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Marine Environmental Research 53 (2002) 425–452

MARINE ENVIRONMENTAL RESEARCH

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Geographical and temporal variation in levels of organochlorine contaminants in marine mammals

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Received 15 November 2000; received in revised form 11 October 2001; accepted 31 October 2001

Abstract

The interpretation of the spatial and temporal patterns of variation in organochlorine concentrations in marine mammal populations is complex because of the lack of wide-scale, long-term surveys. Therefore the results from several surveys must be combined and this causes undesired heterogeneity due to differences in the sampling and analytical techniques used and in the biological characteristics of the individuals sampled. Moreover, information is not homogeneously distributed in either space or in time. Most research is concentrated in western Europe, northern America and certain areas of Asia, while it is extremely limited or non-existent in Africa and most regions of the southern hemisphere. Marine mammals from the temperate fringe of the northern hemisphere, particularly fish-eating species which inhabit the mid-latitudes of Europe and North America, show the greatest organochlorine loads; noteworthy are the extremely high levels found in the Mediterranean Sea and certain locations on the western coasts of the United States. Concentrations in the tropical and equatorial fringe of the northern hemisphere and throughout the southern hemisphere are low or extremely low. The polar regions of both hemispheres showed the lowest concentrations of DDTs and PCBs, although levels of HCHs, chlordanes and HCB were moderate to high in the cold waters of the North Pacific. During recent decades, concentrations have tended to decrease in the regions where pollution was initially high but they have increased in regions located far from the pollution source as a consequence of atmospheric transport and redistribution. It is expected that the Arctic and, to a lesser extent, the Antarctic, will become major sinks for organochlorines in the future; this process may already be significant for some compounds

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PII: S0141-1136(01)00128-3

such as HCB and HCHs. Effort should be devoted to both assessment of organochlorine trends in the now highly polluted populations of the temperate fringe of the northern hemisphere and to the implementation of long-term monitoring of marine mammal populations inhabiting polar regions.

Keywords: Pollution; Organochlorine compounds; PCBs; DDTs; Cetaceans; Pinnipeds; Geographical variation; Temporal trends; Review

1. Introduction

In the mid-1960s, Koeman and van Genderen (1966) reported, for the first time, the presence of organochlorine compound residues in the tissues of a wild marine mammal. In a paper mostly focusing on birds, the results of blubber and fat analyses of three common seals (*Phoca viulina*) from the Wadden Sea showed a few parts per million of dieldrin and various forms of DDTs. Soon after, Jensen, Johnels, Olsson, and Otterlind (1969), in a toxicological survey of Swedish marine fauna, were the first to detect polychlorinated biphenyls (PCBs) in the tissues of marine mammals, this time in common seals, grey seals (*Halichoerus grypus*) and ringed seals (*Pusa hispida*). Since then, the number of references on the subject has increased exponentially, reaching a ceiling of 30–40 refereed articles per year in the 1990s (Aguilar & Borrell, 1996).

Substantial evidence points to organochlorine compounds having, at environmental levels, a number of adverse effects on marine mammal populations. These include depression of the immune system (De Swart et al., 1994, 1995, 1996; Lahvis, Well, Kuehl, Stewart, Rhinehart, & Via, 1995; Ross, De Swart, Timmerman et al., 1996) and the subsequent triggering of infectious diseases (Aguilar & Borrell, 1994; Kannan, Tanabe, Borrell, Aguilar, Focardi, & Tasukawa, 1993b; Ross, De Swart, Addison, Van Loveren, Vos, & Osterhaus, 1996; Simmonds & Mayer, 1997), reproductive impairment (Addison, 1989; Baker, 1989; Béland et al., 1993; Helle, Stenman, Olsson, Helander, & Harkonen, 1990; Reijnders, 1986; Reijnders & Brasseur, 1992), lesions of the adrenal glands and other organs (Olsson, Karlsson, & Ahnland, 1994), cancers (Martineau et al., 1994), and alterations in skeletal growth and ontogenic development as well as the induction of bone lesions (Bergman, Olsson, & Reiland, 1992; Mortensen, Bergman, Hansen, Harkonen, & Olsson, 1992; Zakharov, Valetsky, & Yablokov, 1997; Zakharov & Yablokov, 1990). The assessment of the levels of organochlorine exposure to which populations are exposed, their evolution and their pattern of geographical variation are significant factors in the management and long-term conservation of marine mammal populations. However, published information on this subject is extremely patchy between species, areas and sampling periods, thus hindering such evaluation.

This review aims to: examine the spatial patterns of variation in selected species of marine mammals with a wide geographical distribution for which a comparatively large amount of information is available, summarize results on time-trends found in populations followed over time, and identify gaps and propose specific research fields to complete the environmental picture necessary to assess the impact of pollutants on marine mammal populations.

2. Methods

To collect data on geographical and temporal variation of organochlorine levels in marine mammals we surveyed in detail over 950 references obtained from cross-referencing in papers and from searches in *Wildlife Review*, *Current Contents*, *Biological Abstract*s, and *Aquatic Sciences and Fisheries Abstracts* from 1965 to date. Moreover, the compilations by Risebrough (1978), Wagemann and Muir (1984), O'Shea and Brownell (1994), Aguilar and Borrell (1996), Borrell and Aguilar (1999), Norstrom and Muir (1994), Muir et al. (1999) and the bibliography sections of many papers on pollutants in marine mammals were searched for additional references. To ensure their accessibility, accuracy and scientific quality, only papers published in refereed scientific journals were considered.

Organochlorine concentrations referred to in this paper were always calculated on the basis of the sample content of extractable lipids (lipid basis). This expression was preferred to that calculated on the basis of the fresh weight of the sample because it corrects, at least partially, for heterogeneity of nutritive condition and laboratory methods used for lipid extraction. These two sources of heterogeneity are likely to introduce additional variation in the determination of levels of organochlorine compounds in tissues. When source papers expressed concentrations on a fresh weight basis, we calculated the lipid-based concentrations by dividing them by sample lipid content. If this latter value was not available, the concentrations in fresh weight basis were divided by 0.7, a figure that was considered to be a representative mean lipid richness for blubber. The mean, maximum and minimum values were directly extracted from the source papers or calculated from the raw data when available. This review does not include papers whose authors did not report these measurements or the sufficient data to calculate them.

To be included in the review of geographical variation, a given survey had to be posterior to 1980. Time trends obviously could not be bound by this restriction and were assessed either by collecting results from surveys specifically addressing this subject or by compiling data from several surveys for the same region and species. This introduces heterogeneity (see later) that makes projections for the earlier period (1965–ca. 1980) less certain than those for the later period (post 1980).

3. Scope and limitations of the review

Due the lack of wide-scale surveys, the pattern of spatial variation of pollution loads in a given species had to be necessarily examined by combining the results obtained by several researchers who used different sampling and analytical techniques. This inevitably produces a number of sources of heterogeneity in the comparisons.

Laboratory techniques for the analysis and quantification of organochlorines vary greatly. This is particularly true for two steps: the extraction of the lipid fraction from the tissue and the clean-up of the obtained extract. The use of

relatively polar solvents in the extraction, such as petroleum ether or a mixture of chloroform and methanol, produces a greater contribution of the more polar organochlorine chemicals (the low chlorinated PCBs and chlordanes, HCHs, aldrin, and *p*,*p*′-DDE, among others). Conversely, the use of more non-polar solvents, such as *n*-hexane, produces an extract with a larger proportion of less polar compounds (the highly chlorinated PCBs, Mirex and *p*,*p*′-DDT, among others; Cairns, Doose, Froberg, Jacobson, & Siegmund, 1986). The clean-up may also selectively underscore or even eliminate some compounds. Although all these techniques are equally valid laboratory practices, they are clearly liable to produce different results.

The effect of variation in laboratory techniques is more severe when the assessment of time-trends involves the comparison of results obtained in surveys which are separated by a substantial number of years. It is estimated that packed columns, commonly used before the 1980s, overestimated PCB concentrations by about 50%, while underestimating DDT concentrations by 30–40%, a bias later overcome by the much more precise capillary columns (Norstrom, Simon, Muir, & Schweinsburg, 1988). In the mid-1980s, the formulation of individual PCB standards allowed the quantification of these compounds congener by congener, and not as a mixture, as previously calculated, and this again increased accuracy and refined assessment of toxicological impact. The incidence of such changes in analytical performance on the observed time-trends cannot be determined and is therefore intrinsic to the assessment. To circumvent this problem, the evaluation of spatial variation has only been made on post-1980 data.

Another source of variation when comparing surveys is the heterogeneity of sample composition. Many papers do not detail the sex and age of the individuals examined, two factors that, given the reproductive transfer that occurs in females during pregnancy and lactation, are known to have a recognised effect on organochlorine levels in tissues (Aguilar, Borrell, & Pastor, 1999). Concentrations on the different compounds may not be identically affected by these processes; therefore, not only the overall pollutant load may vary, but also the relative abundance of the various compounds. For example, reproductive transfer is higher for less lipophilic than for more lipophilic compounds. In several delphinid species it has been shown that the transfer rate from mother to calf during lactation is higher for *p*,*p*′-DDE than for *p*,*p*′-DDT, and for tDDT than for tPCB (Aguilar et al., 1999; Borrell, Bloch, & Desportes, 1995; McKenzie, Rogan, Reid, & Wells, 1997; Tanabe, Tatsukawa, Maruyama, & Miyazaki, 1982). This differential transfer has clear effects on several ratios which are commonly used in ecotoxicological assessment, such as DDE/tDDT and tDDT/tPCB, and is likely to affect the pollutant concentration profile of samples of heterogeneous sex and age composition.

Finally, other factors may also contribute undesired variation in samples. Nutritive condition, incidence of disease, preservation state of the tissue, and type of sampling method, are well known sources of variation in the resulting organochlorine concentrations (see Aguilar et al., 1999, for a review). In usual field conditions, these factors are considered to be less influential than those discussed earlier, but they

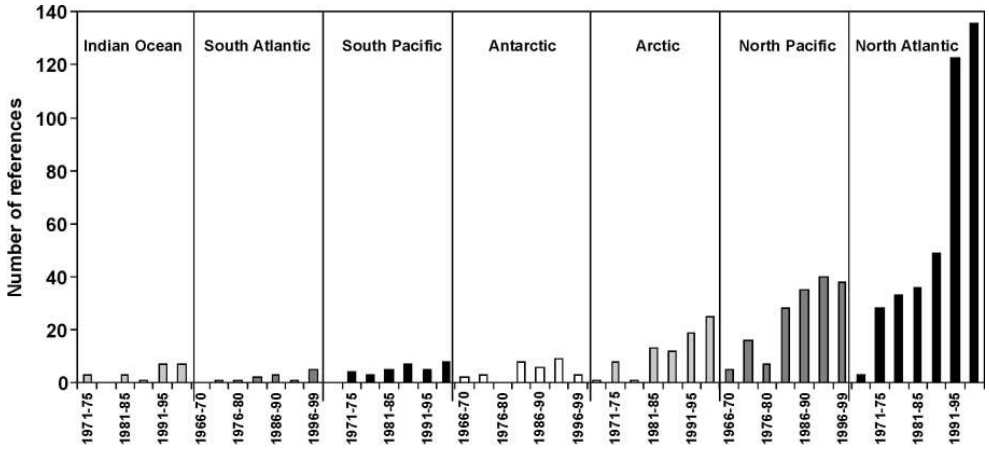

Fig. 1. Distribution frequency of published refereed articles by ocean, and time period.

should not be overlooked, particularly if they are cumulative. For example, samples from naturally stranded cetaceans often present a skewed age composition, include an abnormally large number of individuals in poor condition and contain numerous diseased animals. Consequently such types of samples are particularly difficult to interpret when assessing spatial and temporal trends.

4. Results and discussion

4.1. Patterns of geographical variation

Information on organochlorine concentrations in marine mammals is not homogeneously distributed, either in space or time (Fig. 1). During the 1970s and 1980s, research concentrated in western Europe and northern America, clearly reflecting the technical strength of researchers from these regions. Since the mid 1980s however, the scientific production on the subject in Asia, particularly Japan, and in South America has increased. However, to date, research levels in Australia, the Indopacific region and Africa still remain minimal. An example of the heterogeneous distribution of research can be appreciated in a recent review, which showed that over 45% of the references published to date refer to the North Atlantic (Aguilar & Borrell, 1996).

Even in the most studied regions, most surveys examining the differences in pollutant concentrations between areas limited their scope to only two localities. Surveys including three localities are rare, and to our knowledge none has examined geographical patterns of variation for a single species on a worldwide scale. For the study of geographical variation, we selected four marine mammal species: the bottlenose dolphin (*Tursiops truncatus*), the harbour porpoise (*Phocoena phocoena*), the fin whale (*Balaenoptera physalus*), and the harbour seal (*Phoca vitulina*). These species were not necessarily chosen because the number of surveys focusing on them

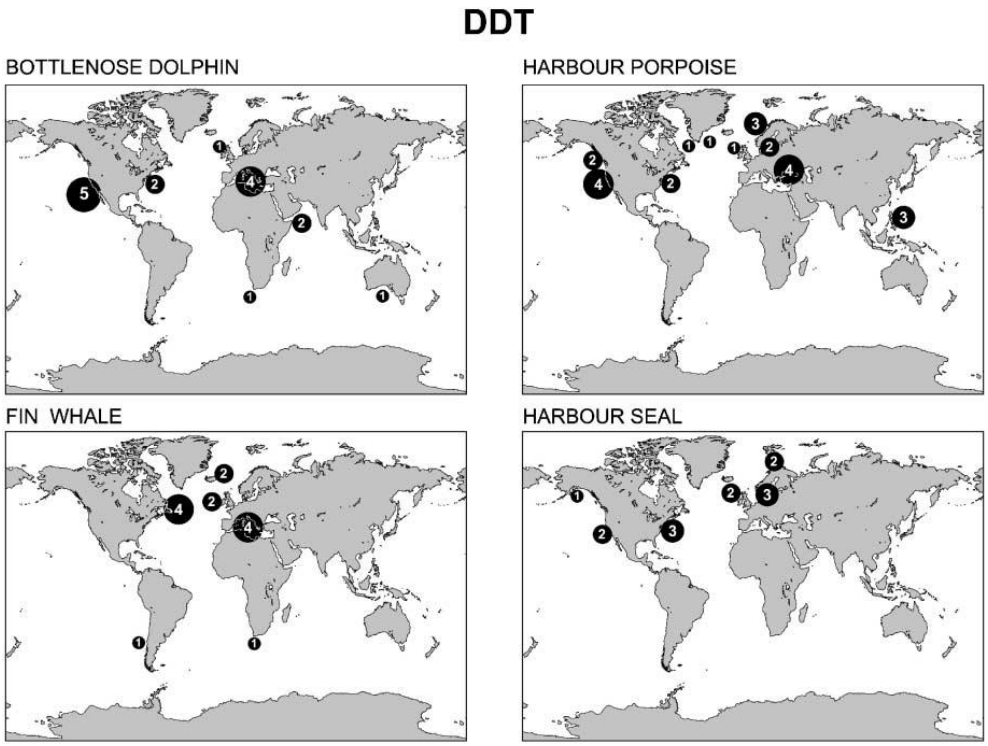

Fig. 2. Relative DDT blubber concentration of selected marine mammal species (see Table 1 for references). Bottlenose dolphin: 1, <10 mg/kg; 2, 10–30 mg/kg; 3, 30–100 mg/kg; 4, 100–500 mg/kg; 5, >500 mg/kg. Harbour porpoise: 1, <5 mg/kg; 2, 5–12 mg/kg; 3, 12–25 mg/kg; 4, 25–50 mg/kg; 5, >50 mg/kg. Fin whale: 1, <1 mg/kg; 2, 1–2.5 mg/kg; 3, 2.5–5 mg/kg; 4, 5–10 mg/kg; 5, >10 mg/kg. Harbour seal: 1, <2 mg/kg; 2, 2–5 mg/kg; 3, 5–10 mg/kg; 4, 10–20 mg/kg; 5, >20 mg/kg.

was particularly large, but because such surveys covered a comparatively wider geographical range. Table 1 details the results of the compilation, and Figs. 2 and 3 summarize in graphical form the levels of DDTs and PCBs, the two most widespread and better known groups of organochlorine compounds, in the different species and areas. Given that the species chosen occupy very distinct trophic niches and also display distinct ecological traits and distribution, their pollutant loads are highly variable, sometimes even of a different order of magnitude. Therefore, geographical patterns cannot be derived from comparisons between several species, but only from intra-specific variation.

Marine mammals from the temperate fringe of the northern hemisphere, particularly fish eating species inhabiting the mid-latitudes of Europe and North America, display the greatest DDT and PCB loads (Figs. 2 and 3). These results are consistent with those from studies undertaken in the early 1980s on the geographical distribution of organochlorines in the atmosphere and surface waters (Tanabe, Mori, Tatsukawa, & Miyazaki, 1983) and with global patterns previously observed in marine mammal tissues (O'Shea & Brownell, 1994; Reijnders & de Ruiter-Dijkman, 1995), and are related to the extensive production and use of organochlorines in industrialized countries in these parts of the world (Reijnders & de Ruiter-Dijkman, 1995).

Table 1
DDT and PCB concentrations and the DDE/tDDT ratio in the blubber of selected marine mammal species from several regions[a]

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
Fin whale														
Mediterranean Sea	Italy	1991/1993	?	23	5.28			3.92						Marsili, Fossi, Notarbartolo di Sciara, Zanardelli, and Focardi (1996)
		1990/1993	BOTH	68	7.87	7.98	6.18	5.62	10.67	4.72				Marsili and Focardi (1996)
	Spain	1998	F	1	24.26			9.76			74.79			Serrano, Lopez, and Hernandez (1999)
NE Atlantic	Spain–France	1982/1984	M	69	1.61	3.48	0.63	1.35	3.39	0.54	36.15	52.00	23.00	Aguilar and Borrell (1988)
			F	97	1.00	4.08	0.17	0.75	3.71	0.15	32.70	50.00	19.00	
	Iceland	1982/1985/1986	M	48	1.26	3.24	0.42	1.29	3.49	0.41	37.70	50.80	9.81	Borrell (1993)
		1985	F	3	0.94	1.08	0.78	0.92	1.20	0.68	37.16	37.69	36.66	
NW Atlantic	Canada	1991/1992	M	9	8.35	34.07	1.52	11.62	43.62	2.04	70.19	87.62	38.31	Gauthier, Metcalfe, and Sears (1997)
			F	6	2.28	4.60	0.33	3.36	6.53	0.94	72.11	94.21	28.88	
SE Pacific	Chile	1983		1				0.08				88.42		Pantoja, Pastene, Becerra, Silva, and Gallardo (1984)
Bottlenose dolphin														
Mediterranean Sea	Italy	1987/1992	M	5	44.63	174.84	0.25	15.00	57.68	0.64	61.00	77.00	20.00	Marsili and Focardi (1997)
			F	2	30.03	31.01	29.04	3.98	4.66	3.30	72.00	77.00	68.00	

(*continued on next page*)

Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
		1992	M	5	1204.00	2100.00	250.00	399.60	1100.00	48.00				Corsolini, Focardi, Kannan, Tanabe, Borrell, and Tatsukawa (1995)
		1992	F	2	590.00	950.00	230.00	140.00	260.00	20.00				
NE Atlantic	Scotland	1989/1991	M	1	30.72									Wells, Campbell, Ross, Thompson, and Lockyer (1994)
			F	5	21.26	39.66	3.89	8.91	15.90	2.20				
NW Atlantic	USA	1987/1988	M	3	138.40	195.00	47.20	38.58[b]	80[b]	3.75[b]				Kuehl, Haebler, and Potter (1991)
			F	9	62.37	157.00	17.40	7.46[b]	21.2[b]	0.52[b]				
	USA	1983/1985	M	8	97.73	195.48	5.72	31.81	63.97	4.30	37.66	49.29	24.88	King (1987)
		1983/1985	F	7	22.47	41.26	2.25	4.62	8.02	0.69	35.97	45.45	29.76	
	Mexico	1990	M	6	93.00	187.00	64.00	38.00	78.00	15.00				Kuehl and Haebler (1995)
		1990	F	10	7.20	18.00	1.50	3.70	14.00	0.60				
	Mexico	?	BOTH	33	36.10	4.10	149.00	15.30	74.60	0.43	83.66			Salata, Wade, Sericano, Davis, and Brooks (1995)
NW Pacific	USA	1978/1984	M	3	100.43	182.86	13.86	1297.62	2745.71	328.57				Schafer, Gossett, Ward, and Westcott (1984)
			F	3	38.77	63.57	13.97	1356.67	2957.14	180.00				
Indian Ocean	India	1990	M	2	1.19	1.68	0.70	9.23	13.21	5.25	94.29	100.00	88.57	Tanabe et al. (1993)
			F	2	0.75	0.94	0.57	14.72	20.90	8.55	72.69	79.66	65.71	
	South Africa	1980/1987	M	52	13.74	67.18	0.00	20.01	85.66	0.45				Cockroft, de Kock, Lord, and Ross (1989)

Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
			F	52	8.45	47.57	0.09	5.70	49.70	0.00				
	South Africa		M	4	3.15	10.02	0.00	11.89	26.25	2.03	63.00	100.00	19.00	Cockroft (1999)
			F	2	2.12	4.23	0.00	5.26	9.60	0.91	46.00	56.00	35.00	
SE Atlantic	South Africa	1980/1987	M	4	4.54	11.84	0.23	6.64	17.56	0.24				Cockroft (1999); de Kock et al. (1994)
			F	1	2.31			2.50						
SW Pacific	Australia	?	?	1/6	0.09				3.44	0.26				Kemper, Gibbs, Obendorf, Marvanek, and Lenghaus (1994)
Harbour porpoise														
Black Sea		1993		8	31.43	48.57	18.57							Minh et al. (2000)
		1993							257.14	5.81	46.00			Tanabe et al. (1997)
NE Atlantic	Poland	1989/1990	F	3	33.93	47.03	25.76	12.59	14.56	11.20				Kannan et al. (1993a)
	Poland	1990/1994	M	4	11.83	22.00	5.00	3.93	8.50	2.20	47.25	62.50	17.73	Kawano, Tanaka, Falandysz, and Tatsukawa (1997)
			F	7	18.44	45.00	5.60	3.64	9.60	1.40	48.66	61.43	3.84	
	Denmark	1980/1981	M	29	116.12	382.02	26.97	35.88	202.25	7.53				Clausen and Andersen (1988)
			F	22	61.49	123.60	4.16	13.89	42.70	1.57				
	North Sea	1988/1995?	BOTH	16				6.35	18.76	1.36	47.00			Vetter, Luckas, Heidemann, and Skirnisson (1996)

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Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
	Netherlands		M	10	36.53	63.61	13.24							Van Scheppingen, Verhoeven, Mulder, Addink, and Smeenk (1996)
			F	12	19.95	39.19	4.13							
	Scotland	1989/1991	M	21	15.98	44.15	0.57	4.88	12.62	0.78				Wells et al. (1994)
			F	15	6.57	32.00	2.71	2.57	10.40	0.91				
	Ireland	1990/1994	M	6	6.15	10.42	2.91	3.15	4.94	1.84	61.76			Smyth, Berrow, Nixon, and Rogan (2000)
			F	6	8.00	12.27	3.04	4.66	5.99	1.64	55.59			
	United Kingdom	1989/1992	M	23	40.10	109.49	0.44	11.07	33.06	0.63	60.23	85.22	33.93	Kuiken et al. (1994)
			F	25	18.69	86.97	1.63	5.73	22.86	0.40	58.72	94.87	37.98	
	United Kingdom	1988/1992	M	50	25.61	109.51	0.13	6.84	33.06	0.04	81.64	100.00	33.93	Law (1994)
		1988/1992	F	47	17.58	138.75	0.13	4.82	33.98	0.06	82.78	100.00	35.48	
	United Kingdom	1990/1996	M	13	61.43									Jepson et al. (1999)
			F	14	20.00									
	Southern/Central Norway	1987/1991	BOTH	27	21.90	65	3.71	15.60	45.09	3.22	44.34	57.47	34.36	Kleivane, Skaare, Bjorge, de Ruiter, and Reijnders (1995)
	Northern Norway	1987/1991	BOTH	7	25.89	44	17.25	18.28	36.18	12.04	43.10	46.51	38.46	Kleivane et al. (1995)
	Faroe Islands	1987/1988	M	3	13.39	15.77	10.13	6.56	8.00	5.77	46.68	55.25	38.50	Borrell (1993)
			F	3	8.83	10.31	7.95	4.43	4.90	3.99	47.65	52.24	45.11	
	Greenland	1989	M	1	7.86									Van Scheppingen et al. (1996)
			F	3	2.76	3.14	2.13							
NW Atlantic	Greenland	1996	M	46	2.42	5.13	0.64	3.45	7.90	0.96	44.89	56.69	36.25	Borrell et al. (1999)
			F	54	1.75	3.33	0.45	2.25	4.94	0.38	42.74	49.78	34.68	

Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
	USA	>1987	BOTH	5	21.14	32.29	10.60	10.40	16.00	6.70	44.78			Becker, Mackey, Demiralp, Schantz, Koster, and Wise (1997)
NE Pacific	Canada	1987	M	3	0.10	0.10	0.10	6.98	9.10	3.45				Langelier, Stacey, Baird, and Marchetti (1988)
			F	3	9.27	16.00	0.10	5.75	12.30	1.84				
	Canada	1986/1989	?	7	9.13			8.91			73.17			Jarman, Norstrom, Muir, Rosenberg, Simon, and Baird (1996)
	USA		?	3	13.70			16.30			80.00			
	USA	1980/1985	M	8	29.18	129.80	3.72	79.61	206.12	9.09				O'Shea et al. (1980)
			F	15	17.48	57.53	2.28	51.96	139.73	9.02				
	USA	1981/1986	M	6	23.03	71.88	3.28	37.35	75.51	12.07				O'Shea et al. (1980)
			F	7	12.29	55.85	1.76	24.17	93.41	2.52				
	USA	1981/1985	M	3	31.80	36.34	23.94	31.01	40.45	20.38				O'Shea et al. (1980)
			F	3	22.83	31.29	10.96	14.08	17.88	9.66				
	USA	1990/1995	BOTH	12	3.00[b]	34.71	0.00	3.57***	13.43[b]	0.71[b]				Hayteas and Duffield (1997)
NW Pacific	Japan	1993	?	3	10.26	15.58	5.32	12.47	27.27	3.25				Minh, Watanabe et al. (2000)
Harbour seal														
NE Atlantic	Sweden	1988	BOTH	9	84.70	141.00	21.20	12.80[b]						Luckas, Vetter, Fisher, Heidemann, and Plötz (1990)
	North Sea	1988/1995?	?	32				3.90	11.48	1.50	73.51			Vetter et al. (1996)

(*continued on next page*)

Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
	North Sea	?	BOTH	87	29.90[c]	148.00	2.50							Van den Brink, Rutter-Dukman, Broekhuizen, Reijnders, and Bosveld (2000)
	Denmark	1988	BOTH	35	14.19	48.39	2.90	5.81	14.19	1.45				Skaare, Hedlund-Markussen, Norheim, Haugen, and Holt (1990)
			BOTH	26	16.88	49.35	5.32	4.29	9.48	1.82				
			BOTH	18	10.00	29.31	24.14	3.10	9.31	0.17				
	Germany	1988	BOTH	32	85.30	250.00	28.70	2.87[b]						Luckas et al. (1990)
	United Kingdom	1988	M	49	47.27	347.75	0.50	5.97	40.75	0.22	66.05	95.07	25.00	Hall et al. (1992)
			F	37	35.91	337.00	2.85	4.67	35.00	0.48	63.04	76.92	52.01	
	United Kingdom	1988	M	5	15.96	47.06	1.67	2.67	5.79	1.06	65.69	75.86	57.54	Law, Allchin, and Harwood (1989)
			F	4	14.97	40.85	2.76	1.70	3.10	0.54	65.33	70.26	55.77	
	Ireland	1988	M	21	59.98	216.91	7.68	5.82	12.17	0.16	63.99	100.00	30.77	Mitchell and Kenedy (1992)
			F	18	39.64	103.09	12.25	3.35	10.63	0.59	58.77	97.06	28.85	
	Iceland	1988	BOTH	7	5.22	13.40	1.08	1.18[b]						Luckas et al. (1990)
Arctic	Norway	1989/1990		7	4.88	11.32	2.24	2.07	7.75	0.62				Ruus, Ugland, Espeland, and Skaare (1999)
NW Atlantic	Canada	1994/1996	M	5	53.90			7.06						Bernt, Hammill, Lebeuf, and Kovacs (1999)
			F	10	14.40			3.48						

Table 1 (*continued*)

Area		Year	Sex	*n*	PCB			tDDT			%DDE/tDDT			References
					x	Max.	Min.	*x*	Max.	Min.	*x*	Max.	Min.	
	USA	1990/1992	BOTH	9	9.51	16.14	3.73	5.89[b]	11.2[b]	2.61[b]				Lake, Lake, Haebler, McKinney, Boothman, and Sadove (1995)
	USA	1980	BOTH	6	17.14	34.71	10.43	15.57[b]	31.29[b]	9.31[b]				Lake et al. (1995)
	USA	>1987	BOTH	4	0.77	0.98	0.53	0.78	2.04	0.01	36.46			Becker et al. (1997)
NN Pacific	Alaska	1992/1993	M	3	0.91	1.55	0.56	0.65	1.10	0.47				Krahn, Becker, Tilbury, and Stein (1997)
			F	2	0.26	0.27	0.25	0.16	0.17	0.15				
NE Pacific	USA	1992/1993	M	5	5.79	12.17	3.20	7.12	16.55	3.69				Krahn et al. (1997)
			F	5	2.42	3.49	1.43	2.69	3.89	1.14				
	USA	1990/1995	BOTH	10	2.43[c]	8.71	0.00	2.71[d]	17.86[b]	0.57[b]				Hayteas and Duffield (1997)

[a] Concentrations are expressed on an extractable lipid basis (mg/kg lipid) and were calculated from original data when not directly expressed in the original source (see text for details).

[b] DDE.

[c] Geometric mean.

[d] Geometric mean of DDE.

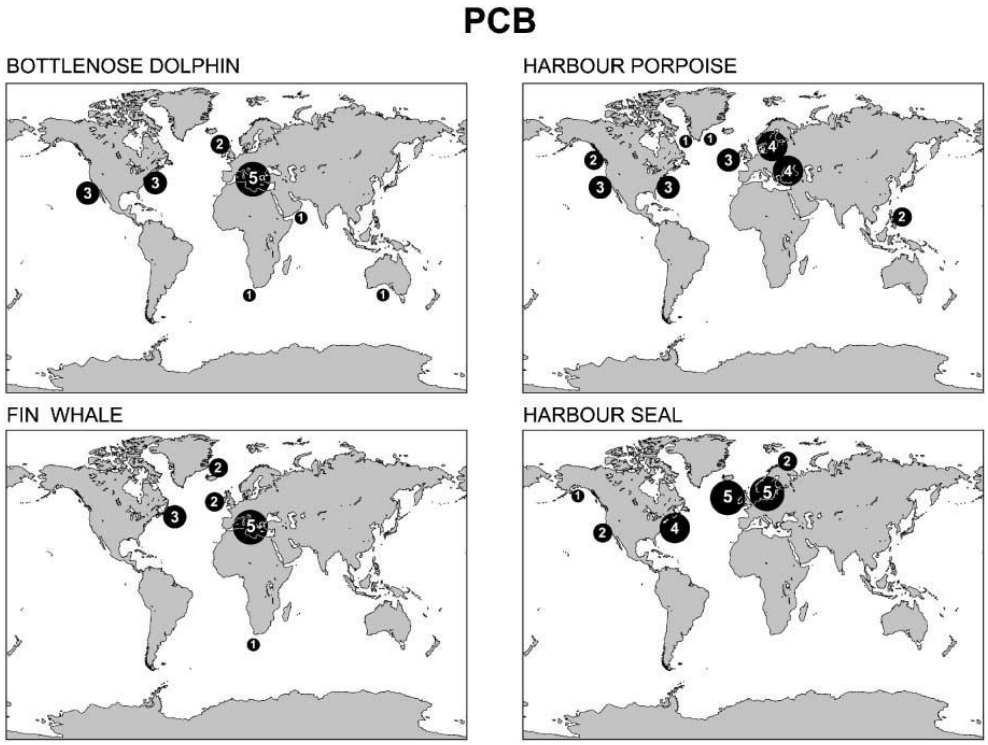

Fig. 3. Relative PCB blubber concentration of selected marine mammal species (see Table 1 for references). Bottlenose dolphin: 1, <10 mg/kg; 2, 10–30 mg/kg; 3, 30–100 mg/kg; 4, 100–500 mg/kg; 5, >500 mg/kg. Harbour porpoise: 1, <5 mg/kg; 2, 5–12 mg/kg; 3, 12–25 mg/kg; 4, 25–50 mg/kg; 5, >50 mg/kg. Fin whale: 1, <1 mg/kg; 2, 1–2.5 mg/kg; 3, 2.5–5 mg/kg; 4, 5–10 mg/kg; 5, >10 mg/kg. Harbour seal: 1, <2 mg/kg; 2, 2–5 mg/kg; 3, 5–10 mg/kg; 4, 10–20 mg/kg; 5, >20 mg/kg.

Noteworthy in Table 1 are the extremely high concentrations of organochlorines found in Mediterranean Sea populations, an observation which is further confirmed by data from other species inhabiting the region, such as the striped (*Stenella coeruleoalba*) and the common dolphin (*Delphinus delphis*; Aguilar & Borrell, 1994; Borrell, Cantos, Pastor, & Aguilar, 2001; Marsili & Focardi, 1997). The western coast of the United States, particularly California, showed similar or even higher concentrations of DDTs although those of PCBs were somewhat lower. The coastal fringe of both areas sustains dense human populations and industrial and agricultural activities are also intense. In both areas DDT production and use was discontinued in the mid-1970s; however, this pesticide had previously been so widely used that the residual levels in the environment have remained extremely high. This trajectory in pesticide use is reflected in the high DDE/tDDT ratio which is characteristic of both areas (see later).

Although less polluted than the above, other areas located along heavily urbanized and/or industrialized coastlines in the northern hemisphere also show relatively high levels of organochlorines (Figs. 2 and 3): the eastern and western temperate North Atlantic, the North Sea, the Caribbean Sea, the waters around Japan and, according to data from other species not included in the compilation, the

tropical and temperate belt of the Indian and Pacific Oceans (Loganathan et al., 1990; Minh, Nakata et al., 2000; Minh, Watanabe, Nakata, Tanabe, & Jeffersson, 1999; Minh, Watanabe et al., 2000; Prudente et al., 1997; Tanabe, Loganathan, Subramania, & Tatsukawa, 1987; Tanabe, Subramanian, Ramesh, Kumaran, Miyazaki, & Tatsukawa, 1993; Tanable et al., 1994). When the toxicity of PCBs, particularly that of the coplanar congeners, is assessed through the TEQ concept (Minh, Nakata et al., 2000), the greatest impact appears to also occur in the mid-latitudes. Similarly, concentrations of HCB, HCH, chlordane, or the recently described *tris*(4-chlorophenyl)methane (TCPMe) and *tris*(4-chlorophenyl) methanol (TCPMOH) in the Indo-Pacific region were relatively high in temperate waters and moderate to high in tropical and cold waters (Minh et al., 1999; Minh, Watanabe et al., 2000; Prudente et al., 1997).

Information on marine mammals from the tropical and equatorial fringe of the northern hemisphere is extremely scarce, but indicates that levels of organochlorine pollutants are low or extremely low, particularly in areas where agriculture and industry are poorly developed. In general, DDTs is the main pollutant, followed closely or equalled by PCBs (Table 1; Borrell, Aguilar, & Pastor, 1997b; Fossi et al., 1997; Tanabe et al., 1993); other organochlorine compounds such as HCB, HCH, chlordane, TCPMe or TCPMOH are usually found only in minor quantities (Minh et al., 1999; Minh, Watanabe et al., 2000; Prudente et al., 1997).

Overall, the southern hemisphere had a low level of pollution (Figs. 2 and 3). Organochlorine concentrations in marine mammals off South America, South Africa and Australia were invariably low, usually in the range of a few parts per million. This trend is confirmed in several other pinnipeds and small cetaceans which were studied but not included in the compilation (Borrell & Aguilar, 1999; Cockcroft, 1999; Corcuera, Monzón, Aguilar, Borrell, & Raga, 1995; de Kock et al., 1994; Fossi et al., 1997; O'Shea & Brownell, 1994). It is likely that, even in a moderately polluted region such as the southern hemisphere, certain restricted areas or localities, like river mouths or semienclosed water masses, may be especially contaminated as a result of point sources of pollution. However, the information necessary to identify such localities is not available yet. Further research is needed in the areas of southwest Asia, south America and Central Africa which most suffer human impact.

However, as expected, the lowest organochlorine concentrations were found in the polar regions of both hemispheres (Figs. 2 and 3). Levels in marine mammals from Greenland, Iceland, Alaska, the Canadian Arctic, and Antarctica were all extremely low and often under detection thresholds. Data on other species also support this observation (Addison & Smith 1998; Aono, Tanabe, Fujise, Kato, & Tatsukawa, 1997; Focardi, Bargagli, & Corsolini, 1995; Metcalfe, Metcalfe, Ray, Paterson, & Koenig, 1999; Muir et al., 1999; Norstrom & Muir, 1994; Schneider, Steinhagen-Schneider, & Drescher, 1985; Skaare, Espeland, Ugland, Bernhoft, Wiig, & Kleivane, 1994). The only exception to this pattern are some species that undergo seasonal migrations, such as the white whale (*Delphinapterus leucas*), which inhabit polluted environments during certain periods of the year and may, even when occupying the higher latitudes of the polar regions, carry relatively high organochlorine

loads (Reijnders & de Ruiter-Dijkman, 1995). However, certain areas may be locally contaminated by certain, not widely dispersed, types of organochlorines. For example, two Toxaphene components, T2 (Parlar 26) and T12 (Parlar 50) are the most prominent organochlorine chemicals observed in white whales and narwhals (*Monodon monoceros*) from certain areas of the Arctic, such as Baffin Bay (Muir, Ford, Grift, Stewart, & Bidleman, 1992; Muir et al., 1999).

4.2. Temporal variation in organochlorine levels

Papers dealing with the evaluation of time trends are equally as limited in scope as those addressing geographical variation; they are few and focus on only one species and area and, often, cover a relatively short period of time. Previous reviews on this subject have been written by Tanabe et al. (1983, 1994), Loganathan and Kannan (1994), Reijnders and de Ruiter-Dijkman (1995), Muir et al., 1999, and Borrell and Reijnders (1999).

Table 2 details the trends in organochlorine concentrations observed in marine mammals from several geographical localities and time periods. Most information relates to DDTs and PCBs although there is also incomplete data for HCB, HCHs and dieldrin from certain regions. The period covered ranges from 1965 to 1996, although most data correspond to the period 1970–1990. The scarcity of information for the latter period undoubtedly reflects the time-lag for publication of more recent surveys. With the exception of a few papers on minke whales (*Balaenoptera acutorostrata*) from the Antarctic and on bottlenose and common dolphins from South Africa, all information focuses on the northern hemisphere, particularly the Atlantic and Pacific Oceans.

Although the environmental presence of all organochlorine compounds has increased since the industrial revolution, their distinct uses and variable persistence and dispersal rates over time produces large variations in past trends of specific compounds in particular areas. Such differences are also likely to continue into the future.

In most parts of the developed world, the production and use of DDTs was discontinued in the early 1970s (Peterle, 1991). Although its use went on for many years in developing countries, and in some, it still appears to be used today (e.g. Shailaja & Singbal, 1994), on a global scale the absolute amounts of DDTs released into the environment have markedly decreased. Moreover, the decomposition rate of DDTs in tropical environments, where most recent use has taken place, is very fast compared to temperate or cold climates (Okla & Larsson, 1987; Wiese, 1976), and this has undoubtedly attenuated the impact of recent releases. In many areas, the observed decrease in DDT levels has been associated with a parallel increase in the relative abundance of its metabolized forms. This differential rate of environmental persistence permits the assessment of the chronology of DDT inputs by using the DDE/tDDT ratio. For example, during the 1970s and 1980s this ratio was found to progressively rise in odontocetes and pinnipeds in the North Atlantic (Aguilar, 1984) and in grey seals from eastern Canada (Addison, Brodie, & Zinck, 1984) concomitantly with a decrease in total DDT loads.

Table 2
Temporal trends observed in the blubber organochlorine pollutant concentrations of several marine mammal species from various geographical regions[a]

	Area	Species	Period	PCB	tDDT	HCB	HCHs	Diel.	Reference
Pinnipedia									
Baltic Sea		*Halichoerus grypus*	1969–1988	nt	d				Blomkvist, Roos, Jensen, Bignert, and Olsson (1992)/Olsson et al. (1994)
	Aland sea	*Halichoerus grypus*	1970,1971–1971,1973	nt	nt				Olsson, Johnels, and Vaz (1974)
		Phoca hispida	1969,1973–1980,1988	d	d				Blomkvist et al. (1992)/Olsson et al. (1994)
	Gulf of Bothnia	*Phoca hispida*	1969,1971–1972,1973	nt	nt				Olsson et al. (1974)
	Gulf of Bothnia	*Phoca hispida*	1972–1977	i	nt				Helle (1981)
	Gulf of Bothnia	*Phoca hispida*	1977–1980	d	d				Helle (1981)
	Gulf of Bothnia	*Phoca hispida*	1980–1983	nt	nt				Helle and Stenman (1984)
	Lake Saimaa	*Phoca hispida*	1970–1977,1981	d	d				Helle, Hyvärinen, Pyysalo, and Wickström (1983)
	Gulf of Finland	*Phoca hispida*	1977,1980–1983	d	d				Helle, Hyvärinen, and Stenman (1985)
North Sea	Farne Islands	*Halichoerus grypus*	1968–1975	nt	d				Holden (1978)
	Scotland	*Halichoerus grypus*	1965–1971	nt	d			d	Holden (1975)
	Netherland	*Phoca vitulina*	1973–1981	d					Van der Zande and De Ruiter (1983)
N W Atlantic Ocean	Sable Island	*Halichoerus grypus*	1974,1976–1982	nt	d				Addison et al. (1984)
	Sable Island	*Halichoerus grypus*	1984–1994	d	d	nt	d		Addison and Stobo (2001)
	N. Baffin	*Phoca hispida*	1972,1976–1984	d	d		nt	nt	Muir, Nostrom, and Simon (1988)
	St.Lawrence Gulf	*Pagophilus groenlandicus*	1971–1973	d	d			nt	Jones, Ronald, Lavigne, Frank, Holdrinet, and Uthe (1976)
	St.Lawrence Gulf	*Pagophilus groenlandicus*	1971–1978	d	d			i	Ronald, Frank, Dougan, Frank, and Braun (1984)
	St.Lawrence Gulf	*Pagophilus groenlandicus*	1971–1982	nt	d				Addison et al. (1984)
	St.Lawrence Gulf	*Pagophilus groenlandicus*	1982–1989	d	nt				Beck, Smith, and Addison(1993)
	New York	*Phoca vitulina*	1980–1990,1992	d	d	nt			Lake et al. (1995)

(*continued on next page*)

Table 2 (*continued*)

	Area	Species	Period	PCB	tDDT	HCB	HCHs	Diel.	Reference
W Arctic Ocean	Holman Island	*Phoca hispida*	1972–1981	d	nt				Addison et al. (1986)
	Holman Island	*Phoca hispida*	1981–1989,1991	nt	d	d	nt		Addison and Smith (1998)
NW Pacific Ocean	Japan	*Callorhinus ursinus*	1971–1976	i	i		i		Tanabe et al. (1994)
	Japan	*Callorhinus ursinus*	1976–1988	d	d		d		Tanabe et al. (1994)
	California	*Zalophus californianus*	1970–1988,1992		d				Lieberg, Bacon, Burns, Jarman, and Le Boeuf (1995)
Cetacea									
Mediterranean Sea	Spain	*Stenella coeruleoalba*	1987–1994	d	d				Borrell, Aguilar, and Pastor (1997a)
North Sea	Scotland	*Phocoena phocoena*	1965–1971	nt	nt			d	Holden (1975)
	Netherland	*Phocoena phocoena*	1970s–1993	d					Van Scheppingen et al (1996)
N W Atlantic Ocean	W. Hudson Bay	*Delphinapterus leucas*	1966,1967–1986	nt	d	nt	i		Muir et al. (1990)
	Bay of Fundy	*Phocoena phocoena*	1969–1973		d				Gaskin, Holdrinet, and Frank (1982)
	Bay of Fundy	*Phocoena phocoena*	1971–1977	nt					Gaskin, Frank, and Holdrinet (1983)
	St.Lawrence Gulf	*Delphinapterus leucas*	1982,1985–1994	d	d				Muir et al. (1996)
	St.Lawrence Gulf	*Delphinapterus leucas*	1986,1990–1994			d	d		Muir et al. (1996)
	Lancaster Sound	*Monodon monoceros*	1982–1994	i	i		i		Muir et al. (1999)
	Mackenzie delta	*Monodon monoceros*	1983–1994	nt	nt		nt		Muir et al. (1999)
NW Pacific Ocean	Japan	*Stenella coeruleoalba*	1978,1979–1986	nt	nt	d	d		Loganathan et al. (1990)
	Japan	*Balaenoptera acutorostrata*	1987–1994	nt or i	nt or d	nt or d	nt or d		Aono et al. (1997)
W Indian Ocean	South Africa	*Tursiops truncatus*	1980,1983–1984,1987	nt	nt			nt	Cockroft et al. (1989)
	South Africa	*Tursiops truncatus*	1980–1987	nt	d				de Kock et al. (1994)
	South Africa	*Delphinus delphis*	1980–1985	nt	nt				de Kock et al. (1994)
N Indian Ocean	Ganges River	*Platanista gangetica*	1988,1992–1994,1996	i	i		i		Senthilkumar, Kannan, Sinha, Tanabe, and Giesy (1999)
Antarctic Ocean		*Balaenoptera acutorostrata*	1984–1991	i	nt	nt	nt		Tanabe, Aono, Fujise, Kato, and Tatsukawa (1995)
Antarctic Ocean		*Balaenoptera acutorostrata*	1984–1993	i	nt	nt	nt		Aono et al. (1997)

[a] i, increase; d, decrease; nt, no trend observed.

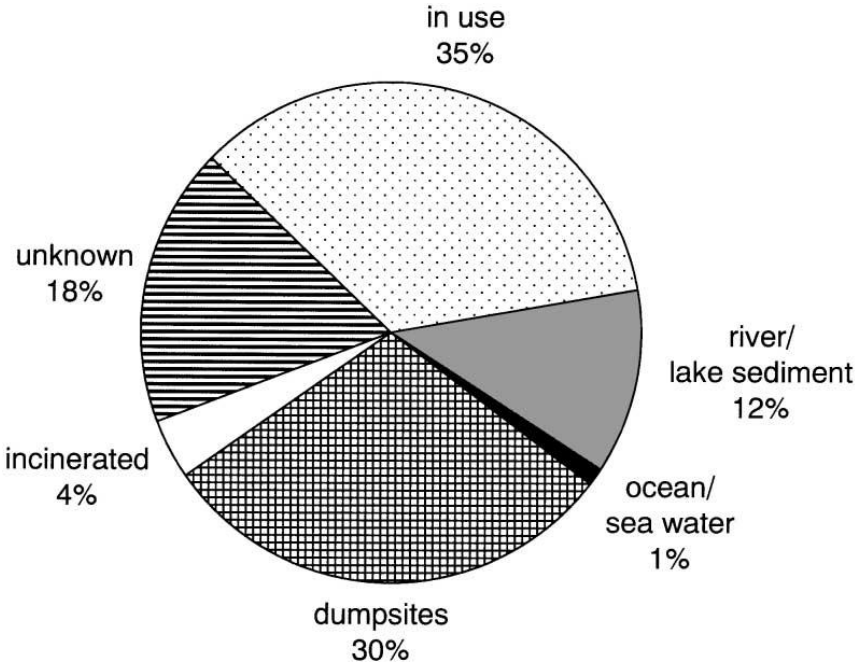

Fig. 4. Global budget of produced PCBs (kilotonnes). After Marquenie and Reijnders (1999).

The other major group of organochlorine compounds which are widely present in marine mammals, PCBs, has followed a somewhat different evolution. The use of these compounds was also discontinued in the 1970s. However, unlike DDTs, which was readily substituted by a new generation of agricultural pesticides, PCBs had no immediate replacement. Consequently, enforcement of controls was poor and the latter compounds have remained in use in many applications. Moreover, the high stability of PCBs and the fact that their disposal is costly and technically difficult, forced industries to store enormous amounts of this product (Safe, Safe, & Mullin, 1987), a problem which still persists today. Also, capacitors containing PCBs are known to have been transported from developed countries to developing ones, therefore facilitating inputs even in non-industrialized regions. This may explain the relatively high levels of PCBs observed in small cetaceans from certain developing countries such as Hong Kong (Minh et al., 1999).

This, together with the redistribution between ecological compartments and the progressive deposition of enormous amounts of PCBs present in the atmosphere, have led Tanabe (1988) and Tateya, Tanabe, and Tatsukawa (1988) to conclude that global PCB levels in marine biota are unlikely to decline in the near future, and certainly not before 2010–2030. Tanabe (1988) calculated that in the mid 1980s only about 30% of all PCBs produced had so far dispersed into the environment. By the end of the 1980s, Marquenie and Reijnders (1989) estimated that only about 1% of all PCBs produced had reached the oceans, while about 30% had accumulated in dump sites and sediment of lakes, coastal zones and estuaries (Fig. 4); their future dispersal into oceans further strengthens the unlikeness of a future decline in the environmental concentrations of these compounds.

The results of the studies detailed in Table 2 reflect the history of the production and use of pollutants. The data shows that there is a decreasing trend in the regions where pollution was initially high, which usually correspond to areas close to point sources of organochlorine compound release, and an increasing trend in regions located far from these sources as a consequence of atmospheric transport and redistribution. In particular, the systematic long-term transfer of airborne pollutants from warmer to colder regions supports the idea that the Arctic and, to a lesser extent, the Antarctic, will become major sinks for organochlorines in the near future (Borrell & Reijnders, 1999; Iwata, Tanabe, Sakal, & Tatsukawa, 1993; Reijnders & de Ruiter-Dijkman, 1995; Tanabe et al., 1994). For example, it is known that pollutants from sources in Europe and the eastern Russian Federation mostly end up in the Arctic region (Ottar, 1981). This process may already be significant for the organochlorine forms that are more movable. Thus, in the North Pacific and Indian Oceans, Prudente et al. (1997) found that concentrations of HCH and HCB were higher in small cetaceans from higher latitudes than in those from temperate or tropical waters. However, this finding should be considered with caution because the comparison involved 11 species and the observed difference could be attributed to interspecific dissimilarities in diet, physiology or other.

In relative terms, the decrease in concentrations in the initially polluted areas was first noticed for DDTs and later for PCBs (Borrell & Reijnders, 1999). The decrease in PCB concentrations is particularly clear in some highly contaminated areas such as Lake Ontario, the Baltic Sea, the Wadden Sea and the North Sea (Addison, Zinck, & Smith, 1986; Borrell & Reijnders, 1999; Olsson & Reutergard, 1996; Reijnders 1996). However, since the mid-1980s this trend appears to be leveling off for both compound groups in the Arctic, an observation which is also found in seabirds (Muir et al., 1999). This suggests that recent PCB and DDT inputs due to geographical redistribution may be large enough in certain areas to maintain concentrations at a steady state. Data on other organochlorine compounds, such as HCH, chlordane, and toxaphene are extremely limited and often do not show any clear pattern with the exception of slight declines in some areas like the Arctic and the North Pacific (Loganathan et al., 1990; Muir et al., 1999), and also some increases in areas located near point sources of pollution (Table 2).

Thus, when comparing data from the 1970s to the 1990s, levels of all organochlorine compounds in the blubber of cetaceans and pinnipeds often showed decreasing trends in the Baltic Sea, the North Sea, the Mediterranean Sea, the Atlantic and Pacific coasts of the USA and Canada, and the Sea of Japan. However, no significant trend was observed on the coasts of South Africa or in some localities of the Canadian Arctic Finally, concentrations, particularly of PCBs, tended to increase in certain areas of the Canadian Arctic, the Ganges River and, especially, in the Antarctic.

It is likely that these patterns of variation will continue in the coming years. Therefore it is essential that stringent policies are developed to ensure the correct collection and disposal of the PCB compounds still in use or stored in improper facilities (Reijnders & de Ruiter-Dijkman, 1995). The monitoring of organochlorine concentrations in the blubber of marine mammals in the temperate fringe of the

northern hemisphere should continue to assess the impact on the currently highly-polluted marine mammal populations. Moreover, given that the information from the southern hemisphere and the Arctic and Antarctic Oceans is extremely scarce and that these water masses are likely to become a major sink for the most persistent forms of organochlorines, particularly PCBs, a special effort should be made to increase their long-term monitoring.

Acknowledgements

This study was possible through funding granted by the *Comisión Interministerial para la Ciencia y Tecnología-CICYT* (projects AMB-399/94 and AMB99-0640), and the *Dirección General de Conservación de la Naturaleza-DGCONA*, Ministry of the Environment of Spain.

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