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Widespread contamination by *tris*(4-chlorophenyl)methane and *tris*(4-chlorophenyl)methanol in cetaceans from the North Pacific and Asian coastal waters

T. Binh Minh[a], M. Watanabe[a], S. Tanabe[a,*], N. Miyazaki[b], T.A. Jefferson[c,d], M.S. Prudente[e], A. Subramanian[f], S. Karuppiah[f]

[a]*Center for Marine Environmental Studies, Ehime University, Tarumi 3-5-7, Matsuyama 790-8566, Japan*
[b]*Otsuchi Marine Research Center, The Ocean Research Institute, The University of Tokyo, Akahama, Otsuchi-cho, Iwate 028-1102, Japan*
[c]*Southwest Fisheries Science Center, National Marine Fisheries Service, NOAA, PO Box 271, La Jolla, CA 92038, USA*
[d]*Ocean Park Conservation Foundation, Ocean Park, Aberdeen, Hong Kong, People's Republic of China*
[e]*Science Education Department, De La Salle University, 2401 Taft Avenue, 1004 Manila, Philippines*
[f]*Centre of Advanced Study in Marine Biology, Annamalai University, Parangipettai 608502, India*

"Capsule": *Accumulation of TCPMe and TCPMOH in cetaceans was similar to DDTs.*

Abstract

For understanding global distribution, transport and behavior of *tris*(4-chlorophenyl)methane (TCPMe) and *tris*(4-chlorophenyl)methanol (TCPMOH), the two newly identified microcontaminants, the present study determined their concentrations and other persistent organochlorines (OCs) in the blubber of nine species of adult male cetaceans collected from various locations in the North Pacific Ocean and coastal waters of some Asian countries, during 1985–97. Concentrations of TCPMe and TCPMOH were found to be highest in northern right whale dolphins, which may be attributable to wide distribution of this species, including some heavily polluted areas such as coastal California. Elevated residue levels of TCPMe and TCPMOH were observed in both off-shore and coastal species, suggesting widespread contamination of these compounds in the marine environment. Higher contamination of TCPMe and TCPMOH was found in cetaceans from temperate and cold waters than those from tropical regions. The latitudinal distribution of TCPMe and TCPMOH in cetaceans from the North Pacific and Asian coastal waters was similar to that of DDTs, suggesting less transportable nature of TCPMe and TCPMOH in the marine environment. Data on the occurrence of TCPMe and TCPMOH further indicated high bioaccumulation potential of these compounds, which was comparable to DDTs. Relative concentrations of TCPMe/TCPMOH in cetaceans were apparently higher than those observed in seals, suggestive of lower metabolic capacity for these compounds in cetaceans than in pinnipeds, which is similar to classic contaminants like polychlorinated biphenyls and DDTs. © 2000 Elsevier Science Ltd. All rights reserved.

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1. Introduction

During the last decade, numerous investigations have been conducted to elucidate the global distribution of persistent organochlorines (OCs). Some OCs, such as DDTs and polychlorinated biphenyls (PCBs), pose potential toxic effects to humans and wildlife due to their persistent and bioaccumulative nature in various environmental compartments, particularly in animal bodies. *Tris*(4-chlorophenyl)methane (TCPMe) and *tris*(4-chlorophenyl)methanol (TCPMOH) are among the most recently identified OC microcontaminants in environmental samples (Buser, 1995). The structural similarity of these two compounds to some well-known and toxic pesticides such as DDTs and dicofol leads to the belief that these chemicals are widely distributed in the environment and could also pose toxic threat. Actually, TCPMOH has been detected in high trophic animals such as dolphins (de Boer et al., 1996), polar bear and herring gull (Jarman et al., 1992) at elevated

* Corresponding author. Tel./fax: +81-89-946-9904.
E-mail address: shinsuke@agr.ehime-u.ac.jp (S. Tanabe).

levels. Regarding toxicity of these compounds, recent in vivo and in vitro studies have shown that TCPMOH may induce hepatic enzymes and pose anti-androgenic effect (Körner et al., 1997; Poon et al., 1997). Furthermore, toxic responses of TCPMOH may also involve endocrine disrupting effect (Aguilar et al., 1997). However, data on the environmental occurrence of TCPMe and TCPMOH are so far limited and, therefore, anthropogenic sources as well as ultimate fate and transport in aquatic ecosystem are still unclear.

In the present study, concentrations of TCPMe and TCPMOH were determined in the blubber of nine species of adult male odontoceti cetaceans collected from various locations in the North Pacific Ocean and Asian coastal waters. For comparison, concentrations of classic OCs such as DDT and its metabolites (DDTs), PCBs, hexachlorocyclohexane isomers (HCHs), hexachlorobenzene (HCB) and chlordane compounds (CHLs) were also determined. In order to understand the behavior and transport of TCPMe and TCPMOH on a global scale, we also examined their latitudinal distributions and compared with those for other persistent OCs such as DDTs, PCBs, and HCHs, which were characterized to have low, moderate and high potential to migrate around the globe, respectively (Iwata et al., 1993; Wania and Mackay, 1996). Furthermore, existing data on global contamination by TCPMe and TCPMOH were compiled and interpreted to understand the bioaccumulation feature in marine mammals.

2. Materials and methods

Blubber samples were taken from seven species of adult male cetaceans collected from different locations in the North Pacific Ocean and along coastal waters of Japan, Philippines and India during 1985–97. Data for Indo-Pacific hump-backed dolphin (*Sousa chinensis*) and finless porpoise (*Neophocaena phocaenoides*) from Hong Kong coastal waters are also cited based on the results of our previous study (Minh et al., 1999). Geographical locations of cetaceans collected are shown in Fig. 1. Blubber samples were obtained from fresh strandings along the coastlines, accidental catches by fishermen and specimens collected by Whaling for Commercial and Scientific Purposes that were accepted by an International Convention for the Regulation of Whaling. In the framework of this research, species with large population size were collected and endangered species were ruled out. Blubber samples were taken from adult male individuals and immediately after dissection, samples were frozen in clean plastic bags, transported to the laboratory and stored at −20°C until analysis.

Chemical analyses of TCPMe and TCPMOH and other OCs were followed by the method previously described (Watanabe et al., 1999). Briefly, about 5 g of blubber samples were homogenized with anhydrous Na_2SO_4 and extracted using a Soxhlet apparatus with a mixture of hexane and diethyl ether. Fat content was determined from an aliquot of the extract. The extract was then added into a dry Florisil column to remove fat. OCs were eluted with 150 ml of 20% water in acetonitrile to a separatory funnel containing hexane and water. After partitioning, the hexane layer was concentrated and then passed through 8 g of activated Florisil column for fractionation. The first fraction eluted with hexane contained PCBs, *p,p'*-DDE, *trans*-nonachlor and HCB, while the second fraction eluted with 20% dichloromethane in hexane contained other OC pesticides and TCPMe. The third fraction eluted with 50% dichloromethane in hexane contained TCPMOH. Each fraction was concentrated and injected into a gas chromatograph with electron capture detector (GC–ECD) and a gas chromatograph with a mass selective detector (GC–MSD) for quantification.

OCs (except TCPMe and TCPMOH) were quantified by a GC–ECD (Hewlett Packard 5890 Series II) equipped with a moving needle-type injection port. The GC column employed was DB-1 (J & W Scientific Inc., USA) fused silica capillary (0.25 mm×30 m) coated with 100% dimethylpolysiloxane at 0.25 μm film thickness. The column oven temperature was programmed from 60 to 160°C, held for 10 min, then increased to 260°C at a rate of 20°C/min and held for 20 min. Injector and detector temperatures were set at 260 and 280ºC, respectively. Helium and nitrogen were used as carrier and make-up gases, respectively. OC concentrations were calculated from the peak area of the sample to the corresponding external standard. The PCB standard used for quantification was an equivalent mixture of Kanechlor preparations (KC-300, KC-400, KC-500, KC-600) with known PCB composition and content. Concentrations of individually resolved peaks of PCB isomers and congeners were summed to obtain total PCB concentrations. For the quantification of TCPMe and TCPMOH, a GC–MSD (Hewlett-Packard 5890 series II GC coupled with 5972 mass selective detector) was employed. Data acquisition was performed by a Hewlett-Packard 5972C data system, in which the cluster ions were monitored at m/z 311, 313, 346, 348 for TCPMe and 139, 251, 253, 362, 364 for TCPMOH. TCPMe standard was synthesized by Tri Chemical Laboratory Inc., Yamanashi, Japan, while TCPMOH standard was obtained from Lancaster Synthesis, Morecambe, UK. The purity of both standards was >95%. Recoveries of this analytical method were 95±1.1% for TCPMe, 100±2.1% for TCPMOH, 99±2.0% for PCBs, 95±7.5% for DDTs, 96±7.7% for HCHs, 100±4.7% for CHLs, 94±5.9% for HCB. Concentrations were not corrected for recovery rates. DDTs represent the sum of *p,p'*-DDE, *p,p'*-DDD and

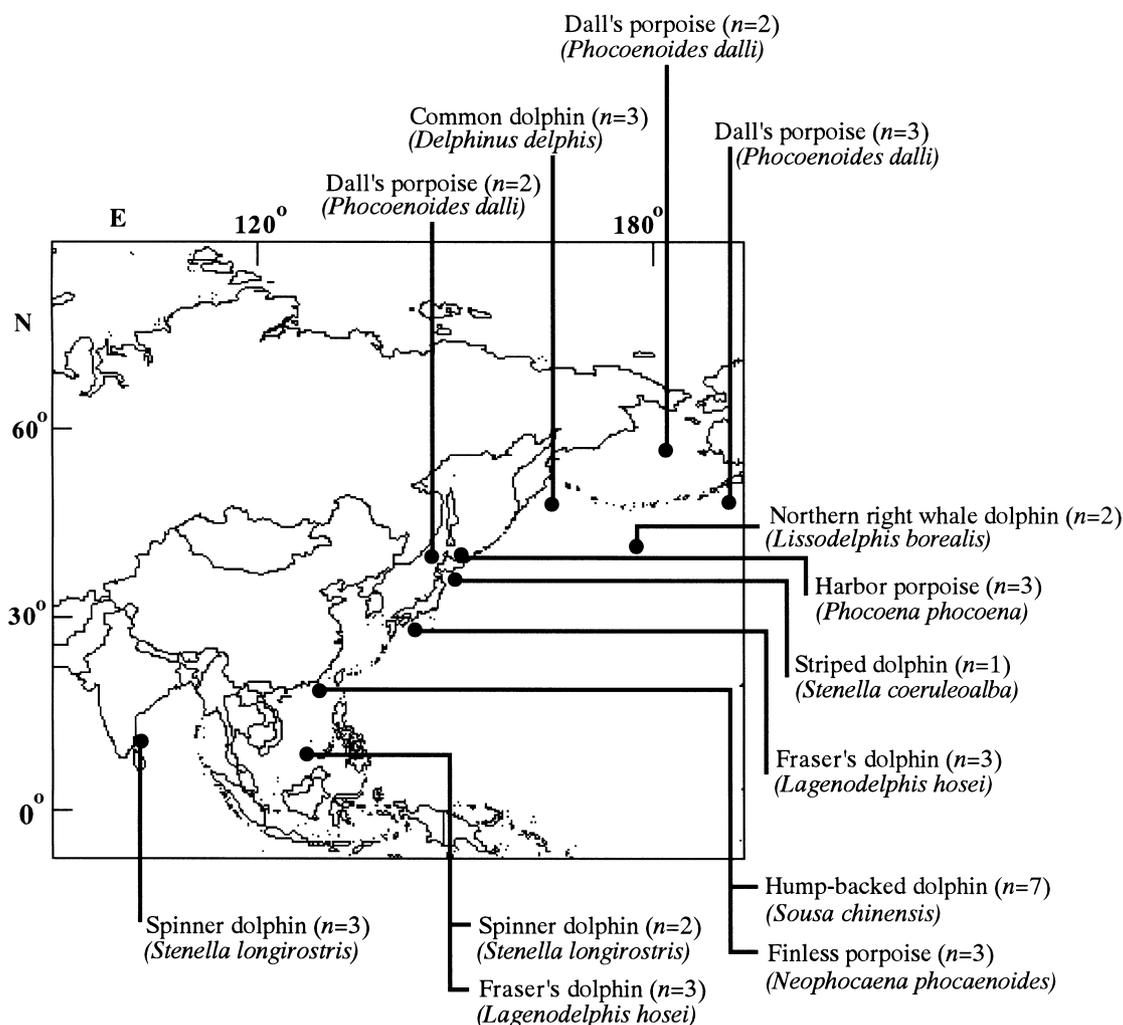Dall's porpoise (*n*=2)
(*Phocoenoides dalli*)

Common dolphin (*n*=3)
(*Delphinus delphis*)

Dall's porpoise (*n*=3)
(*Phocoenoides dalli*)

Dall's porpoise (*n*=2)
(*Phocoenoides dalli*)

Northern right whale dolphin (*n*=2)
(*Lissodelphis borealis*)

Harbor porpoise (*n*=3)
(*Phocoena phocoena*)

Striped dolphin (*n*=1)
(*Stenella coeruleoalba*)

Fraser's dolphin (*n*=3)
(*Lagenodelphis hosei*)

Hump-backed dolphin (*n*=7)
(*Sousa chinensis*)

Finless porpoise (*n*=3)
(*Neophocaena phocaenoides*)

Spinner dolphin (*n*=3)
(*Stenella longirostris*)

Spinner dolphin (*n*=2)
(*Stenella longirostris*)

Fraser's dolphin (*n*=3)
(*Lagenodelphis hosei*)

Fig. 1. Map showing geographical sampling locations of cetaceans from the North Pacific and Asian coastal waters. Values in parentheses indicate number of samples analyzed.

p,p'-DDT, while CHLs include *cis*-chlordane, *trans*-chlordane, *cis*-nonachlor, *trans*-nonachlor and oxy-chlordane. HCHs include α-, β-, and γ-isomers.

3. Results and discussion

3.1. Residue levels

Concentrations of TCPMe, TCPMOH and other OCs in the blubber of cetaceans collected from the North Pacific and Asian coastal waters are given in Table 1. In general, residue pattern was in the order of DDTs $\geqslant$ PCBs > CHLs > HCHs > HCB > TCPMOH > TCPMe. Elevated concentrations of PCBs and DDTs were found in some species collected from Japanese and Hong Kong coastal waters, indicating serious pollution by persistent OCs in Asian industrialized nations. PCB residue levels in some individuals of Fraser's dolphins collected off Kii Peninsula, Japan, and hump-backed

dolphins from Hong Kong exceed 50 µg/g wet wt. in the blubber.

In general, TCPMe and TCPMOH residue levels were the lowest among OCs analysed. This can be explained by the origin of these compounds, which has been, so far, considered to be impurities of technical pesticide preparations such as DDT (Buser, 1995) and dicofol (de Boer, 1997) as well as synthetic high polymer and dye products (Jarman et al., 1992). However, TCPMe and TCPMOH were detected in most of the samples ana-lysed, ranging from 0.5–470 ng/g to <0.1–440 ng/g on a wet weight basis, respectively, which may indicate widespread contamination by these compounds in the marine ecosystem. A previous investigation also repor-ted global occurrence of TCPMOH in various marine mammals and sea birds from Canadian Arctic, North America, Australia and Antarctica (Jarman et al., 1992). Among the cetaceans analyzed, northern right whale dolphins collected from the North Pacific Ocean contained the highest concentration of TCPMe and

Table 1
Mean and range concentrations (wet wt.) of TCPMe, TCPMOH and other OCs in cetaceans from the North Pacific and Asian coastal waters[a]

Location	Species	Collection year	n	Lipid content (%)	TCPMe (ng/g)	TCPMOH (ng/g)	DDTs (μg/g)	PCBs (μg/g)	HCHs (ng/g)	HCB (ng/g)	CHLs (ng/g)
Bering Sea	Dall's porpoise (*Phocoenoides dalli*)	1985	2	87 (84–89)	4.3 (1.3–7.2)	16 (3.0–28)	6.5 (5.4–7.5)	13 (11–14)	780 (650–910)	620 (560–680)	3000 (2200–3800)
Northeastern North Pacific	Dall's porpoise (*P. dalli*)	1987	4	87 (85–88)	4.1 (2.1–7.0)	14 (1.1–25)	6.7 (6.2–7.1)	19 (16–21)	900 (790–1100)	380 (350–420)	2700 (2500–3100)
Northwestern North Pacific	Common dolphin (*Delphinus delphis*)	1987	3	67 (64–68)	25 (6.0–63)	14 (2.0–30)	8 (6.9–9.0)	22 (19–26)	150 (25–430)	140 (97–170)	1400 (1300–1600)
Hokkaido, Japan	Harbor porpoise (*Phocoena phocoena*)	1993	3	77 (70–90)	2.0 (0.5–3.9)	75 (35–130)	9.6 (2.5–21)	7.9 (4.1–12)	1300 (420–3100)	370 (150–490)	1200 (570–1700)
Japan Sea	Dall's porpoise (*P. dalli*)	1989	3	87 (86–89)	4.5 (3.5–5.4)	<0.1	18 (13–23)	29 (19–34)	2200 (1900–2400)	580 (460–740)	3300 (2800–4000)
Northern North Pacific	Northern right whale dolphin (*Lissodelphis borealis*)	1991	2	84 (82–85)	330 (180–470)	300 (150–440)	32 (30–33)	30 (30–30)	590 (490–690)	210 (150–260)	3100 (2300–3800)
Off Sanriku, Japan	Striped dolphin (*Stenella coeruleoalba*)	1992	1	50	5.8	28	17	37	310	120	2900
Off Kii Peninsula, Japan	Fraser's dolphin (*Lagenodelphis hosei*)	1991	3	70 (61–78)	39 (13–82)	34 (15–51)	27 (20–32)	51 (45–57)	140 (110–160)	120 (110–130)	3000 (2100–3400)
Hong Kong	Hump-backed dolphin (*Sousa chinensis*)	1993–97	7	46 (31–76)	100 (43–240)	180 (66–270)	56 (31–80)	31 (13–50)	1100 (300–2200)	88 (13–240)	430 (260–580)
Hong Kong	Finless porpoise (*Neophocaena phocaenoides*)	1993–97	3	64 (29–87)	120 (27–260)	150 (35–300)	74 (26–160)	20 (5.9–48)	440 (54–1100)	74 (26–110)	310 (94–720)
Mindanao Sea, Philippines	Spinner dolphin (*Stenella longirostris*)	1996	2	66 (63–68)	32 (27–37)	34 (30–38)	4.9 (4.4–5.4)	2.5 (2.4–2.6)	48 (48–48)	42 (35–48)	360 (250–460)
Mindanao Sea, Philippines	Fraser's dolphin (*L. hosei*)	1996	3	65 (64–67)	21 (19–44)	19 (3.0–31)	7.1 (5.4–9.1)	4.1 (3.8–8.6)	47 (32–67)	36 (32–39)	330 (300–390)
Bay of Bengal, India	Spinner dolphin (*S. longirostris*)	1990	3	66 (53–73)	31 (8.7–63)	36 (10–58)	18 (12–27)	2.2 (1.6–3.0)	120 (56–220)	15 (7.1–23)	96 (71–130)

[a] Data on hump-backed dolphins and finless porpoises from Hong Kong were cited from Minh et al. (1999). Values in parentheses indicate the range.

TCPMOH (Table 1). This species mainly inhabits deep off-shore waters. The distribution area of this species is quite large, extending from the Kamchatka in the western North Pacific to Japan and also from British Columbia in the eastern North Pacific to California coasts (Carwardine, 1995). Elevated concentrations of TCPMe and TCPMOH as well as other OCs in this species may be attributable to exposures in some heavily polluted areas such as California coastal waters. Actually, elevated TCPMOH residues were reported in peregrine falcon eggs from British Columbia and California (Jarman et al., 1992). Furthermore, a high level of this compound was also detected in the milk of California sea lion (Jarman et al., 1992) as well as in livers of sea otters (Bacon et al., 1999) collected along California coasts. Similar to other OCs, relatively high accumulation of TCPMe and TCPMOH was found in cetaceans from Hong Kong.

Comparing residue levels found in this study to those reported from other parts of the world, contamination by TCPMe and TCPMOH were recorded to a lesser extent. As shown in Fig. 2, concentrations of TCPMe in cetaceans from the North Pacific and Asian coastal waters were lower than those reported in beluga whales from the St. Lawrence River, Canada (Muir et al., 1996a, b), comparable to higher than those in harbor porpoises from the Baltic south coast (Falandysz et al., 1999). For TCPMOH, elevated concentrations were observed in killer whales from the Pacific coast of Japan, similar to those reported for cetaceans from the Wadden Sea and Dutch coastal waters (de Boer et al., 1996). TCPMOH residues in cetaceans from the coastal waters of the Philippines, India, Hong Kong, the North Pacific and the Bering Sea were comparable to those observed in beluga whales from the St. Lawrence River,

Canada, but greater than those in harbor porpoises from the Baltic Sea (Falandysz et al., 1999), whitebeaked dolphin and common dolphin from the Dutch Wadden Sea (de Boer et al., 1996). Nevertheless, the levels are still significantly lower than those in harbor porpoise and whitebeaked dolphin from the North Sea (de Boer et al., 1996). Moreover, lipid-normalized concentrations of TCPMe and TCPMOH in the blubber of cetaceans analyzed in this study were apparently lower than those reported in the liver of small cetaceans stranded along coastal Florida, USA (Watanabe et al., 2000). Relatively high residues of TCPMOH were also reported in the liver of polar bears from Churchill, Canada (Jarman et al., 1992). In general, residue levels of TCPMe and TCPMOH in cetaceans from temperate and cold waters were higher than those in cetaceans from tropical regions. Variations in concentrations might be attributable to the differences in sampling time, age, sex, trophic level and metabolic capacity among marine mammals examined. In addition, greater residue levels of TCPMe and TCPMOH were found in highly industrialized areas such as North Sea, St. Lawrence River, Canada and Hong Kong coastal waters, indicating that industrial and human activities may influence the extent of environmental contamination by these compounds.

3.2. Transport and behavior in global terms

In order to understand the transport and behavior of TCPMe and TCPMOH on a global scale, latitudinal distributions of these compounds detected in cetaceans were examined in comparison with other persistent organochlorines (Fig. 3). It is clear that the distribution pattern of TCPMe and TCPMOH is quite different from that of HCHs, which showed higher residue levels
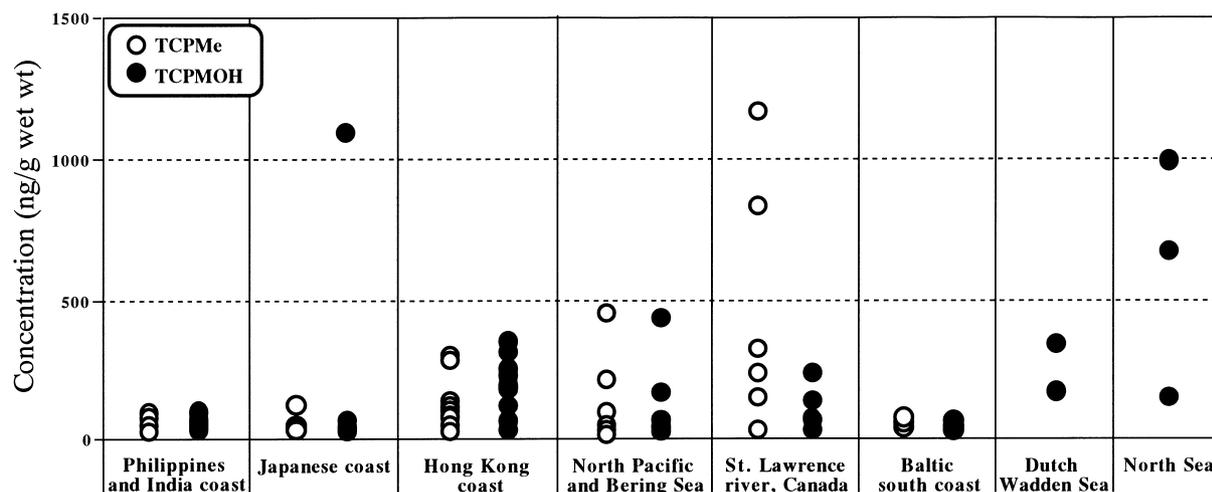

Fig. 2. Comparison of TCPMe and TCPMOH residue levels in cetaceans collected from various regions. Data from St. Lawrence River, Canada, were cited from Muir et al. (1996a, b); Baltic south coast from Falandysz et al. (1999); North Sea and Dutch Wadden Sea from de Boer et al. (1996); Japanese coast from the present study and Watanabe et al. (1999); Philippines, India, Hong Kong coast, North Pacific and Bering Sea from the present study.
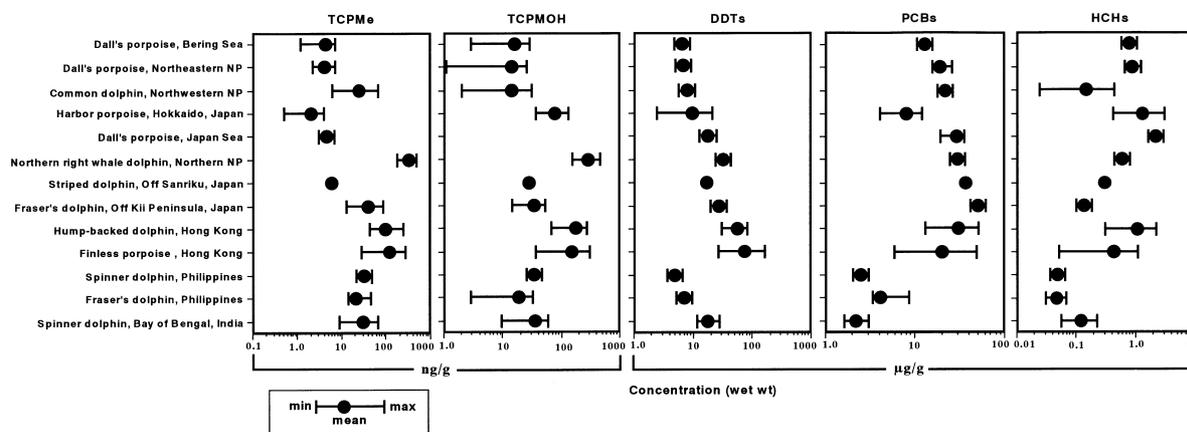
Fig. 3. Latitudinal distributions of TCPMe, TCPMOH and other persistent OCs in cetaceans from the North Pacific (NP) and Asian coastal waters.

in high-latitude regions, such as the Bering Sea and northeastern North Pacific. When compared with PCBs, different residue patterns were also observed, wherein residue levels in tropical cetacean species (Phillipines, India) were relatively higher for TCPMe and TCPMOH than PCBs. Interestingly, distribution pattern of TCPMe and TCPMOH is similar to that of DDTs, which showed an increasing trend in the mid-latitude region and decreasing trend towards the tropical region as in the Philippines and in India. The highest concentrations of TCPMe, TCPMOH and DDTs were found in northern right whale dolphins from the northern North Pacific and hump-backed dolphins and finless porpoises from Hong Kong, while the lowest concentrations were observed in colder regions. Our previous study on marine mammals from Russia and Japan indicated that the bioaccumulation potential of TCPMe and TCPMOH is comparable to DDTs (Watanabe et al., 1999). A recent investigation has also suggested high biomagnification of TCPMe and TCPMOH in marine food chain, even to a greater extent than DDTs (Falandysz et al., 1999). Similar bioaccumulation potential of TCPMe and TCPMOH to DDTs might be explicable from their distribution. However, it is noted that the differences in OC residues between individual cetaceans, which may influence the distribution, cannot be ruled out.

The atmospheric transport processes of semivolatile organic compounds are mainly influenced by their physico-chemical properties, particularly Henry's Law constant (HLC) (Iwata et al., 1993). DDT compounds, with low HLCs (low mobility), tend to remain in the vicinities of the pollution sources. On the other hand, HCH isomers, with high vapor pressures, tend to be transported to long distances, primarily towards the polar regions. PCBs are considered to have moderate mobility as compared with DDT compounds and HCH isomers. Considering these properties, results of the present study suggest that similar to DDT, TCPMe and TCPMOH may have less mobility to migrate through long-range atmospheric transport.

3.3. Bioaccumulation feature in marine mammals

Existing data on the occurrence of TCPMOH in different kinds of biological samples facilitate further understanding about the bioaccumulation feature of this compound. TCPMe residues were scarcely reported since its analytical standard was synthesized only recently (de Boer et al., 1996). Data available for fish, seals, polar bears, cetaceans and birds collected from various locations in the world clearly indicated that TCPMOH has a strong biomagnification potential along the aquatic food chain (Fig. 4). Results on Caspian seals from Russia and larga seals from northwestern Japan suggested comparable biomagnification factors (BMFs) of TCPMe ($\log$BMF = 2.3), TCPMOH ($\log$BMF = 2.4) and DDTs ($\log$ BMF = 2.4) (Watanabe et al., 1999). TCPMe and TCPMOH residues in southern Baltic Sea food web indicated a high bioaccumulation potential (Falandysz et al., 1999). The above findings suggest the highly bioaccumulative nature of these two compounds. High biomagnification potential of TCPMe was also suggested in a recent investigation on physico-chemical property and dietary bioaccumulation relationships (Fisk et al., 1998). TCPMe and TCPMOH were hydrophobic OCs with high values of octanol–water partition coefficients (K_{ow}). The $\log K_{ow}$ of TCPMe was estimated to be 7.6, comparable to nonachlorobiphenyls ($\log K_{ow} \approx 7.5$) (Mackay et al., 1992) but higher than p,p'-DDT ($\log K_{ow} = 6.36$) (Schwarzenbach et al., 1993). TCPMOH ($\log K_{ow} = 5.0$) was considered to be less hydrophobic than TCPMe (Fisk et al., 1998). Data from field studies generally agreed with those observed in laboratory experiments, indicating the highly hydrophobic and bioaccumulative nature of TCPMe and TCPMOH.

In order to evaluate the capacity of marine mammals to metabolize TCPMe and TCPMOH, data on seals and cetaceans collected from different locations were compiled. As shown in Fig. 5, relative concentrations of TCPMe/TCPMOH in cetaceans were significantly
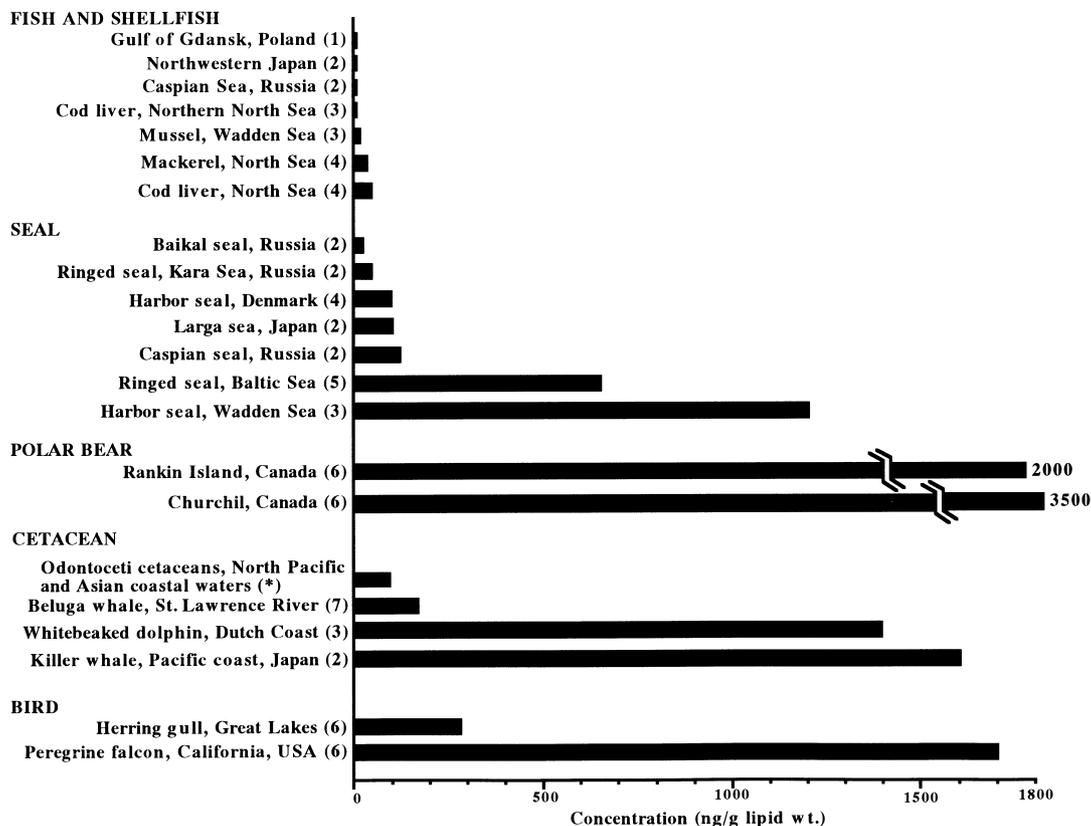
Fig. 4. Comparison of TCPMOH residue levels in biological samples from various locations. Data were cited from: (1) Falandysz et al. (1998); (2) Watanabe et al. (1999); (3) de Boer et al. (1996); (4) Rahman et al. (1993); (5) Zook et al. (1992); (6) Jarman et al. (1992); (7) Muir et al. (1996a); (*) present study.
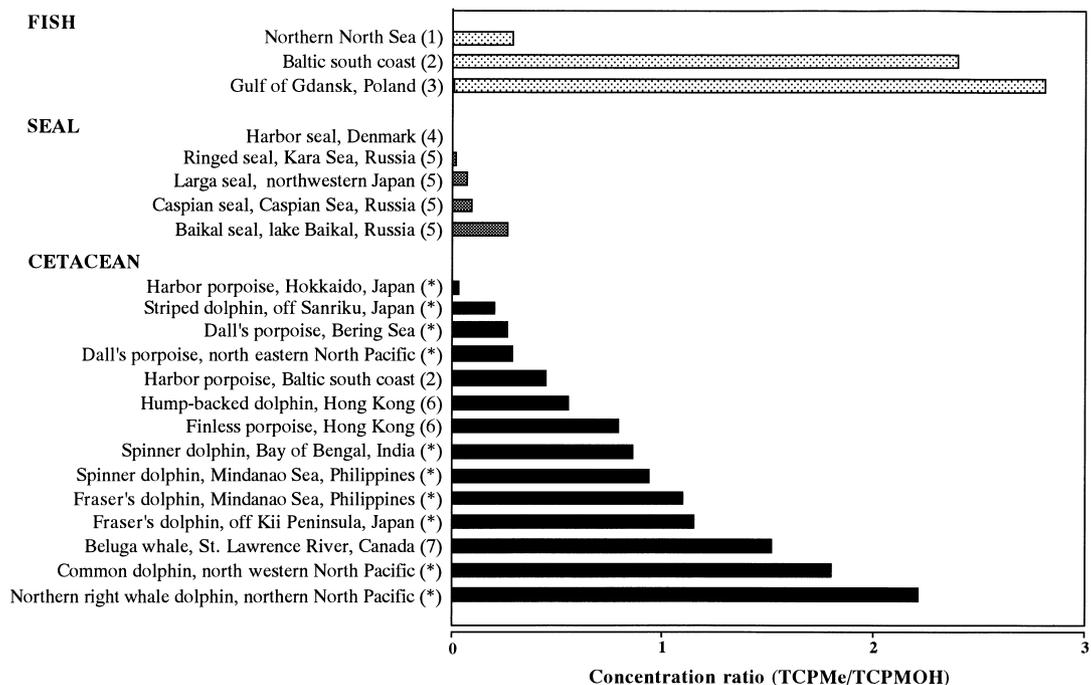
Fig. 5. Comparison of concentration ratios TCPMe/TCPMOH in different biological samples from various locations. Data were cited from: (1) de Boer et al. (1996); (2) Falandysz et al. (1999); (3) Falandysz et al. (1998); (4) Rahman et al. (1993); (5) Watanabe et al. (1999); (6) Minh et al. (1999); (7) Muir et al. (1996a); (*) present study.

higher than those observed in seals. This fact may suggest the differences in metabolic capacity to TCPMe and TCPMOH in cetaceans and in pinnipeds. It is well known that cetaceans have lower ability to metabolize persistent OCs compared with other mammals due to the specific mode of cytochrome P-450 enzyme system (Tanabe et al., 1994). In the case of TCPMe and TCPMOH, considerably higher residue levels in cetaceans than in seals may also suggest lower capacity to degrade these two compounds in cetaceans. In addition, local source of pollution may also be a reason for the variations in concentration ratio. However, existing data so far demonstrated that, in general, concentration ratios of TCPMe to TCPMOH in cetaceans were apparently greater than in seals, even in some polluted areas such as lake Baikal and Caspian Sea (Fig. 5). Considering these observations, it can be suggested that the differences in metabolic capacity among different marine mammals play an important role in the accumulation feature and kinetics of TCPMe and TCPMOH in their bodies.

3.4. Pollution sources

Information regarding the sources of TCPMe and TCPMOH is rather limited. An experimental investigation conducted by Buser (1995) provided a link between environmental existence of TCPMe and DDT. The author detected trace levels of TCPMe in two technical DDT preparations. When these levels in technical DDT were compared with those in different kinds of biological samples, it was noted that concentration ratios of TCPMe/DDTs in environmental samples were apparently higher (Fig. 6). Moreover, considering the similar bioaccumulation potential of TCPMe/TCPMOH and DDTs as discussed above, and the different concentration ratios of TCPMe/DDTs in technical DDT and environmental samples, it is believed that technical DDT may not be the only source of TCPMe. However, the present results suggest higher enrichment rate of TCPMe than DDTs in food chain. Since K_{ow} of TCPMe is greater than that of DDT as mentioned earlier, TCPMe could be more persistent for metabolism than DDT and as a result, TCPMe/DDTs ratio in biological samples are expected to be higher than that in technical DDT.

Some evidences for the possible sources of TCPMe and/or TCPMOH from technical DDT were indicated by significant correlations between DDTs and TCPMe/TCPMOH concentrations found in various biological samples. Concentrations of TCPMOH were reported to be highly correlated with DDT concentrations in harbor seals from Pudget Sound (Walker et al., 1989) and in various marine mammals such as beluga whales, harp seals, northern fur seals, polar bears as well as in eggs of herring gulls (Jarman et al., 1992). In the Baltic coast food web, TCPMe/TCPMOH was correlated with DDTs in flounders, perches, lampreys, but in higher trophic animals such as white-tailed sea eagles and harbor porpoises, only p,p'-DDE concentrations were found to be highly correlated with TCPMe and/or TCPMOH (Falandysz et al., 1999). Our recent investigation in
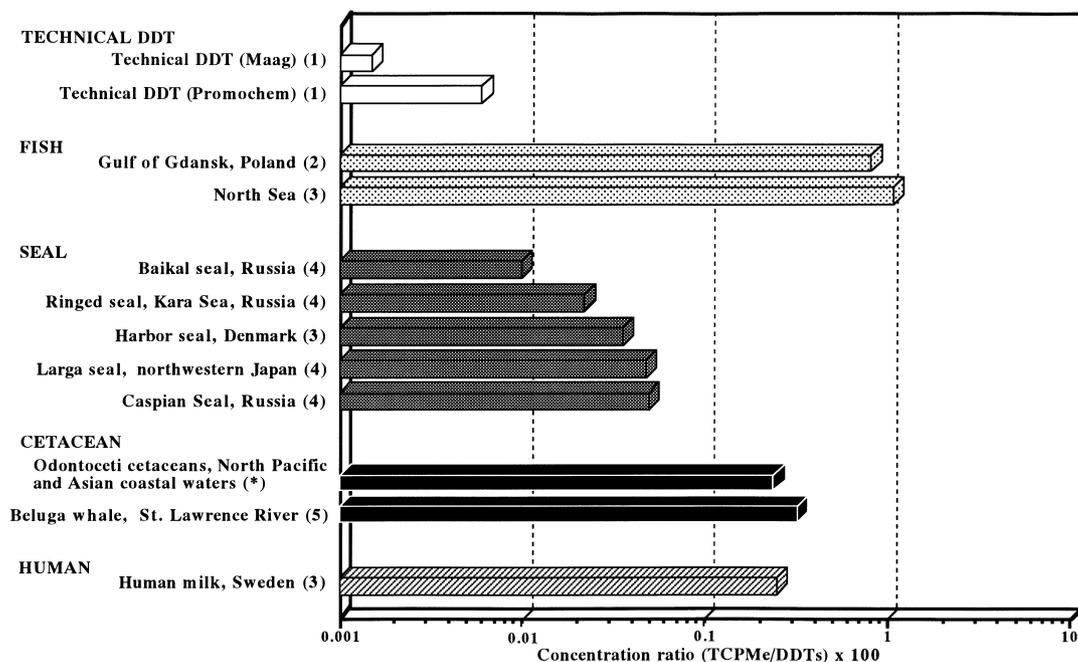

Fig. 6. Comparison of concentration ratios TCPMe/DDTs in technical DDT preparations with those in biological samples from various locations. Data were cited from: (1) Buser (1995); (2) Falandysz et al. (1998); (3) Rahman et al. (1993); (4) Watanabe et al. (1999); (5) Muir et al. (1996a); (*) present study.

Caspian seals has reported a significant relationship of TCPMe and TCPMOH with DDT concentrations (Watanabe et al., 1999). In the present survey on cetaceans from the North Pacific and Asian region, significant correlation was found in hump-backed dolphins and finless porpoises from Hong Kong and various species collected from the North Pacific (Fig. 7). Thus, the existing data on the occurrence of TCPMe and/or TCPMOH suggest that technical DDT is still a known source for these compounds.

Jarman et al. (1992) suggested that TCPMOH may be a starting product for the manufacture of dyes for acrylic fibers and synthetic high polymers. Information on the use of TCPMOH in synthetic high polymers and dyes for acrylic fibers and worldwide contamination by this compound in open ocean marine mammals may support the possible transport of polymers, synthetic dyes and agrochemicals used in the terrestrial environment to oceans.
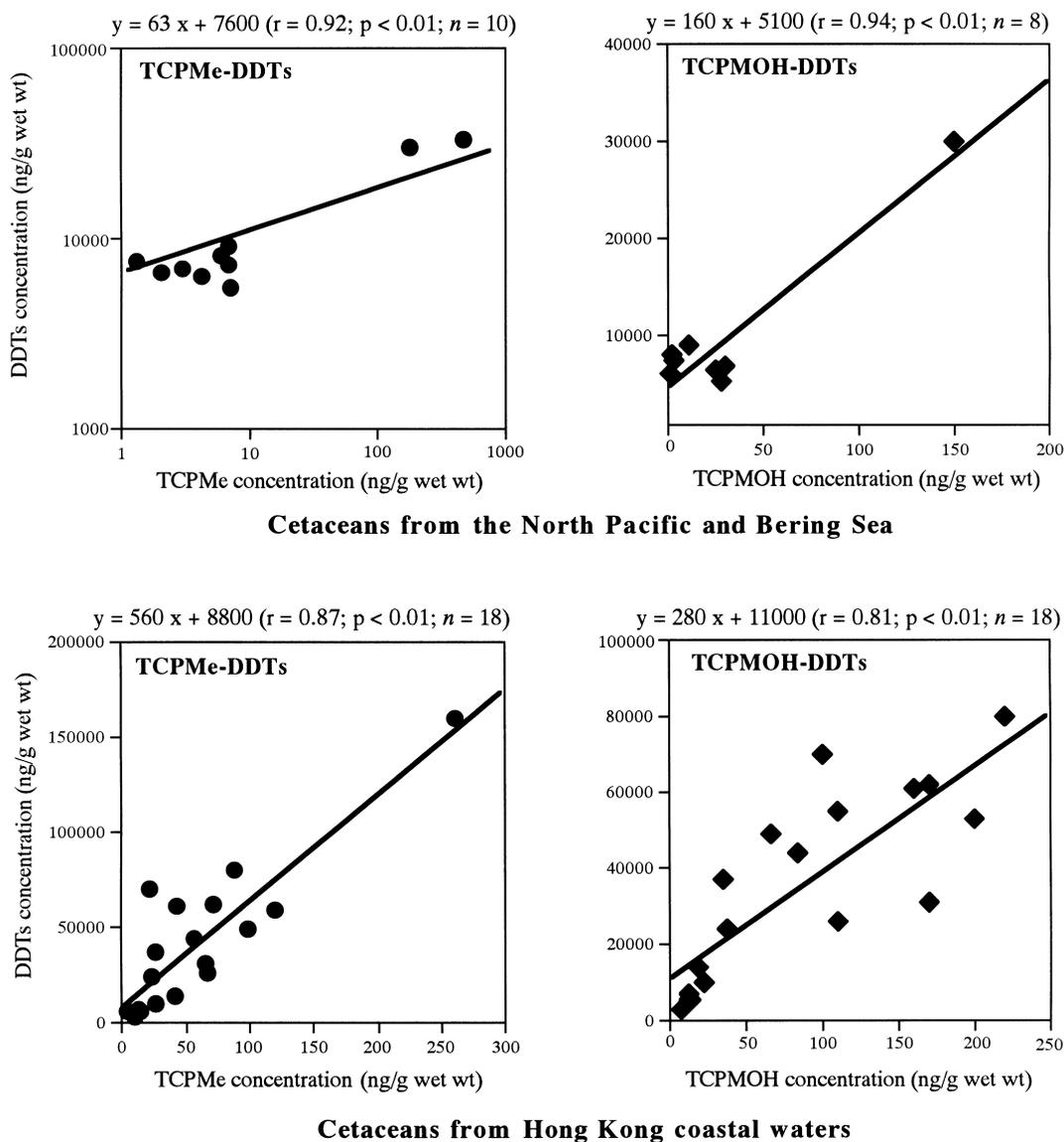
Fig. 7. Correlation between TCPMe, TCPMOH and DDT concentrations in cetaceans from North Pacific, Bering Sea and Hong Kong coastal waters.

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