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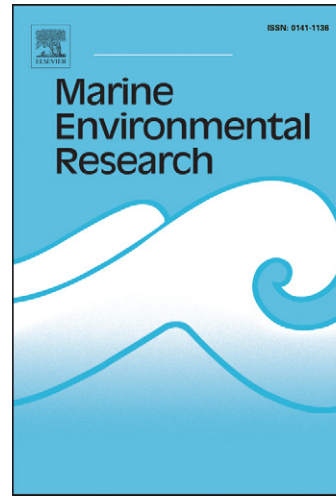

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Persistent Organic Pollutants (POPs) in killer whales (*Orcinus orca*) from the Crozet archipelago, southern Indian Ocean

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Abstract

Persistent Organic Pollutants (POPs), including polychlorinated biphenyls (PCBs), polychlorinated dibenzo-*p*-dioxins (PCDDs), and dibenzofurans (PCDFs), are ubiquitous environmental contaminants of which significant concentrations are reported in upper trophic level animals. In 1998, we collected blubber biopsy samples (n=11) from killer whales (*Orcinus orca*) inhabiting the coastal waters around Possession Island, Crozet Archipelago, southern Indian Ocean, for contaminant analyses. Despite inhabiting an isolated region far removed from industrial activities, these killer whales can presently be considered among the most PCB-contaminated cetaceans in the southern hemisphere, with concentrations ranging from 4.4 to 20.5 mg/kg lipid weight (lw). PCDD levels ranged from below the detection limit (5 ng/kg) to 77.1 ng/kg lw and PCDF levels from below the detection limit (7 ng/kg) to 36.1 ng/kg lw. Over 70 % of our study animals had PCB concentrations which exceeded a 1.3 mg/kg PCB threshold established for endocrine disruption and immunotoxicity in free-ranging harbour seals, suggesting that organic contaminants cannot be ruled out as an additional threat to this declining population.

Keywords: killer whale, Crozet Archipelago, PCBs, PCDDs, PCDFs, TEQs

1. Introduction

Polychorinated biphenyls (PCBs) and polychlorinated dibenzo-*p*-dioxins / dibenzofurans (PCDDs/Fs) are persistent organic pollutants (POPs). They are hydrophobic, have moderate vapour pressure, and low reactivity that allow them to be readily transported in the environment. “Global distillation” was described as the process by which POPs evaporate in the warmer regions and are then transported in the atmosphere towards the poles where they condense and are deposited. Cold regions, such as the Arctic and the Antarctic, have thus been described as important sinks for these contaminants (Wania and Mackay, 2001).

PCBs were extensively used by industrialized nations until the late 70s when they were largely banned. They were detected for the first time in the Antarctic environment in the early 70s. Atmospheric transport is believed to be the dominant pathway for PCB contamination in Antarctica where local sources are thought to be restricted to electrical equipment at isolated research stations (Risebrough et al., 1990).

PCDD/Fs are formed as by-products during different industrial and thermal processes such as emissions from metallurgical industries, municipal incinerators, pulp and paper mills, and the manufacture of chlorinated chemicals. Atmospheric transport is also thought to be the dominant pathway for the delivery of dioxins and furans to the Antarctic along with minor local sources such as incinerators at research stations (Lugar et al., 1996).

As long lived, high level trophic feeders, marine mammals tend to bioaccumulate POPs in lipid tissues. Marine mammals from Europe, North America, Asia, and the Arctic have been found to be particularly contaminated with PCBs (Bergman et al., 2001;

Kajiwara et al., 2006; Muir et al., 2000; Ross et al., 2000). Marine mammal-eating transient killer whales (*Orcinus orca*) from the Northeastern Pacific Ocean are reported as among the most PCB-contaminated marine mammals in the world (Ross et al., 2000) and killer whales from Norway are the most PCB-contaminated animals in the Arctic (Wolkers et al., 2007).

The biology of killer whales from the coastal waters of Possession Island, Crozet Archipelago, southern Indian Ocean, has been studied since the late 1980s, but no information exists on their exposure to POPs. Killer whales are sighted in the area year round but some photo-identified individuals have been seen at the nearby Kerguelen Island and close to the Antarctic continent. Killer whales in the Crozet Archipelago feed on a variety of prey including fish, penguins (*Eudyptes sp.*), southern elephant seals (*Mirounga leonina*) and, occasionally, large cetaceans (Guinet, 1991). Between 1988 and 2002, this population declined by 50% due to the decline of an important prey species (the southern elephant seal), interactions with the Patagonian toothfish (*Dissostichus eleginoides*) longline fishery, and/or emigration of individuals or groups from the coastal waters of the Crozet Archipelago (Poncelet et al., 2008).

POPs are known to cause hormone disruption, immunosuppression, and reproductive failure in other marine mammals (Mos et al., 2007a; Reijnders, 1986; Ross et al., 1995; Tabuchi et al., 2006). Such persistent contaminants can represent a population-level conservation threat to killer whales (Ross, 2006), with heavy POP accumulation largely due to their high trophic position and long lifespan (Hickie et al., 2007). In analysing PCB, PCDD, and PCDF concentrations in blubber biopsies taken from the declining Crozet Archipelago population, we aimed to provide a much-needed baseline for these priority contaminants, and to characterize associated health risks.

2. Materials and methods

2.1 Sampling

The study was carried out in the Crozet Archipelago, southern Indian Ocean (46º25’S; 51º59’E) (Figure 1). Lightweight pneumatic darts were used to biopsy killer whales inhabiting the coastal waters of Possession Island. The variable-power dart projector and its stainless-steel, 6.4 mm diameter tip, as well as a full description of the sampling procedure, is described elsewhere (Barrett-Lennard et al., 1996). This method was adapted to enable shore-based, rather than ship-based, collections.

Briefly, biopsy sampling was conducted from shore with a fine fishing line attached to the dart in order to retrieve biopsies taken posterior to, and below, the dorsal fin of the animal. Samples of epidermal, dermal, and hypodermal tissue typically weighed approximately 0.5g. Skin from the biopsy samples was preserved in dimethyl sulphoxyde and a saturated salt solution and stored at 4ºC for DNA extraction. Blubber from the biopsies was placed in pesticide grade hexane-rinsed glass vials with aluminium foil-covered caps and stored at -20ºC for contaminant analyses. 11 blubber biopsies were collected from nine different killer whales. Genetic results from the present samples, combined with the photo-identification work conducted previously (Guinet, 1991), enabled a determination of sex as well as age class for each of the nine individuals.

2.2 Contaminant analyses

Blubber samples were analyzed for PCDDs, PCDFs, and mono-ortho, di-ortho, and non-ortho (planar) PCBs. Thawed blubber was ground in a porcelain mortar and pestle with 200 g of anhydrous sodium sulphate and spiked with a mixture of ^{13}C-labelled PCBs, PCDDs, and PCDFs. The blubber-sodium sulphate mixture was transferred to an extraction column and extracted with 250 mL of 1:1 dichloromethane/hexane (DCM/hex) from a glass column by gravity flow. The extract was evaporated to dryness and the lipid content of the samples was determined. Analyses of cleaned-up samples were conducted by high resolution gas chromatography / high resolution mass spectrometry (HRGC/HRMS). Procedural blanks were analyzed along with samples to assess the integrity of the analytical procedures. Details on the sample clean-up, instrumental analysis condition used, quantification protocols, criteria used for congener identification and the quality assurance / quality control (QA/QC) measures undertaken for the HRGC/HRMS analysis of all the analytes of interest are described elsewhere (Ikonomou et al., 1999; Ikonomou et al., 2001; Rantaleinen et al., 1998).

2.3 Data Analysis

Two killer whales were biopsied twice allowing for replicate comparison. For one of these two whales, contaminant concentrations differed between the two samples. However, in the case of this individual, one of the biopsies was taken on the dorsal ridge just behind the dorsal fin. This sample was low in lipid content, not representative of a typical biopsy and therefore eliminated from our analysis. For the other whale, the two biopsies provided similar results (2.3% and 8.6% difference between the two samples for PCBs and PCDDs/Fs, respectively). Average contaminant concentrations were considered for this whale. Although we were unable to age sampled individuals, we were able to divide our study animals into three categories for POP comparisons: juvenile females (n = 3), adult females (n = 5) and adult male (n = 1).

All the concentrations were expressed on a lipid weight basis (lw) and were blank-corrected as well as recovery-corrected using the stable isotope dilution method based on ^{13}C-labelled PCB, PCDD, and PCDF spikes (Ikonomou et al., 2001) (the recovery ranges were a mean of 60% for PCBs (range 20-95%), 82% for PCDDs (50-105%), and 81% for PCDFs (60-100%)).

Many congeners were not detected. When congeners were undetected, substitutions were applied according to the following rule: 1) when congeners were detected in less than 70% of the samples, concentrations of 0 was substituted; and, 2) when congeners were detected in more than 70% of the samples, a detection limit value substitution was applied.

Total Toxic Equivalents (TEQ) to 2,3,7,8-tetrachlorodibenzo-*p*-dioxin (TCDD) were calculated for all dioxin-like PCBs measured (mono-ortho PCBs 105, 114, 118, 123, 156, 157, 167, and 189; non-ortho PCBs 77, 81, 126, and 169) and 2,3,7,8-Cl substituted PCDDs (n = 7) and PCDFs (n=10) using the international Toxic Equivalency Factors (TEF) (Van den Berg et al., 2006).

2.4 Statistical analysis

Differences in contaminant levels were assessed using an unpaired t-test between juvenile and adult females. The only adult male was considered separately due to evidence in the literature of sex-based differences in POP concentrations (Ross et al., 2000).

3. Results and discussion

PCB and PCDD/F data were available for 10 blubber biopsies taken from nine killer whales inhabiting the coastal waters of Possession Island, Crozet Archipelago. This represents the first information on POP contamination for this declining population of killer whales.

3.1 PCBs

3.1.1 Concentrations

Of the 209 PCB congeners sought, we detected 124 ± 8 (range: 119 – 134) congeners in our killer whale samples. PCB levels ranged from 2.7 to 20.5 mg/kg lw (Table 1). Concentrations in juvenile and adult females did not differ ($p > 0.05$), in contrast to reported findings for killer whales from the Northeastern Pacific Ocean, where immature whales had higher PCB concentrations than adult females (Ross et al., 2000). While the only male killer whale in the present study had the highest PCB levels of all individuals sampled in our study (Figure 2, Table 1), this could not be statistically compared with our other age/sex categories. Higher POP concentrations in males compared to females have been reported in various species of marine mammals including killer whales from the Northeastern Pacific Ocean (Ross et al., 2000) and in pilot whales (*Globicephala melaena*) (Borrell et al., 1995). In marine mammals, a continuous increase of POP concentrations with age is observed in immature individual of both sexes and in adult males. In adult females, concentrations start to decline when they reach sexual maturity because of the offload of fat-soluble contaminants to their young during gestation and nursing (Borrell et al., 1995; Ross et al., 2000; Ylitalo et al., 2001).

The Possession Island killer whales were approximately 10 times more contaminated than the type C killer whales sampled recently from the southern Ross Sea adjacent to Antarctica (Krahn et al., 2008), likely reflecting a difference in diet and possibly the remote location of the Ross Sea. While PCB concentrations in our whales were lower than those reported in killer whales from industrialized areas (Table 2), they were similar to those observed in resident killer whales from Alaska (Ylitalo et al., 2001), and only slightly lower than the levels in northern residents from the Northeastern Pacific Ocean (Ross et al., 2000) and Norwegian killer whales (Wolkers et al., 2007). As the latter killer whales are considered the most contaminated Arctic marine mammal, our results again underscore the vulnerability of high trophic level marine mammals in polar and otherwise remote regions.

However, the Norwegian killer whales and the resident killer whales of the Northeastern Pacific Ocean are fish-eating, while the Possession Island killer whales appear to consume a mixed diet of mostly marine mammals but also fish and penguins (Guinet, 1991). Recently, a juvenile male was found stranded on Possession Island and the stomach content revealed remains of elephant seals, fur seals (*Arctocephalus sp)*, and king penguins but no fish. Dietary preferences are a major factor affecting contaminant concentrations in marine mammals. In the Northeastern Pacific Ocean, transient marine mammal-eating killer whales are far more contaminated than the resident fish-eating killer whales (Ross et al., 2000). Based on the limited data available, the diet of the Possession Island killer whales seems trophically comparable to transient killer whales of the Northeastern Pacific Ocean and thus comparing contaminant burdens between these two populations provides at least a crude evaluation of differences in regional contamination. The present levels in the

Possession Island killer whales were one fifth to one tenth of the levels recorded in transients from the Northeastern Pacific Ocean (Ross et al., 2000) and Alaska (Ylitalo et al., 2001). These differences could be the result of dietary preferences but may also be due to the remote nature of Possession Island.

PCB concentrations in Crozet killer whales were higher than levels reported in marine mammals from Antarctica, including southern elephant seals (Miranda-Filho et al., 2007), and minke whales (*Balaenoptera acutorostrata*) (Aono et al., 1997). The Crozet killer whales were less PCB-contaminated than only two other southern hemisphere species, namely the estuarine dolphins (*Sotalia guianensis*) and Atlantic spotted dolphins (*Stenella frontalis*) inhabiting the nearshore waters of Brazil (Kajiwara et al., 2004). The Crozet whales had higher PCB concentrations than Arctic belugas (*Delphinapterus leucas*) (Helm et al., 2002) that are at the receiving end of PCB dispersion from northern hemispheric industries.

3.1.2 Homologue group and congener patterns

No significant differences ($p > 0.05$) in PCB homologue group and congener patterns were observed between juvenile and adult females (Figure 3). The adult male also revealed a similar pattern. In all the three groups, di-ortho PCBs were highly dominant (97.5 ± 1.1 % of ∑PCBs) (Figure 2). Hexa-CBs was the dominant homologue group, accounting for 55 ± 3 % of total PCBs, followed by hepta- (23 ± 2 %) and penta-CBs (11 ± 3 %). A similar pattern was found in southern elephant seals but they had a more even contribution of hepta and penta-CBs (Miranda-Filho et al., 2007). The six dominant congeners in our killer whales accounted for 55 ± 1 % of ∑PCBs (Table 1).

Similar to our results, PCBs-153 and 138 were among the dominant congeners in the three populations of killer whales from the Northeastern Pacific Ocean (Ross et al., 2000), in killer whales from Alaska (Ylitalo et al., 2001), and Norway (Wolkers et al., 2007). In addition, PCB 153 and 138 dominated PCB patterns in southern elephant seals (Miranda-Filho et al., 2007) as well as in lower trophic levels of the Antarctic killer whale food web (Goerke et al., 2004). This is likely due to the fact that PCB-153 was a major component of the most heavily used PCB technical mixture, and, as it has no vicinal hydrogen and is Cl-substituted at both para positions, is resistant to metabolism (Boon and Eijgenraam, 1988).

3.2 Dioxins and furans

3.2.1 Concentrations

Dioxin and furan concentrations were very low with many congeners being undetected. Out of the 75 possible dioxin congeners, only up to 3 congeners were detected. The total PCDD concentrations ranged from below the mean detection limit (6.8 ± 2.3 ng/kg) to 77.1 ng/kg lw with a mean concentration of 17.5 ± 7.8 ng/kg lw (Table 2). Out of the 135 possible furan congeners, only up to two congeners were detected in our whales and the total PCDF concentrations ranged from below the mean detection limit (3.4 ± 1.4 ng/kg) to 113.1 ng/kg lw with a mean concentration of 5.1 ± 3.6 ng/kg lw.

There were no significant differences in PCDD/F concentrations between juvenile and adult female killer whales from Possession Island (Table 2). The adult male was the most PCDD/F contaminated, as observed in killer whales from the Northeastern Pacific Ocean (Ross et al., 2000), but the small sample size precluded statistical evaluation. The apparently higher dioxin and furan concentrations in adult females compared to juvenile females were not supported statistically ($p > 0.05$). The high standard error reported for adult females was due to only one highly PCDD contaminated individual. Due to our limited sample size and therefore lack of strong statistical basis, it was not clear whether or not the higher concentrations reported in adult females compared to the juveniles could be an indication that dioxins and furans are not transferred as efficiently as PCBs from mother to calf.

Data on dioxins and furans in killer whales are limited (Table 2). PCDD/F concentrations in our study were higher than those reported for killer whales from Japan (Kajiwara et al., 2006), but lower than levels reported for the three different communities of killer whales living in the Northeastern Pacific Ocean (Ross et al., 2000). The higher PCDD/F concentrations observed in the latter killer whales may reflect local contamination by pulp and paper mills (Ross et al., 2004). Despite these observations, however, PCDD/F concentrations in all killer whales are relatively low, something that has been attributed to the minimal biomagnification of planar "dioxin-like" compounds in aquatic food webs as a consequence of metabolism and excretion (Ross et al., 2000). The duality of distance from source (remote location) and limited biomagnification in aquatic food webs likely explains the low levels of dioxins and furans in Crozet killer whales.

Total PCDD/F concentrations in killer whales from Possession Island were higher than levels previously reported for other Antarctic marine mammals such as Weddell seals (Corsolini et al., 2002) and Antarctic fur seals (*Arctocephalus gazella*) (Oehme et al., 1995). However, they were lower than levels reported for south polar skuas (*Catharacta maccomicki*) which migrate to more contaminated areas in lower latitudes (Senthilkumar et al., 2002). Compared to Arctic marine mammals, our observed concentrations were in the same range as reported in ringed seals (*Pusa hispida*) (Norstrom and Simon, 1990), but higher than levels in belugas from Baffin Island (Norstrom et al., 1992).

3.2.2 Homologue group and congener patterns

Only congeners from the hexa and octa homologue groups were detected, accounting for 30 ± 35 % and 56 ± 37 % of ∑PCDD, respectively. 1,2,3,6,7,8 HxCDD accounted on average for 65 ± 15 % of ∑HxCDD. This particular congener was previously reported as being dominant in several species of marine mammals including killer whales from Japan (Kajiwara et al., 2006), Weddell seals from Antarctica (Senthilkumar et al., 2002), harbour seals (*Phoca vitulina*) from British Columbia (Addison et al., 2005). This congener has, in part, been attributed to the historical use of elemental chlorine by pulp mills, and to woodchips contaminated by pentachlorophenol (Addison et al., 2005). OCDD was also detected in Arctic ringed seals (Norstrom and Simon, 1990). OCDD originates from pulp mills, combustion, or high temperature industrial processes in which there is a chlorine source (Norstrom and Simon, 1990). The remote nature of Possession Island thus suggests that PCDD contamination is most likely due to long range atmospheric transport but could also be in part due to small-scale combustion-related activities nearby, such as shipping or research operations (Lugar et al., 1996).

No PCDF congeners were detected in juvenile females. 1,2,3,4,7,8 HxCDF, 1,2,3,4,6,7,8 HpCDF, 1,2,3,4,7,8,9 HpCDF, and OCDF were the only congeners detected in the adult females. 2,3,7,8 TCDF was the only congener detected in the adult male. Even though the patterns appeared different among the three groups, no clear trend can be concluded as the sample size was small and the number of analytes detected very limited.

3.3 Toxic Equivalents (TEQ)

Consistent with the POP concentration patterns, juvenile females had similar TEQ to adult females (76.5 ± 5.0 ng/kg lw and 44.2 ± 8.4 ng/kg lw, respectively) ($p > 0.05$), while the adult male had the highest TEQ (109.1 ng/kg lw) (Table 3). Again, however, the small sample size precluded statistical interpretation of the single male sample. PCBs contributed up to 99.9% of the total TEQ in the killer whale samples, underscoring the recalcitrance of this contaminant class. The adult male appeared to have a proportionately lower contribution of PCBs to TEQ (88.3%) than did the immature (99.8 ± 0.1%) and adult females (97.3 ± 1.7%), although the limited sample size precludes a rigorous evaluation. Non-ortho PCB congeners contributed to most of the PCB TEQ (Table 3). This pattern differed to those previously reported for other cetaceans species including killer whales from the Northeastern Pacific Ocean (Ross et al., 2000), in which non-ortho and mono-ortho PCBs contributed more equally to the ∑PCB TEQ. However, our use of new TEF values (2005) partly explains this difference in observations; similar non-ortho and mono-ortho PCB contributions to the ∑PCB TEQ was observed when we applied TEFs from 1998 to this dataset.

CB-118, 169, 126, and 105 were the dominant contributors to the total TEQ, consistent with previous studies of cetaceans (Corsolini et al., 2003; Focardi et al., 1995; Ylitalo et al., 2001).

3.4 Toxicological implications

Organic contaminants are a concern to the health of marine mammals, with observations of impaired reproduction, skeletal lesions, kidney damage, tumors, premature birth and skin lesions in populations inhabiting contaminated areas (Beckmen et al., 1997; Béland et al., 1993; Bergman et al., 2001; Delong et al., 1973; Olsson et al., 1992). High PCB concentrations have been linked to decreased immune function in field studies of harbour seals (Mos et al., 2006), bottlenose dolphins (*Tursiops truncatus*) (Lahvis et al., 1995) and polar bears (Lie et al., 2005), as well as captive feeding studies of harbour seals (De Swart et al., 1994; Ross et al., 1996). Contaminant-related immunotoxicity has been, in part, blamed for serious outbreaks of infectious disease outbreaks in marine mammals (Osterhaus et al., 1995). In addition, PCBs have been implicated in the disruption of vitamin A and thyroid hormone systems in harbour seals, which could lead to adverse effects on growth and development (Mos et al., 2007a; Tabuchi et al., 2006).

Different thresholds for adverse health effects have been established for PCBs in marine mammals. Immunotoxicity and endocrine disruption were observed in harbour seals at a concentration of 209 ng/kg total TEQ (De Swart et al., 1994; Ross et al., 1995). This value was adjusted to 255 ng/kg TEQ for killer whales from the Northeastern Pacific Ocean owing to a broader analytical evaluation (Ross et al., 2000). None of our Crozet killer whales surpass this threshold.

Thresholds have also been determined for PCBs in marine mammals. A threshold value of 17 mg/kg lw was established for immunotoxicity in captive harbour seals (Ross et al., 1996), while a 10 mg/kg lw threshold was associated with an increase in calf mortality in bottlenose dolphins (Hall et al., 2006). Recently, a more protective value of 1.3 mg/kg was proposed for the protection of marine mammal health based on the 95% confidence interval for endocrine disruption and immunotoxicity in multiple studies of free-ranging harbour seals (Mos et al., 2007b). Even though the PCB concentrations in killer whales from Possession Island were among the lowest reported in killer whales world-wide, 70% of our study animals surpassed this latter threshold level, suggesting that contaminants cannot be excluded as a possible risk factor in the decline of this population.

4. Conclusion

Despite our limited number of samples, this study provided baseline organic contaminant data for the Possession Island killer whales. While these killer whales were less contaminated than some of their northern hemispheric counterparts, they can be counted among the most PCB-contaminated marine mammals in the southern hemisphere. Despite a PCB ban in the industrialized world in the 1970s, the dispersion of this chemical has attained the far reaches of the planet, including the killer whales that ply the remote waters around Possession Island. While concentrations of POPs are decreasing in the industrialized regions and in the Arctic, the global PCB distribution has not yet reached a steady state and concentrations are expected to increase in the Antarctic before reaching a state of equilibrium (Goerke et al., 2004). While we did not measure the organochlorine pesticide dichloro diphenyl trichloroethane (DDT), other studies of marine mammals from the southern hemisphere have found their concentrations to exceed that of PCBs by 2 - 9 times (Krahn et al., 2008; Aono et al., 1997). The results of our study provide another reminder that POPs know no borders, and can significantly contaminate wildlife inhabiting remote corners of the world.

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FIGURE CAPTIONS

Figure 1: Killer whales were sampled around Possession Island, Crozet Archipelago, in the southern Indian Ocean.

Figure 2: Juvenile and adult females have similar PCB (total, di-ortho, mono-ortho and coplanar), PCDD, and PCDF concentrations (mean ± standard error). The adult male shows the highest PCB, PCDD and PCDF concentrations (mg/kg lw), although limited sample size precluded statistical evaluation.

Figure 3: The homologue group contribution to the total PCB concentrations was similar across age/sex categories. (Mean ± standard error is available only for juvenile and adult female killer whales).

Table 1: The PCB composition was generally similar across age/sex categories as evidenced by the top six PCB congeners (concentrations in mg/kg lw). The percentage contribution to the total PCB contamination is added in brackets.

	Juvenile Females (n = 3)		Adult Females (n = 5)		Adult male (n = 1)	
∑ PCBs	11.4 ± 1.2		10.1 ± 3.1		19.5	
	153[a]	2.2 ± 0.2 *(19.3 ± 0.4%)*	153[a]	2.0 ± 0.6 *(19.9 ± 0.2%)*	153[a]	3.8 *(19.2%)*
	138[a]	1.5 ± 0.1 *(12.9 ± 0.8%)*	138[a]	1.4 ± 0.4 *(13.1 ± 0.2%)*	138[a]	2.7 (13.8%)
	180	0.9 ± 0.2 *(7.5 ± 1.2%)*	149	0.8 ± 0.3 *(7.4 ± 0.3%)*	180	1.5 *(7.8%)*
	149	0.8 ± 0.2 *(7.2 ± 0.6%)*	180	0.7 ± 0.2 *(7.2 ± 0.1%)*	149	1.3 (6.9%)
	187[a]	0.7 ± 0.1 *(5.8 ± 0.6%)*	187[a]	0.6 ± 0.2 *(5.8 ± 0.2%)*	187[a]	1.1 *(5.6%)*
	201	0.5 ± 0.2 *(3.9 ± 1.0%)*	201	0.4 ± 0.1 *(3.4 ± 0.1%)*	118[a]	0.6 *(3.1%)*
∑ top 6	**6.5 ± 0.3** ***(57.4 ± 0.6%)***		**5.9 ± 0.3** ***(54.6 ± 0.2%)***		**11.0** ***(56.4%)***	

[a]: dominant congener of a co-eluting group (CB-153 was co-eluting with 132; 138 with 163 and 164; 187 with 182; 118 with 106)

Table 2: PCB concentrations in the largely marine mammal-eating Crozet killer whales exceed concentrations in the fish-eating Antarctic killer whales. PCB levels approach those observed in the relatively contaminated northern resident killer whales from the Northeastern Pacific Ocean.

Location	Year	Type	Sex	Lifestage	PCBs (mg/kg lw)	PCDDs (ng/kg lw)	PCDFs (ng/kg lw)	TEQ (ng/kg lw)	Reference
Crozet Archipelago	1998		F	Juvenile	11.4 ± 1.1	3.9 ± 1.8	ND[a]	76.5 ± 5.0	Our study
			F	Adult	10.1 ± 3.0	20.6 ± 12.1	6.6 ± 5.9	44.2 ± 8.4	
			M	Adult	19.5	38.79	11.4	120.4	
Ross Sea, Antarctica	2005-2006	Type C[b]	M	Adult	1.6 ± 1.1	n.a[c]	n.a	n.a	Krahn et al.,2008
			M	Subadult	2.1 ± 2.3	n.a	n.a	n.a	
			F	Adult	0.6 ± 0.4	n.a	n.a	n.a	
Norway	2002		M	Adult/Juvenile	16-44	n.a	n.a	n.a	Wolkers et al., 2007
Japan	2005		M	Calf	47	0.62	0.77	300	Kajiwara et al., 2006
			F	Calf	51	N.D	0.96	405	
			M	Adult	57	1.50	3.90	350	
			F	Adult	30	3.50	5.80	220	
British Columbia, Canada	1993-1996	Northern Resident	M	Adult	37 ± 6			435 ± 60	Ross et al., 2000
			F	Adult	9 ± 3			111 ± 29	
		Southern Resident	M	Adult	146 ± 33	1050 ± 258	55 ± 6	845 ± 195	
			F	Adult	55 ± 19			456 ± 27	
		Transient	M	Adult	251 ± 55			699 ± 142	
			F	Adult	59 ± 21			338 ± 53	
	2004-2006	Southern Resident	M	Adult	66 ± 26	n.a	n.a	n.a	Krahn et al.,2007
			F	Adult	45	n.a	n.a	n.a	
Alaska, USA	1994-1999	Resident	M/F	Adult/Juvenile	14 ± 13	n.a	n.a	100 ± 98	Ylitalo et al., 2001
		Transient	M/F	Adult/Juvenile	230 ± 130	n.a	n.a	860 ± 640	

[a]: Non detected
[b]: Type C killer whales are thought to prey mainly on marine fish
[c]: Non available

Table 3: In juvenile female, adult female and adult male killer whales, PCBs are the dominant contributor to the total toxic equivalents to 2,3,7,8-TCDD.

		Mean TEQ ± st error (pg/g lw)		
	PCB congeners	Juvenile Female (n=3)	Adult female (n=6)	Adult male (n=1)
Non-ortho PCBs	77	0.01 ± 0.00	0.01 ± 0.00	0.02
	81	0.01 ± 0.00	0.01 ± 0.00	0.01
	126	10.61 ± 2.53	7.69 ± 1.11	16.63
	169	53.39 ± 7.07	31.38 ± 7.02	60.56
∑ non-ortho PCBs		64.01 ± 8.72	39.07 ± 7.65	77.21
% of ∑PCB TEQ		*83.42 ± 8.71%*	*87.30 ± 2.06%*	*70.82%*
Mono-ortho PCBs	105	1.17 ± 0.25	0.81 ± 0.18	4.74
	114	0.09 ± 0.02	0.06 ± 0.01	0.32
	118	3.77 ± 0.74	2.71 ± 0.66	17.89
	123	0.04 ± 0.01	0.02 ± 0.01	0.20
	156	0.57 ± 0.03	0.38 ± 0.09	2.16
	157	0.15 ± 0.01	0.11 ± 0.03	0.69
	167	0.51 ± 0.04	0.39 ± 0.12	2.68
	189	0.08 ± 0.02	0.06 ± 0.02	0.40
∑ mono-ortho PCBs		6.38 ± 0.98	4.55 ± 1.12	29.08
% of ∑PCB TEQ		*8.28 ± 0.97%*	*9.97 ± 1.17%*	*26.68%*
Sum PCB TEQ		76.26 ± 4.88	43.62 ± 8.66	106.29
		99.77 ± 0.15%	*97.27 ± 1.70%*	*88.31%*
Sum PCDD TEQ		0.19 ± 0.13	0.45 ± 0.34	1.59
		0.23 ± 0.15%	*1.99 ± 1.56%*	*1.46%*
Sum PCDF TEQ		ND	0.17 ± 0.14	1.14
		-	*0.72 ± 0.66%*	*1.04%*
TOTAL TEQ		**76.45 ± 5.00**	**44.24 ± 8.37**	**109.02**

Figure 1:

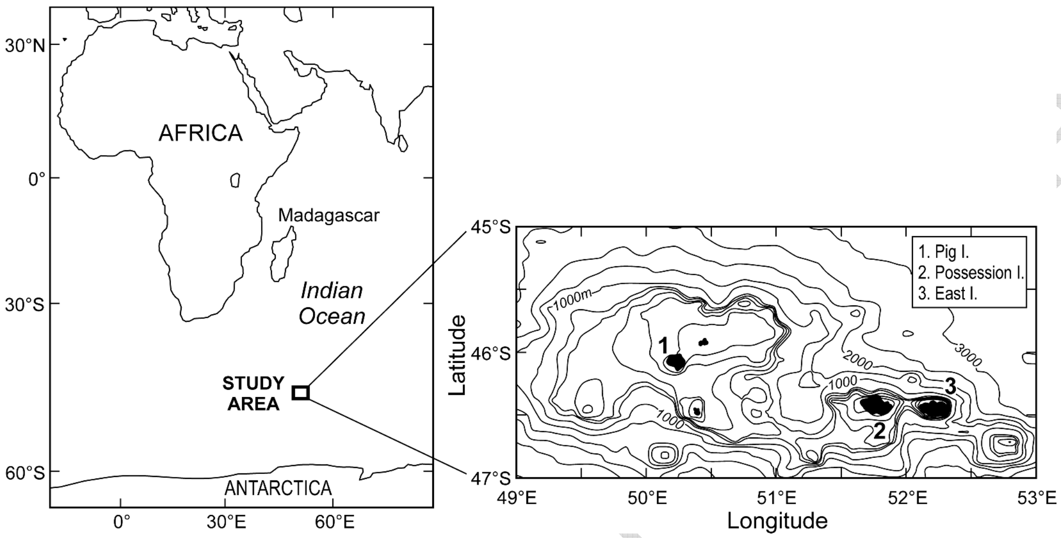

30°N
0°
30°S
60°S
0°
30°E
60°E
AFRICA
Madagascar
Indian
Ocean
STUDY
AREA
ANTARCTICA
45°S
46°S
47°S
49°E
50°E
51°E
52°E
53°E
Latitude
Longitude
1. Pig I.
2. Possession I.
3. East I.
1000m
1000
2000
3000
1
2
3

Figure 2:

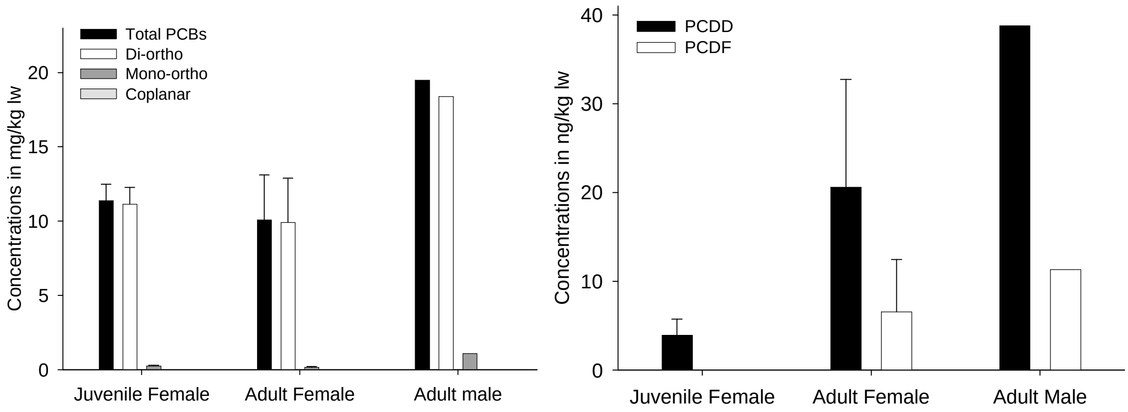
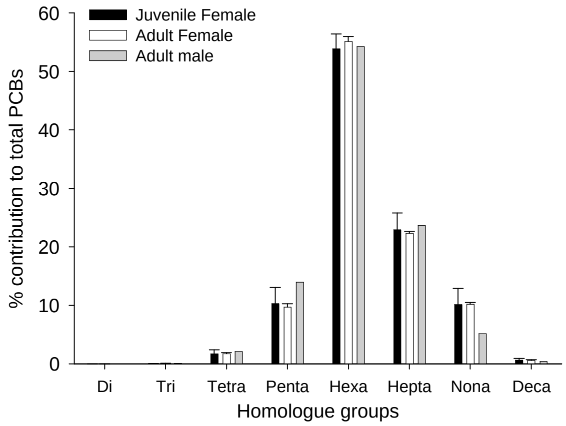

Total PCBs
Di-ortho
Mono-ortho
Coplanar
Concentrations in mg/kg lw
0
5
10
15
20
Juvenile Female
Adult Female
Adult male
PCDD
PCDF
Concentrations in ng/kg lw
0
10
20
30
40
Juvenile Female
Adult Female
Adult Male

Figure 3:


Juvenile Female
Adult Female
Adult male
% contribution to total PCBs
0
10
20
30
40
50
60
Di
Tri
Tetra
Penta
Hexa
Hepta
Nona
Deca
Homologue groups