg and muscle. *Chemosphere* 65 (9), 1591–1599.
- Martinson, S.C., Bird, D.M., Shutt, J.L., Letcher, R.J., Ritchie, I.J., Fernie, K.J., 2010. Multi-generational effects of polybrominated diphenylethers exposure: embryonic exposure of male American kestrels (*Falco sparverius*) to DE-71 alters reproductive success and behaviors. *Environ. Toxicol. Chem.* 29 (8), 1740–1747.
- Miller, A., Nyberg, E., Danielsson, S., Faxneld, S., Haglund, P., Bignert, A., 2014. Comparing temporal trends of organochlorines in guillemot eggs and Baltic herring: advantages and disadvantage for selecting sentinel species for environmental monitoring. *Mar. Environ. Res.* 100, 38–47.
- Morales, L., Martrat, M.G., Olmos, J., Parera, J., Vicente, J., Bertolero, A., Abalos, M., Lacorte, S., Santos, F.J., Abad, E., 2012. Persistent organic pollutants in gull eggs of two species (*Larus michahellis* and *Larus audouinii*) from the Ebro delta Natural Park. *Chemosphere* 88 (11), 1306–1316.
- Morales, L., Martrat, M.G.R., Parera, J., Bertolero, A., Ábalos, M., Santos, F.J., Lacorte, S., Abad, E., 2016. Dioxins and dl-PCBs in gull eggs from Spanish Natural Parks (2010–2013). *Sci. Total Environ.* 550, 114–122.
- Nordlöv, U., Helander, B., Zebühr, Y., Bignert, A., Asplund, L., 2012. Polychlorinated dibenzo-p-dioxins, polychlorinated dibenzofurans and non-ortho-PCBs in eggs of white-tailed sea eagles collected along the Swedish coast in the Baltic Sea. *Sci. Total Environ.* 438, 166–173.
- Pastor, D., Ruiz, X., Barceló, D., Albaigés, J., 1995. Dioxins, furans and AHH-active PCB congeners in eggs of two gull species from the western Mediterranean. *Chemosphere* 31 (6), 3397–3411.
- Peck, L.E., Gilchrist, H.G., Mallory, C.D., Braune, B.M., Mallory, M.L., 2016. Persistent organic pollutant and mercury concentrations in eggs of ground-nesting marine birds in the Canadian high Arctic. *Sci. Total Environ.* 556, 80–88.
- Polder, A., Venter, B., Skaare, J.U., Bouwman, H., 2008. Polybrominated diphenyl ethers and HBCD in bird eggs of South Africa. *Chemosphere* 73 (2), 148–154.
- Polder, A., Müller, M.B., Brynildsrud, O.B., De Boer, J., Hamers, T., Kamstra, J.H., Sandvik, M., 2016. Dioxins, PCBs, chlorinated pesticides and brominated flame retardants in free-range chicken eggs from peri-urban areas in Arusha, Tanzania: levels and implications for human health. *Sci. Total Environ.* 551, 656–667.
- Quinn, L.P., Roos, C., Pieters, R., Løken, K., Polder, A., Skaare, J.U., Bouwman, H., 2013. Levels of PCBs in wild bird eggs: considering toxicity through enzyme induction potential and molecular structure. *Chemosphere* 90 (3), 1109–1116.
- Rappe, C., Andersson, R., Bergqvist, P.A., Brohede, C., Hansson, M., Kjeller, L.O., Tysklind, M., 1987. Overview on environmental fate of chlorinated dioxins and dibenzofurans. Sources, levels and isomeric pattern in various matrices. *Chemosphere* 16 (8–9), 1603–1618.
- Rice, C.P., O'Keefe, P., Kubiak, T., 2003. Sources, pathways, and effects of PCBs, dioxins, and Dibenzofurans. In: Hoffman, D.J., Rattner, B.A., Burton Jr., G.A., Cairns Jr., J. (Eds.), *Handbook of Ecotoxicology*. Lewis Publishers, London, New York, pp. 501–573.
- Rigét, F., Vorkamp, K., Bossi, R., Sonne, C., Letcher, R.J., Dietz, R., 2016. Twenty years of monitoring of persistent organic pollutants in Greenland biota. A review. *Environ. Pollut.* 217, 114–123.
- Roscales, J.L., Vicente, A., Muñoz-Arnanz, J., Morales, L., Abad, E., Aguirre, J.I., Jiménez, B., 2016. Influence of trophic ecology on the accumulation of dioxins and furans (PCDD/Fs), non-ortho polychlorinated biphenyls (PCBs), and polybrominated diphenyl ethers (PBDEs) in Mediterranean gulls (*Larus michahellis* and *L. audouinii*): a three-isotope approach. *Environ. Pollut.* 212, 307–315.
- Schröter-Kermani, C., Schmolz, E., Herrmann, T., Päpke, O., 2005. PCDD, PCDF, and Dioxin-like PCB in herring gull eggs from the North Sea and Baltic Sea: levels, patterns and temporal trends. *Organohalogen Compd.* 67, 1295–1299. Sellström et al., 2003.
- Sellström, U., Bignert, A., Kierkegaard, A., Häggberg, L., De Wit, C.A., Olsson, M., Jansson, B., 2003. Temporal trend studies on tetra- and pentabrominated diphenyl ethers and hexabromocyclododecane in guillemot egg from the Baltic Sea. *Environ. Sci. Technol.* 37 (24), 5496–5501.
- Suzuki, K., Nakai, K., Sugawara, T., Nakamura, T., Ohba, T., Shimada, M., Murata, K., 2010. Neurobehavioral effects of prenatal exposure to methylmercury and PCBs, and seafood intake: neonatal behavioral assessment scale results of Tohoku study of child development. *Environ. Res.* 110 (7), 699–704.
- Szlinder-Richert, J., Barska, I., Usydus, Z., Ruczyńska, W., Grabic, R., 2009. Investigation of PCDD/Fs and dl-PCBs in fish from the southern Baltic Sea during the 2002–2006 period. *Chemosphere* 74 (11), 1509–1515.
- Szlinder-Richert, J., Usydus, Z., Drgas, A., 2012. Persistent organic pollutants in sediment from the southern Baltic: risk assessment. *J. Environ. Monit.* 14 (8), 2100–2107.
- Tard, A., Gallotti, S., Leblanc, J.C., Volatier, J.L., 2007. Dioxins, furans and dioxin-like PCBs: occurrence in food and dietary intake in France. *Food Addit. Contam.* 24 (9), 1007–1017.
- Thyen, S., Becker, P.H., Behmann, H., 2000. Organochlorine and mercury contamination of little terns (*Sterna albifrons*) breeding at the western Baltic Sea, 1978–96. *Environ. Pollut.* 108 (2), 225–238.
- Toms, L.M.L., Hearn, L., Kennedy, K., Harden, F., Bartkow, M., Temme, C., Mueller, J.F., 2009. Concentrations of polybrominated diphenyl ethers (PBDEs) in matched samples of human milk, dust and indoor air. *Environ. Int.* 35 (6), 864–869.
- Toschik, P.C., Rattner, B.A., McGowan, P.C., Christman, M.C., Carter, D.B., Hale, R.C., et al., 2005. Effects of contaminant exposure on reproductive success of ospreys (*Pandion haliaetus*) nesting in Delaware River and Bay, USA. *Environ. Toxicol. Chem.* 24 (3), 617–628.
- US EPA Method 1613, 1994. Tetra-Through Octa-Chlorinated Dioxins and Furans by Isotope Dilution HRGC/HRMS. Washington, DC.
- Usydus, Z., Szlinder-Richert, J., Polak-Juszczak, L., Komar, K., Adamczyk, M., Malesa-Cieciwierz, M., Ruczyńska, W., 2009. Fish products available in Polish market—Assessment of the nutritive value and human exposure to dioxins and other contaminants. *Chemosphere* 74 (11), 1420–1428.
- Van den Berg, M., Birnbaum, L., Bosveld, A.T., Brunström, B., Cook, P., Feeley, M., et al., 1998. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for humans and wildlife. *Environ. Health Perspect.* 106 (12), 775–792.
- Wan, Y., Hu, J., Yang, M., An, L., An, W., Jin, X., et al., 2005. Characterization of trophic transfer for polychlorinated dibenzo-p-dioxins, dibenzofurans, non- and mono-ortho polychlorinated biphenyls in the marine food web of Bohai Bay, North China. *Environ. Sci. Technol.* 39 (8), 2417–2425.
- Wang, Y., Lam, J.C., So, M.K., Yeung, L.W., Cai, Z., Hung, C.L., Lam, P.K., 2012. Polychlorinated dibenzo-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), dioxin-like polychlorinated biphenyls (PCBs) and polybrominated diphenyl ethers (PBDEs) in waterbird eggs of Hong Kong, China. *Chemosphere* 86 (3), 242–247.
- Waszak, I., Dabrowska, H., Góra, A., 2012. Bioaccumulation of polybrominated diphenyl ethers (PBDEs) in flounder (*Platichthys flesus*) in the southern Baltic Sea. *Mar. Environ. Res.* 79, 132–141.
- Wohlfahrt-Weje, C., Audouze, K., Brunak, S., Antignac, J.P., le Bizet, B., Juul, A., Main, K.M., 2014. Polychlorinated dibenzo-p-dioxins, furans, and biphenyls (PCDDs/PCDFs and PCBs) in breast milk and early childhood growth and IGF1. *Reproduction* 147 (4), 391–399.
- Zicus, M.C., Rave, D.P., 2004. DDE, PCB, and mercury residues in Minnesota common goldeneye and hooded merganser eggs: a followup. *Summaries of Wildlife Research Findings* 129.