

LEVELS AND HUMAN HEALTH SIGNIFICANCE OF DIOXINS AND COPLANAR PCBS IN CETACEAN PRODUCTS SOLD IN JAPAN

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Introduction

Whale meat and products are available in retail markets in Japan. Most of them are believed to be from legal sources; "scientific" whaling of minke whales in the Antarctic and North Pacific, small cetacean hunting; and stockpiles from past whaling operations. According to the Fisheries Agency, the current supply of whale products is "a total of less than 4,500 tons per year," including 2,000 tons from "scientific" whaling.

In a recent survey¹, we found that the cetacean products collected from cities across Japan in 1999 contained high levels of mercury and organochlorines (PCBs, DDTs and other pesticides), in which there were large differences in contaminant levels between products from toothed whales and baleen whales. For example, the products from toothed whales and dolphins contain PCB residues of 2.80 µg/g wet wt (n=19), whereas the products from North Pacific minke whales have the residue of 0.76 µg/g wet wt (n=19), much higher than the products from southern hemisphere minke whales.

Our primary concern is the pollutant load being ingested by human consumers of whale products, especially from small cetaceans in Japanese coastal waters which may contain higher levels of dioxins and coplanar PCBs.

In this study, we calculated 2,3,7,8-TCDD toxic equivalency (TEQ) levels of dioxins (PCDDs/DFs) and coplanar PCBs (Co-PCBs) in meat and bacon/blubber of whales and dolphins sold in Japan to evaluate the risk for human consumption of foods contaminated with such toxic compounds.

Materials and methods

Products, including sashimi, canned and cooked meat, whale bacon, sliced blubber strips, were purchased from retail outlets of six cities or towns across Japan in February 1999. The sub-samples were subjected to DNA analysis by Dr Cipriano and Dr Palumbi of San Francisco and Harvard universities to identify the species². The 38 products used in this study were classified into three groups, toothed whales and dolphins (n=19), North Pacific minke whales (*Balaenoptera acutorostrata*, n=12) and southern hemisphere minke whales (n=7). The species of toothed whales and dolphins were Dall's porpoise (*Phocoenoides dalli*), Striped dolphin (*Stenella coeruleoalba*), Bottlenose dolphin (*Tursiops truncatus*), Risso's

dolphin (*Grampus griseus*), Short-finned pilot whale (*Globicephala macrorhynchus*), Baird's beaked whale (*Berardius bairdii*), and Sperm whale (*Physeter macrocephalus*).

An aliquot of the extract from each product was spiked with an internal standard mixture of ^{13}C -PCDD/PCDFs and coplanar ^{13}C -PCBs (Wellington), and the fat was removed by alkali saponification and conc. H_2SO_4 treatment. Dioxin fraction was separated from PCBs using a column of activated carbon mixed with silica gel. Analyses were carried out by GC/MS (Finnigan MAT95XL/95S) on a Thermo Quest TRACE GC2000 operating in the SIM mode. Resolution for dioxins/Co-PCBs was 10,000. Extracts were chromatographed on DB-5 (PCDDs/DFs and Co-PCBs) and DB-17 (PCDDs/DFs) capillary columns. The concentrations of dioxins and Co-PCBs were converted to TEQs using WHO-TEFs³.

Results and Discussion

Table 1 shows the TEQ levels (pg/g wet wt) of PCDD/DFs and Co-PCBs in the meat and bacon/blubber products from the toothed whales and dolphins, North Pacific minke whales and southern hemisphere minke whales. Mean total TEQs in products from the toothed whales and dolphins were 2.97 pg/g wet wt (n=6) in meat, and 232 pg/g wet wt (n=13) in bacon/blubber. Mean TEQs in products from North Pacific minke whales were 5.2 pg/g wet wt (n=3) in meat, and 57.4 pg/g wet wt (n=9) in bacon/blubber. Mean TEQs in products from southern hemisphere minke whales were 1.5 pg/g wet wt (n=2) in meat and 4.9 pg/g wet wt (n=5) in bacon/blubber.

Relative contribution of PCDDs, PCDFs and Co-PCBs to the total TEQs in bacon/blubber from toothed whales and dolphins accounted for 0.5%, 18% and 82%, respectively. The most important PCDF congeners were 2,3,4,7,8-PentaCDF and 1,2,3,4,7,8-HexaCDF, which accounted for 34% and 32% of total TEQs for PCDFs, respectively. Among Co-PCBs, the most dominating congeners were 3,3',4,4',5-pentaCB (#126) and 2,3',4,4',5-pentaCB (#118), representing about 60% and 20% of the total TEQs for Co-PCBs, respectively. Mean TEQs in bacon/blubber from North Pacific minke whales were 2.2 pg/g for PCDFs (3.5% contribution) and 55 pg/g for Co-PCBs (96% contribution), which were estimated to be 10-fold higher than those in products from southern hemisphere minke whales.

The data thus indicated that the TEQs for Co-PCBs in most extracts exceeded the TEQs calculated for the PCDDs/DFs. Such results have been documented in some previous studies⁴⁻⁷, but in this study it should be noted that PCDFs in bacon/blubber from toothed whales and dolphins contributed relatively highly to the total toxic potency, compared to those from minke whale products. High accumulation of organochlorine residues including Co-PCBs has also been observed in toothed whales and dolphins from Japanese coastal waters^{4,5}. This would be explained by various aspects of cetacean physiology, including the fact that cetaceans have a low activity of 3-MC-type enzymes and lack PB-type enzymes necessary for rapidly metabolizing PCBs into excretable metabolites^{8,9}. The higher aquatic predators such as cetaceans therefore receive a greater toxic threat from non- and mono-*ortho* Co-PCBs, especially #126 and #118^{6,7}.

This study also confirmed that cetacean products such as bacon/blubber sold in Japan are contaminated to an unacceptable degree for human consumption. Even one meal (about 50 g) of cetacean bacon/blubber per month may exceed the tolerable daily intake (200 pg-TEQ/50kg bw/day set by Ministry of Health and Welfare, Japan). The US FDA advised that

in the case of fish materials from the Great Lakes where the average TEQ levels exceeded 50 pg/g wet wt, serious consideration should be given to limiting the taking of fish from the Great Lakes¹⁰. The present study showed that products exceeded such advisory level amounted to 12 (92%) of 13 blubber products from toothed whales, and to 5 (56%) of 9 blubber products from North Pacific minke whales. According to the Fishery Agency of Japan, annual consumption of whale products for general population in Japan is estimated to be less than 30 g per year, but some individuals may be eating far more than this and, in particular where a tradition of eating cetacean products still exists in local areas, some individuals may consume relatively large amounts. Therefore, consumers should be advised that cetacean products, especially fatty products from North Pacific mammals could cause adverse health effects.

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References

1. Simmonds MP, Hanly K, and Dolman SJ. (1999) Report to IWC, SC/51/E13.
2. Cipriano F, and Palumbi SR. (1999) Report to IWC, SC/51/09.
3. Van den Berg M, Birnbaum L, Bosveld ATC, Brunström B, Cook P, Feely M, Giesy JP, Hanberg A, Hasegawa R, Kennedy SW, Kubiak T, Larsen JC, Van Leeuwen FXR, Liem AKD, Nolt C, Peterson RE, Poellinger L, Safe S, Schrenk D, Tillitt D, Tysklind M, Younes M, Waern F, Zacharewski T. (1998) *Environ. Health Perspect.*, 106, 775-792.
4. Kannan N, Tanabe S, Ono M, and Tatsukawa R. (1989) *Arch. Environ. Contam. Toxicol.* 18, 850-857.
5. Prudente M, Tanabe S, Watanabe M, Subramnian A, Miyazaki N, Suarez P, and Tatsukawa R. (1997) *Mar. Environ. Res.*, 44, 415-427.
6. Kamrin MA, and Ringer RK. (1994) *Toxicol. Environ. Chem.*, 41, 63-84.
7. Kuehl DW, Haebler R, and Potter C. (1994) *Chemosphere*, 28, 1245-1253.
8. Tanabe S, and Tatsukawa R. (1992) in: *Persistent Pollutants in Marine Ecosystem* (Walker CH, and Livingstone DR, Ed), Pergamon Press, Chpt. 7; 161-177.
9. Reijnders PJH. (1996) in : *Organochlorine and heavy metal contamination in cetacean* (Simmonds MP, and Hutchinson JD, Ed), John Wiley & Sons Publishers, Chpt 8; 206-217.
10. Boyer IJ, Kokoski CJ, and Bolger PM. (1991) *J. Toxicol. Environ. Health.*, 33, 93-101.

Table 1. TEQ levels of PCDDs/DFs and coplanar PCBs in cetacean products sold in Japan.

Congener	Toothed whale & dolphin products		North Pacific minke whale products		Southern Hemisphere minke whale products	
	meat n=6	bacon/blubber n=13	meat n=3	bacon/blubber n=9	meat n=2	bacon/blubber n=5
PCDD						
Total PCDDs	0	0-14.6 (1.15)	0	0	0	0
PCDF						
Total PCDFs	0-0.41 (0.08)	1.61-71.8 (42.2)	0.09-0.43 (0.19)	0-12.6 (2.22)	0	0-0.75 (0.18)
Co-PCB						
non-ortho-	0.40-2.46 (1.38)	18.8-452 (118.6)	0.30-6.27 (2.36)	11.2-95.9 (41.0)	0.26-1.70 (0.98)	1.62-6.97 (3.61)
mono-ortho-	0.15-2.35 (1.46)	4.9-148 (70.7)	0.25-7.1 (2.69)	9.7-29.7 (14.2)	0.24-0.79 (0.52)	0.2-2.47 (1.09)
Total Co-PCBs	0.15-4.63 (2.84)	23.7-569 (189)	0.8-13.4* (5.0)	15.6-115 (55.2)	0.5-2.5 (1.5)	2.0-7.7 (4.7)
Total TEQs	0.85-5.04 (2.97)	27.1-691 (232)	0.84-13.8 (5.2)	15.6-127.2 (57.4)	0.5-2.5 (1.5)	2.1-8.4 (4.9)

Values are ranges and averages (in parentheses) in pg-TEQ/g wet weight; Not detected = 0