

# Pilot Assessment of Status and Trends of Persistent Chemicals in Marine Mammals





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#### **OSPAR** Convention

The Convention for the Protection of the Marine Environment of the North-East Atlantic (the "OSPAR Convention") was opened for signature at the Ministerial Meeting of the former Oslo and Paris Commissions in Paris on 22 September 1992. The Convention entered into force on 25 March 1998. The Contracting Parties are Belgium, Denmark, the European Union, Finland, France, Germany, Iceland, Ireland, Luxembourg, the Netherlands, Norway, Portugal, Spain, Sweden, Switzerland and the United Kingdom

#### Convention OSPAR

La Convention pour la protection du milieu marin de l'Atlantique du Nord-Est, dite Convention OSPAR, a été ouverte à la signature à la réunion ministérielle des anciennes Commissions d'Oslo et de Paris, à Paris le 22 septembre 1992. La Convention est entrée en vigueur le 25 mars 1998. Les Parties contractantes sont l'Allemagne, la Belgique, le Danemark, l'Espagne, la Finlande, la France, l'Irlande, l'Islande, le Luxembourg, la Norvège, les Pays - Bas, le Portugal, le Royaume - Uni de Grande Bretagne et d'Irlande du Nord, la Suède, la Suisse et l'Union européenne.

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# Key Message

Polychlorinated biphenyls (PCBs) are present in marine mammals living in all five OSPAR Regions. Toothed-cetaceans and some subpopulations of pinnipeds present moderate to high ranges of PCB concentrations, often surpassing the estimated toxicity thresholds for the onset of reproductive incapacity. The ranges of PCB concentrations of baleen whales are always below the estimated toxicity thresholds.

# Background (brief)

Due to their long lifespan, high trophic level and large blubber reservoirs, marine mammals can accumulate extremely high concentrations of persistent organic pollutants (POPs) such as organochlorinated pesticides (i.e. DDTs and dieldrin), polybrominated diphenyl ethers (PBDEs) and PCBs. In marine mammals, PCBs concentrations can surpass the levels of PCB toxicity above which adverse consequences for the health of the animals are expected. Between the 1990s and the beginning of the 2000s, PCBs trends were found to decrease in time in sediments and marine biota. However, levels of PCBs were still associated with toxic effects in marine mammals, such as immunosuppression, endocrine disruption, reproductive impairment and reduced lifespan. PCBs were originally produced as technical mixtures such as Aroclors (U.S.). Today, the use of new analytical methods allows consistent detection in the tissues of several marine mammal species like the white-beaked dolphin and Risso's dolphin of over 145 out of the known 209 Aroclor and non-Aroclor PCB congeners<sup>1</sup>.

Data gathered from the existing literature was used as the basis for this report. As such, discrepancies may arise given the variability of data reporting methods, the species and areas considered and the list of PCB congeners analysed. This issue was considered when interpreting these data and in reaching conclusions. A broader approach at the ecosystem level is needed in order to assess past and present trends of organic pollution in top predators of the North-East Atlantic Ocean.

# Background (extended)

#### Bioaccumulation of persistent chemicals in marine mammals: a timely problem.

Persistent organic pollutants (POPs) are a subset of hydrophobic lipophilic and non-lipophilic chemicals that persist in the environment, bioaccumulate through the food web, and pose a risk of causing adverse effects to human health and the environment<sup>2</sup>. Marine mammals are sentinels for oceans and human health, as many species have long lifespans that permit the expression of chronic diseases, abnormalities in growth and development, cancer formation, immune suppression, endocrine disruption and reproductive failure<sup>3</sup>. Marine mammals have unique large blubber reservoirs, in which the lipid concentration can be over 90% in healthy and well-fed animals, that can serve as depots for lipophilic contaminants<sup>4</sup>. Additionally, some species are often long-term coastal residents and closely exposed to terrestrial point sources of pollution. Several marine mammal species (i.e. seals and toothed-cetaceans) are apex predators, feeding at the top of marine food web<sup>5</sup>. As such, they are vulnerable to higher rates of bioaccumulation, biomagnification and lactational transfer of specific POPs such as organochlorinated pesticides (i.e. DDTs and dieldrin), polybrominated diphenyl ethers (PBDEs), toxaphene and industrial polychlorinated biphenyls (PCBs), mono-methylmercury (MMHg)<sup>6,7</sup>. This makes them suitable for monitoring pollutants that are already considered of relevance for the Water Framework Directive (WFD) or Marine Strategy Framework Directive (MSFD)<sup>8</sup>. As such, marine mammals act as sentinel species for monitoring long-term trends in chemical contaminant exposure in the marine environment, the protection of the highest trophic level being the main goal of most environmental protective legislation. Decades of experimental studies across the entire OSPAR area have shown that exposure to i.e. PCBs have a range of **dose-dependent toxic effects** such as immunosuppression, endocrine disruption, reproductive impairment and reduced lifespan in mammalian species, including humans<sup>9–11</sup>.

Although levels of these pollutants are declining in some fish and marine mammal species, it is not always clear if such temporal trends are due to a decrease of POPs concentrations in the environment or a change in diet due to

environmental change<sup>10,11,14</sup>. For example, mean PCB concentrations were found to be 6-to-9-fold greater in Icelandic killer whales *Orcinus orca* with a mixed diet including marine mammals than in fish specialist individuals<sup>15</sup>. Despite these decreasing trends, toxic effects are still associated to moderate to high levels of POPs in marine mammals' tissues<sup>4,16</sup>. For example, Williams *et al.* (2022) found that PCBs levels below the estimated toxicity thresholds still caused important toxic effects in harbour porpoises *Phocoena phocoena*<sup>16</sup>. Additionally, combined toxic effects of multiple exposures to pollutants at low dose levels cannot be ruled out<sup>9</sup>. This highlights the need to pursue research about the levels and potential toxic effects of all (persistent) pollutants in marine mammals. Existing literature shows discrepancies with regards to data reporting (i.e. lipid or fresh weight) as well as studied species and area. The global assessment of POPs contamination in the North-East Atlantic is strongly biased towards few species (i.e. harbour porpoises) and accessible geographical regions. This leads to an underrepresentation of remote areas and rarer marine mammal species. However, the large variability in marine mammal's life styles, distribution and ecologies **necessitates a broader approach**, in order to effectively understand the status and trends of pollution at the North-East Atlantic scale.

# Potential and Goals of a Polychlorinated Biphenyls (PCBs) indicator for marine mammals

Until now the efforts of creating a valuable indicator of the levels of POPs pollution in marine mammals have concentrated on PCBs (See PCBs general introduction in "Status and Trends of Polychlorinated Biphenyls (PCB) in Fish and Shellfish and Sediment"). PCBs are considered a good starting point, as they can provide crucial information on the bioaccumulation and biomagnification of POPs in these predators. PCBs are some of the most widely studied POPs, they are ubiquitous in the environment, and as a result are routinely detected in marine environmental samples<sup>1</sup>.

Some species, such as the harbour seal *Phoca vitulina* or the coastal bottlenose dolphin *Tursiops truncatus* ecotype, live in localised colonies with high site fidelity and limited home ranges<sup>17,18</sup>. As such, they act as good indicators of local contamination from source regions. Other species predominantly feed in the deeper ocean far from the mainland, such as the long-finned pilot whale *Globicephala melas* and sperm whale *Physeter macrocephalus*<sup>19,20</sup>. These species are excellent indicators for the transport of POPs in the deeper regions of the ocean. Finally, species like the short-beaked common dolphins have wider offshore home range. These species are excellent indicators away from the major source and sink regions.

**The first goal** of the "PCBs indicator for marine mammal" is to assess the spatial and temporal trends of PCB levels in the North-East Atlantic Ocean, in order to infer the general spatial and temporal trends of marine top predators' pollution.

The most common PCBs analysed in marine mammals' tissues originally derive from technical mixtures such as Aroclors (U.S.), Clophen (Germany), Phenochlor (France), Sovol (former Soviet Union). Although "Aroclor" PCBs are still found in the environment, several studies on marine invertebrates (e.g. bivalves) increasingly focus on non-Aroclor (inadvertently produced) PCBs (i.e. PCB11, 206, 207, 208 and 209)<sup>21</sup>. Although new analytical methods allow the determination of all theorical 209 PCBs also in marine mammals<sup>1</sup>, there are no evidences that all congeners (included non-Aroclor ones) might be of concern to marine mammals or should be included in future studies. **The second goal** of the PCBs indicator for marine mammals is to assess the status of PCB toxicity in marine mammals, in order to infer the general health status of North-East Atlantic populations. Due to the difficulties for experimental and captive studies on marine mammals, PCBs toxicity thresholds have only been estimated in few captive mammals (seals, mink and sea otters) or dead animals<sup>26,28</sup>. As such, effective cause-effect relationships between PCB levels and profile and marine mammals' health are wildly missing. Additionally, these estimated thresholds consider all PCBs together as no congener-specific approach was investigated. Finally, no empirical evidence for the effects of chronic exposure to PCBs was ever assessed in different species, tissues, sexes or age classes. For the purposes of the status assessment we will focus on those thresholds that were available related to reproduction<sup>27</sup> - and not immunological ones, for which other thresholds exist<sup>22</sup>.

# Assessment Method

This preliminary assessment constitutes a first effort to collect all available PCBs data across all OSPAR Contracting Parties (CPs) with the aim of defining the criteria to use within both the Trend and Status assessments.

# Description of the indicator

The bioindicator of PCBs in marine mammals will follow the approach taken for the OSPAR indicator "<u>Status and Trends</u> of Polychlorinated Biphenyls (PCB) in Fish and Shellfish".

In assessing contaminants both 'relative' and 'absolute' aspects will be analysed:

- The 'Trend assessment' will focus on *relative* differences and changes on spatial and temporal scales providing information about the rates of change;
- The **'Status assessment'** will focus on the significance of the (risk of) pollution, defined as the level where chemical exposure becomes hazardous. This usually requires assessment criteria (See "Fish and Shellfish PCB indicator" Assessment Method for definition of Environmental Assessment Criteria (EAC)) to assess the extent of the impacts and, hence, the ecotoxicology of the contaminant. The metric "Total blubber PCB concentrations (as lipid weight)" will be determined separately for each Assessment Unit (AU) that will be species-specific, as well as an OSPAR-wide indicator. Total blubber PCB concentrations will be represented as the sum of the seven ICES (International Council for the Exploration of the Sea) PCBs (Σ7PCBs, IUPAC number: 28, 52, 101, 118, 153, 138, and 180), as well as the sum of 18-25 PCBs, for which toxicity thresholds have been calculated<sup>22</sup> (Σ18-25PCBs, See "Criteria for Status Assessment"). The Σ7PCBs was recommended for monitoring by the European Union Community Bureau of Reference. As such, the seven ICES congeners are routinely measured by CPs and easily available on a spatial and temporal scale. The sum of 18-25 PCBs has mostly been measured starting the 2010s, but will allow to assess the risk of PCB toxicity for each subpopulation from each AU.

# **PCBs data collection**

This preliminary assessment presents the ranges of PCBs concentrations measured in the blubber of marine mammals living in the North-East Atlantic Ocean and are expressed as mg/kg on a lipid basis (lw). At this stage, raw data of PCBs congeners were not available for all species and OSPAR regions, and PCBs concentration ranges (as Min – Max) were either obtained directly from CPs, or extracted from the existing literature. PCBs concentration ranges are here shown as the sum of total PCB congeners ( $\Sigma$ PCB<sub>tot</sub>) analysed in each study. The PCBs congeners included in the  $\Sigma$ PCB<sub>tot</sub> varied from one study to the other (See Table 3 to 7). We did not transform the data to get a single common sum of congeners, because we did not have access to the raw data, but only PCBs ranges. Each  $\Sigma$ PCB<sub>tot</sub> derives therefore from the given range of the specific congeners presented in each study, separately. Based on previous studies seven PCB congeners (PCB118, -138, -149, -153, -170, -180, and -187) contributed to 79% of the  $\Sigma$ PCB content in female harbour porpoises blubber samples<sup>11</sup>. Although these are results for just one species (and only female porpoises) and need to be considered with caution, the  $\Sigma$ PCB<sub>tot</sub> was considered comparable between the different studies, as long as these 7 PCBs were included into the measurement. The ranges of  $\Sigma$ PCB<sub>tot</sub> of each species include data measured on both males and females as well as adults and subadult individuals together. Data are organised in different functional groups as defined by the MSFD and habitat use, as: coastal small, toothed-cetaceans (Table 3), offshore odontoces(Table 4), deep-diving cetaceans (Table 5), seals (Table 6) and baleen whales (Table 7).

# **Preliminary Status Assessment**

The status of PCBs toxicity is inferred qualitatively comparing the ranges of  $\Sigma PCB_{tot}$  with the toxicity thresholds estimated for reproductive impairment.

A lower PCB toxicity threshold is used for the onset of physiological endpoints in marine mammals, equal to:

- 17 mg/kg lw as Aroclor 1254 (IUPAC numbers: 105, 118, 156, 99, 138, 153) as determined by Kannan et al. (2000); or
- **9.0 mg/kg lw** as ∑18-25PCBs (IUPAC numbers: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, 194), as determined by Jepson et al. (2016).

A higher PCB toxicity threshold, the highest reported in marine mammal toxicology studies, is used for the onset of reproductive impairment:

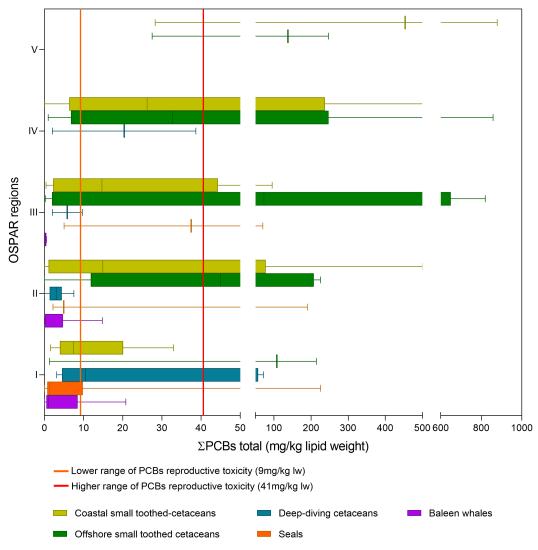
• 77 mg/kg lw as Clophen 50 as determined by Helle et al. (1976);

or

**41 mg/kg lw** as ∑18-25PCBs (IUPAC numbers: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, 194), as determined by Jepson et al. (2016).

# Results (brief)

- Small toothed cetaceans present the highest concentration range of the ΣPCB<sub>tot</sub> (0,20 820 mg/kg lw), followed by pinnipeds (0,01 226 mg/kg lw), deep-diving cetaceans (0,01 72 mg/kg lw), and baleen whales (0,04 21 mg/kg lw), all ages and sexes together. The harbour porpoise is the most represented species in PCBs studies. Much PCBs data are available for most of harbour porpoise AUs. For certain CPs continuous data are available since 2009. A moderate number of PCBs data is available for short-beaked common dolphin, coastal bottlenose dolphin and killer whales, covering most of their AUs or known geographical range (for killer whales);
- A moderate to low number of PCBs data is available for long-finned pilot whales, sperm whales and harbour seals;
- Little PCBs data are available for baleen whales and other pinnipeds;
- For harbour porpoises and long-finned pilot whales the ranges of PCBs concentrations appear overall higher in the 80s (3 72 mg/kg lw) than the 2010-2020 decade (0.10 50 mg/kg lw);
- Major gaps in PCBs data exist for OSPAR regions I (Arctic) and V.



**Figure 1.** Ranges of Polychlorinated biphenyls (PCBs) in the blubber of marine mammals living in the Northeast Atlantic Ocean (OSPAR area). Marine mammals are separated in five categories: coastal small toothed-whales (light green), offshore small toothed cetaceans (dark green), deep-diving cetaceans (blue), seals (orange) and baleen whales (purple). Data (in mg/kg lw) are shown as boxplots, representing the Minimum, 25<sup>th</sup> percentile, Average, 75<sup>th</sup> percentile and Maximum values of PCBs ranges obtained by the CPs and literature. PCBs levels are compared

to existing reproductive toxicity thresholds estimated for the sum of 25-18 PCBs<sup>23</sup>. OSPAR regions are defined as: *I* Arctic Waters, *II* Greater North Sea, *III* Celtic Seas, *IV* Bay of Biscay and Iberian Coast, *V* Wider Atlantic.

# Results (extended)

In the following paragraphs concentration ranges of  $\Sigma PCBs_{tot}$  of marine mammals living in the North-East Atlantic Ocean are presented by functional group (as described by the MSFD) and habitat use (offshore, coastal, deep-diving). Due to the limited availability of data for most species and regions, results are discussed qualitatively, without performing any statistical analysis.  $\Sigma PCBs_{tot}$  varied a lot between regions and functional groups. However, this high variability is mostly due to the heterogeneity of the type and origin of available PCB data. This has been considered in the interpretation of both spatial and temporal assessments.

Table 1 and 2 describe the geographical distribution of available PCB data in the OSPAR regions for small, toothedcetaceans (**Table 1**) and seals (**Table 2**). Table 3 to 7 show the range of  $\Sigma PCBs_{tot}$  of marine mammals by functional group and species level. They also report the CP who provided the data, the OSPAR region and the years the data is available for and the list of congeners measured.

# **PCBs Data Availability**

#### Most represented areas

Data on PCBs concentration were collected from various OSPAR CPs (Belgium, Denmark, France, Germany, Ireland, the Netherlands, Portugal, Spain, Sweden and UK) by species, area and time period. Additional information is also collected (i.e. age, sex, cause of death) by the aforementioned CPs.

For cetaceans, data are largely available for the Greater North Sea and the Celtic Sea (**Table 1**). Moderate amounts of data are available for the Bay of Biscay and the Iberian coast, while few data are available for Arctic waters. For pinnipeds, PCBs data are largely available for the Greater North Sea and the Celtic Sea (**Table 2**). No data are available for all the other regions. This ensemble of data does not reflect the available data, but only the data submitted.

#### Most represented species

At this stage, PCBs concentration data were collected for more than 24 species of cetaceans and seals. This nonexhaustive list includes: Harbour porpoise *Phocoena phocoena*, Short-beaked common dolphin *Delphinus delphis*, Striped dolphin *Stenella coeruleoalba*, White-beaked dolphin *Lagenorhynchus albirostris*, Long-finned pilot whale *Globicephala melas*, Atlantic white-sided dolphin *Lagenorhynchus acutus*, Bottlenose dolphin *Tursiops truncatus*, Minke whale *Balaenoptera acutorostrata*, Risso's dolphin *Grampus griseus*, Sowerby's beaked whale *Mesoplodon bidens*, Sperm whale *Physeter macrocephalus*, Northern bottlenose whale *Hyperoodon ampullatus*, Fin whale *Balaenoptera physalis*, Killer whale *Orcinus orca*, Pygmy sperm whale *Kogia breviceps*, Cuvier's beaked whale Ziphius cavirostris, Humpback whale *Megaptera novaeangliae*, Sei whale *Balaenoptera borealis*, Blainville's beaked whale *Mesoplodon densirostris*, beluga *Delphinapterus leucas*, harbour seal *Phoca vitulina*, grey seal *Halichoerus grypus*, ringed seal *Pusa hispida*, hooded seal *Cystophora cristata*, bearded seal *Erignathus barbatus*, harp seal *Pagophilus groenlandicus* and walrus *Odobenus rosmarus*. This is not exhaustive.

The largest set of PCBs data are available for harbour porpoise, although the northernmost limits of its distribution (Region I, Arctic Waters) are scarcely represented within the dataset (**Table 3**). Other species for which PCB data from OSPAR CPs and existing literature cover most of the spatial distribution range are the short-beaked common dolphin, coastal common bottlenose dolphin and killer whale (**Table 3 and 4**). Data for pinnipeds lack coverage in time and space (**Table 6**), while data on baleen whales are few (**Table 7**).

#### Most represented tissues

Most of the analyses of samples collected between 2009 and 2021 were carried out in blubber and results expressed on a lipid weight basis. Other tissues like liver, whole blood and muscle are scarcely and randomly represented.

#### Most represented temporal trends

The aim of this assessment is to evaluate the variation of PCBs levels in marine mammals in time from the North-East Atlantic Ocean and compare it with the timeframe of the QSR 2010 (1998 – 2008), the IA 2017 (2009 - 2015) and QSR 2023 report (2009 – 2021).

The preliminary analysis carried out with PCB concentration ranges provided by CPs shows that PCBs analysis are scattered in time and highly heterogenous. In general, most data are available for the time period between 2000 and 2008. Moderate amounts of data are available between 2008 and 2015. Few data are available between 2016 and 2021, although this number is on the rise. The harbour porpoise is the most represented species and the only one for which a temporal trend analysis could be efficiently run at large scale. This suggests that harbour porpoise could be a valid model species for the PCB indicator, at least at a first stage and for Regions II and III.

#### **Preliminary PCBs Assessment**

In this assessment the comparison of the ranges of  $\Sigma PCBs_{tot}$  with toxicity thresholds (9-41 mg/kg lw) is only indicative of a lower or higher level of PCBs toxicity potential in the concerned subpopulation or region. Because of the large variability in the number of congeners constituting each  $\Sigma PCBs_{tot}$ , as well as differences in sample individuals' age and gender, comparison between regions has to be taken with caution. Since in most cases raw data were not available, it was not possible to calculate the proportion of marine mammals at risk.

This preliminary assessment of PCBs level ranges in marine mammals from the North-East Atlantic suggests that the ranges of  $\Sigma PCBs_{tot}$  are higher in offshore, small toothed cetaceans, followed by coastal, small toothed cetaceans, deepdiving cetaceans, pinnipeds and finally baleen whales.

With regards to **coastal, small toothed cetaceans** the ranges of  $\Sigma PCBs_{tot}$  seem to be higher in the period between the 70s and the 90s than after the 90s (Table 3). However recent papers on UK porpoises have shown that the overall rate of decline is slow in comparison to other pollutants, and that the declining trends are not homogenous across all OSPAR regions. For example, the sum of 25-18 PCBs in harbour porpoises show a faster decrease in East England (region II) than West England and Wales (region III)<sup>16</sup>. This suggests that PCBs temporal trend might vary geographically, based on local pollution histories.

In most of the OSPAR regions small toothed cetaceans present  $\Sigma PCBs_{tot}$  between the lower and higher toxicity threshold (9 – 41 mg/kg lw). The higher bound of ranges of  $\Sigma PCBs_{tot}$  surpass the higher toxicity threshold mostly in OSPAR region II and IV (Table 3). In region IV, the upper range of PCBs data surpasses 41 mg/kg in bottlenose dolphins studied between 2000 and 2012. In region II, the upper range of PCBs data surpasses 41 mg/kg in harbour porpoises studied between 2006 and 2019 in the Netherlands and between 1990 and 2014 in the UK and Belgium. It is possible that this is linked with higher PCBs concentration in these two regions compared to the others, but it can't be ruled out that the higher ranges of PCBs are also linked to species-specific traits (i.e. trophic level).

Ranges of  $\Sigma PCBs_{tot}$  for **offshore, small toothed cetaceans** are above the lower toxicity threshold (9 mg/kg lw) in all regions for which data were available, with the exception of short-beaked common dolphins studied between 2001 and 2003 in the Celtic and Irish Sea<sup>24</sup> (Table 4). This could be related to the fact that offshore toothed cetaceans are represented mostly by the killer whale, which is at the top of the marine food web, with several observations of feeding on other species of marine mammals <sup>25</sup>. The maximum values of killer whales' range of  $\Sigma PCBs_{tot}$  surpass 41 mg/kg lw in all areas.

Moderate ranges of  $\Sigma PCBs_{tot}$  are found in **deep-diving cetaceans**. Higher values are found in studies from the 1970s (Table 5). The ranges of  $\Sigma PCBs_{tot}$  do not surpass the higher toxicity threshold of 41 mg/kg lw in any region.

With regards to **pinnipeds**, the maximal values of the  $\Sigma PCBs_{tot}$  ranges surpass the 41 mg/kg lw threshold in harbour seals from the Southern North Sea and ringed seal from Arctic waters (OSPAR regions I and II, Table 6).

Ranges of  $\Sigma PCBs_{tot}$  for **baleen whales** are generally lower than both PCBs toxicity thresholds (Table 7).

# **Putative Criteria for the Trend assessment**

Following existing marine mammals and PCBs indicators (See <u>Abundance and distribution of cetaceans and Status and</u> <u>Trends of Polychlorinated Biphenyls (PCB) in Fish and Shellfish</u>) the criteria for the Trend assessment will be selected based on the availability of data (See "Data availability" paragraph and Table 3 to 7). They include i.e. baselines for species-specific AUs, minimum number of years in a time series, number of years included within each reporting period, sample size required for detecting trends between reporting periods, etc.

#### Temporal trends

Late efforts of temporal trends assessment mostly on mercury in marine mammals from Arctic waters (OSPAR Region I) evaluated the number of years required to detect a statistical decline at  $\alpha = 0.05$ , with a power of 80% for the particular time-series<sup>26</sup>. This study suggested that a minimum of 10 continuous years of data is necessary to infer temporal variation in long-lived animals like marine mammals<sup>27</sup>. It is recommended that juveniles or males with known age (in years) should be studied, in order to correct for the potential effects of age on PCB bioaccumulation levels, and offloading in mature females.

Provisional assessment criteria could include:

- Decreasing trend: the mean concentration is statistically significantly decreasing (p < 0.20), given sufficient data to achieve an estimated 80% power;
- Stable trend: there is no statistically significant change in mean concentration (p > 0.20), given sufficient data to achieve an estimated 80% power;
- Increasing trend: the mean concentration is statistically significantly increasing (p < 0.20), given sufficient data to achieve an estimated 80% power;
- Inconclusive trend: no trend can be assumed, due to insufficient data to achieve an estimated 80% power.

In other OSPAR indicators, a significance level  $\alpha$ -value of 0.20 has been used to provide equal probability of type I and type II errors based on ICES advice (See i.e., <u>seal abundance indicator</u>). This approach is will be considered for this PCB indicator, for consistency across indicators.

Some CPs (i.e. UK and Ireland) have been monitoring the list of 25 PCBs (IUPAC N°: 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, 194) from 1990 to 2018. In these areas two different trends will be analysed: one including only the 7 ICES PCBs and another considering the 25 PCBs. These trends will be subsequently compared to select the best list of congeners.

#### Model species

For the Trend Assessment the harbour porpoise *Phocoena phocoena* will be used as model species, since: (1) a lot of samples and data are available in space and time, which will allow to cover most of OSPAR regions (I to IV); (2) it is a coastal and uniformly dispersed species with a generalist diet, which will allow to cover all potential local PCB sources.

#### Assessment areas

The analysis of PCB trends will be conducted at the local scale of harbour porpoise species-specific estimated AUs (See <u>Abundance and distribution of cetaceans</u>), and potentially the OSPAR broader scale, combing the information across all five OSPAR regions, to get a general overview of the state of the North-East Atlantic

#### Sum of PCBs

The seven ICES (International Council for the Exploration of the Sea) PCBs (Σ7PCBs, IUPAC number: 28, 52, 101, 118, 153, 138, and 180) will be the priority congeners of both spatial and temporal assessments. Recently, it was observed that other PCB congeners (i.e. 187 and 149) consist in the top 5 congeners found in blubber of harbour porpoises from Ireland (Williams *et al., In prep.*)<sup>28</sup>. This suggests that two of the top 5 congeners (187 and 149) for porpoises in UK waters are not on the ICES7 list. As such, additional PCBs might be included accordingly.

# Putative criteria for the Status Assessment

#### **OSPAR Commission 2022**

Differently from the Trend Assessment, the selection of criteria for the Status Assessment will focus on: (1) biological and physiological reasons, (2) the availability of pathological data, and (3) the necessity of covering the entire range of PCBs concentrations in marine mammals form the North-east Atlantic.

#### **Toxicity thresholds and sum of PCBs**

So far, Environmental Assessment Criteria (EACs) have not been developed for marine mammals. At this stage, PCBs toxicological risk can be assessed only by comparing PCBs levels measured in species tissues to the estimated marine mammal toxicity thresholds (see "Preliminary Status Assessment" paragraph) extracted from studies on the effect of PCBs on few marine and terrestrial mammals <sup>21,22,30</sup>. As such, at a first stage the sum of 25-18 PCBs will be used, following Jepson *et al.* (2016).

#### **Model species**

In order to cover the entire range of PCB concentrations, species like harbour porpoise *Phocoena phocoena* (for which more data are available) and the very contaminated killer whale *Orcinus orca* and the coastal bottlenose dolphin *Tursiops truncatus* will be used. Our preliminary assessment show how these two species are the most contaminated in PCBs in the OSPAR area (Table 3 and 4).

#### **Assessment areas**

The analysis of PCB levels will be conducted at the local scale of bottlenose dolphin and killer whale species-specific delineated AUs or populations geographical ranges (See <u>Abundance and distribution of cetaceans</u> and <u>Pilot</u> <u>Assessment on Abundance and Distribution of Killer whales</u>)</u>, as well as at the OSPAR broader scale, combing the information across all five OSPAR regions. Bottlenose dolphins will be used for the Status Assessment of coastal marine mammals, whilst killer whales will be used for the Status Assessment of offshore marine mammals.

#### **Pathological information**

General pathological information such as the possible cause of the death or presence of parasites is routinely gathered when samples are collected from dead animals (i.e. stranded, bycaught or hunted) of all age groups (juveniles, adult males and females). However, efforts to collect more detailed information like the presence of lesions, infectious diseases or malformations (i.e. ear bone) are conducted only by few Contracting Parties (CPs), because of funding limitations. As such, the Status Assessment will prioritize the integration of available pathological data with animals' reproductive status and age.

**Table 1.** Spatial distribution of Polychlorinated Biphenyls (PCBs) measured in blubber of marine mammals living in the North-East Atlantic Ocean by OSPAR Regions (I to V) and the MSFD subregions for small toothed cetaceans. Data availability is shown in blue palette. Underlined are the species for which PCBs concentrations are available for the majority of their distribution range. \*OSPAR region II includes the Swedish/Danish Sound, Roskilde fjord and Isefjord.

Assessment group		Assessment Units							
OSPAR	Т	*	III	IV					
MSFD Small-toothed cetaceans	I	Greater North Sea	Celtic Sea	Bay of Biscay and Iberian Coas	Maca	ronesia	v		
Species: Harbour porpoise, Short-beaked common dolphin, Striped dolphin,         Data availability         High           White-beaked dolphin, Long-finned pilot whale, Atlantic white-sided dolphin,         Moderate									
Bottlenose dolphin, Minke whale, Risso's dolphin, Sowerby's beaked whale, Sperm							N		
whale, Northern bottlenose whale, Fin whale, <u>Killer whale</u> , Pygmy sperm whale, Cuvier's beaked whale, Humpback whale, Sei whale, Blainville's beaked whale							ilable		

**Table 2.** Spatial distribution of Polychlorinated Biphenyls (PCBs) data measured in blubber of pinnipeds living in the North-East

 Atlantic Ocean by OSPAR Region (I to V) and MSFD subregions. Data availability is shown in blue palette as indicated by the QSR

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guidelines. Underlined are the species for which PCBs concentrations are available for the majority of their distribution range. \*OSPAR region II includes the Swedish/Danish Sound, Roskilde fjord and Isefjord.

Assessment group		Assessment Units							
OSPAR	Т	II <i>*</i>	III	IV					
MSFD Seals	I	Greater North Sea	Celtic Sea	Bay of Biscay and Iberian Coast Mac		Macaronesia			
Species: <u>harbour seal</u> , grey seal, hooded seal, harp seal, Data a ringed seal, walrus						Hig Mode			
						Low			
						Not Available			

**Table 3.** Levels of Polychlorinated biphenyl (PCBs) in blubber of coastal small toothed cetaceans living in the OSPAR regions (I to V) and Marine Strategy Framework Directive (MSFD) subregions of the North-East Atlantic Ocean. Data are shown as sum of total

PCBs measured (ΣPCBs<sub>tot</sub>) as Min – Max (mg/kg, lw). Data presented here derive from the following species: harbour

porpoise *Phocoena phocoena*, bottlenose dolphin *Tursiops truncatus*, beluga *Delphinapterus leucas*. The list of specific congeners is given after Table 7.

OSPAR/MSFD Region	Species	Data contributor	Sampling method	Sampling period	IUPAC N.*	<b>ΣPCBs</b> tot
		Iceland <sup>31</sup>	S; B	1987 - 1988	10	7,78 - 15,8
I		Germany <sup>32</sup>	В	1988 - 1990	6	7,20 - 33,0
Arctic waters		Germany <sup>33</sup>	S; B	1998 - 2005	6	1,55 - 4,71
		Norway (B. M. Jenssen)	FR	1995 - 1997	27	3.20 - 10.1
		France <sup>34</sup>	S; B	2002 - 2017	48	2,34 - 41,6
		Belgium <sup>35</sup>	S; B	1999 - 2004	28	1,33 - 22,6
		The Netherlands <sup>36</sup>	S	2006 - 2019	17	0,20 - 90,2
		Germany; Sweden <sup>32</sup>	В	1978 - 1993	6	2,20 - 67
II		Germany <sup>38</sup>	S; B	1998 - 2005	6	7,66 – 8,2
Greater North Sea,		Sweden <sup>39</sup>	S	2006 - 2019	7	0,20 - 24,2
Kattegat and Skagerack		UK <sup>23</sup>	S; B	1990 - 2012	18-25	0,40 - 81,
		UK <sup>23</sup>	S; B	1991 - 2001	18-25	7,21 - 358
		UK <sup>23</sup>	S; B	1999 - 2014	48	0,76 - 500
		Belgium <sup>40</sup>	S; B	2010 - 2012	6	4,50 - 393
		UK <sup>23</sup>	S; B	2004 - 2012	18-25	4,11 - 125

	UK <sup>23</sup>	S; B	2000	18-25	1,52 - 95,1
	Ireland <sup>41</sup>	FR	2000	7	0,98 – 48,8
III Celtic and Irish Sea	France <sup>42</sup>	S; B	2001 - 2017	48	7,71 - 23,6
insii Sea	UK <sup>23</sup>	S; B	1990 - 1998	18-25	0,40 - 65,7
	UK <sup>23</sup>	S; B	1999 - 2012	18-25	2,45 - 37,2
	France <sup>43</sup>	S; B	2004 - 2008	32	0,10 - 40,9
	France	S; B	2001 - 2003	18	3,10 - 10
IV	France <sup>24</sup>	S; B	2005 - 2008	32	18 - 102
Bay of Biscay and	France <sup>24</sup>	S; B	2003 - 2010	48	10,8 - 578
lberian Coast	Spain, Portugal, UK <sup>23</sup>	S; B	1995 - 2006	18-25	34,4 - 282
	Spain, Portugal, UK <sup>22</sup>	S; B	2008 - 2012	18-25	5,08 - 445
V Macaronesia	Spain, Portugal, UK <sup>23</sup>	S; B	2000 - 2012	18-25	28,3 - 879

**Table 4,** Levels of Polychlorinated biphenyl (PCBs) in blubber of **offshore toothed cetaceans** living in the OSPAR regions (I to V) and Marine Strategy Framework Directive (MSFD) subregions of the North-East Atlantic Ocean. Data are shown as sum of total

PCBs measured (ΣPCBs<sub>tot</sub>) as Min – Max (mg/kg, lw). Data presented here derive from the following species:

Short-beaked

common dolphin Delphir	nus delphis,	killer whale O	<i>rcinus orca</i> . The list of s	pecific congeners is g	iven after	Table 7.
OSPAR/MSFD Region	Species	Data contributor	Sampling method	Sampling period	IUPAC N.*	<b>ΣPCBs</b> tot
		Iceland <sup>15</sup>	FR	2014 - 2016	62	1,3 - 214
I Arctic Waters		Denmark⁵	FR	1995 - 2013	18-25	6,5 - 436
		USA <sup>44</sup>	FR	2005 - 2014	40	9,01 - 356
П	-	UK <sup>23</sup>	FR	1994 - 2012	18-25	23,6 - 189,2
Greater North Sea, Kattegat and		Scotland <sup>1</sup>	S	2010 - 2013	150	0,01 - 45,0
Skagerack		Germany <sup>45</sup>	S	2019	28	225
III Celtic and Irish Sea	-	UK <sup>23</sup>	FR	1997 - 2001	18-25	139 - 820
		Ireland <sup>46</sup>	S	2010 - 2017	16	10,6 - 49,3
		France <sup>24</sup>	S; B	2001 - 2003	18	0,25 - 7,04

	4					
		Spain, Portugal, UK <sup>23</sup>	FR	2006 - 2008	18-25	43,3 - 858
IV Bay of Biscay and Iberian Coast		France <sup>24</sup>	S; B	2001 - 2003	18	0,97 - 26,41
		France <sup>42</sup>	S; B	2004 - 2008	32	8,7 - 38,9
V Macaronesia	*	Spain, Portugal, UK <sup>23</sup>	FR	2009	18-25	27,53 - 247

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**Table 5,** Levels of Polychlorinated biphenyl (PCBs) in blubber of **deep-diving cetaceans** living in the OSPAR regions (I to V) and Marine Strategy Framework Directive (MSFD) subregions of the North-East Atlantic Ocean. Data are shown as sum of total PCBs

measured (SPCBstot) as Min – Max (mg/kg, lw). Data presented here derive from the following species:

long-finned pilot

whale *globicephala melas*, sperm whale *Physeter macrocephalus*. The list of specific congeners is given after Table 7.

OSPAR/MSFD Region	Species	Data contributor	Sampling method	Sampling period	IUPAC N*	ΣPCBs <sub>tot</sub>
Ι		Faroe Islands <sup>31</sup>	S	1987	10	3,15 - 71,94
Arctic waters		Iceland <sup>31</sup>	S	1982	10	8,44 -12,58
II Greater North Sea,		Scotland <sup>1</sup>	S	2010 - 2013	150	0,01 - 7,17
Kattegat and Skagerack		Germany <sup>47</sup>	S	2016	7	0,31 - 7,53
III Celtic and Irish Sea		Scotland <sup>1</sup>	S	2010 - 2013	150	2 - 9,71
IV Bay of Biscay and Iberian Coast	$ \checkmark$	France <sup>42</sup>	S; B	2006 - 2008	32	2 - 38,7

Table 6, Levels of Polychlorinated biphenyl (PCBs) in blubber of pinnipeds living in the OSPAR regions (I to V) and Marine Strategy Framework Directive (MSFD) subregions of the North-East Atlantic Ocean. Data are shown as sum of total PCBs measured

(ΣPCBs<sub>tot</sub>) as Min – Max (mg/kg, lw). Data presented here derive from the following species:

harbour seal Phoca vitulina,

bearded	seal Erignathus L	parbatus,	grey seal <i>Halichoerus</i>	grypus,	harp se	eal <i>Pagophilus</i>
groenlandicus and	ringed se	al <i>Pusa hispida</i> . The list	t of specific congeners i	is given after Table 7		
OSPAR/MSFD Region	Species	Data contributor	Sampling method	Sampling period	IUPA C N*	ΣPCBs <sub>tot</sub>
I		Norway <sup>48</sup>	S	1990 - 1991	5	0,71 - 1,34
Arctic waters		Barent Sea <sup>49</sup>	S	1993	21	0,75 - 9,81

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		Scotland <sup>1</sup>	S	2010 - 2012	150	0,01 - 8,69
		Norway <sup>48</sup>	S	1990 - 1991	5	0,712 - 1,34
	-	Norway <sup>49</sup>	Н	1993	21	0,75 - 9,81
		East Greenland <sup>50</sup>	Н	1994	10	25,7 - 226
II		Belgium, The Netherlands <sup>35</sup>	S	2000 - 2005	21	2,21 - 190
Greater North Sea, Kattegat and Skagerack		Sweden <sup>51</sup>	н	2016 - 2020	7	1,2 – 17,6
		Germany <sup>52</sup>	S	2002	13	4,95
III Celtic and Irish Sea	<b>~</b>	Ireland <sup>53</sup>	S	1988	6	5 - 70

 Table 7, Levels of Polychlorinated biphenyl (PCBs) in blubber of baleen whales living in the OSPAR regions (I to V) and Marine

 Strategy Framework Directive (MSFD) subregions of the North-East Atlantic Ocean. Data are shown as sum of total PCBs measured

(ΣPCBstot) as Min – Max (mg/kg, lw). Data presented here derive from the following: fin whale Balaenoptera physalis,

Sei whale Balaenoptera borealis, minke whale Balaenoptera acutorostrata, blue whale

Balaenoptera musculus and the Table 7.

humpback whale Megaptera novanglicae. The list of specific congeners is given after

OSPAR/MSFD Region	Species	Data contributor	Sampling method	Sampling period	IUPAC N*	<b>ΣPCBs</b> tot
		Norway <sup>54</sup>	FR	2014 - 2018	10	0,079 - 0,522
		Norway <sup>54</sup>	FR	2014 - 2018	10	0,055 - 0,334
		Iceland <sup>31</sup>	Н; В	1982 - 1986	10	0,65 - 1,87
		Iceland <sup>31</sup>	Н; В	1982	10	8,44 - 12,6
Ι		Norway <sup>55</sup>	Н	1992	21	0,60 - 20,8
North Atlantic Ocean		East Greenland <sup>55</sup>	н	1998	102	0,710 - 1,88
		Jan Mayen55	Н	1998	102	0,518 - 8,63
		Norwegian Sea <sup>55</sup>	Н	1998	102	0,422 - 8,10
		White Sea <sup>55</sup>	Н	1998	102	1,13 - 5,25
		Bering Sea <sup>55</sup>	Н	1998	102	1,02 - 11,9

		North Sea <sup>55</sup>	Н	1998	102	0,224 - 14,8
II North Atlantic Ocean		Scotland <sup>1</sup>	S	2010 - 2013	150	0,3 - 0,135
		Scotland <sup>1</sup>	S	2011 - 2013	150	0,2 - 1,36
III North Atlantic Ocean		Scotland <sup>1</sup>	S	2011 - 2013	150	0,1 - 0,532
	T	Ireland <sup>56</sup>	FR	2009 - 2012	10	0,04 - 0,337

List of measured PCB congeners:

**Σ6PCBs:** 52, 101, 118, 138, 153, 180;

**27PCBs:** 118, 153, 138, 180, 28, 52, 101;

**Σ10PCBs:** 31; 28; 52; 101; 149; 118; 153; 105; 138; 187; 156; 180; 170; 194;

**217PCBs:** 47, 49, 52, 101, 105, 118, 128, 138, 149, 151, 153, 156, 170, 180, 187, 194, 202; **218PCBs:** 28, 49, 52, 99, 101, 118, 128, 138; 141, 149, 151, 153, 170, 177, 180, 183, 187, 194;

**218PCBS:** 28, 49, 52, 99, 101, 118, 128, 138; 141, 149, 151, 153, 170, 177, 180, 183, 187, 194; **225PCBs:** 18, 28, 31, 44, 47, 49, 52, 66, 101, 105, 110, 118, 128, 138, 141, 149, 151, 153, 156, 158, 170, 180, 183, 187, 194;

**Z25PCBS:** 18, 28, 51, 44, 47, 45, 52, 66, 101, 105, 110, 112, 118, 128, 136, 141, 145, 131, 135, 136, 136, 136, 170, 180, 183, 187, 194, **Z28PCBs:** 8, 18, 28, 44, 52, 66, 70, 87, 95, 101, 105, 110, 112, 118, 128, 138, 149, 153, 156, 170, 180, 183, 187, 194, 195, 206, 209;

**Z32PCBs:** 28, 31, 52, 49, 44, 74, 70, 101, 99, 97, 110, 123, 118, 105, 114, 149, 153, 132, 137, 138, 158, 128, 156, 167, 157, 187, 183, 180, 170, 189, 194, 209;

**EX8PCBs**: 18, 28, 31, 44, 47, 49, 52, 66, 74, 77, 81, 87, 99, 101, 105, 110, 114, 118, 123, 126, 128, 138, 141, 146, 149, 151, 153, 156, 157, 158, 167, 169, 170, 172, 177, 178, 180, 183, 187, 189, 194, 195, 196, 199, 201, 203, 206, 209.

# Conclusion (brief)

- PCBs are always found in the blubber of North-East Atlantic Ocean marine mammals, with concentrations often in the range (9 – 41 mg/kg lw) of potential reproductive toxicity;
- No conclusion can be drawn with regard to spatial differences, as ΣPCBs<sub>tot</sub> range seems to be also related to species-specific traits (i.e. trophic level and prey type);

Ranges of the sum of total PCBs seem overall higher in marine mammals studied in the 70s - 80s (3 - 72 mg/kg lw) than today (0.10 - 50 mg/kg lw); A greater effort needs to be made to fill in the spatial, species and temporal gaps existing in the literature (i.e. data in grey literature or databases), as well as a structural homogenisation of analytical and data reporting protocols.

# Conclusion (extended)

This assessment aimed at assessing PCBs pollution in marine mammals from the OSPAR region, in order to set the base for the development of an indicator of North Atlantic chemical pollution in marine predators.

The main findings of this assessment underline the urgency for evaluating the trends and effects of PCBs (and other chemicals) pollution in marine mammals at the species-specific level as well as at larger spatial and temporal scales. Such efforts should also include information on age, sex, reproductive capacity, trophic position and diet into the interpretation and application of toxicity thresholds.

Despite the importance of using the sum of several PCBs (seven or 18-25) for the monitoring of pollution trends, the toxicological analysis must use a congener-specific approach. This might include assessment of PCBs trends in all congeners separately as well as studying the potential interactions between congeners. This is of key importance to harmonize the decision of CPs about which congener to prioritize in future monitoring studies, based on its specific toxic potential.

# Knowledge Gaps (brief)

The main limitation to the development of a marine mammals' "persistent chemicals indicator" is the large variability of available data with regards to species, areas, tissue, age and sex groups and list of congeners. OSPAR contracting parties recommend to:

- 1. Structure and harmonise pollutants data acquisition (i.e. species, age and sex group, list of congeners), which could be undertaken via the use of international databases (i.e. ICES);
- 2. Structure and harmonise pathological data acquisition;
- 3. Analyse old samples not included in previous studies;
- 4. Validate the existing estimated reproductive toxicity thresholds;

Push for international and national funding of toxicity studies including several health indicators (i.e. rate of infectious disease, general health).

# Knowledge Gaps (extended)

This preliminary overview of the current PCBs data available underlines the following crucial gaps:

- Major gaps exist on a temporal scale regarding the concentrations of PCBs at a congener level, since most studies until 2008 focused on the sum of the ICES 7 PCBs (118, 153, 138, 180, 28, 52, 101);
- Only few toxicity thresholds exist, which focus only on reproductive or immunotoxicological impairment;
- One of the major problems underlined in this preliminary assessment is the large variability of laboratory technics for the analysis of PCBs (and other pollutants). However, the literature has shown how the measurement of PCBs is strongly influenced by blubber extraction, pre-treatment and storage technics. Future efforts should bring the harmonisation across CPs of: (1) reference material (fish or marine mammals' tissues); (2) blubber extraction, pre-treatment and quantification limits. The same homogenate of marine mammals' blubber should be analysed in the laboratories of all CPs to assess interlaboratory repeatability;
- The CPs have underlined how the temporal coverage of PCBs data does not represent the entirety of samples, which are effectively available in tissue banks and could be analysed for persistent chemicals pollution. Indeed, a large quantity of samples from the 80s and 90s and numerous species are available and could be partially analysed to acquire PCB data for that time periods underrepresented by the available dataset. The costs of analyses have limited the acquisition of data from a wide range of samples, years, species, areas and coverage of both sexes. This data gap can still be filled in if sufficient research funding is available. Some CP (i.e. Germany) has already started to run PCBs (and other pollutants) analysis in old samples available in the tissue banks. National and international funding should push for homogenise this effort at the broad scale of the OSPAR region.

#### Next steps:

- There is a need for structural and harmonised protocols for PCBs data acquisition, including the list of congeners and the tissue matrix analysed, as well as integration on a spatial scale, in order to allow geographical comparison;
- Although very pertinent at the moment the existing estimated toxicity thresholds for reproductive impairment should undertake further work on dose response curves, in order to include different age and sex groups or species as well as should be extended to include other health aspects (i.e. immunological response and general health);
- Scientific efforts should target the analysis of excluded PCB congeners with the larger aim to get datasets over a continuous time span of 20+ years and to evaluate how well different congeners represent total contaminant loads. Environmental monitoring should then prioritize on measuring PCB congeners that are relevant for marine mammal's health and the current situation;
- Some CP (i.e. Germany) has already started to run PCBs (and other pollutants) analysis in old samples available in the tissue banks. National and international funding should push for homogenise this effort at the broad scale of the OSPAR region.

Future research should not be limited to the analysis of PCB congeners, but also include other classes of legacy (PBDEs, OCPs, HBCDs, MeHg) or emerging (PCNs, SCCPs) persistent chemicals and aim at assessing potential interactions and potential synergic effects.

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# Assessment Metadata

Field	Data Type	
Assessment type	List	Pilot Assessment
SDG Indicator	List	14.1 By 2025, prevent and significantly reduce marine pollution of all kinds, in particular from land-based activities, including marine debris and nutrient pollution
Thematic Activity	List	Hazardous Substances
<b>Conditions applying</b>	URL	https://oap.ospar.org/en/data-policy/
to access and use		



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Our vision is a clean, healthy and biologically diverse North-East Atlantic Ocean, which is productive, used sustainably and resilient to climate change and ocean acidification.

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