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Michielsen, R.J.; Shamoun-Baranes, J.; Parsons, J.R.; Kraak, M.H.S.

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Rosanne J. Michielsen, Judy Shamoun-Baranes, John R. Parsons, and Michiel H.S. Kraak

Contents

Abbreviations

DDE	1,1′-(2,2-Dichloro-1,1-ethenediyl)bis(4-chlorobenzene)
DDT	1,1′-(2,2,2-Trichloro-1,1-ethanediyl)bis(4-chlorobenzene)
DecaBDE	Decabromodiphenyl ether
OCP	Organochlorine pesticide
PBDE	Polybrominated diphenyl ether
PCB 118	2,3′,4,4′,5-Pentachlorobiphenyl
PCB 153	2,2′,4,4′,5,5′-Hexachlorobiphenyl
PCB 52	2,2′,5,5′-Tetrachlorobiphenyl

R. J. Michielsen (✉) · J. Shamoun-Baranes · J. R. Parsons · M. H. S. Kraak
Institute for Biodiversity and Ecosystem Dynamics, University of Amsterdam, Amsterdam, The Netherlands
e-mail: J.Z.Shamoun-Baranes@uva.nl; J.R.Parsons@uva.nl; M.H.S.Kraak@uva.nl

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PCB Polychlorinated biphenyl
POP Persistent organic pollutant

1 Introduction

Persistent organic pollutants (POPs) are highly bioaccumulative chemicals that are present in almost all environments, despite the ban on the production of most of these substances (Stockholm Convention 2009, 2011, 2013, 2015). Animals that inhabit contaminated environments may contain high concentrations of POPs due to bioaccumulation and biomagnification within a food web, which might lead to an array of adverse effects such as the disruption of their endocrine homeostasis (Gould et al. 1999; MacKay and Fraser 2000; Borgå et al. 2004; Fernie et al. 2005). Recently, Jamieson et al. (2017) reported high concentrations of POPs in arthropods living on the bottom of the Mariana trench, highlighting the global extent of the pollution by these chemicals. Particularly high concentrations of these substances were detected in leachate and dust from landfills (Hansen et al. 1997; Öman and Junestedt 2008; Li et al. 2012, 2014; Melnyk et al. 2015). Hence, animals that inhabit or regularly visit contaminated landfills or other contaminated areas might be exposed to high POP concentrations (Gould et al. 1999; Fernie et al. 2005; Técher et al. 2016). It is therefore alarming that several bird species increasingly forage on landfills and waste treatment areas and have subsequently altered their foraging and even migration behavior. Studies from Canada, Western and Central Europe, and Asia report that landfills are utilized by raptors, gulls (*Larus* sp.) corvids, and white storks (*Ciconia ciconia*) (Baxter and Allan 2006; Elliott et al. 2006; Kruszyk and Ciach 2010; de la Casa-Resino et al. 2014; Patenaude-Monette et al. 2014; Fazari and Mcgrady 2016; Tauler-Ametller et al. 2017). In Western and Central Europe, the concern is increasing that due to the overabundance of anthropogenic food provided by landfills, white storks are short stopping their migration (Blanco 1996; Massemin-Challet et al. 2006; Kruszyk and Ciach 2010; de la Casa-Resino et al. 2014). Similarly, the accessibility of landfills influences the distribution of gull species in Europe (Sol et al. 1995; Arizaga et al. 2014). The harmful effect of POP contamination on the reproductive success of species that live or forage in contaminated areas has been reported for several bird species, like tree swallows (*Tachycineta bicolor*), ring-billed gulls (*Larus delawarensis*), and European starlings (*Sturnus vulgaris*) (Halbrook and Arenal 2003; Gilchrist et al. 2014; Técher et al. 2016). Thus, POPs are very widespread but heterogeneous in their distribution. Hence, in order to take effective measures to mitigate the effects on bird populations, it is important to identify the main sources of POP contamination in bird populations.

Birds have long been suggested to function as suitable monitors of environmental pollutants although drawbacks to using certain species and ethical objections have also been noted (Furness 1993, 1997). Gulls are known for decades to

opportunistically utilize anthropogenic resources (Bosch et al. 1994; Belant et al. 1998; Duhem et al. 2003; Christel et al. 2012; Caron-Beaudoin et al. 2013; Scott et al. 2014; van Donk et al. 2017). This behavior has led this species group to be involved in many types of human-wildlife conflicts, like collisions with aircrafts and wind turbines, changing EU policies regarding fishery discards, and landfills and nuisance due to increased urban gull populations (Belant et al. 1993; Dolbeer et al. 1993; Sol et al. 1995; Belant 1997; Garthe and Hüppop 2004; Hüppop et al. 2006; Soldatini et al. 2008; Bernhardt et al. 2010; Bicknell et al. 2013; Abdennadher et al. 2014; Arizaga et al. 2014; Tyson et al. 2015; Sommerfeld et al. 2016). Simultaneously, the successful adaptation to human activities could have adverse effects on gull populations, by enhancing their exposure to harmful substances like POPs (Técher et al. 2016). Many gull populations have been declining during recent years, and this may in part be attributed to the adverse effects of POP contamination (Hario and Rintala 2016; Poprach et al. 2016; Técher et al. 2016). The close connection of gull populations with human activities and the associated exposure to POP contamination makes them suitable species to study regarding the effects of POP contamination in foraging habitats on omnivorous seabirds.

In order to assess the effects of POP contamination on the functioning of gull populations, it is important to pinpoint the different sources of contamination. Since gulls are highly opportunistic and versatile foragers and individuals specialize in certain foraging tactics, individual gulls of the same colony could visit very different foraging habitats, ranging from landfills to the open sea (Camphuysen et al. 2015; Tyson et al. 2015; van Donk et al. 2017). Hence, the source of POP contamination, and thus the degree and nature of the exposure to POPs, is expected to vary greatly between individual gulls. Therefore, to clarify the effect of different POP-contaminated areas on gull populations, it is important to also identify the source of contamination in individual gulls.

Conventional sampling methods applied when studying POP contamination, such as taking liver or fat samples, are destructive and ethically undesired. A less destructive method could be the use of feathers, as it is likely that POPs are deposited in and onto feathers, through, for example, preen oil, blood, or contaminated dust. In fact, feather sampling has been applied for decades to assess the exposure to heavy metals and POPs (Goede and De Voogt 1985; Abbasi et al. 2015). Thus, analyzing differences in POP concentrations in feathers could be a nondestructive way to identify POP sources of individual gulls.

The aim of this literature review was therefore to evaluate the potential of using feathers to determine different sources of POP contamination in individual gulls. This aim was translated into two research questions. The first question was to what extent feathers reflect internal and environmental levels of contamination. Since until now, feather analysis was mainly used to determine the degree of the POP contamination of species inhabiting certain areas, and not to determine where the contamination originated from, the second question was whether it would be possible to distinguish between POP contaminations that originate from different foraging habitats visited by gulls. If this is indeed the case, there are many means to develop similar approaches for studies in other bird species.

2 The Reflection of Internal and Environmental Contaminant Concentrations in Feathers

During the last years, many have studied the possibility to use feathers as a nondestructive biomonitoring tool for persistent organic pollutants (reviewed by García-Fernández et al. 2013). The most commonly studied pollutants are polychlorinated biphenyls (PCBs), organochlorine pesticides (OCPs), and polybrominated diphenyl ethers (PBDEs). These contaminants have been studied all over the world in feathers of a wide variety of bird species, such as predatory birds in Greenland (Jaspers et al. 2011), Norway (Eulaers et al. 2011a, b), Pakistan (Abbasi et al. 2016), Belgium (Jaspers et al. 2006, 2007b; Eulaers et al. 2014), and Argentina (Martínez-López et al. 2015); non-predatory aquatic and terrestrial birds from Iran (Rajaei et al. 2011), the USA (Summers et al. 2010), Belgium (Dauwe et al. 2005; Jaspers et al. 2007b), Spain (Espín et al. 2012), and Romania (Matache et al. 2016); and even poultry in Slovenia (Zupancic-Kralj et al. 1992). Perfluoroalkyl substances (PFAS) have also been detected in feathers of several birds from different trophic levels (Meyer et al. 2009), but the most frequently reported substances are PCBs, OCPs, and PBDEs. Therefore, we will focus on these substances in this section.

Although contaminant concentrations differ among different types of feathers, all studied feather types seem to be adequate biomonitoring tools (reviewed by García-Fernández et al. 2013). There are different pathways of POP deposition into and onto feathers (summarized in Fig. 1). One way is the internal allocation of substances, mainly from the bloodstream, and it has been suggested that this could be a way to sequester harmful substances (Van den Steen et al. 2007). Internal allocation of contaminants to feathers probably occurs during the growth of the feather, when the feather is still connected to the bloodstream (Fig. 1). This implies that especially concentrations of contaminants in newly grown feathers of adult birds and nestlings are related to concentrations in blood and blood plasma (Van den Steen et al. 2007; Eulaers et al. 2011a, b). Concentrations in muscle tissue and fat are also correlated with those in feathers (Dauwe et al. 2005; Jaspers et al. 2006, 2007b; Rajaei et al. 2011; Eulaers et al. 2014). In addition, some studies observed a correlation between POP concentrations in feathers and liver tissue (Rajaei et al. 2011; Eulaers et al. 2014). However, Meyer et al. (2009) only found a correlation between concentrations in feathers and liver tissue when five bird species of different trophic levels were pooled, but not for individual bird species, probably due to a small sample size. Therefore, despite some exceptions, it is concluded that especially newly grown feathers of adult birds and nestlings may reflect the internal contamination profile.

After the feather is fully grown, it is disconnected from the bloodstream, and hence POP concentrations in feathers are less affected by the internal contamination (Fig. 1) (Dauwe et al. 2005). Contamination profiles in fully grown feathers seem to remain rather stable, as it was possible to analyze POP concentrations in feathers of stuffed birds more than 10 years after they were collected (Behrooz et al. 2009).

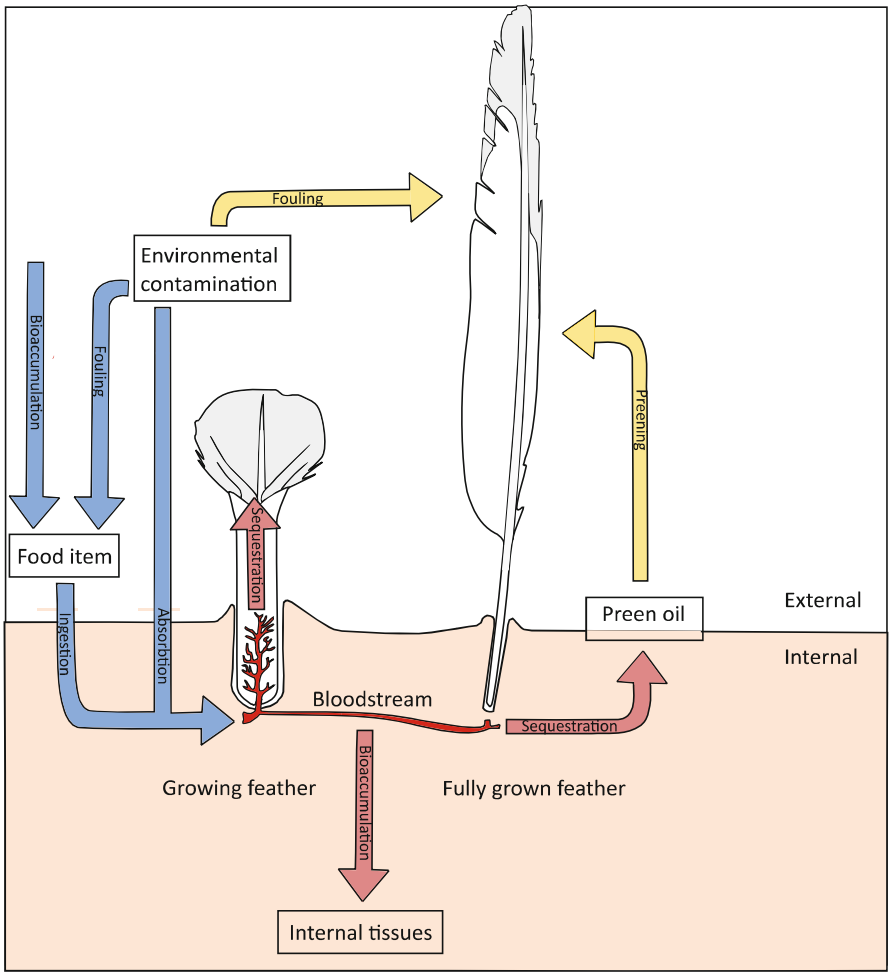

Fig. 1 Schematic overview of the major internal and external deposition pathways of POPs into growing feathers that are connected to the bloodstream (left), and fully grown feathers that are no longer connected to the bloodstream (right). The blue pathways show how environmental contamination enters the bloodstream. POPs can be taken up from the environment directly, via absorption through the skin or in the lungs, or indirectly by ingesting food items that are contaminated by bioaccumulation through the food chain or by fouling with contaminated dust or liquids. In red is the internal pathway that shows the sequestration of POPs from the bloodstream into growing feathers and preen oil. In addition, POPs from the bloodstream can bioaccumulate in internal tissues. In yellow is the external pathway of POP deposition onto feathers that could be by preening with contaminated preen oil and by fouling of the feathers with contaminated dust or liquids

Thus, contamination that is present in the feather at the time of sampling, was probably acquired during feather growth, which can be up to 1 year earlier for flight feathers (Harris 1971). On the contrary, internal POP concentrations could change frequently, as a result of tissue-specific metabolic processes and changed exposure (Jaspers et al. 2006). Therefore, internal body contaminant concentrations represent more recent exposure, and as these concentrations change, the correlation between feather concentrations and internal body concentrations could be weakened in older feathers (Jaspers et al. 2006).

Another pathway of POP deposition on feathers is the external deposition by preening with preen oil (Fig. 1). As older feathers are preened more often than newly grown feathers, this effect changes with the age of the feather (Jaspers et al. 2011). Due to the hydrophobicity of PCBs, PBDEs, and OCPs, preen oil contains relatively high concentrations of POPs (Burreau et al. 2004; Yamashita et al. 2007). Consequently, when preen oil was removed from the feathers, the total POP concentration was significantly reduced in white-tailed eagle (*Haliaeetus albicilla*) and common magpie (*Pica pica*) feathers (Jaspers et al. 2008, 2011). Concentrations of POPs in preen oil correlated with internal POP concentrations in white-tailed eagles (Eulaers et al. 2011a; Jaspers et al. 2011), although this correlation was not observed in water birds by Kocagöz et al. (2014). Nevertheless, preening activity of birds probably enhances the correlation of internal levels of contamination with the contamination levels in older feathers. Hence, after feathers are disconnected from the bloodstream, their contaminant concentrations could remain correlated to the internal concentrations due to preening with contaminated preen oil.

Finally, dust particles could also cause the deposition of POPs onto feathers (Fig. 1). Jaspers et al. (2014) suggested that external contamination by dust at a local point source led to a different ratio between perfluorooctanoic acid (PFOA) and perfluorooctane sulfonate (PFOS) in the feathers of barn owls (*Tyto alba*). In addition, white-tailed eagle and common magpie feathers washed with water showed significantly reduced POP concentrations, possibly due to the removal of dust and preen oil (Jaspers et al. 2008, 2011). Moreover, it has been suggested that contaminated dust on feathers is a source of internal PBDEs in ring-billed gulls (*Larus delawarensis*) that forage on landfills, as a result of dust ingestion when gulls preen their feathers (Gentes et al. 2015). Yet, this seemed of little importance for common buzzards (*Buteo buteo*) and great tits (*Parus major*), possibly due to less exposure to highly polluted dust in their habitats (Dauwe et al. 2005; Jaspers et al. 2007a). The contribution of pollution by dust particles on feathers is probably especially high for adult birds foraging in highly contaminated areas, like landfills. The contribution of this pathway will probably be lower for nestlings, since they are not yet visiting these contaminated areas.

In conclusion, during the growth of the feather, POPs are probably mainly deposited internally, via the bloodstream. Subsequently, when the feather is fully grown and disconnected from the bloodstream, most POPs are probably deposited externally by preening with preen oil and by dust particles (summarized in Fig. 1). These pathways of deposition overlap up to a certain extent, as preen oil is

excreted from internal tissues and thus reflects internal contamination levels, and dust particles can be inhaled or ingested and thus also have an effect on the internal levels of contamination. Nevertheless, newly grown feathers reflect recent exposure through internal sequestration and external deposition of contaminated dust particles and preen oil, while older feathers reflect recent exposure through external deposition only.

3 Identification of the Source of Contamination Based on the POP Concentrations in Feathers

One way to distinguish between birds foraging in marine areas or at landfills could be the difference in POP concentrations. Even though the production of PCBs and most PBDEs and OCPs is banned, high concentrations of PCBs and PBDEs are detected in leachate (Öman and Junestedt 2008; Li et al. 2012, 2014) and dust from landfills (Hansen et al. 1997; Melnyk et al. 2015). As shown in Table 1, several studies indicated that concentrations of OCPs, PCBs, and PBDEs in animals are elevated when their habitat is contaminated. However, the source of contamination differs for each POP. OCPs like DDTs were used in European agriculture until the late 1970s and early 1980s (FAO/UNEP 1991), and nowadays elevated concentrations of OCPs are still measured in eggs of great tits in a rural area in Flanders (Van den Steen et al. 2008) (Table 1). Also in common magpie feathers from Flanders, DDE concentrations were higher in rural samples compared to urban samples (Jaspers et al. 2009) (Table 1). Therefore, high concentrations of OCPs in feathers could indicate a rural foraging area. However, several raptors collected in a Chinese urban area contained high DDT concentrations, up to 158,700 ng g^{-1} DDT in Eurasian sparrowhawks (*Accipiter nisus*), that could be the result of highly contaminated wintering or stopover sites in southeast China (Chen et al. 2009). Although the use of DDT in China has been banned in 1983, no apparent decline in DDT concentrations has been observed in the field, and large amounts of DDT are still produced and probably discharged as a result of export demands and the production of dicofol (Qiu et al. 2005; Zhao et al. 2018).

Table 1 shows that, in contrast to OCPs, elevated concentrations of PCBs and PBDEs in birds were mostly linked to urban areas (Jaspers et al. 2009; François et al. 2016; Zeng et al. 2016), industry (Batty et al. 1990; Smith et al. 2003; Van den Steen et al. 2008; Zeng et al. 2016), and landfills (Johnson et al. 1996; Halbrook and Arenal 2003; Chen et al. 2013; Gilchrist et al. 2014), and the study of Van den Steen et al. (2008) showed that PCB and PBDE concentrations were highly correlated to each other. Moreover, Ito et al. (2013) measured elevated PCB concentrations in preen oil of GPS-tracked streaked shearwaters (*Calonectris leucomelas*) that foraged in an inland sea surrounded by urbanized coast, compared to shearwaters that foraged in the Pacific Ocean. On the contrary, De la Casa-Resino et al. (2015) did not measure any detectable concentrations of PCBs in white stork chicks (*Ciconia*

Table 1 Overview of reported mean or median DDT, PCB, and PBDE concentrations (ng g^{-1}) detected in samples collected from birds inhabiting different areas

POP	Species	Animal	Landfill (ng g^{-1})	Urban (ng g^{-1})	Industry (ng g^{-1})	Rural (ng g^{-1})	Sample	Median/ mean	Reference
Σ DDTs	*Pica pica*	Common magpie	–	3.07	–	34.2	Feathers	Median	(Jaspers et al. 2009)
	Parus major	Great tit	–	–	400–1100	500–2700	Eggs	Median	(Van den Steen et al. 2008)
	Falco tinnunculus	Common kestrel	–	8600	–	–	Liver	Median	(Chen et al. 2009)
	Athene noctua	Little owl	–	23,200	–	–	Liver	Median	(Chen et al. 2009)
	Otus sunia	Scops owl	–	16,900	–	–	Liver	Median	(Chen et al. 2009)
	Asio otus	Long-eared owl	–	1100	–	–	Liver	Median	(Chen et al. 2009)
	Accipiter nisus	Eurasian sparrowhawk	–	158,700	–	–	Liver	Median	(Chen et al. 2009)
	Accipiter gularis	Japanese sparrowhawk	–	62,700	–	–	Liver	Median	(Chen et al. 2009)
	Buteo buteo and Buteo hemilasius	Common and upland buzzard	–	1900	–	–	Liver	Median	(Chen et al. 2009)
*	*Ciconia ciconia*	White stork	ND	–	–	0.11	Nestling blood	Median	(de la Casa-Resino et al. 2015)
Σ PCBs	*Pica pica*	Common magpie	–	140	–	4.24	Feathers	Median	(Jaspers et al. 2009)
	Corvus macrorhynchos	Jungle crow	540	–	–	2100	Breast muscle	Mean	(Watanabe et al. 2005)
*	*Corvus splendens*	House crow	1200	–	–	820	Breast muscle	Mean	(Watanabe et al. 2005)
	Parus major	Great tit	–	–	2000–6000	1000–3700	Eggs	Median	(Van den Steen et al. 2008)
	Sturnus vulgaris	European starling	4100–27,700	–	–	300	Eggs	Mean	(Halbrook and Arenal 2003)
	Falco tinnunculus	Common kestrel	–	3500	–	–	Liver	Median	(Chen et al. 2009)
	Accipiter nisus	Eurasian sparrowhawk	–	19,900	–	–	Liver	Median	(Chen et al. 2009)
	Accipiter gularis	Japanese sparrowhawk	–	3900	–	–	Liver	Median	(Chen et al. 2009)

	Buteo buteo and Buteo hemilasius	Common and upland buzzard	–	200	–	–	Liver	Median	(Chen et al. 2009)
	Athene noctua	Little owl	–	1500	–	–	Liver	Median	(Chen et al. 2009)
	Otus sunia	Scops owl	–	500	–	–	Liver	Median	(Chen et al. 2009)
	Asio otus	Long-eared owl	–	100	–	–	Liver	Median	(Chen et al. 2009)
	Gallinago gallinago	Common snipe	–	–	2200	–	Pectoral muscle	Median	(Luo et al. 2009)
*	*Ciconia ciconia*	White stork	ND	–	–	ND	Nestling blood	Median	(de la Casa-Resino et al. 2015)
	Ardeola bacchus	Chinese-pond heron	–	–	2200	–	Pectoral muscle	Median	(Luo et al. 2009)
	Calonectris leucomelas	Streaked shearwaters	–	1600	–	200	Preen oil	Median	(Ito et al. 2013)
	Amaurornis phoenicurus	White-breasted waterhen	–	–	600	–	Pectoral muscle	Median	(Luo et al. 2009)
	Gallirallus striatus	Slaty-breasted rail	–	–	820	–	Pectoral muscle	Median	(Luo et al. 2009)
	Porzana fusca	Ruddy-breasted crake	–	–	37	–	Pectoral muscle	Median	(Luo et al. 2009)
Σ PBDEs	*Pica pica*	Common magpie	–	0.41	–	0.27	Feathers	Median	(Jaspers et al. 2009)
	Parus major	Great tit	–	–	55–80	35–60	Eggs	Median	(Van den Steen et al. 2008)
	Sturnus vulgaris	European starling	30–280	–	15–102	6.7–44	Eggs	Median	(Chen et al. 2013)
	Tachycineta bicolor	Tree swallow	–	205.5–590.1	–	83.6	Eggs	Mean	(Gilchrist et al. 2014)
	Larus delawarensis	Ring-billed gull	–	128	–	–	Liver	Mean	(François et al. 2016)
	Gallus gallus	Chicken	–	–	7700	3100	Eggs	Median	(Zeng et al. 2016)
*	*Anser anser*	Goose	–	700	–	400	Eggs	Median	(Zeng et al. 2016)

Areas are divided into four categories: landfills, urban areas, industrial areas, and rural areas. Also the sample type and whether the provided value is a median or a mean are provided. Within chemical compounds, species are ordered taxonomically (Laurin and Gauthier 2012). Concentrations specified with ND (no data) were analyzed, but were below detection limit. Concentrations specified with a dash (−) were not analyzed. Rows marked with an asterisk (*) indicate that contaminant concentrations at different locations did not differ significantly ($p > 0.05$)

ciconia) in a nest close to a landfill, even though white storks in this rural area visited the landfill frequently (Table 1). In addition, magpie feathers from urban, rural, and industrial sites did not exhibit different concentrations of PBDEs (Table 1) (Jaspers et al. 2009). Watanabe et al. (2005) did not observe a significantly different PCB concentration in the breast muscles of house crows (*Corvus splendens*) living on an Indian landfill compared to rural house crows, while the PCB concentration in the breast muscles of rural jungle crows (*Corvus macrorhynchos*) was even significantly higher than in crows on landfills. However, both crow species from landfills exhibited significantly higher concentrations of the more harmful dioxin-like PCBs (Watanabe et al. 2005). Finally, no significantly different PBDE concentrations were reported for urban domestic goose eggs (*Anser anser*), although concentrations appeared to be higher (Zeng et al. 2016). Nevertheless, as is shown in (Table 1), there is substantial evidence in the literature that elevated PCB and PBDE concentrations in birds can be linked to urban or industrial areas and landfills.

Therefore, gulls that forage in PCB-, PBDE-, or OCP-contaminated areas are likely to contain elevated concentrations of these contaminants in their body, eggs, and feathers. In this regard, PCBs and PBDEs could be especially useful, since elevated concentrations of these POPs are linked to landfills and urban and industrialized areas. The next step would therefore be to evaluate if specific PCB and PBDE profiles could give information about specific POP sources.

4 Identification of the Source of Contamination Based on the POP Congener Profile in Feathers

4.1 Linking the POP Congener Profile to the Source of Contamination

The POP congener composition could also provide valuable information regarding the source of contamination in gulls. In total, there are 209 PCB and 209 PBDE congeners, numbered after the position and number of chlorine (in PCBs) or bromine (in PBDEs) atoms [Fig. 2: PCB (a) and PBDE (b) molecules. The numbers indicate the positions that can be halogenated with chlorine (PCB) or bromine (PBDE) atoms]. The degree of biomagnification between trophic levels is determined by the bioavailability, the uptake, the excretion, and the ability of the animal to metabolize or dehalogenate the congener (biotransformation) (Arnot and Gobas 2003; Burreau et al. 2004). These factors depend greatly on the halogenation of the congener and the physiology and metabolic capacity of the animal (Hawker and Connell 1988; Boon et al. 1989, 1994; Fisk et al. 1999; Arnot and Gobas 2003; Voorspoels et al. 2007). Highly chlorinated PCBs ($\geq$6 chlorines) are more hydrophobic than the lightly chlorinated PCBs ($\leq$5 chlorines) and are also metabolized more slowly than the lightly chlorinated (<5 chlorines) compounds (Boon et al. 1989), making them highly bioaccumulative (Arnot and Gobas

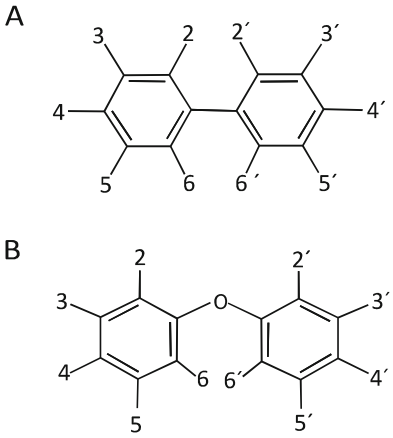

Fig. 2 PCB (**a**) and PBDE (**b**) molecules. Numbers indicate positions that can be halogenated with chlorine (PCB) or bromine (PBDE) atoms

2003; Burreau et al. 2004). In contrast to PCBs, for PBDEs especially the lightly brominated congeners (≤5 bromines) bioaccumulate strongly, but the bioaccumulation of highly brominated PBDEs (≥6 bromines) is restricted by their slow uptake due to their large size and high molecular weight and their metabolic debromination after uptake (de Wit 2002; Burreau et al. 2004, 2006; Van den Steen et al. 2007; Voorspoels et al. 2007; Letcher et al. 2014; François et al. 2016). Therefore, highly chlorinated (≥6 Cl) PCBs and lightly brominated (≤5 Br) PBDEs have a comparably high biomagnification potential, in contrast to lightly (≤5 Cl) chlorinated PCB and highly (≥6 Br) brominated PBDE congeners (Burreau et al. 2004).

Congener-specific biomagnification rates and site-specific contamination sources are likely to result in different PCB and PBDE congener profiles. A gull foraging at sea is mainly exposed to POPs through the bioaccumulation of POPs in its prey, and hence the trophic position of the gull and the prey plays an important role in the exhibited congener composition in the body and feathers (Borgå et al. 2001; Ruus et al. 2002). A higher proportion of more bioaccumulative congeners will probably occur in these gulls due to biomagnification and metabolism via the food chain (Strandberg et al. 1998; Dietz et al. 2000; Borgå et al. 2005). As we explained in the section above, highly chlorinated PCBs and lightly brominated PBDEs are more bioaccumulative, and, therefore, higher proportions of these congeners are likely to be present in gulls foraging at sea.

In contrast to gulls foraging at sea, gulls foraging at landfills predominantly feed on anthropogenic food. The food itself has in general relatively low POP concentrations but can be covered by leachate and dust containing high POP concentrations (Brousseau et al. 1996; Duhem et al. 2003, 2005; Schecter et al. 2010; Huwe and Larsen 2005; Li et al. 2012; McFarland and Clarke 1989; Öman and Junestedt 2008; Hansen et al. 1997; Persson et al. 2005). Gulls are thus probably mainly exposed to POPs by eating food items or preening feathers that are fouled with contaminated dust or leachate (Persson et al. 2005; Gentes et al. 2015). In addition, substantial amounts of POPs could be absorbed when lungs, skin, or

feathers are regularly exposed to contaminated dust, leachate, and aerosols. Due to the relatively high proportion of less bioaccumulative congeners in these substances, gulls that foraged on landfills are likely exposed to a higher proportion of less bioaccumulative congeners, which could be reflected by the congener profile of their feathers (Hansen et al. 1997; Öman and Junestedt 2008; Melnyk et al. 2015). A higher proportion of less bioaccumulative congeners in birds inhabiting contaminated areas is supported by several studies. European starlings (*Sturnus vulgaris*) nesting on a landfill and common magpies living in urban areas exhibited higher proportions of lightly chlorinated and thus less bioaccumulative PCBs in their eggs or feathers, compared to starlings or magpies living in a less contaminated area (Halbrook and Arenal 2003; Jaspers et al. 2009). In addition, a high proportion and concentration of the fully brominated and therefore less bioaccumulative decabromodiphenylether (DecaBDE or BDE 209) was found in 25% of male ring-billed gulls that visited refuse tips at least once (Gentes et al. 2015) and elevated concentrations of highly brominated PBDEs were found in eggs of great tits and tissue of ring-billed gulls that inhabit urban areas (Van den Steen et al. 2008; François et al. 2016). Nevertheless, in contrast to what would be expected based on the level of bromination, the tetrabrominated PBDE 47 was more prominent in urban common magpie feathers, while the pentabrominated PBDE 99 was more prominent in rural magpie feathers (Jaspers et al. 2009). However, despite this last exception, a higher proportion of less bioaccumulative congeners is in general exhibited in birds inhabiting contaminated areas.

Thus, based on the combined evidence described in this section, we conclude that the analysis of the congener profiles in gull feathers could be a promising approach to determine the likely source of contamination in gulls. The trophic position of gulls foraging at sea will likely cause a higher proportion of more bioaccumulative POPs, such as highly chlorinated PCBs and lightly brominated PBDEs. Gulls that forage on landfills will probably exhibit a higher proportion of less bioaccumulative congeners, due to a relatively high availability of these congeners in these areas. This approach will be further demonstrated by means of a case study in the next section.

4.2 Case Study: The [PCB 153]/[PCB 52] and [PCB 118]/[PCB 52]-Ratios

To demonstrate the feasibility of the analysis of differences in congener profiles to assess the contamination source, we performed a case study regarding the ratio between the concentrations of the highly bioaccumulative PCB 153 and PCB 118 (2,2′,4,4′,5,5′-hexachlorobiphenyl and 2,3′,4,4′,5-pentachlorobiphenyl, respectively) and the less bioaccumulative PCB 52 (2,2′,5,5′-tetrachlorobiphenyl) (Borgå et al. 2004). Based on the theory explained in the previous paragraph, we

hypothesized that relatively low ratios are exhibited in birds that foraged on landfills and in urban areas, and relatively high ratios are exhibited in birds that foraged at sea.

To test this hypothesis, we calculated the [PCB 118]/[PCB 52] and [PCB 153]/[PCB 52] ratios for animals inhabiting natural areas or urban areas and landfills, from concentrations obtained from studies that measured PCB concentrations in feather, liver, and preen oil/gland samples in birds (references in Table 3). In addition, to assess the bioavailability of these congeners at landfills, we calculated the ratio from concentrations obtained from studies that measured PCBs in different landfill samples (references in Table 2). The results from this calculation are summarized in Tables 2 and 3 and Fig. 3.

As we hypothesized, relatively high ratios were mostly obtained for feather, preen oil, and liver samples from birds that foraged in a natural environment (Fig. 3 and Table 3). This is presumably due to a higher degree of biomagnification of the more bioaccumulative PCBs 118 and 153 in their prey, which is indicated by the high ratios obtained for fish and crustaceans in natural environments (Table 3) (Duhem et al. 2005; Abdennadher et al. 2014). Simultaneously, usually low ratios were obtained for feather, liver, and preen gland samples from birds that forage in urban areas or at landfills, which is presumably due to the availability of similar proportions of PCBs 52, 118, and 153 in these areas (Tables 2 and 3 and Fig. 3). Indeed, the low ratios obtained in different landfill samples indicate a similar or even higher availability of PCB 52 compared to PCBs 118 and 153 (Table 2).

The ratios in liver tissue in particular for birds from natural areas were higher than the ratios for birds from landfills (Fig. 3). This pattern was also generally observed in feathers and preen gland tissue/oil, although there were some exceptions (Fig. 3). First of all, relatively low ratios were calculated for feathers of white-tailed eagles from a natural environment, compared to the high [PCB 153]/[PCB 52] ratio obtained from preen oil of the same species (Table 3, Fig. 3) and to the ratios that were obtained from feathers of common magpies from an industrial urban area (Table 3, Fig. 3). Furthermore, a relatively high [PCB 153]/[PCB 52] ratio was obtained from the preen gland of common magpies inhabiting an industrial urban area (Flanders, Belgium), especially compared to the [PCB 118]/[PCB 52] ratio in the preen gland from the same individuals. This might be due to a local source of PCB 153 (Table 3, Fig. 3) (Jaspers et al. 2008). However, no such high [PCB 153]/[PCB 52] ratio was obtained from common magpie feathers from the same area (Table 3, Fig. 3) (Jaspers et al. 2008).

Table 2 [PCB 153]/[PCB 52] and [PCB 118]/[PCB 52] ratios in different landfill samples, calculated from concentrations obtained from different studies

Sample	Country	[PCB 153]/[PCB 52]	[PCB 118]/[PCB 52]	Reference
Leachate sediment	Canada	1.33	0.56	(Öman and Junestedt 2008)
Dust	USA	0.98	–	(Hansen et al. 1997)
Surface soil	Poland	0.68	0.75	(Melnyk et al. 2015)

Table 3 [PCB 153]/[PCB 52] and [PCB 118]/[PCB 52] ratios for different species from different trophic levels and habitats, calculated from concentrations obtained from different studies

Species	Tissue	Trophic level	Area	[PCB 153]/[PCB 52]	[PCB 118]/[PCB 52]	Diet	Reference
Zooplankton	Homogenized	Primary/secondary consumer	Natural area, northern Baltic Sea	3.25	1.06	Phytoplankton, zooplankton, detritus	(Strandberg et al. 1998)
Zooplankton	Homogenized	Primary/secondary consumer	Natural area, mid Baltic Sea	1.36	1.23	Phytoplankton, zooplankton, detritus	(Strandberg et al. 1998)
Mysis sp.	Homogenized	Secondary consumer	Natural area, northern Baltic Sea	6.62	3.69	Zooplankton	(Strandberg et al. 1998)
Mysis sp.	Homogenized	Secondary consumer	Natural area, mid Baltic Sea	4.15	2.31	Zooplankton	(Strandberg et al. 1998)
Clupea harengus	Homogenized	Secondary consumer	Natural area, northern Baltic Sea	7.33	3.17	Crustaceans, zooplankton, fish	(Strandberg et al. 1998)
Clupea harengus	Homogenized	Secondary consumer	Natural area, mid Baltic Sea	9.17	4.58	Crustaceans, zooplankton, fish	(Strandberg et al. 1998)
Cepphus grylle	Liver	Omnivore	Natural area, Barents Sea	12.5	8.38	Fish, crustaceans, and molluscs, insects, plants	(Borgå et al. 2005)
Uria lomvia	Liver	Secondary consumer	Natural area, Barents Sea	25	22.5	Fish, crustaceans, and molluscs	(Borgå et al. 2005)
Haliaeetus albicilla	Feathers	Top predator	Natural area, Trøndelag, NO	6.88	2.8	Fish, birds, and mammals	(Eulaers et al. 2011b)
Haliaeetus albicilla	Preen oil	Top predator	Natural area, Trøndelag, NO	31	8	Fish, birds, and mammals	(Eulaers et al. 2011b)
Larus audouinii	Eggs	Secondary consumer	Mediterranean Sea	–	3.78–25	Fish	(Goutner et al. 2001)
Sturnus vulgaris	Chicks	Omnivore	Landfill, Illinois, USA	2.5	2.55	Invertebrates, seeds, fruit	(Halbrook and Arenal 2003)
Sturnus vulgaris	Chicks	Omnivore	Landfill, Illinois, USA	4.18	3.5	Invertebrates, seeds, fruit	(Halbrook and Arenal 2003)
Pica pica	Feathers	Omnivore	Urban area, Antwerp, BE	2.59	3.89	Young birds and eggs, small mammals, insects, scraps, acorns	(Jaspers et al. 2008)
Pica pica	Preen gland	Omnivore	Urban area, Antwerp, BE	48	3.4	Young birds and eggs, small mammals, insects, scraps, acorns	(Jaspers et al. 2008)

Sample type, animal type, and a brief diet overview are also provided

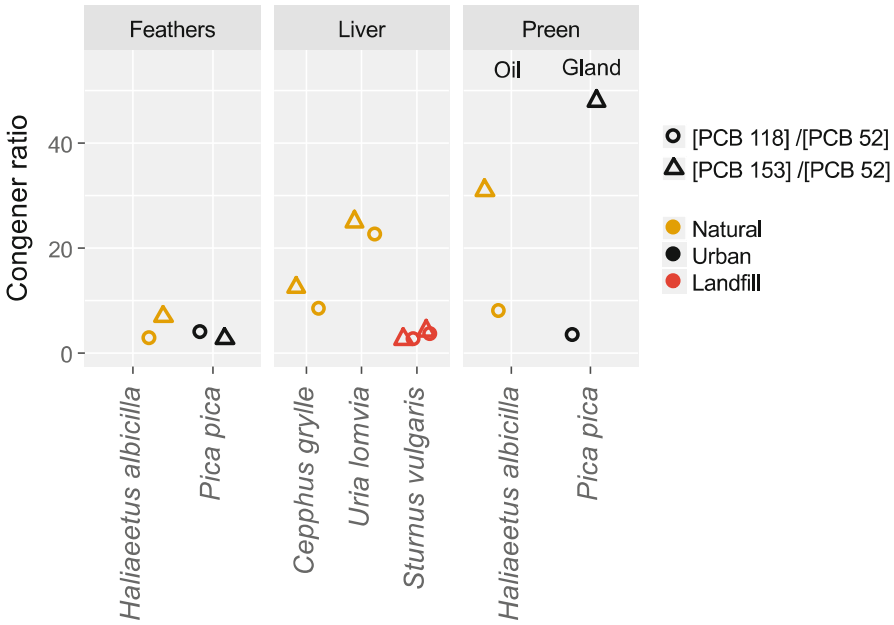

Fig. 3 [PCB 153]/[PCB 52] ratio (circles) and [PCB 118]/[PCB 52] ratio (triangles) for from either natural habitats (yellow) or urban areas (black) and landfills (red). Please note that preen oil and preen gland samples from *Haliaeetus albicilla* and *Pica pica*, respectively, are compared. Exact values, a more detailed description of the data and references can be found in Table 3

These exceptions could be due to species- and tissue-specific characteristics. Most likely, the higher chlorinated congeners have a higher affinity for preen oil than for feathers. These observations clearly indicate that preen oil PCB concentrations cannot be simply compared to concentrations and ratios in feathers, and when analyzing feathers, the influence of the congener profile of preen oil spread on the feathers should be taken into account.

Nevertheless, despite some exceptions, the ratios calculated for birds that inhabit natural areas, especially those for different seabird species, were generally substantially higher than the ratios for birds that foraged on landfills or in urban areas. Therefore, the analysis of POP congener profile in feathers and calculation of the ratios between more and less-accumulative congeners could be a promising approach to determine the source of contamination in gulls and is worth further investigation. Further species-specific analysis of a wide variety of PCB and PBDE congeners in feathers, combined with the analysis of PCB and PBDE congers in leachate, dust, and surface soil samples from landfills visited by the birds, according to Watanabe et al. (2005) and Table 2, could provide a stronger empirical basis for this approach. In addition, combining congener profile analysis in feathers with the analysis of stable isotopes signatures (Hobson and Clark 1992; Hobson 1993; Moreno et al. 2010; Auman et al. 2011; Caron-Beaudoin et al. 2013) on a subsample

from the same feathers and GPS tagging of the birds to quantify the time spent in different habitats (e.g., Camphuysen et al. 2015) could make this method very powerful.

5 Discussion and Conclusions

Based on the literature we studied for this review, we conclude that it is most likely possible to distinguish between POP contamination that originates from different foraging areas, like landfills or marine environments, based on the congener concentrations and profiles in gull feathers. Environmental and internal concentrations were to a certain extent reflected by the concentrations in feathers of adult birds and nestlings. In addition, it is likely possible to distinguish between different foraging habitats by combining the analysis of the total POP concentrations with the determination of the ratios between more-accumulative and less-accumulative PCB and PBDE congeners. However, this conclusion was drawn from the combined evidence of different studies, concerning a wide variety of species and tissues. Although PCB and PBDE concentrations in feathers are to a certain extent related to internal tissues, caution is necessary when comparing different tissues. Therefore, more insight is required into the establishment of POP congener concentrations and profiles in feathers in relation to the source of POP contamination, before using this approach.

Several aspects of this approach should be taken into account. First of all, the age of the birds greatly affects the exposure, since nestlings are unlikely to come in direct contact with environmental contamination, and young adults need to develop a specialization. Moreover, the type and age of the feather determine to what extent the POP concentrations in the feather reflect recent exposure and internal contaminant concentrations. Hence, especially newly grown feathers of adults are very suitable for this analysis, as they reflect both recent exposure through internal sequestration of contaminants and external deposition of dust particles from contaminated foraging areas. Sampling shed feathers is not possible for this approach, because external contamination might be worn off. Secondly, individual specializations in foraging strategies can lead to a large variety of foraging habitats and degrees of POP exposure, due to the large diversity in POP bioavailability between foraging habitats and even specific foraging locations. Furthermore, preening, washing, and swimming also affect the outer POP concentration, and thus influence to what extent POP concentrations on the feather reflect environmental and internal contamination. Finally, the concentrations of PCB and PBDE congeners in feathers are determined by the local bioavailability and the chemical properties of the congeners. These complications should be taken into account when analyzing POPs in gull feathers to identify contamination source.

In order to gain more insight into the complications of POP analysis in gull feathers and to provide a stronger basis to implement this approach, more research is required. We advise to further investigate the differences in POP concentrations

between recently grown and older feathers from adult or preadult gulls and to study the concentrations and composition of a wide variety of PCB and PBDE congeners inside the feather as well as on the outer surface, for example, conform the method of Jaspers et al. (2008). We also advise investigating the relation between the POP concentrations and congener compositions in preen oil and the concentrations and congener profiles in and on the feathers.

A case study of Jaspers et al. (2014) showed that the analysis of POP signatures in animal tissue may identify a specific point source of contamination, in this case the manufacturer of these POPs. However, when the possible source of contamination is a landfill, the larger variety of POPs that originate from this source complicates the analysis (Hansen et al. 1997; Öman and Junestedt 2008; Melnyk et al. 2015). Therefore, POP analysis could be combined with the analysis of stable isotopic signatures and GPS tracking. The identification of foraging habitats by tracking bird movements with bird-borne GPS loggers could be a crucial step in directly linking individual contamination profiles to a contamination source (Ito et al. 2013; Gentes et al. 2015). In addition, the analysis of stable isotopic signatures of carbon, nitrogen, and sulfur isotopes in feathers can provide information regarding the foraging area, trophic level, and diet composition (Hobson and Clark 1992; Hobson 1993; Moreno et al. 2010; Auman et al. 2011; Caron-Beaudoin et al. 2013). This would allow for an evaluation of the strength of the relationship between the sources of contamination obtained from POP analysis and the foraging habitats derived from GPS tracking and the analysis of stable isotopic signatures. For example, in white storks breeding in the vicinity of a landfill (1.5 and 4.9 km) in Spain, no PCBs but high concentrations of DDTs were detected (de la Casa-Resino et al. 2015). This could indicate that these birds forage in agricultural areas rather than on landfills, despite the close proximity of the landfill to their nests. GPS tracking, to determine their actual habitat use, and stable isotope analysis, to determine their trophic level and distinguish direct POP exposure in landfills from POP uptake via the food chain, would provide important complementary information (Abdennadher et al. 2014; Sommerfeld et al. 2016). Gentes et al. (2015) successfully related individual contamination in gulls to foraging habitat use, by combining GPS tracking with the analysis of PBDE concentrations in blood plasma. However, a strong link between habitat use and contamination in and on feathers instead of in blood plasma or certain tissues has not yet been made. This could be a crucial step in testing and further develop the proposed nondestructive approach. Moreover, when fully developed, this approach could be applied to many more individuals and on a far broader scale than would ever be possible when using GPS tags.

Finally, in this review gulls were the focal species, but the analysis of POPs in feathers could probably also be applied when studying other bird species. However, other bird species might have a totally different ecology that induces complications that are not discussed in this review. For example, not all bird species produce preen oil to treat their feathers, some species use powder down or do not use any substance for preening (Wetmore 1920; Kenyon Ross 1976). This will almost certainly affect the POP concentrations in and on their feathers and therefore should be taken into account.

In conclusion, despite some uncertainties that might be reduced by future research, enough evidence was obtained from the reviewed literature to propose the analysis of POPs in newly grown feathers of adult gulls and nestlings as a promising nondestructive approach to analyze the exposure of gulls to POPs and to identify the source of contamination. It could probably be extended to analyze sources of POP contamination in other bird species, provided that complications regarding the biology of the species are taken into account. Especially when integrated with other methods, like GPS tagging and stable isotope analysis, our proposed approach could prove to be very powerful.

6 Summary

Persistent organic pollutants (POPs) are present in almost all environments due to their high bioaccumulation potential. Especially species that adapted to human activities, like gulls, might be exposed to harmful concentrations of these chemicals. The nature and degree of the exposure to POPs greatly vary between individual gulls, due to their diverse foraging behavior and specialization in certain foraging tactics. Therefore, in order clarify the effect of POP-contaminated areas on gull populations, it is important to identify the sources of POP contamination in individual gulls. Conventional sampling methods applied when studying POP contamination are destructive and ethically undesired. The aim of this literature review was to evaluate the potential of using feathers as a nondestructive method to determine sources of POP contamination in individual gulls. The reviewed data showed that high concentrations of PCBs and PBDEs in feathers together with a large proportion of less bioaccumulative congeners may indicate that the contamination originates from landfills. Low PCB and PBDE concentrations in feathers and a large proportion of more bioaccumulative congeners could indicate that the contamination originates from marine prey. We propose a nondestructive approach to identify the source of contamination in individual gulls based on individual contamination levels and PCB and PBDE congener profiles in feathers. Despite some uncertainties that might be reduced by future research, we conclude that especially when integrated with other methods like GPS tracking and the analysis of stable isotopic signatures, identifying the source of POP contamination based on congener profiles in feathers could become a powerful nondestructive method.

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