A Review of Organochlorine Contaminants in Nearshore Marine Mammal Predators

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Abstract
Nearshore vertebrate predators such as marine mammals and otters are apex predators, and as such, act both as drivers of the ecosystems in which they live and as sentinels of environmental health. As apex predators, these wild animals at the top of the food chain often bioaccumulate persistent organic pollutants (termed POPs) widespread throughout the environment. Organochlorines are a type of POP that tends to be lipophilic and hydrophobic which accumulate in the fatty tissues of marine mammals and other vertebrate predators over time. There has been growing concern about these POPs in the marine environment and within wildlife, as they can potentially cause health problems. Since the 1950s there have been 81,000 papers published on organochlorines in nearshore predators and potential negative effects to the environment, wildlife and humans. Here, we review organochlorines reported in the tissues of marine mammals and other nearshore vertebrate predators since 1995. We focus on five organochlorines that have been studied the most within nearshore vertebrate predators: 2,2-bis-(p-chlorophenyl)-1,1,1-trichloroethane or dichlorodiphenyltrichloroethane (DDT), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), toxaphene, and polychlorinated naphthalene (PCNs).

Keywords: Organochlorine; Contaminants; Persistent organic pollutants; Nearshore vertebrate marine mammal predators; Wildlife

Introduction
Life history characteristics such as long life spans and limited home ranges make marine mammals living indicators and sentinels of environmental health. By monitoring wildlife health, we can infer the relative health of the environment in which they live and use data from these living sentinels to investigate shifts and changes in the ecosystem. Environmental contaminants from man-made chemicals are found to be widespread, many of which have unknown toxic effects at the ecosystem, population and individual level. Some environmental contaminants are short lived and cause acute rather than chronic damage. Others are persistent in the environment and may cause chronic environmental and wildlife health problems long after they are manufactured and even banned. In addition, some may also accumulate, increasing over time in individuals (bioaccumulation) or in orders of magnitude with each trophic level (biomagnification). The most damaging to wildlife, and in turn their ecosystem, are those contaminants that are persistent and bioaccumulate.

Contaminants may enter the environment through air, soil, and water. They are easily spread throughout the nearshore marine environment primarily due to the proximity of dense human populations that facilitate the transfer of contaminants from land to water via runoff. Contaminants may also travel farther offshore from their origins via atmospheric deposition through atmospheric currents and precipitation. Because of their widespread and persistent nature, there are rising concerns on the effects of environmental contaminants on ecosystems. Organochlorines are of particular concern, as they bioaccumulate through the food chain, are lipophilic, hydrophobic and persistent in the environment. In the past two decades there has been increasing concern about the effects of persistent and bioaccumulated toxicaants in long lived apex predators in the marine environment. These apex predators include nearshore vertebrate and marine mammals. Aquatic organisms are good bio-indicators of environmental pollution because they concentrate bioaccumulative pollutants in their bodies from water and sediment, in addition to uptake from diet. There are numerous publications on marine mammal toxicology, but vary widely in methods used and variables measured. Determination of the potential impacts to marine animal health is still not completely understood. Differences in species, age, diet, geographical location, health, and proximity to environmental contaminants all can affect the chemical load an individual carries. In this review, we have compiled and reviewed nearshore marine mammal toxicology literature from the last 25 years (1990’s to present), to establish the current state of knowledge and priorities for future research.

Toxins in nearshore vertebrate predators
Species included in this review were limited to marine nearshore vertebrate predators including pinnipeds (sea lions, fur seals, and true seals), cetaceans (porpoises, dolphins, and other odontocete or toothed whales), polar bears and mustelids (sea otters and river otters). We are defining nearshore inhabitants as species that spend most or all of their life near coastal environments. Orcinus Orca, or killer whales, are the most widely ranging mammal on this list, but are also commonly found in nearshore areas [1].

Contaminants were limited to organochlorines including 2,2-bis-(p-chlorophenyl)-1,1,1-trichloroethane or dichlorodiphenyltrichloroethane (DDT), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), chlordane (CHL), hexachlorocyclohexan (HCH), polychlorinated dibenz-p-dioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), mirex, toxaphene, hexachlordobenzene (HCBs), Dieldrin, and polychlorinated naphthalene (PCNs). All organochlorines are POPs, are persistent and bioaccumulate. The organochlorines that have been studied the most...
with nearshore vertebrate predators include: DDT, PCBs, PBDEs, Toxaphene and PCNs, and are the main focus of this review.

After adhering to these definitions of marine nearshore vertebrate predators and limiting the toxins reviewed, a total of 47 papers and 36 species were utilized for this review (Table 1). Of the total species, there were 14 different pinnipeds, 19 cetaceans, two mustelids and one polar bear species. Of the total papers, 21 studied pinnipeds, 21 studied cetaceans, two studied mustelids and four focused on polar bears. The number of publications on toxins in marine mammals has increased dramatically in the last 25 years starting from zero in 1950 to 81,000 through 2016 (Figure 1). Sampling of toxins and analytical methods varied between papers making comparisons between studies and general conclusions difficult. Tissue samples include blubber, liver, muscle, blood and skin biopsy (Table 1). Analytical tools include high performance mass spectrometry/liquid chromatography or mass spectrometry/gas chromatography. Geographical locations varied, with most research conducted in the United States, followed by Canada and Norway (Figure 2), with the vast majority of the research on marine mammal toxicology conducted in the northern hemisphere (Figure 3).

Organochlorines are the most heavily studied environmental contaminant [2]. They are a group of industrial and agricultural compounds that are lipophilic and hydrophobic [3]. They were designed for chemical stability and thus persist in the environment. These contaminants enter the marine environment through municipal and industrial wastewater outfalls, landfill leachate, and atmospheric deposition [4].

**Dichlorodiphenyltrichloroethane (DDT):**

Dichlorodiphenyltrichloroethane (DDT) and its metabolites are the most widely described organochlorine contaminant. It has been used worldwide as a potent chemical pesticide. It is banned in most developed countries, but is still used in other parts of the world. It is a neurotoxin and can cause pathological changes to the liver and reproductive system in animals [3]. Even though it has been in widespread use worldwide, there are relatively few values for levels found within nearshore vertebrate predators presumably because DDT was banned for use in the United States in the 1970s. The highest DDT level found in nearshore marine mammals was measured in transient killer whale (Orca orca) blubber from Kenai Fjords/Prince William Sound with a very high value of 320,000 ng/g lipid weight [5]. The highest pinniped value was measured in Antarctic fur seal (Arctocephalus gazella) blubber at a value of 168 ng/g lipid weight [6]. In Nearshore vertebrate predators such as the Eurasian river otter (Lontra canadensis) blubber was found to have high values of 3,859 ng/g liver in England and Wales [7]. Lastly eastern Greenland polar bear (Ursus maritimus) blubber was found to have a moderate value of 559 ng/g lipid weight [8].

**Polychlorinated biphenyls (PCBs):** Polychlorinated biphenyls (PCBs) include 209 congeners with a multitude of uses including transformers, plastics and inks [2]. Blubber concentrations have been reported for a variety of marine mammals worldwide [3]. Manufacturing of PCBs occurred from 1929 through the late 1970s [9] but was halted in most industrial nations in the 1970s-1980s [3]. Despite this, as machinery systems leak, degrade or are disposed of, there is an expectation that levels in the environment will continue to increase [3]. Dose response relationships for many of the congeners have not been well established [9]. The highest PCB values reported within marine mammals was 18,135 μg/kg within Puget Sound harbor seal (Phoca vitulina) blubber [10]. Of the cetaceans, transient Kenai Fjords/Prince William Sound killer whale blubber had the highest value of 230,000 ng/g [5]. While Eurasian river otter liver from England and Wales had an average value of 12,928 ng/g liver tissue [7], and Polar bear blubber from East Greenland had 9,100 ng/g tissue [8].

**Polybrominated diphenyl ethers (PBDEs):** Polybrominated diphenyl ethers (PBDEs) are compounds that have been widely used as flame retardants in textiles, furniture, upholstery, plastics and electronics [9]. Two of the three commercial forms (penta and octa) were removed from the European (1998) and North American (2004) marketplaces. Deca-BDE remains on the market in both countries [9]. Although most compounds have been banned, it has been rapidly accumulating in the environment since its introduction in the 1970’s. Levels reached their peak in 2001 at 67,000 tons emitted. Their toxic effects persist and they are thought to be neurotoxic and hormone disruptors [11-13]. Concentrations in human blood and tissue have increased exponentially almost 100-fold since the 1970s, doubing about every five years [14,15]. Sources of contamination into the environment occur at production of the product and throughout the life of the product as it degrades. Since 2001 Asia has been the major producer of PBDEs and most come from e-recycling and waste that is often collected in developed countries and sent to developing countries for processing. PBDEs are then released into air, sediment, sewage sludge and water in the environment.

These compounds have been documented in many marine mammals. The presence of PBDEs was first reported in archived northern fur seal (Callorhinus ursinus) samples from the Pacific coast of Japan between 1972 and 1998 [16]. A total of eight congeners of dio- to hepta-BDEs were detected in all the samples, with concentrations ranging from 0.33 to 100 ng/g lipid weight in 1972 and 1994, with average concentration increasing about 150-fold from 1972 to 1994. However, the levels decreased by about 50% in 1998 which coincided with the withdrawal of penta-BDEs from use in Japan.

Ramu et al. measured the levels, profiles, and distribution of PBDEs in small cetaceans from Hong Kong waters. They analyzed ten targeted PBDE congeners in the blubber, liver and kidney of Indo-Pacific humpback dolphins (Sousa chinensis) and finless porpoises (Neophocaena phocaenoides) that were stranded in Hong Kong coastal waters between 1995 and 2001. The total concentrations of PBDEs ranged from 230 to 980 ng/g lipid weight in the blubber of finless porpoises and from 280 to 6000 ng/g lipid weight in the blubber of humpback dolphins. Recently, the geographical distribution of PBDEs in small cetaceans from Asian waters ranged from 6.0 to 6000 ng/g lipid weight. In contrast levels within human blood taken in Chinese workers ranged from 8.61 to 46.05 ng/g of lipid weight, with an average value of 19.33 ng/g lipid weight [17].

The highest PBDE value in pinnipeds was reported from harbor seal (Phoca vitulina concolor) liver from the Northwest Atlantic Ocean at a value of 2,670 ng/g [18]. In cetaceans, southern resident killer whale blubber had between 1,200-15,000 ng/g of PBDEs [19]. Two musteld species, California sea otter (Enhydra lutris nereis) and the Eurasian river otter, were also tested and were found to have levels of 2,170 ng/g [20] and 3,242 ng/g [7] within livers respectively. Finally polar bears from East Greenland had values of 68-75 ng/g blubber [21].

**Toxaphene:** Toxaphene is an organochlorine insecticide and is deposited all over the world via atmospheric transport. It has even been found in arctic marine mammal blubber far from its targeted use [2]. Toxaphene was primarily used in the 50s through 70s, and was banned in the US in 1982. It was the most widely used pesticide before restrictions for its use were put in place [3]. Gues et al. suggests...
Table 1: Summary of selected organochlorine residue surveys in pinnipeds, cetaceans, mustelids and polar bears from 1991-2012. Contaminant concentration includes mean or range (ng/g lipid weight, unless otherwise stated) [6-71].

| Reference | Year       | Location           | Species                                      | Sample Type | N  | Chemical compound                  | Amount (average or range lipid weight until otherwise stated) |
|-----------|------------|---------------------|----------------------------------------------|-------------|----|-------------------------------------|-------------------------------------------------------------|
| [39]      | 1991-1997  | California          | California sea lion (Zalophus californianus)  | Blubber, Liver | 15 | DDTs, PCBs, CHL, HCH              | Blubber: DDT: 13-2,900 µg/g PCBs: 7.2-1,300 µg/g CHL: 0.15-7.0 µg/g HCH: 0.17-4.5 µg/g | Liver: DDT: 12-970 µg/g PCBs: 11-410 µg/g Chlordanes: 0.44-37 µg/g HCH: Not detectable-0.040 µg/g |
| [39]      | 1991-1997  | California          | Harbor seal (Phoca vitulina)                  | Liver       | 10 | DDTs, PCBs                  | Blubber: DDT: 2.8-85 ng/g PCBs: 5.350 ng/g Chlordanes: 0.11-4 µg/g HCH: Not detectable-0.040 µg/g |
| [30]      | 1991-1997  | California          | Elephant seal (Mirounga angustirostris)       | Blubber, Liver | 15 | DDTs, PCBs                  | Blubber: DDT: 8.3-110 µg/g PCBs: 5.0-58 µg/g Chlordanes: 0.83-8.6 µg/g HCH: 0.12-0.90 µg/g |
| [40]      | 2009       | Galapagos Islands   | Galapagos sea lion (Zalophus wollebaeki)      | Muscle-Blubber | 21 | PCBs, PBDEs, PCDDs, PCDFs      | PBDEs: Trace PCDDs & PCDFs: Not detected |
| [41]      | 2012       | Strait of Georgia, BC | Stellar sea lions (Eumetopias jubatus)       | Blubber     | 22 | PBDEs, PCBs                  | PBDEs: 50-3780 µg/kg PCBs: 272-14280 µg/kg                |
| [42]      | 1999-2003  | Hudson Bay, Canada  | Ringed Seal (Pusa hispida)                    | Blubber     | 11 Males | PCBs, DDTs, HCHs, CBz, Cyclodienes, PBDEs | PCBs: 602 ng/g DDTs: 413 ng/g HCHs: 145 ng/g CBz: 78 ng/g Cyclodienes: 157 ng/g PBDEs: 11 ng/g |
| [43]      | 2001-2002  | San Francisco Bay   | Harbor seal (Phoca vitulina)                  | Blood       | 33 | PBDEs                  | PBDEs: 760 ng/g                                       |
| [44]      | 1993-2003  | California          | California sea lion (Zalophus californianus)  | Blubber     | 25 Males | PBDEs, PCBs                  | PBDEs: 5036 mg/g                                    |
| [43]      | 2000-2001  | Bristol Bay, Alaska | Spotted seal (Phoca largha)                  | Blood       | 5 Females | 2 Males | PBDEs                  | PBDEs: Females: 0.280 ng/g Males: 0.456 ng/g |
| [45]      | 2003       | Bering Sea, Alaska  | Bearded seal (Erignathus barbatus)            | Blubber     | 5  | PBDEs                  | PBDE: 3.42 ng/g                                    |
| [45]      | 2003       | Bering Sea, Alaska  | Ringed seal (Phoca hispida)                  | Blubber     | 6  | PBDEs                  | PBDEs: 5.85 ng/g                                    |
| [45]      | 2003       | Bering Sea, Alaska  | Spotted seal (Phoca largha)                  | Blubber     | 3  | PBDEs                  | PBDEs: 12.4 ng/g                                    |
| [45]      | 2003       | Bering Sea, Alaska  | Ribbon seal (Phoca fasciata)                 | Blubber     | 6  | PBDEs                  | PBDEs: 16.5 ng/g                                    |
| [10]      | 1996-1997  | Queen Charlotte Strait, Canada | Harbor seal (Phoca vitulina) | Blubber | 60 pups | PCBs, PCDDs, PCDFs | PCBs: 1,143 ± 262 µg/kg PCDDs: 96 ± 10 ng/kg PCDFs: 26 ± 4 ng/kg |
| [10]      | 1996-1997  | Strait of Georgia, Canada | Harbor seal (Phoca vitulina) | Blubber | 60 pups | PCBs, PCDDs, PCDFs | PCBs: 2,475 ± 174 µg/kg PCDDs: 279 ± 32 ng/kg PCDFs: 25 ± 13 ng/kg |
| [10]      | 1996-1997  | Puget Sound, WA     | Harbor seal (Phoca vitulina)                  | Blubber     | 60 pups | PCBs, PCDDs, PCDFs | PCBs: 18,135 ± 3082 µg/kg PCDDs: 119 ± 16 ng/kg PCDFs: 10 ± 1 ng/kg |
| [46]      | 1989-1998  | San Francisco Bay, CA | Harbor seal (Phoca vitulina Richards)         | Blubber     | 34 | PBDEs                  | All adults: 1730 ng/g Females: 449 ng/g Males: 2583 ng/g |

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| Year     | Location          | Species                  | Sample Type | Age | PCBs, PCBDeS, PCDEs, PCNs |
|----------|-------------------|--------------------------|-------------|-----|--------------------------|
| 2001 & 2003 | Hornby Island, BC | Harbor seal (Phoca vitulina) | Blubber     | 32 pups | PCBs, PBDEs, PCDEs, PCNs |
| 2001 & 2003 | Vancouver, BC     | Harbor seal (Phoca vitulina) | Blubber     | 32 pups | PCBs, PBDEs, PCDEs, PCNs |
| 2003     | Smith Island, WA  | Harbor seal (Phoca vitulina) | Blubber     | 32 pups | PCBs, PBDEs, PCDEs, PCNs |
| 2003     | Gertrude Island, WA| Harbor seal (Phoca vitulina) | Blubber     | 32 pups | PCBs, PBDEs, PCDEs, PCNs |
| 2009     | Puget Sound, WA   | Harbor seal (Phoca vitulina) | Blubber     | 24 pups | PCBs, PBDEs                |

| Year     | Location          | Species                  | Sample Type | Age | PCBs, PCBDeS, PCDEs, PCNs |
|----------|-------------------|--------------------------|-------------|-----|--------------------------|
| 2009-2010 | Svalbard, Norway  | Harbor seal (Phoca vitulina) | Plasma      | 12  | PCBs, CHLs, DDT, HCH, HCB, Mirex |
| 2004-2006 | King George Island, Antarctica | Weddell seal (Leptonychotes weddellii) | Blubber     | 2   | PBDEs, PCBs, PBDEs, HCBS |
| 2004-2006 | King George Island, Antarctica | Antarctic fur seal (Arctocephalus gazella) | Blubber     | 3   | PCBs, DDTs, HCBS, Mirex |
| 2004-2006 | King George Island, Antarctica | Crab-eater seal (Lobodon carcinophagus) | Blubber     | 2   | PCBs, DDTs, HCBS |
| 2004-2006 | King George Island, Antarctica | Southern elephant seal (Mirounga leonina) | Liver       | 1   | PCBs, DDTs, HCBS |
| 2002-2007 | Svalbard, Norway  | Ringed seal (Phoca hispida) | Liver       | 18  | DDE, HCB, Toxaphene |
| 2002-2007 | Baltic            | Ringed seal (Phoca hispida) | Liver       | 31  | DDE, HCB, Toxaphene |
| 1997-1998 | Baltic            | Gray seal (Halichoerus grypus) | Liver       | 12  | PCBs, DDT |
| 1997-1998 | Canada            | Gray seal (Halichoerus grypus) | Liver       | 18  | PCBs, DDT |
| 1997-1998 | Baltic            | Ringed seal (Phoca hispida botonica) | Liver       | 20  | PCBs, DDT |

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| Year Range  | Location          | Species/Region                        | Sampling Type | Gender(s) | Organics Measured | Concentration Range |
|------------|-------------------|---------------------------------------|---------------|------------|-------------------|---------------------|
| 1997-1998  | Canada            | Ringed seal (Phoca hispida botnica)    | Liver         | 26         | PCBs, DDT         | PCBs: 0.81 µg/g, DDT: 0.34 µg/g |
| 2001-2002  | East Greenland    | Ringed seal (Phoca hispida botnica)    | Blubber       | 6 Females  | PCBs, CHLs, PBDEs | PCBs: 686 ng/g, Chlordanes: 241 ng/g, PBDEs: 149 ng/g |
| 1999-2004  | East Greenland    | Ringed seal (Phoca hispida)            | Blubber       | 20         | PCBs, PBDEs       | PCBs: 596-680 ng/g, PBDEs: 31-38 ng/g |
| 1997-1998  | East Greenland    | Ringed seal (Phoca hispida)            | Blubber       | 5          | PCBs, DDTs, Toxaphene, CHL, HCH, HCB, PBDEs | PCBs: 1,370 ng/g, DDTs: 1,200 ng/g, Toxaphene: 38 ng/g, CHL: 400 ng/g, HCH: 67 ng/g, HCB: 16 ng/g, PBDEs: 38 ng/g |
| 1991-2005  | Northwest Atlantic| Harbor seal (Phoca vitulina concolor)  | Liver         | 56         | PBDE             | PBDE: 2,670 ng/g   |
| 1991-2005  | Northwest Atlantic| Harbor seal (Phoca vitulina concolor)  | Blubber       | 42         | PBDE             | PBDE: 2,403 ng/g   |
| 2001-2002  | San Francisco Bay, CA | Harbor seal (Phoca vitulina)      | Blood         | 35         | PCBs, PBDEs, DDE | PCBs: 5,335 ng/g, PBDEs: 760 ng/g, DDE: 6,806 ng/g |
| 1994-2009  | Brazilian Coast   | Franciscana dolphin (Pontoporia blainvillii) | Liver         | 53         | PBDEs            | PBDEs: 7-1797 ng/g |
| 2003-2004  | Indian River Lagoon, FL | Bottlenose dolphin (Tursiops truncatus) | Blubber       | 58         | PBDEs            | PBDEs: 5,860 ng/g   |
| 2003-2004  | Charleston Harbor estuary, SC | Bottlenose dolphin (Tursiops truncatus) | Blubber       | 53         | PBDEs            | PBDEs: 1,260 ng/g   |
| 2000-2002  | Mediterranean     | Striped dolphin (Stenella coeruleoalba) | Blubber       | 6 (2000), 31 (2001), 5 (2002) | DDT, PCBs | DDT: 56.22 µg/g, PCB: 90.4 µg/g, 2001: DDT: 38.11 µg/g, PCB: 54.42 µg/g, 2002: DDT: 55.09 µg/g, PCB: 75.90 µg/g |
| 1997-2011  | Pacific Islands (Hawaiian, Mariana, Micronesia) | Killer whale (Orcinus Orca) | Blubber       | 1          | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 171,000 ng/g, PCBs: 93,200 ng/g, Chlordanes: 13,600 ng/g, Toxaphenes: 6,890 ng/g, PBDEs: 938 ng/g, Mirex: 6330 ng/g, HCB: 580 ng/g, HCHs: 187 ng/g, HBCDs: 213 ng/g |
| 1997-2011  | Pacific Islands (Hawaiian, Mariana, Micronesia) | False killer whale (Pseudorca crassidens) | Blubber       | 1          | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 28,200 ng/g, PCBs: 26,200 ng/g, Chlordanes: 4,900 ng/g, Toxaphenes: 2,610 ng/g, PBDEs: 1650 ng/g, Mirex: 1080 ng/g, HCB: 364 ng/g, HCHs: 133 ng/g, HBCDs: 353 ng/g |
| 1997-2011  | Pacific Islands (Hawaiian, Mariana, Micronesia) | Pygmy killer whale (Feresa attenuata) | Blubber       | 2          | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 24.9-43.8 ng/g, PCBs: 2,350-58,200 ng/g, Chlordanes: 498-7,590 ng/g, Toxaphenes: 518-6,280 ng/g, PBDEs: 45.1-512 ng/g, Mirex: 136-1620 ng/g, HCB: 145-696 ng/g, HCHs: <5.86-52.4 ng/g, HBCDs: 48.4-248 ng/g |
| Reference | Year | Region | Species | Tissue | Concentration |
|-----------|------|--------|---------|--------|---------------|
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Melon-headed whale (*Peponocephala electra*) | Blubber | 4 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 31,100 ng/g PCBs: 15,600 ng/g Toxaphenes: 2,510 ng/g PBDEs: 409 ng/g Mirex: 1,190 ng/g HCB: 315 ng/g HCHs: 119 ng/g HBCDs: 108 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Striped dolphin (*Stenella coeruleoalba*) | Blubber | 6 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 20,000 ng/g PCBs: 13,800 ng/g Toxaphenes: 2,880 ng/g PBDEs: 258 ng/g Mirex: 725 ng/g HCB: 419 ng/g HCHs: 281 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Rough-toothed dolphin (*Steno bredanensis*) | Blubber | 1 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 16,100 ng/g PCBs: 13,800 ng/g Toxaphenes: 1,230 ng/g PBDEs: 192 ng/g Mirex: 829 ng/g HCB: 98 ng/g HCHs: 33.4 ng/g HBCDs: 80 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Bottlenose dolphin (*Tursiops truncatus*) | Blubber | 3 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 15,000 ng/g PCBs: 11,800 ng/g Toxaphenes: 1,490 ng/g PBDEs: 1,070 ng/g Mirex: 605 ng/g HCB: 378 ng/g HCHs: 136 ng/g HBCDs: 81.7 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Longman’s beaked whale (*Indopacetus pacificus*) | Blubber | 1 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 12,000 ng/g PCBs: 7,610 ng/g Toxaphenes: 1,430 ng/g PBDEs: 118 ng/g Mirex: 139 ng/g HCB: 360 ng/g HCHs: 121 ng/g HBCDs: 62.4 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Spotted dolphin (*Stenella attenuata*) | Blubber | 3 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 12,100 ng/g PCBs: 4,610 ng/g Toxaphenes: 850 ng/g PBDEs: 559 ng/g Mirex: 348 ng/g HCB: 78.9 ng/g HCHs: 21.8 ng/g HBCDs: 20 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Spinner dolphin (*Stenella longirostris*) | Blubber | 10 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 2,530 ng/g PCBs: 2,090 ng/g Toxaphenes: 533 ng/g PBDEs: 222 ng/g Mirex: 107 ng/g HCB: 134 ng/g HCHs: 29 ng/g HBCDs: 20 ng/g |
| [58]      | 1997-2011 | Pacific Islands (Hawaiian, Mariana, Micronesia) | Dwarf sperm whale (*Kogia sima*) | Blubber | 1 |
|           |      |        |         | DDT, PCBs, CHL, Toxaphenes, PBDEs, Mirex, HCB, HCHs, HBCDs | DDT: 3,030 ng/g PCBs: 1,900 ng/g Toxaphenes: 313 ng/g PBDEs: 25.5 ng/g Mirex: 106 ng/g HCB: 79.9 ng/g HCHs: <3.24 ng/g HBCDs: 17.2 ng/g |
| Year | Location                          | Species                        | Sample Type | Sex | Pollutants            | Concentration(s) |
|------|-----------------------------------|--------------------------------|-------------|-----|-----------------------|------------------|
| 1994-1999 | Kenai Fjords/ Prince William Sound | Killer whale (Orcinus Orca) | Blubber     | 77  | PCBs, DDT             |                  |
| 2002  | Ligurian and Ionian Sea           | Striped dolphin (Stenella Coerulea) | Skin Biopsy | 18  | PCBs, DDT             |                  |
| 2002  | Ligurian and Ionian Sea           | Bottlenose dolphin (Tursiops Truncatus) | Skin Biopsy | 1   | PCBs, DDT             |                  |
| 2002  | Ligurian and Ionian Sea           | Common dolphin (Delphinus Delphis) | Skin Biopsy | 3   | PCBs, DDT             |                  |
| 2002  | Ligurian and Ionian Sea           | Fin whale (Balaenoptera Physalus) | Skin Biopsy | 14  | PCBs, DDT             |                  |
| 2004-2006 | Puget Sound, Washington and Georgia Basin, Canada | Southern resident killer whale (Orcinus Orca) | Blubber | 5 Males (L-Pod) | PCBs, PBDEs, DDT, CHL, HCH, HCB |                  |
| 2007  | Puget Sound, WA                   | Southern resident killer whale (Orcinus Orca) | Blubber     | 2   | PCBs, DDT, PBDEs, CHL, HCH |                  |
| 2007  | Puget Sound, WA                   | Southern resident killer whale (Orcinus Orca) | Blubber     | 5   | PCBs, DDT, PBDEs, CHL, HCH |                  |
| Reference | Year | Location | Species | Organ (L) | Contaminants Assayed | Concentration Range (ng/g) |
|-----------|------|----------|---------|-----------|----------------------|--------------------------|
| [19]      | 2007 | Puget Sound, WA | Southern resident killer whale (Orcinus orca) | Blubber | 5 (L-pod) | PCBs, DDT, PBDEs, CHL, HCH |
|           |      |           |         |           |                      | PCBs: 5,600-55,000 ng/g | DDT: 4,300-99,000 ng/g |
|           |      |           |         |           |                      | PBDEs: 680-4,400 ng/g   | CHL: 730-9,500 ng/g   |
|           |      |           |         |           |                      | HCH: 150-750 ng/g       |                         |
| [42]      | 1999-2003 | Hudson Bay, Canada | Beluga whale (Delphinapterus leucas) | Blubber | 9 Calves, 21 males | PCBs, DDT, HCHs, CBz, Cyclodienes, PBDEs |
|           |      |           |         |           |                      | PCBs: 5,600-55,000 ng/g | DDT: 1030 ng/g |
|           |      |           |         |           |                      | PBDEs: 457 ng/g          | CHL: 9,500 ng/g |
|           |      |           |         |           |                      | HCHs: 377 ng/g           |                         |
|           |      |           |         |           |                      | Calves: 112 ng/g         |                         |
|           |      |           |         |           |                      | Males: 147 ng/g          |                         |
|           |      |           |         |           | HCBs: 497 ng/g        |                         |
|           |      |           |         |           | Dieldrin: 1005 ng/g   |                         |
|           |      |           |         |           | CHLs: 377 ng/g        |                         |
|           |      |           |         |           | HCHs: 119 ng/g        |                         |
|           |      |           |         |           | PBDEs: 377 ng/g       |                         |
|           |      |           |         |           | Calves: 27 ng/g       |                         |
|           |      |           |         |           | Males: 16 ng/g        |                         |
|           |      |           |         |           | 34 ng/g               |                         |
| [62]      | 1995-1997 | Svalbard, Norway | Beluga whale (Delphinapterus leucas) | Blubber | 10 Males | PCBs, HCB, HCH, Dieldrin, PCB |
|           |      |           |         |           |                      | PCBs: 430 ng/g          | Dieldrin: 1005 ng/g   |
|           |      |           |         |           |                      | HCB: 430 ng/g           |                         |
|           |      |           |         |           |                      | DDT: 5083 ng/g          |                         |
|           |      |           |         |           |                      | PCBs: 4,680 ng/g        |                         |
| [63]      | 2002-2003 | St. Lawrence River Estuary, Canada | Beluga whale (Delphinapterus leucas) | Liver | 6 | PBDEs, PCBs, DDT, HCB, Mirex, HCBs, Dieldrin |
|           |      |           |         |           |                      | PBDEs: 2, 210 ng/g      |                         |
|           |      |           |         |           |                      | PCBs: 31,937 ng/g       |                         |
|           |      |           |         |           |                      | DDT: 5,536 ng/g         |                         |
|           |      |           |         |           |                      | HCH: 111 ng/g           |                         |
|           |      |           |         |           |                      | PCBs: 2,549 ng/g        |                         |
|           |      |           |         |           |                      | Mirex: 47 ng/g          |                         |
|           |      |           |         |           |                      | HCBs: 121 ng/g          |                         |
|           |      |           |         |           |                      | Dieldrin: 757 ng/g      |                         |
| [63]      | 2002-2003 | Western Hudson Bay, Canada | Beluga whale (Delphinapterus leucas) | Liver | 11 | PBDEs, PCBs, DDT, HCH, Mirex, HCBs, Dieldrin |
|           |      |           |         |           |                      | PBDEs: 53 ng/g          |                         |
|           |      |           |         |           |                      | PCBs: 1,737 ng/g        |                         |
|           |      |           |         |           |                      | DDT: 284 ng/g           |                         |
|           |      |           |         |           |                      | HCH: 45 ng/g            |                         |
|           |      |           |         |           |                      | Chlorodanes: 808 ng/g   |                         |
|           |      |           |         |           |                      | Mirex: 0.59 ng/g        |                         |
|           |      |           |         |           |                      | HCBs: 144 ng/g          |                         |
|           |      |           |         |           |                      | Dieldrin: 275 ng/g      |                         |
| [64]      | 2001-2004 | Gulf of Mexico | Bottlenose dolphin (Tursiops truncatus) | Blubber | 14 | PBDEs |
|           |      |           |         |           |                      | PBDEs: 817 ng/g         |                         |
| [64]      | 2001-2004 | Atlantic Coast | Bottlenose dolphin (Tursiops truncatus) | Blubber | 6 | PBDEs |
|           |      |           |         |           |                      | PBDEs: 1130 ng/g        |                         |
| [65]      | 2002-2004 | Atlantic Coast (FL) | Bottlenose dolphin (Tursiops truncatus) | Blubber | 6 Females | PCBs, DDT, CHL, PBDEs |
|           |      |           |         |           |                      | PCBs: 891 ng/g          |                         |
|           |      |           |         |           |                      | DDT: 97 ng/g            |                         |
|           |      |           |         |           |                      | CHL: 39 ng/g            |                         |
|           |      |           |         |           |                      | PBDEs: 21 ng/g          |                         |
| [65]      | 2002-2004 | Atlantic Coast (FL) | Bottlenose dolphin (Tursiops truncatus) | Blubber | 31 Males and Juveniles | PCBs, DDT, CHL, PBDEs |
|           |      |           |         |           |                      | PCBs: 19,900 ng/g       |                         |
|           |      |           |         |           |                      | DDT: 2,980 ng/g         |                         |
|           |      |           |         |           |                      | CHL: 1,070 ng/g         |                         |
|           |      |           |         |           |                      | PBDEs: 394 ng/g         |                         |
| [56]      | 2003-2004 | Atlantic Coast (FL) | Bottlenose dolphin (Tursiops truncatus) | Blubber | 16 Females 25 Males 11 Juveniles | PBDEs |
|           |      |           |         |           |                      | PBDEs: Females: 718 ng/g |                         |
|           |      |           |         |           |                      | Males: 1,690 ng/g       |                         |
|           |      |           |         |           |                      | Juveniles: 979 ng/g     |                         |
| [56]      | 2003-2004 | Atlantic Coast (SC) | Bottlenose dolphin (Tursiops truncatus) | Blubber | 9 Females 31 Males 13 Juveniles | PBDEs |
|           |      |           |         |           |                      | PBDEs: Females: 1,153 ng/g |                         |
|           |      |           |         |           |                      | Males: 6,830 ng/g       |                         |
|           |      |           |         |           |                      | Juveniles: 7,055 ng/g   |                         |
| Study | Year | Location | Species | Tissue | Number | Contaminants | Concentration |
|-------|------|----------|---------|--------|--------|-------------|---------------|
| [64]  | 1991-1996 | Florida, western coast | Bottlenose dolphin (Tursiops truncatus) | Blubber | 11 Males 9 Females | PCBs, PBDEs | PCBs: 36,200 ng/g PBDEs: 363 ng/g |
| [64]  | 2000-2001 | Florida, western coast | Bottlenose dolphin (Tursiops truncatus) | Blubber | 14 | PCBs, PBDEs | PCBs: 240,000 ng/g PBDEs: 1,270 ng/g |
| [64]  | 2001-2004 | Florida, eastern coast | Bottlenose dolphin (Tursiops truncatus) | Blubber | 6 | PCBs, PBDEs | PCBs: 96,300 ng/g PBDEs: 1,130 ng/g |
| [64]  | 2001-2004 | Florida, western coast | Striped dolphin (Stenella coeruleoalba) | Blubber | 1 | PCBs, PBDEs | PCBs: 5,700 ng/g PBDEs: 660 ng/g |
| [66]  | 2006 | Korea | Long-beaked common dolphin (Delphinus capensis) | Blubber | 22 | PCBs, DDTs, CHL, HCH, HCB, PBDEs | | |
| [67]  | 2001-2003 | Gulf of Alaska, Aleutian Island and Prince William Sound | North Pacific killer whale (Orcinus orca) | Blubber | 14 Residents | PCBs, DDT, CHL, HCHs | PCBs: 15,000 ng/g DDT: 25,000 ng/g Chlordane: 5,700 ng/g HCHs: 470 ng/g |
| [67]  | 2001-2003 | Gulf of Alaska, Aleutian Island and Prince William Sound | North Pacific killer whale (Orcinus orca) | Blubber | 2 Offshores | PCBs, DDT, CHL, HCHs | PCBs: 66,000 ng/g DDT: 170,000 ng/g Chlordane: 6,600 ng/g HCHs: 120 ng/g |
| [67]  | 2001-2003 | Gulf of Alaska, Aleutian Island and Prince William Sound | North Pacific killer whale (Orcinus orca) | Blubber | 5 Transients | PCBs, DDT, CHL, HCHs | PCBs: 150,000 ng/g DDT: 270,000 ng/g Chlordane: 72,000 ng/g HCHs: 11,500 ng/g |
| [68]  | 2003 | Korean coast | Finless porpoise (Neophocaena phocaenoides) | Blubber | 7 Males 10 Females | PCBs, DDTs, CHLs, HCHs, HCB, PBDEs | | |
| [20]  | 1992-2002 | California | California sea otter (Enhydra lutris nereis) | Liver | 80 Females | PBDEs | PBDEs: 2,170 ng/g |
| [7]   | 1988-2008 | England and Wales | Eurasian river otter (Lutra lutra) | Liver | 129 PBDE 86 (DDT and PCB) | PBDEs, DDT, PCBs | PBDE: 3,241.9 ng/g DDT: 3,859 ng/g PCB: 12,928.2 ng/g |
| [8]   | 1999-2001 | East Greenland | Polar bear (Ursus maritimus) | Blubber | 50 Subadult | PCBs, HCH, DDT, CHL, Mirex, Dieldrin | PCBs: 6,470 ng/g HCH: 198 ng/g DDT: 462 ng/g CHL: 2,010 ng/g Mirex: 4.06 ng/g Dieldrin: 218 ng/g |
[8] 1999-2001 East Greenland Polar bear (Ursus maritimus) Blubber 25 Adult females PCBs, HCH, DDT, CHL, Mirex, Dieldrin PCBs: 8,240 ng/g HCH: 263 ng/g DDT: 462 ng/g CHL: 2,220 ng/g Mirex: 2.79 ng/g Dieldrin: 208 ng/g

[8] 1999-2001 East Greenland Polar bear (Ursus maritimus) Blubber 16 Adult males PCBs, HCH, DDT, CHL, Mirex, Dieldrin PCBs: 9,100 ng/g HCH: 218 ng/g DDT: 559 ng/g CHL: 1,710 ng/g Mirex: 6.59 ng/g Dieldrin: 245 ng/g

[21] 1999-2001 East Greenland Polar bear (Ursus maritimus) Blubber 52 Subadult females 23 Adult females 17 Adult males PBDEs PBDE: Subadult: 68 ng/g Female: 69 ng/g Male: 75 ng/g

[69] 1999-2001 East Greenland Polar bear (Ursus maritimus) Liver 20 PCBs, PBDEs PCBs: 3,125 ng/g PBDEs: 40 ng/g

[69] 1999-2001 East Greenland Polar bear (Ursus maritimus) Blubber 20 PCBs, PBDEs PCBs: 5,387 ng/g PBDEs: 83 ng/g

[70] 2003 Alaskan Beaufort Sea Polar bear (Ursus maritimus) Blubber 57 PCBs, DDT, CHL, HCH PCBs: 7,818 ng/g DDT: 165 ng/g CHL: 1,478 ng/g HCH: 490 ng/g

[71] 2007 Hudson Bay Polar bear (Ursus maritimus) Blubber 12 PBDEs, PCBs, CHL, DDT, Mirex PBDEs: 35.4-38.8 ng/g PCB: 2,559-3,652 ng/g CHL: 2,697 ng/g DDT: 105 ng/g Mirex: 44.7 ng/g

Figure 1: Significant increase in publications using keywords “toxicology of marine mammals” in Google Scholar over time. Zero papers were published in 1950, increasing to 81,000 through 2015.
that reported toxaphene levels are either over- or under-estimated, due to differences between sample patterns and standards. As the number and pattern of congener mixtures in the environment is much different than the technical mixtures when made, they suggest that total values given should be considered "only indicative." The LD50 of lab mammals ranged from 5 to 1075 mg/kg depending on species and route of exposure [22]. The highest toxaphene value over the last twenty years measured in pinnipeds was reported as 38 ng/g in East Greenland ringed seal blubber [23] and the highest value in cetaceans was found to be 8,206 ng/g in killer whale blubber in Norway [24]. Only two publications reported values for pinnipeds [23], and two publications were found for cetaceans [24]. All four publications collected samples either in the Pacific Island chain or between East Greenland and Norway. There were no toxaphene values given for polar bear and mustelid samples in our analysis.

**Polychlorinated naphthalenes (PCNs):** Polychlorinated naphthalenes (PCNs) were originally used as flame retardants and dielectric fluids for capacitors as early as 1900. Today, manufacture of PCNs is thought to have ended, although illegal importation of PCN-containing products into Japan was reported after 2000 [25,26]. Sources to the environment include evaporation from old or in-use products containing PCNs and PCBs, and release of PCNs during combustion [27]. Polychlorinated naphthalenes are globally distributed via air, sediments, water and biologically through the food chain. They are easily transported through the atmosphere and have been found in remote areas of the arctic and subarctic regions. They are lipophilic, hydrophobic and bioaccumulate in inverts, fish, seabirds and marine mammals.

Average PCNs have been reported in a variety of arctic marine mammals: Helm et al. reported values within eastern Canadian ringed seal (*Phoca hispida*) of 0.045 ng/g within three males and 0.051 ng/g within three females collected in 1993 [28]. A follow up study from...
1999-2003 found much higher values of 0.33 ng/g within two males and 0.25 ng/g within 19 females [29]. Seven seals in Svalbard tested in 1981 had 0.038 ng/g [30]. Vorkam et al. reported an upper limit of 0.13 ng/g for east and west Greenland seals [31]. Wang et al. reported 39 harbor seals in gulf of Alaska (prince William sound and Kodiak island) between 2000-2001 having an average of 4.8 ng/g blubber, 1.1 ng/g liver, and 0.59 ng/g kidney [32]. Weddell seals from terra nova bay, Antarctica had levels of 0.077 ng/g blubber and 1.63 ng/g liver [33]. Total PCN concentrations in seal blubber were positively and significantly correlated to age [34]. Canadian arctic beluga sampled in 1994 was found to be 0.33 ng/g within males and 0.18 ng/g within females [28]. While levels found in blubber of Arctic beluga collected later between 1999-2000 averaged 0.25 pg/g for males and 0.14 pg/g for females [29]. Killer whales from the northeastern pacific had levels of 21.6 ng/g within northern residents, 20.4 ng/g within southern residents and 167 ng/g within transient whales [35].

Other organochlorines: Other organochlorines contaminants have been tested in nearshore vertebrate predators and marine mammals, however data is lacking and non-comprehensive (Table 1). The following chemicals have been investigated within nearshore mammals for which there is very limited data: Mirex is an insecticide used for ants and termites, and as a flame retardant in plastics and paints. Hexachlorocyclohexan (HCH) is an insecticide and is neurotoxic, structurally related to Mirex. Polychlorinated dibenz-p-dioxins (PCDDs) and polychlorinated dibenzofurans (PCDFs) are by products of industrial processes. Hexachlorobenzene (HCB) is a fungicide for seed grain and is used to produce solvents and dyes. Chlordane is an organochlorine pesticide, which is now banned in Canada. It was primarily used in the 60s and 70s. Dieldrin is an organochlorine insecticide originally produced in 1948, was widely used until the 1970s and now is banned because of its persistence in the environment.

Conclusion

There are still major limitations to our knowledge of contaminants in nearshore vertebrate predators and marine mammals. Analysis of patterns remains complex due to lack of wide scale long term results. Lab studies often expose captive animals to a single chemical at high doses for a short amount of time (acute high doses), making it difficult to extrapolate what effect low chronic doses from environmental exposure have on wildlife [36]. Free ranging wildlife is also exposed to not only one but often a complex cocktail of persistent organic pollutants. In addition uncontrolled or unknown variation in the environment (exposure to degraded mixtures that differ from original products) and the individuals such as differences in bioaccumulation potentials and excretion capabilities of nearshore vertebrate predators and marine mammals may produce vastly different physiological results compared to controlled lab studies [37]. Differences in species, age, health, sex, and proximity to human populations also complicate patterns in the data. Adding further complexity to interpreting existing data is that sampling and analytical methodology for many organochlorines is not standardized making the results not easily comparable and long term studies building on previous work challenging. An internationally standardized set of analytical methods and quality assurance procedures has yet to be developed making all existing studies using different methods and reporting in different units difficult to compare (3, Table 1). In the future it is essential that methods are standardized to ensure a better understanding of how much POPs are in wildlife and what the corresponding health and environmental effects are.

Organochlorines are manufactured and used worldwide, however there is a geographic skew in past and current research. Vos et al. draws attention to the fact that 90% of toxin samples are from the northern hemisphere, primarily Europe, Canada and the United States [38]. Although it is understood that data is geographically specific, more research is needed in varying parts of the world to analyze variance between locations and identify toxic hotspots. Since many of these compounds may travel far distances because of atmospheric deposition, it is imperative that data is collected and understood from all over the world.

Of the 36 species selected for this review (text and Table 1), three are listed as endangered on the ICUN Red List of Threatened Species. This includes the Galapagos sea lion (Zalophus wollebaeki), fin whale (Balaenoptera physalus), and California sea otter. Three species are listed as vulnerable, Franciscana dolphin (Pontoporia blainvillii), finless porpoise, and polar bear. The three species that are near threatened are Steller sea lion (Eumetopias jubatus), beluga whale, and Eurasian river otter. Many were of least concern including California sea lion (Zalophus californianus), harbor seal, ringed seal, bearded seal (Erignathus barbatus), weddell seal, Antarctic fur seal, crabeater seal (Lobodon carcinophagus), southern elephant seal (Mirounga leonina), grey seal, bottlenose dolphin (Tursiops truncates), striped dolphin (Stenella coeruleoalba), melon-headed whale (Peponocephala electra), rough toothed dolphin (Steno bredanensis), and common dolphin (Delphinus delphis). What is important to note are those species that are listed as data deficient including spotted seal (Phoca largha), ribbon seal (Histiophoca fasciata), killer whale, false killer whale (Pseudorca crassiden), pygmy killer whale (Feresa attenuata), Longman’s beaked whale (Indopacetus pacificus), spinner dolphin (Stenella longirostris), dwarf sperm whale (Kogia sima), Blainville’s beaked whale (Mesoplodon densirostris), Guiana dolphin (Sotalia guianensis), and long beaked common dolphin (Delphinus capensis). These data gaps only highlight the need for continued research into potential health and reproductive effects of contaminants on these species. Due to the fact that new chemicals and compounds are being synthesized every day, it is crucial not only to track legacy contaminants but also new ones within nearshore vertebrate predators including marine mammals.

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