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Organotin Compounds in Liver Tissue of Harbour Porpoises (*Phocoena phocoena*) and Grey Seals (*Halichoerus grypus*) from the Coastal Waters of England and Wales

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Common (harbour) porpoises (*Phocoena phocoena*) are small cetaceans which are widely distributed throughout the temperate and sub-Arctic waters in the Northern Hemisphere. They are essentially coastal animals, generally bottom-feeding on small schooling fish such as herring or anchovy, although they are also sighted offshore in deep-water areas (Martin, 1990; Northridge *et al.*, 1995). They are under threat in a number of areas around the North Atlantic as a result of the scale of incidental capture in fishing gear (Clausen and Andersen, 1988; Hammond *et al.*, 1995; International Whaling Commission, 1996; NMFS, 1992; Read and Gaskin, 1990; Trippel *et al.*, 1996). Grey seal (*Halichoerus grypus*) populations occur in the Baltic Sea and the NE and NW Atlantic Ocean, with the greater part of the NE Atlantic population being found around the British Isles (Bonner, 1989). Grey seals feed on a wide variety of fish, with some invertebrates including cephalopods, sandeels (*Ammodytidae*) and gadoid fish being of prime importance around the UK (Hammond and Prime, 1990; Hammond *et al.*, 1994a; Hammond *et al.*, 1994b; Prime and Hammond, 1990). The major part of the NE Atlantic population is found around Scotland and NE England (Bonner, 1989), and is currently increasing in numbers (Hiby *et al.*, 1996).

Tributyltin (TBT) compounds have been used extensively as the active component of antifouling paints for ships and marine structures since the 1960s. Their use on small craft <25 m in length has been restricted in many countries since the late 1980s as TBT caused shell thickening and spat failure in oysters, and imposex in gastropods (Alzieu, 1991; Bryan *et al.*, 1987; Waite *et al.*, 1991). At that time the open sea was not considered to be at risk, and the use of TBT has continued on large vessels. However, a recent study in the southern North Sea has demonstrated the presence of imposex in whelks (*Buccinum undatum*) as a result of TBT contamination from shipping (Ten Hallers-Tjabbes *et al.*, 1994), causing its local extinction within the Dutch Wadden Sea (Cadee *et al.*, 1995). The imposition of wider controls on the use of TBT-based antifouling on shipping is currently under discussion within the International Maritime Organisation. Following release to the environment, TBT degrades by dealkylation to form dibutyl- and monobutyltin (DBT and MBT). These compounds are also used industrially as stabilizers for chlorinated polymers, and as catalysts for silicones and polyurethane foams (Evans and Karpel, 1985). Triphenyl tin compounds (as the acetate and hydroxide) are used in Great Britain in the control of potato blight, with 52 t of fentin being applied in 1994 (Garthwaite *et al.*, 1995).

Many studies of organotin contamination have been conducted, but to date few have included determinations in the tissues of marine mammals. As top predators, cetaceans and pinnipeds accumulate high concentrations of lipophilic and persistent organic compounds through their diet, due to a low metabolic capacity for degradation both in the mammals themselves and components of their food-chain (Suzuki *et al.*, 1992; Tanabe and Tatsukawa, 1992). TBT is bioaccumulative, with an octanol–water partition coefficient in the range 10^3–10^4 (Laughlin and Lindén, 1987) and the ability of cetaceans to metabolise TBT is thought to be low as a result of the limited activity of some cytochrome P450 enzymes within these animals (Kannan *et al.*, 1996; Tanabe *et al.*, 1988). This suggests that butyltin compounds can accumulate in marine mammals, and this has been demonstrated in bottlenose dolphins from the Italian coast (Kannan *et al.*, 1996) and the USA (Kannan *et al.*, 1997a), porpoises from the Baltic Sea (Kannan and Falandysz, 1997), and seals, Steller sea lions, and a range of cetacean species from Japan and the north Pacific Ocean (Iwata *et al.*, 1994; Iwata *et al.*, 1995; Kim *et al.*, 1996). No comparable information has yet been presented for the United Kingdom. In this study, concentrations of MBT, DBT and TBT have been determined in the livers of porpoises and grey seals stranded or bycaught around England and Wales, in order to establish the current levels of contamination by these compounds, and to assess the need for further work.

Samples of liver tissue were taken from 29 porpoises and five seals bycaught or stranded around England and Wales from 1992 to 1996 according to previously established protocols (Kuiken and Baker, 1993; Law, 1994), with only those carcases considered freshly dead or slightly decomposed being sampled. The tissues were stored in glass at −20°C prior to analysis. Previous studies have shown no measurable breakdown of TBT to have occurred after 6 months storage at this temperature. After thawing and homogenization, the

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samples were analysed for butyltin compounds using a method established for use in a long-term monitoring programme operated by the Burnham on Crouch laboratory and validated during that study (Waldock *et al.*, 1989; Waldock and Waite, 1994). Briefly, subsamples were extracted by shaking with 0.1% sodium hydroxide and methanol after the addition of a known quantity of an internal (surrogate) standard (tripropyltin chloride) used for quantification. The organotin compounds were then back-extracted into *n*-hexane, and converted to their respective hydrides with sodium borohydride. They were analysed using capillary gas chromatography with flame-photometric detection at 610 nm. The recovery of TBT relative to the internal standard was 99.6% ($n = 13$, s.d. = 5.6%) and the limit of detection was around 10 μg kg^{-1} wet weight. The performance of the method was evaluated by routine analysis of an in-house quality control sample (a wet oyster tissue derived from our own studies) within each sample batch. Over the last 2 years this has been analysed 90 times for DBT (mean value 34 ± 13 μg kg^{-1}) and TBT (mean value 226 ± 110 μg kg^{-1}); MBT is not routinely determined. In a recent European certification exercise for a reference mussel sample (BCR477) our data were: MBT, 2.03 mg kg^{-1}; DBT, 1.35 mg kg^{-1}; TBT, 2.16 mg kg^{-1}; compared with the assigned values of 1.96, 1.58 and 2.14 mg kg^{-1}, respectively. Figure 1 shows the distribution of sampling sites around England and Wales, and gas chromatograms of a standard solution and sample extract are given as Fig. 2.

The results of analyses are given in Table 1, and other data reported in the literature are listed in Table 2. All data are given as micrograms per kilogram on a wet weight basis. Triphenyltin is also detected using our methodology (although with less sensitivity than for butyltins), but none of these samples contained detectable concentrations (limit of detection about 100 μg kg^{-1} wet weight).

Although the use of TBT-based antifouling paints on deep-sea vessels results in some direct inputs offshore, the major inputs (including those from shipping) occur in coastal areas. Industrial inputs of MBT and DBT also occur to coastal areas, and concentrations of these compounds will be supplemented by breakdown of TBT after its release. From the data on TBT concentrations in blubber (Iwata *et al.*, 1994, 1995; Table 2), it is clear that organotin residues can be found at low levels in the blubber of dolphins, porpoises, and a beaked whale from offshore locations in the Pacific Ocean, the Bay of Bengal and the Japan Sea, although they were not detectable in a minke whale from the Antarctic Ocean. In the Southern Hemisphere, minke whales feed mainly upon euphausiids in temperate and polar waters (Martin, 1990), and are apparently not exposed to significant concentrations of organotin compounds in their diet. The highest concentrations of organotins were, however, found in finless porpoises inhabiting the coastal waters and rivers of Japan, believed to feed mainly upon sandlance (*Ammodytes*) at the seabed (Martin, 1990). Comparable concentrations have also been found in the blubber of bottlenose

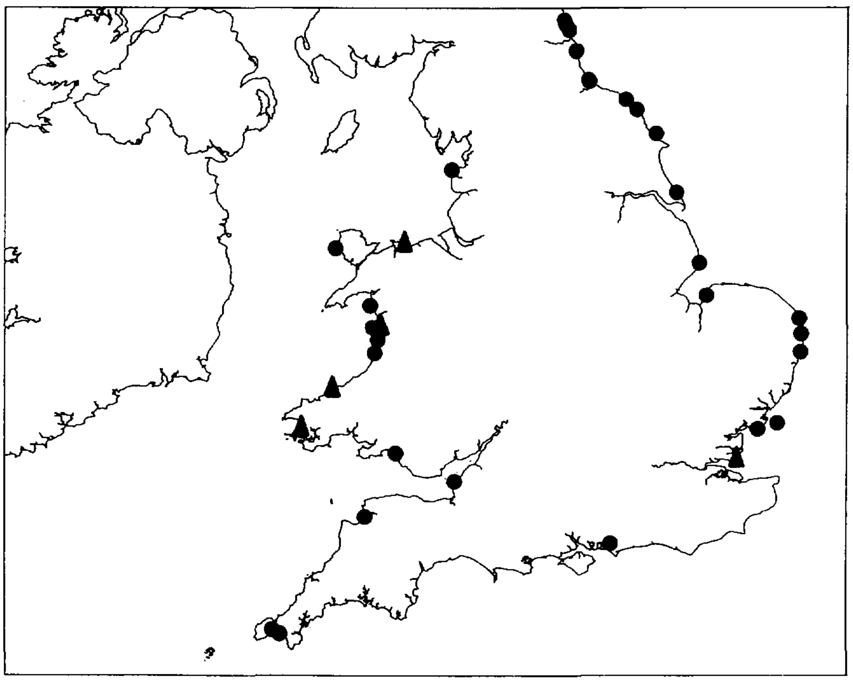

Fig. 1 Map of England and Wales, showing the stranding locations of porpoises (circles) and grey seals (triangles).

dolphins (*Tursiops truncatus*) from the coasts of Italy and the USA (Kannan *et al.*, 1996, 1997a; Table 2), which feed on a wide variety of fish, crustaceans and squid (Evans, 1987).

Organotin compounds have been detected in all the tissues and organs of the finless porpoise (Iwata *et al.*, 1995), with the highest concentrations occurring in the liver. Muscle contained the major burden of TBT (71%), whilst MBT and DBT were retained predominantly in the blubber and liver (41% and 50%, respectively; Table 2 in Iwata *et al.*, 1995). In overall terms, muscle, blubber, and liver retained almost 90% of the total organotin body burden. In Steller sea lions, 40% of the total organotin burden was in the liver, but 26% accumulated in hair, implying that these animals (and presumably also seals) may eliminate a considerable proportion of ingested organotins through the shedding of fur (Kim *et al.*, 1996). Organotin compounds were also found in the brain of finless porpoises (Iwata *et al.*, 1995) and bottlenose dolphins (Kannan *et al.*, 1997a), indicating that these compounds can pass through the blood–brain barrier, possibly by binding to sulphydryl residues as for methylmercury (Kannan and Falandysz, 1997).

Concentrations of total butyltins (ΣBTs) in porpoise liver from the present study (Table 1) were within the range 22–640 μg kg^{-1} wet weight, lower than those reported in finless porpoises from Japan (1120–10 200 μg kg^{-1} wet weight; Table 2) and bottlenose dolphins from the Adriatic Sea (1200 and 2200 μg kg^{-1} wet weight) or the USA (110–11 340 μg kg^{-1} wet weight). On average, only about 20% of the total butyltin tin in these individuals was present as TBT, with DBT the major component. The livers from two neonatal harbour porpoises from the Baltic Sea yielded concentrations at the lower end of the range of the samples analysed here, at 18 and 27 μg kg^{-1} wet weight (Kannan and Falandysz, 1997), and three pygmy sperm whales from the USA exhibited similar ΣBT levels to those in UK porpoises (350–410 μg kg^{-1} wet weight). Concentrations of total butyltins in the liver tissues of grey seals were lower than those seen in the porpoises (Table 1; ND to 22 μg kg^{-1} wet weight), similar to that reported for a Largha seal from Hokkaido, Japan

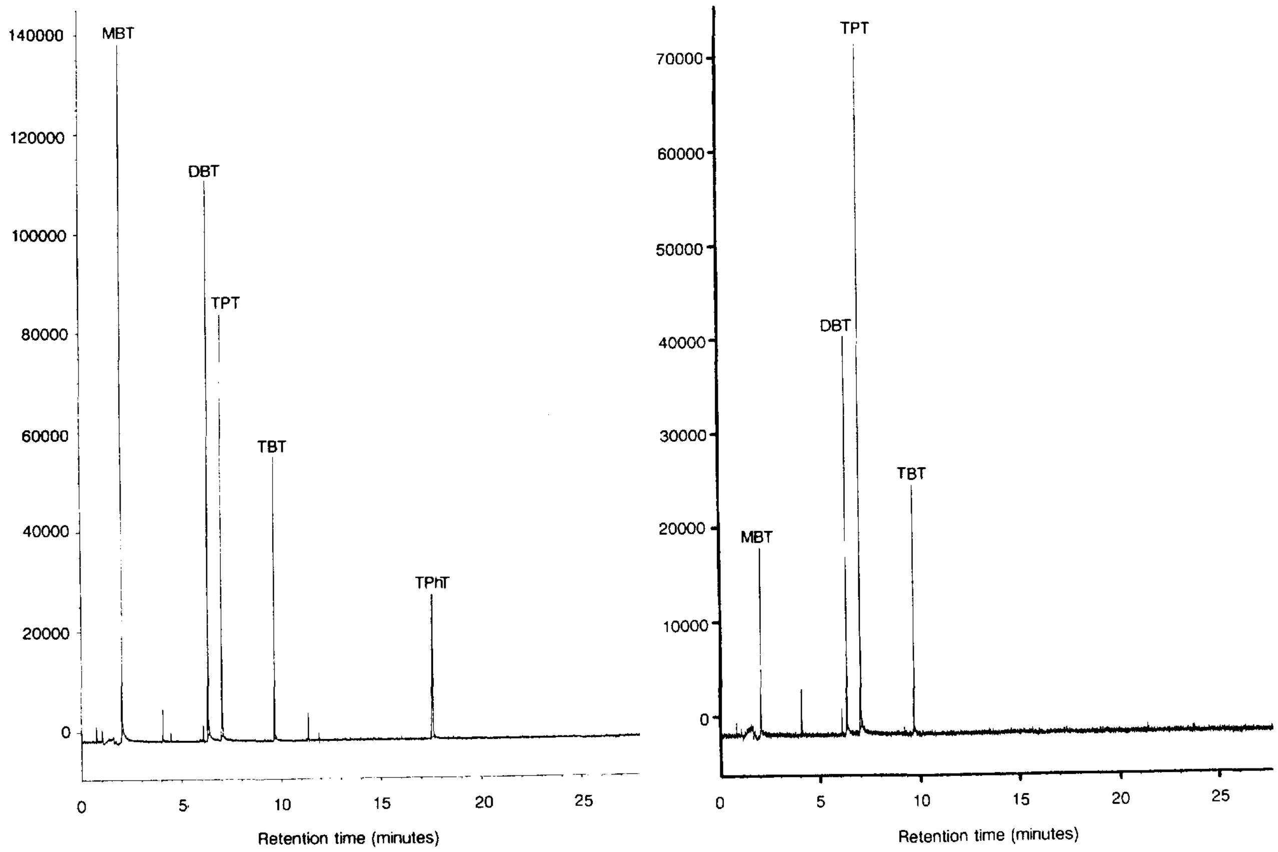

Fig. 2 Gas chromatograms (flame-photometric detection) of (a) a standard solution of organotin compounds, and (b) the extract of liver from a harbour porpoise (SW1992/202; Table 1). The capillary column used was a 25 m × 0.32 mm ID fused silica column coated with 5% phenylmethylsilicone; the column temperature was raised after injection from 40 to 225°C at 15°C min^{-1}. MBT, monobutyltin; DBT, dibutyltin; TPT, tripropyltin; TBT, tributyltin; TPhT, triphenyltin; all as the hydrides.

($4.4\ \mu g\ kg^{-1}$) and lower than concentrations in Steller sea lions from the same area ($170–460\ \mu g\ kg^{-1}$). This suggests that the seals have either a lower intake of organotins from their diet or a higher catabolic capacity for these compounds than sea lions or small cetaceans.

The data reported here demonstrate that low-level organotin contamination in marine mammal predators of upper trophic levels extends to the United Kingdom in addition to those areas previously reported, and tend to suggest that these compounds may be ubiquitous in coastal areas frequented by shipping. However, the significance of these results is harder to assess. The presence of organotins at relatively high concentrations in exposed animals suggests a limited catabolic capacity for such compounds in both the mammals themselves and their prey, although biomagnification seems to be relatively modest (1.8 times in the finless porpoise, Iwata *et al.*, 1995; 0.2 to 7.5-fold in the Ganges river dolphin *Platanista gangetica*, Kannan *et al.*, 1997b; 1.0 to 6.8-fold in the bottlenose dolphin, Kannan *et al.*, 1997a). Preferential enrichment of organotins in the liver may be associated with the presence of metal-binding proteins such as glutathione, and could indicate the operation of a detoxification mechanism (Kannan *et al.*, 1996). The occurrence of DBT as the major component in liver, whereas TBT predominates in blubber, also indicates that some active metabolism does occur in all the marine mammal species studied to date.

The reported effects of butyltins in mammals include immunosuppression (Vos *et al.*, 1984; Snoeij *et al.*, 1989), and it has been suggested that a number of mass mortalities in marine mammal populations in European and North American waters in recent years resulting from morbillivirus infections may have been rendered more severe in animals whose immune function had been impaired by environmental pollutants (Dietz *et al.*, 1989; Heide-Jørgensen *et al.*, 1992; Ross *et al.*, 1995). Interest in this area has mainly centred on organochlorine contaminants (including PCBs, dieldrin and DDT and its metabolites) which are accumulated to high concentrations in the blubber of many marine mammals (Hall *et al.*, 1992), but high

TABLE 1

Concentrations of organotin compounds in livers of porpoises and grey seals ($\mu g\ kg^{-1}$ wet weight).

Ref. no.	Location	Date found	Sex	Age	Length (cm)	TBT	DBT	MBT	ΣBTs
Harbour porpoise									
SW1992/202	Sunderland, Tyne & Wear	13/10/92	F	ND	133	180	350	110	640
SW1993/12	Cambois, Northumberland	27/1/93	M	ND	144	39	150	63	252
SW1994/32	Westward Ho!, Devon	4/3/94	M	2	127	21	99	<10	120
SW1994/45A	Rossall Point, Lancashire	2/4/94	M	8	151	9	25	19	53
SW1994/53	Morfa Dyffryn, Gwynedd	10/4/94	F	3	167	19	46	9	74
SW1994/68	6 miles off Walton pier, Essex	29/4/94	F	ND	112	53	150	50	253
SW1994/80	Port Talbot, Glamorgan	6/5/94	M	4	122	23	82	16	121
SW1994/99	Tywyn, Gwynedd	2/6/94	M	1	120	<10	14	8	22
SW1994/108	Weston-Super-Mare, Avon	25/6/94	M	0	75	<10	13	100	113
SW1994/148	Sandsend, Whitby, N. Yorkshire	1/9/94	M	ND	130	40	160	94	294
SW1994/153	Ynyslas, Dyfed	7/9/94	M	ND	153	21	97	26	144
SW1994/171	Martello Bay, Clacton, Essex	22/11/94	M	ND	146	57	210	<10	267
SW1994/175	Marazion, Cornwall	4/12/94	F	ND	NR	8	71	49	128
SW1994/185	Bosham Harbour, W. Sussex	30/12/94	F	5	145	36	460	130	626
SW1995/52	Seaton sluice, Teeside	20/4/95	M	3	NR	41	140	<9	181
SW1995/55	Seaton Sands, Teeside	1/5/95	M	8	153	47	190	14	251
SW1995/76	Newport, Norfolk	1995[1]	M	0	NR	47	58	<7	105
SW1995/78	Filey Brigg, N. Yorkshire	7/7/95	M	ND	NR	120	440	18	578
SW1995/85	Gorleston, Norfolk	5/7/95	F	0	86	19	71	7	97
SW1995/86	Trearddur Bay, Anglesey, Gwyndedd	7/7/95	F	5	172	46	210	9	265
SW1995/94	St. Mary's Island, Whitley Bay, Tyne & Wear	31/7/95	F	ND	112	33	79	8	120
SW1995/102	Aberystwyth, Dyfed	1/10/95	F	ND	167	14	150	16	180
SW1995/120A	Snettisham, Norfolk	5/10/95	F	ND	107	16	120	<10	136
SW1995/126	Withernsea, E. Yorkshire	13/10/95	F	0	95	64	150	<390	214
SW1996/2	Robin Hood's Bay, N. Yorkshire	3/1/96	M	5	140	41	21	<7	62
SW1996/27(1)	Praa Sands, Cornwall	22/1/96	M	ND	153	39	240	12	291
SW1996/29	Skegness, Lincolnshire	25/1/96	M	> 1	126	42	91	<6	133
SW1996/30	Blyth, Northumberland	26/1/96	M	2	117	32	65	<6	97
SW1996/37	Kessingland, Suffolk	7/2/96	M	ND	137	76	300	15	391
Grey seal									
SS1993/243	Colwyn Bay, Clwyd	25/12/93	F	ND	176	6	5	<10	11
SS1994/63	Poppitt Sands, Cardigan	24/5/94	F	ND	141	<4	<5	<3	nd
SS1994/277	Tywyn, Gwynedd	7/10/94	F	ND	179	<4	6	14	20
SS1995/80	Broad Haven, Pembrokeshire	13/7/95	M	ND	224	<4	<4	11	11
SS1995/143	River Thames	11/9/95	M	ND	226	<4	11	11	22

[1]Precise date not known.
TBT, tributyltin; DBT, dibutyltin; MBT, monobutyltin; ΣBTs, sum of MBT, DBT and TBT.
nd, not detected; ND, not determined; NR, not reported.

concentrations of organotins could have an additive effect (Kannan *et al.*, 1997a). TBT (though not DBT or MBT) also acts as an endocrine disrupter in gastropods, causing an increase in testosterone levels which leads to the development of imposex (Bettin *et al.*, 1996; Spooner *et al.*, 1991), and could interact with the endocrine system in marine mammals. Whilst the levels of organotins in porpoises and seals stranded around England and Wales are lower than some of those reported for small cetaceans from other areas, further study is needed of the possible toxic effects of these compounds and the risk that their accumulation poses to these animals. Data are also needed for other coastal mammals in the United Kingdom, such as the common seal (*Phoca vitulina*) and the bottlenose dolphin.

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TABLE 2

Concentrations of organotin compounds in blubber and liver tissues of marine mammals reported in the literature ($\mu g\ kg^{-1}$ wet weight). Key as for Table 1. Where a range of years is indicated, the date of sampling of individual animals was not given in the original paper.

Species	Location	Year	Sex	Tissue	TBT	DBT	MBT	ΣBTs	Ref.[1]
Dall's porpoise	NE North Pacific	1987	M	Blubber	4.7	<1	<14	4.7	1
Dall's porpoise	NW North Pacific	1987	M	Blubber	6.2	<2	<20	6.2	1
Common dolphin	NW North Pacific	1987	M	Blubber	15	4.3	<20	19	1
Atlantic spotted dolphin	Southeastern USA	1991–94	M	Blubber	80	12	110	200	2
Atlantic spotted dolphin	Southeastern USA	1991–94	M	Liver	76	380	170	630	2
Finless porpoise	Pacific coast of Chiba, Japan	1981	M	Blubber	36	<5	91	130	1
Finless porpoise	Pacific coast of Chiba, Japan	1981	M	Liver	200	790	130	1120	3
Finless porpoise	Seto-inland Sea, Japan	1985	M	Blubber	460	74	210	740	1
Finless porpoise	Seto-inland Sea, Japan	1985	M	Liver	1100	6100	3000	10200	3
Finless porpoise	Western coast of Nagasaki, Japan	1992	M	Blubber	31	<5	64	95	1
Finless porpoise	South China Sea	1990	M	Blubber	6.7	<1	<14	6.7	1
Finless porpoise	Ise Bay, Japan	1994	F	Blubber	120	20	73	213	3
Finless porpoise	Ise Bay, Japan	1994	F	Liver	810	1800	680	3290	3
Harbour porpoise	Baltic Sea, coast of Poland	1991	F	Liver	NR	NR	NR	18[2]	4
Harbour porpoise	Baltic Sea, coast of Poland	1991	F	Liver	NR	NR	NR	27[2]	4
Bottlenose dolphin	NW Adriatic Sea, coast of Italy	1992	M	Blubber	41	16	55	110	5
Bottlenose dolphin	NW Adriatic Sea, coast of Italy	1992	M	Blubber	14	9	25	48	5
Bottlenose dolphin	NW Adriatic Sea, coast of Italy	1992	M	Blubber	210	42	65	320	5
Bottlenose dolphin	NW Adriatic Sea, coast of Italy	1992	M	Liver	250	800	150	1200	5
Bottlenose dolphin	NW Adriatic Sea, coast of Italy	1992	M	Liver	400	1600	200	2200	5
Bottlenose dolphin	Southeastern USA	1989–94	M	Blubber	240	310	78	630	2
Bottlenose dolphin	Southeastern USA	1989–94	M[3]	Liver	5.8–770	450–8300	120–2260	570–11340	2
Bottlenose dolphin	Southeastern USA	1989–94	F[3]	Liver	26–170	290–1570	100–760	420–2500	2
Bottlenose dolphin	Southeastern USA	1989–94	M[4]	Liver	22	230	68	320	2
Bottlenose dolphin	Southeastern USA	1989–94	F[4]	Liver	94–130	67–720	310–360	1120–1160	2
Bottlenose dolphin	Southeastern USA	1989–94	M[5]	Liver	40–50	54–370	44–290	140–710	2
Bottlenose dolphin	Southeastern USA	1989–94	F[5]	Liver	10–110	70–880	32–350	110–1260	2
Killer whale	Pacific coast of Taiji, Japan	1986	M	Blubber	12	3.4	<20	15	1
Spinner dolphin	Bay of Bengal	1990	M	Blubber	2.0	<1	<14	2.0	1
Minke whale	Antarctic Ocean	1985	M	Blubber	<1	<1	<10	nd	1
Gingko-toothed beaked whale	Japan Sea	1993	M	Blubber	33	10	<20	43	1
Pygmy sperm whale	Southeastern USA	1991–94	M	Liver	5–12	240–290	86–160	350–410	2
Largha seal	Western coast of Hokkaido, Japan	1992	M	Blubber	4.4	<1	<14	4.4	1
Steller sea lion	Coast of Hokkaido, Japan	1994	M	Blubber	5.6	5.2	7.1	18	6
Steller sea lion	Coast of Hokkaido, Japan	1994	M	Liver	20	89	63	170	6
Steller sea lion	Coast of Hokkaido, Japan	1995	M	Liver	16	130	100	250	6
Steller sea lion	Coast of Hokkaido, Japan	1995	F	Liver	22	120	52	190	6
Steller sea lion	Coast of Hokkaido, Japan	1995	F	Liver	19	98	67	180	6
Steller sea lion	Coast of Hokkaido, Japan	1995	F	Liver	26	110	100	240	6
Steller sea lion	Coast of Hokkaido, Japan	1995	F	Liver	16	51	110	180	6
Steller sea lion	Coast of Hokkaido, Japan	1995	F	Liver	85	370	<6.6	460	6

[1]1, Iwata *et al.* (1994); 2, Kannan *et al.* (1997a); 3, Iwata *et al.* (1995); 4, Kannan and Falandysz (1997); 5, Kannan *et al.* (1996); 6, Kim *et al.* (1996).
[2]Neonates.
[3]Adults.
[4]Juveniles.
[5]Calves.
Dall's porpoise, *Phocoenoides dalli*; Spinner dolphin, *Stenella longirostris*; Common dolphin, *Delphinus delphis*; Minke whale,*Balaenoptera acutorostrata*; Atlantic spotted dolphin, *Stenella frontalis*; Gingko-toothed beaked whale, *Mesoplodon ginkgodens*; Finless porpoise, *Neophocoena phocaenoides*; Pygmy sperm whale, *Kogia breviceps*; Harbour porpoise, *Phocoena phocoena*; Largha seal, *Phoca largha*; Bottlenose dolphin, *Tursiops truncatus*; Steller sea lion, *Eumetopias jubatus*; Killer whale, *Orcinus orca*.

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