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Organochlorine contaminants in the muscle, liver and brain of seabirds (*Larus*) from the coastal area of the Southern Baltic

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ABSTRACT

The presence of persistent organic pollutants in the environment manifests itself most strongly in the marine trophic chain, where the highest link is comprised of seabirds. At the same time, seabirds are excellent indicators of contamination in their habitat. The present study concentrates on toxic substances: polychlorinated dibenzo-p-dioxin (PCDDs), polychlorinated dibenzofurans (PCDFs) and chlorinated organic pesticides (OCPs) accumulated in the livers, pectoral muscles and brains of dead gulls collected along the Polish coast of the Baltic Sea in the years 2010–12. The highest toxic equivalence was determined in the livers of *Larus argentatus* ($TEQ_{(birds\ TEF)}$ – 28.3 pg g^{-1} ww) and *Larus marinus* ($TEQ_{(birds\ TEF)}$ – 29.9 pg g^{-1} ww.). However, the toxic equivalence of muscles was lower and amounted to 3.9 pg g^{-1} ww. and 7.8 pg g^{-1} ww. respectively for the two species. The lowest toxic equivalence was found in the brains of birds, where only one, the most toxic, 2,3,7,8 TCDD congener was found ($TEQ_{(birds\ TEF)}$ 0.87 pg g^{-1} ww). The highest concentration of chloroorganic pesticides was determined in the brains of the birds (total OCP 167.8 pg g^{-1} ww.), lower concentrations were found in the livers (total OCP 92.1 pg g^{-1} ww.) and muscles (total OCP 43.1 pg g^{-1} ww.). With regard to pesticides, the highest proportion in the total OCP content was constituted by DDT and its isomers (liver 81%, muscles 77% and brain 55%).

High concentrations of the studied pollutants in the livers of gulls found dead on the coast of the Southern Baltic could have been effected by levels of contamination in the birds' last meals, which resulted in a seven-fold increase of the liver's toxic equivalence and a two-fold increase in OCP concentration in relation to muscles.

1. Introduction

Seabirds, being an integral part of aquatic ecosystems, have become sensitive biomonitors of the changes occurring there under the influence of both natural and anthropogenic factors. It is in birds that we can find ecological responses to changes in environmental conditions and the productivity of the basin (Buckman et al., 2004; Burger and Gochfeld, 2004; Carere et al., 2010). The condition and reproductive success of seabirds are shaped both in breeding areas and in remote places where they live outside the breeding season. This is why they can be used as proxies to assess the impact of many variables affecting their environment in temporal and spacial terms (Vander Pol and Becker, 2007; Mallory et al., 2010).

Fish – consumed by mammals, seabirds and humans – are recognized bioindicators of sea pollution. The marine diet of birds, although valuable in terms of protein, vitamin and omega 3 acid content, is burdened with dangerous endocrine active compounds (Staniszewska et al., 2014; Reindl et al., 2013; Miller et al., 2013). This group includes, among others, organochlorine compounds purposefully produced by humans (pesticides, fungicides) and those formed as by-products of herbicide and fungicide production, as well as products of incomplete combustion of debris or wood (polychlorinated dibenzo-p-dioxins and polychlorinated dibenzo-furans – PCDDFs) (Armitagea et al., 2009). The main ways in which organochlorine compounds reach the sea are atmospheric transportation, deposition and surface run-off (Bartnicki et al., 2011; Bojakowska and Gliwicz, 2005; Miller et al., 2013; Niemirycz, 2008; Sellström et al., 2009; Shelepchikov et al., 2008).

Constant exposure of fish to PCDD/Fs and other endocrine active substances leads to the accumulation and biomagnification of such compounds on the next link in the food chain (OSPAR, 2007). Since the middle of the last century, an increase in PCDD/F concentrations in Baltic fish has been observed (Isosaari et al., 2006; Kiviranta et al., 2003; Shelepchikov et al., 2008). In the last decade of the 20th century the concentrations of these pollutants

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stabilized (Karl et al., 2010), and in some parts of the Baltic Sea (Bothnian Bay, Southern Baltic Proper), there was even a distinctive downward trend in PCDD/Fs in herring (Miller et al., 2013). The drop in toxic equivalence factor in herring did not occur on the southern coast of Sweden, while there was a clear downward trend discovered in the eggs of pescivorous guillemots (*Uria aalge*). This is testified to by the studies by Miller et al. (2014), which have been conducted over the last 10 years. The successive downward tendencies in chlorinated organic pesticides (OCP) in various elements of the Southern Baltic environment are confirmed by a series of earlier studies spanning the last fifteen years (Falandysz et al., 1999, 2001; Sapota, 2006; HELCOM, 2010; Reindl et al., 2013; Falkowska et al., 2013; Reindl et al., 2015). This is directly related to the cessation of OCP usage in agrotechnology. Nonetheless these organochlorine pollutants are still detectable in the environment, which suggests that marine organisms are chronically exposed to chlorinated hydrocarbons.

As they consume food from various trophic levels at sea and on land, seabirds make it possible to assess the contamination of the coastal zone environment more broadly. They are good indicators of pollution in a given area, and at the same time allow us to evaluate the health conditions of habitats and feeding areas (Furnes and Camphuysen, 1997; Braune et al., 2005). Spacial and temporal changes in the status of environmental pollution of the coast and sea result in, for example, changes in bird colony population, breeding success and survival. The Baltic Seabirds Project (www.balticseabird.se), conducted in the Gottland Basin area in the years 1997–2013, broadened the knowledge of seabird ecology, even though it did not include chemical analyses. The observation results pointed to a drop in the breeding success of the Common Murre (*Uria aalge*) and the successively decreasing weight of hatched chicks as well as a drop in the number of breeding pairs of the Herring Gull (*Larus argentatus*) and the Lesser Black-backed Gull (*Larus fuscus)*. These observations testify to the need for studies into seabirds and their habitats burdened with life-threatening substances. The research carried out so far in various parts of the world has indicated a process of internal distribution of xenobiotics which, beside tissues and organs (Debacker et al., 2003; Braune et al., 2005; Wan et al., 2006; Rattner and McGowan, 2007; Kubota et al., 2013; Ferreira and Wermelinger, 2013) in birds also includes feathers and eggs (Choi et al., 2001; HELCOM, 2010; Corsolini et al., 2011; Morales et al., 2012; Nordlof et al., 2012; Falkowska et al., 2013; Miller et al., 2014).

Studies which have been carried out for many years point to the presence of xenobiotics in the fish of the Southern Baltic (e.g. Falandysz, 1999, Falandysz et al., 2001; Sapota, 2006; Szlinder-Richert et al., 2011; Reindl et al., 2013; Staniszewska et al., 2014). However, there are few publications on POPs in birds that feed on the fish in this region of the Baltic (Falkowska et al., 2013; Reindl and Falkowska, 2014; Staniszewska et al., 2014; Falkowska and Reindl, 2015; Reindl et al., 2015). These concern mainly the bioaccumulation and biomagnification of chlorinated organic pesticides, fungicides, herbicides and phenol derivatives. The study results presented here are the first to inform on dibenzodioxins, dibenzo-furans and chlorinated organic pesticides found in the muscles, livers and brains of gulls inhabiting (permanently or seasonally) the coast of the Southern Baltic. These birds, as they obtain their food from various trophic levels, are contamination monitors of the environments of the Southern Baltic coastal zone, as well as North-Eastern and Central Europe which are covered by gull migration. These birds can therefore help us to assess the condition and healthiness of a large area surrounding the Baltic Sea. Moreover, the results of studies on seabirds can be useful for humans who, just like birds, obtain some of their food from the sea.

2. Materials and methods

2.1. Study species

The Herring Gull (*Larus argentatus*) is widely distributed in the North of Europe. These birds inhabit seaside areas, islands and sand bars in river estuaries, as well as the vicinity of in-land water basins. The gulls breeding on the Polish coast, particularly mature specimens, are mainly sedentary. Young birds are also partly sedentary, but some of them migrate to the west (Germany being the furthest destination). In winter, herring gulls also arrive to the Polish coast from the East of Scandinavia and the West of Russia. The Herring Gull is one of the largest birds and most numerous species found on the Polish coast in winter (Meissner and Betleja, 2007), and feeds on organisms from various trophic levels with a diet including: fish, invertebrae, crustaceans, small amphibians, eggs and chicks of other gulls, as well as carrion and communal waste. The latter, supplementary source of food can be found by the birds in landfills located close to the so-called „Tricity" agglommeration of Gdansk, Sopot and Gdynia. The total number of gulls found at such sites can range from a few to over 30,000 specimens.

The migratory Great Black-backed Gull (*Larus marinus*) is the largest gull, and does not breed on the Southern Baltic but arrives from Scandinavia (Sweden, Finland) and from Western Russia to winter on the Polish coast. This gull is placed at a higher trophic level than the Herring Gull, as it feeds mainly on fish, but a few percent of birds seen at communal landfills will nevertheless be of this species (Meissner et al., 2007). In Poland, birds of the *Laridae* family are a protected species. At the age of 4 years the birds are ready to procreate, and the breeding period lasts from the middle of April until the end of June (Cramp and Simmons, 1983).

2.2. Sampling area

These studies were conducted on dead herring gulls (*Larus argentatus*) and great black-backed gulls (*Larus marinus*) with the permission of the General Director of Environmental Protection (permit no. DOPozg1z-4200/II1–169/2015″/10/km) and the Regional Director of Environmental Protection in Gdansk (permit no. RDOŚ – 22-PN. II-6631–4-42/2010/ek) (Table 1).

The dead herring gulls were collected between February 2010 and March 2012 along the Southern Baltic coast: in winter close to the fishing port in Wladyslawowo, ($\varphi=54°47'$, $\lambda=18°25'$), and during the breeding period in Gdynia Harbour ($\varphi=54°30'$, $\lambda=18°33'$) and on the beach in Gdansk ($\varphi=54°22'$, $\lambda=18°33'$). After the breeding season, some dead herring gulls were found in the "Mewia Lacha" nature reserve, situated in the Vistula Estuary, and in a small fishing port in Swibno ($\varphi=54°21'$, $\lambda=18°57'$). In winter 2012, the largest gull – *Larus marinus* – was found at Gdynia Harbour. The cause of death was not analyzed for any of the birds, but the condition of the dead seabirds was assessed in analogy with Daoust et al. (1998) and Debacker et al. (2003). The body condition and cachectic status of the dead specimens was assessed during autopsy on the basis of weight of body and pectoral muscle, and also on the abundance of subcutaneous and internal fatty tissue (e.g. around the heart). Two mature specimens and one immature specimen were found to be in a cachectic condition (Table 1).

2.3. Samples

Dioxins, furans and chlorinated organic pesticides were assayed in gull livers and muscles as listed in Table 1. Additional PCDD/Fs assay was carried out in one combined brain sample, composed of material from immature (n=2) and mature specimens (n=3).