

- Jensen, S., Johnels, A. G., Olsson, M. and Otterlind, G. (1969) DDT and PCB in marine animals from Swedish waters. *Nature* **224**, 247.
- Kannan, K., Tanabe, S., Ramesh, A., Subramanian, A. N. and Tatsukawa, R. (1992a) Persistent organochlorines in foodstuffs from India and their implications on human dietary exposure. *Journal of Agricultural and Food Chemistry* **40**, 518–524.
- Kannan, K., Falandysz, J., Yamashita, N., Tanabe, S. and Tatsukawa, R. (1992b) Temporal trends of persistent organochlorine residues in cod-liver oils from the southern Baltic Proper, 1971–1989. *Marine Pollution Bulletin* **24**, 358–363.
- Martineau, D., Beland, P., Desjardins, C. and Lagace, A. (1987) Levels of organochlorine chemicals in tissues of beluga whales (*Delphinapterus leucas*) from the St. Lawrence Estuary, Quebec, Canada. *Archives of Environmental Contamination and Toxicology* **16**, 137–147.
- Muir, D. C. G., Norstrom, R. J. and Simon, M. (1988) Organochlorine contaminants in Arctic marine food chains: accumulation of specific polychlorinated biphenyls and chlordane-related compounds. *Environmental Science and Technology* **2**, 1071–1079.
- Murphy, P. G. (1972) Sulfuric acid for the cleanup of animal tissues for analysis of acid-stable chlorinated hydrocarbon residues. *Journal of Association of Official Analytical Chemists* **55**, 1360–1362.
- Reijnders, P. J. H. (1986) Reproductive failure in common seals feeding on fish from polluted coastal waters. *Nature* **324**, 456–457.
- Sawhney, B. L. (1986) Chemistry and properties of PCBs in relation to environmental effects. In *PCBs and the Environment*, ed. J. S. Waid, vol. 1, pp. 48–61. CRC Press, Boca Raton, Florida.
- Swart, R. L., Ross, P. S., Vedder, L. J., Timmermann, H. H., Heisterkamp, S., Loveren, H. V., Vos, J. G., Reijnders, P. J. H. and Osterhaus, A. D. M. E. (1994) Impairment of immune function in harbour seals (*Phoca vitulina*) feeding on fish from polluted waters. *Ambio* **23**, 155–159.
- Tanabe, S., Tatsukawa, R., Tanaka, H., Maruyama, K., Miyazaki, N. and Fujiyama, T. (1981) Distribution and total burdens of chlorinated hydrocarbons in bodies of striped dolphin (*Stenella coeruleoalba*) *Agricultural and Biological Chemistry* **45**, 2569–2578.
- Wagemann, R. and Muir, D. C. G. (1984) Concentrations of heavy metals and organochlorines in marine mammals of northern waters: overview and evaluation. *Canadian Technical Report of Fisheries and Aquatic Sciences* **1272**, 97.



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High Levels of PCB and *p,p'*-DDE Found in the Blubber of Killer Whales (*Orcinus orca*)

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Organochlorine pollutants in the marine environment represent a threat to marine organisms in general, but especially to marine mammals (Tanabe *et al.*, 1994). The homogeneity of the ocean environment, coupled with the potential of long-range transport, allows for widespread effects from sources that are often small and localized (Tatton and Ruzicka, 1967; Loganathan and Kannan, 1991; Norstrom and Muir, 1994). Because they are usually highly persistent as well as lipophilic, organochlorines tend to accumulate in the fat deposits of animals, especially those with poor metabolic and excretory capabilities for these pollutants, such as marine mammals (Brooks, 1974; Hutzinger *et al.*, 1974; Tanabe *et al.*, 1988). In addition, the greatest accumulations will be found in those animals at the top of the food chain, which is also the case for many species of marine mammals (Tanabe *et al.*, 1984; Loganathan and Kannan, 1994).

The most prevalent of the organochlorines currently identified in marine mammals are the polychlorinated biphenyls (PCBs) and *p,p'*-DDE, the major metabolite of insecticide DDT (Calambokidis and Francis, 1994; O'Shea and Brownell, 1994; Jarman *et al.*, 1996). Although its use is banned in most countries, DDT is still utilized in some areas of the world (Goldberg, 1991). Production of PCBs has declined worldwide, but entry of substantial amounts into the marine environment is probably still occurring (Tanabe, 1988; Peterle, 1991).

The killer whale (*Orcinus orca*) represents the top of many marine food webs. This species, which feeds primarily on fish and/or other marine mammals, is found in many of the world's oceans in either localized resident populations, or in highly migratory groups (Matkin and Leatherwood, 1986). As with other cetaceans, these animals possess thick blubber layers and accumulate PCBs and *p,p'*-DDE in these fat deposits (Calambokidis *et al.*, 1984; Ono *et al.*, 1987; Kemper *et al.*, 1994; Jarman *et al.*, 1996). Although killer whales have been harvested and utilized by humans in the past, they are now rarely killed intentionally (Kirkevold, 1986; Oien, 1988). Blubber residue determination in wild specimens has, therefore, depended largely on dead stranded animals. We report here the residue levels of total PCBs and *p,p'*-DDE for five killer whales stranded on the Oregon coast from 1988 to 1997. These levels were determined by high performance liquid chromatography (HPLC) using the method of Hayteas and Duffield (1998). Contaminant levels are given in Table 1, and all data are quoted here as $\mu\text{g g}^{-1}$ wet weight.

All but one of the stranded animals were carrying high levels of PCBs and/or *p,p'*-DDE. The adult female (length, 622 cm) exhibited concentrations of $276 \mu\text{g g}^{-1}$ of PCBs and $494 \mu\text{g g}^{-1}$ of *p,p'*-DDE. In the juvenile male (length, 385 cm), the levels were also quite high, $415 \mu\text{g g}^{-1}$ of PCBs and $368 \mu\text{g g}^{-1}$ of *p,p'*-DDE. In one of the calves (length, 212 cm), the level of *p,p'*-DDE was

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TABLE 1

Levels of PCBs and *p,p'*-DDE ($\mu\text{g g}^{-1}$ wet weight) found in the blubber of killer whales (*Orcinus orca*) stranded on the Oregon coast.

Date	Location	Length (cm)	Sex	PCBs	<i>p,p'</i> -DDE
2/08/88	Central OR	385	M	415	368
7/24/93	Central OR	235	M	19.5	151
5/13/95	Central OR	212	F	117	519
4/13/96	North OR	622	F	276	494
4/25/97	North OR	256	M	3.9	4.4

519 $\mu\text{g g}^{-1}$, the highest seen in this study. In this same calf, the level of PCBs was somewhat lower, 117 $\mu\text{g g}^{-1}$. An additional calf (length, 235 cm) exhibited a relatively high concentration of *p,p'*-DDE, 151 $\mu\text{g g}^{-1}$, but a much lower level of PCBs, 19.5 $\mu\text{g g}^{-1}$. In contrast, the calf stranded in 1997 (length, 256 cm) carried very low levels of both pollutants, 3.9 $\mu\text{g g}^{-1}$ of PCBs and 4.4 $\mu\text{g g}^{-1}$ of *p,p'*-DDE.

Comparison of these results with those previously reported for killer whales reveals that all but one of the stranded Oregon animals exhibited PCB and DDE levels at the high to extremely high end of the reported range for both pollutants. Kemper *et al.* (1994) reported a concentration of 28.4 $\mu\text{g g}^{-1}$ of total DDT for a killer whale stranded in Australia, while Calambokidis *et al.* (1984) found levels of 38 $\mu\text{g g}^{-1}$ of PCBs and 59 $\mu\text{g g}^{-1}$ of DDE in an animal stranded in Puget Sound. The levels reported by Jarman *et al.* (1996) for killer whales stranded in British Columbia from 1986 to 1989 were relatively low, 22 $\mu\text{g g}^{-1}$ (geometric mean) for total PCBs and 28 $\mu\text{g g}^{-1}$ (geometric mean) for *p,p'*-DDE. At the other end of the range, Ono *et al.* (1987) reported PCB concentrations of 350–410 $\mu\text{g g}^{-1}$ in three adult killer whales caught off the coast of Japan, while Calambokidis *et al.* (1984) reported levels of 250 $\mu\text{g g}^{-1}$ PCBs and 640 $\mu\text{g g}^{-1}$ DDE in a male killer whale stranded in British Columbia in 1979.

Occurrence of the high levels found in most of the stranded Oregon killer whales may be due to different factors. One possibility is that these whales were members of migratory populations ranging along the west coast of North America, distinct from resident populations that have a more localized range (Heimlich-Boran, 1986; Matkin and Leatherwood, 1986). With an extensive range, migrant killer whales would feed in a variety of areas. Some of these areas may have experienced high contaminant inputs in the past, such as the waters of southern California, which were heavily impacted by both DDT and PCB pollution (MacGregor, 1974; Calambokidis and Francis, 1994), compared with other feeding areas that seem to have experienced lower contamination, such as waters off the Oregon coast (Hayteas and Duffield, 1997). Three of the Oregon-stranded killer whales may represent individuals from groups with feeding ranges that are concentrated off southern California, as indicated by the higher levels of *p,p'*-DDE relative to PCBs (Calambokidis and Barlow, 1991). A fourth exhibited elevated PCB concentrations relative to

p,p'-DDE, perhaps indicating a different primary feeding area (Calambokidis and Barlow, 1991).

Another factor contributing to these observed high levels of organochlorines may relate to the observation that transient (migratory) killer whales feed primarily on other marine mammals, in contrast to resident individuals which feed mostly on fish (Bigg *et al.*, 1987; Baird *et al.*, 1992; Barrett-Lennard *et al.*, 1996). Transient whales are, therefore, one trophic level higher on the food chain, leading to the potential for greater organochlorine accumulation through biomagnification. It has been determined that transient killer whales do strand on the Oregon coast (Stevens *et al.*, 1989). This would be consistent with the high levels of pollutants found in the killer whales reported here, as opposed to the much lower levels previously reported for killer whales presumably stranded from resident populations in British Columbia (Jarman *et al.*, 1996). The one killer whale in our study with low levels of contaminants was a young calf (with fresh umbilical scarring). Such low levels might indicate that this animal was not a first-born, but the offspring of a female who had already passed a large part of her organochlorine load to previous calves (Addison and Smith, 1974; Addison and Brodie, 1977; Donkin *et al.*, 1981; Gaskin, 1982; Wagemann and Muir, 1984; Agiular and Borrell, 1988; Subramanian *et al.*, 1989). On the other hand, resident populations of killer whales may occur in Oregon and this calf may have been a member of such a population.

The high levels of PCBs and *p,p'*-DDE found in killer whales stranded on the Oregon coast is definitely cause for concern. It has been hoped that areas which had been heavily contaminated in the past were becoming less so (Lieberg-Clark *et al.*, 1995). Even though this may be the case, there is evidence that organochlorine compounds are being concentrated in remote latitudes, such as polar environments, due to the processes of global fractionation and distillation, associated with long-range air transport (Oehme, 1991; Wania and Mackay, 1993; Tanabe *et al.*, 1994). The occurrence of high levels of organochlorine contaminants in killer whales may reflect the accumulated effects of contaminants moving through latitudinally related food chains on a wide-ranging, higher trophic level species. The presence of these substances has been linked to many health problems in marine mammals, including dysfunctions in the reproductive and immune systems (Reijnders, 1986, 1994; Beland *et al.*, 1992; de Swart *et al.*, 1994). It is essential that these animals continue to be monitored for the presence of organochlorines, both as indicators of their health and that of the marine environment.

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- Addison, R. F. and Brodie, P. F. (1977) Organochlorine residues in maternal blubber, milk, and pup blubber from grey seals (*Halichoerus grypus*) from Sable Island, Nova Scotia. *Journal of the Fisheries Board of Canada* **34**, 937–941.
- Addison, R. F. and Smith, T. G. (1974) Organochlorine residue levels in Arctic ringed seals: variation with age and sex. *Oikos* **25**, 335–337.
- Aguilar, A. and Borrell, A. (1988) Age- and sex-related changes in organochlorine compound levels in fin whales (*Balaenoptera physalus*) from the eastern North Atlantic. *Marine Environmental Research* **25**, 195–211.
- Baird, R. W., Abrams, P. A. and Dill, L. M. (1992) Possible indirect interactions between transient and resident killer whales – implications for the evolution of foraging specializations in the genus *Orcinus*. *Oecologia* **89**, 125–132.
- Barret-Lennard, L. G., Ford, J. K. B. and Heise, K. A. (1996) The mixed blessing of echolocation: differences in sonar use by fish-eating and mammal-eating killer whales. *Animal Behaviour* **51**, 553–565.
- Beland, P., De Guise, S. and Plante, R. (1992) *Toxicology and Pathology of St. Lawrence Marine Mammals, Project Report*. World Wildlife Fund, Washington, DC.
- Bigg, M. A., Ellis, G. M., Ford, J. K. B. and Balcomb III, K. C. (1987) *Killer Whales: A Study of their Identification, Genealogy, and Natural History in British Columbia and Washington State*. Phantom Press, Nanaimo, British Columbia.
- Brooks, G. T. (1974) *Chlorinated Insecticides: Technology and Application*. CRC Press, Cleveland.
- Calambokidis, J. and Barlow, J. (1991) Chlorinated hydrocarbon concentrations and their use for describing population discreteness in harbor porpoises from Washington, Oregon and California. In *Marine Mammal Strandings in the United States: Proceedings of the Second Marine Mammal Stranding Workshop*. NOAA Technical Report National Marine Fisheries Service 98, eds. J. E. Reynolds III and D. K. Odell, pp. 101–110. National Technical Information Service, Rockville, Maryland.
- Calambokidis, J. and Francis, J. (1994) *Marine Mammal Exposure to PCB and DDT Contamination in the Southern California Bight*. Cascadia Research, Olympia, Washington.
- Calambokidis, J., Peard, J., Steiger, G., Cabbage, J. and DeLong, R.L. (1984) *Chemical Contaminants in Marine Mammals from Washington State*. NOAA Technical Memorandum NOS OMS6. National Technical Information Service, Rockville, Maryland.
- de Swart, R., Ross, P. S., Vedder, L. J., Timmerman, H. H., Heisterkamp, S., Van Loveren, H., Vos, J. G., Reijnders P. J. H. and Osterhaus, A. D. M. E. (1994) Impairment of immune function in harbour seals (*Phoca vitulina*) feeding on fish from polluted waters. *Ambio* **23**, 155–159.
- Donkin, P., Mann, S. V. and Hamilton, E. I. (1981) Polychlorinated biphenyl, DDT and dieldrin residues in grey seals (*Halichoerus grypus*) males, females and mother-foetus pairs sampled at the Farne Islands, England, during the breeding season. *The Science of the Total Environment* **19**, 121–142.
- Gaskin, D. E. (1982) *The Ecology of Whales and Dolphins*. Heinemann, London.
- Goldberg, E. D. (1991) Halogenated hydrocarbons: past, present and near-future problems. *The Science of the Total Environment* **100**, 17–28.
- Hayteas, D. L. and Duffield, D. A. (1997) The determination by HPLC of PCB and *p,p'*-DDE residues in marine mammals stranded on the Oregon coast, 1991–1995. *Marine Pollution Bulletin* **34**, 844–848.
- Hayteas, D. L. and Duffield, D. A. (1998) Use of high-performance liquid chromatography for the estimation of polychlorinated biphenyls and *p,p'*-DDE residues in marine mammals. *Journal of Chromatography B* **705**, 362–366.
- Heimlich-Boran, S. L. (1986) Cohesive relationships among Puget Sound killer whales. In *Behavioral Biology of Killer Whales* eds. B. C. Kirkevoid and J. S. Lockard, pp. 251–284. Alan R. Liss, New York.
- Hutzinger, O., Safe, S. and Zitko, V. (1974) *The Chemistry of PCBs*. CRC Press, Cleveland, Ohio.
- Jarman, W. M., Norstrom, R. J., Muir, D. C. G., Rosenberg, B., Simon, M. and Baird, R. W. (1996) Levels of organochlorine compounds, including PCDDs and PCDFs, in the blubber of cetaceans from the west coast of North America. *Marine Pollution Bulletin* **32**, 426–436.
- Kemper, C., Gibbs, P., Obendorf, D., Marvanek, S. and Lenghaus, C. (1994) A review of heavy metal and organochlorine levels in marine mammals in Australia. *The Science of the Total Environment* **154**, 129–139.
- Kirkevoid, B. C. (1986) Introduction and management issues of wild and captive killer whales. In *Behavioral Biology of Killer Whales*, eds. B. C. Kirkevoid and J. S. Lockard, pp. 3–16. Alan R. Liss, New York.
- Lieberg-Clark, P., Bacon, C. E., Burns, S. A., Jarman, W. M. and LeBoeuf, B. J. (1995) DDT in California sea-lions: a follow-up study after twenty years. *Marine Pollution Bulletin* **30**, 744–745.
- Loganathan, B.G. and Kannan, K. (1991) Time perspectives of organochlorine contamination in the global environment. *Marine Pollution Bulletin* **22**, 582–584.
- Loganathan, B.G. and Kannan, K. (1994) Global organochlorine contamination trends: an overview. *Ambio* **23**, 187–191.
- MacGregor, J. S. (1974) Changes in the amount and proportions of DDT and its metabolites, DDE and DDD, in the marine environment off southern California. *Fisheries Bulletin* **72**, 275–293.
- Matkin, C.O. and Leatherwood, S. (1986) General biology of the killer whale, *Orcinus orca*: a synopsis of knowledge. In *Behavioral Biology of Killer Whales*, eds. B. C. Kirkevoid, and J. S. Lockard, pp. 35–68. Alan R. Liss, New York.
- Norstrom, R. J. and Muir, D. C. G. (1994) Chlorinated hydrocarbon contaminants in arctic marine mammals. *The Science of the Total Environment* **154**, 107–128.
- Oehme, M. (1991). Further evidence for long-range air transport of polychlorinated aromatics and pesticides: North America and Eurasia to the Arctic. *Ambio* **20**, 293–297.
- Oien, N. (1988). The distribution of killer whales (*Orcinus orca*) in the North Atlantic based on Norwegian catches, 1938–1981, and incidental sightings, 1967–1987. *Rit Fiskideildar* **11**, 65–78.
- Ono, M., Kannan, N., Wakimoto, T., Tatsukawa, R. (1987) Dibenzofurans a greater global pollutant than dioxans? Evidence from analyses of open ocean killer whale. *Marine Pollution Bulletin* **18**, 640–643.
- O'Shea, T. J. and Brownell Jr., R. L. (1994) Organochlorine and metal contaminants in baleen whales: a review and evaluation of conservation implication. *The Science of the Total Environment* **154**, 179–200.
- Peterle, T. J. (1991) *Wildlife Toxicology*. Van Nostrand Reinhold, New York.
- Reijnders, P. J. H. (1986) Reproductive failure in common seals feeding on fish from polluted coastal waters. *Nature* **324**, 456–457.
- Reijnders, P. J. H. (1994) Toxicokinetics of chlorobiphenyls and associated physiological responses in marine mammals, with particular reference to their potential for ecotoxicological risk assessment. *The Science of the Total Environment* **154**, 229–236.
- Stevens, T. A., Duffield, D. A., Asper, E. D., Hewlett, K. G., Bolz, A., Gage, L. J. and Bossart, G. D. (1989) Preliminary findings of restriction fragment differences in mitochondrial DNA among killer whales (*Orcinus orca*). *Canadian Journal of Zoology* **67**, 2592–2595.
- Subramanian, A. N., Tanabe, S. and Tatsukawa, R. (1998) Use of organochlorines as chemical tracers in determining some reproductive parameters in *Dalli*-type Dall's porpoise (*Phocoenoides dalli*). *Marine Environmental Research* **25**, 161–174.
- Tanabe, S. (1988) PCB problems in the future: foresight from current knowledge. *Environmental Pollution* **50**, 5–28.
- Tanabe, S., Tanaka, H. and Tatsukawa, R. (1984) Polychlorobiphenyls, tDDT, and hexachlorocyclohexane isomers in the western North Pacific ecosystem. *Archives of Environmental Contamination and Toxicology* **13**, 731–738.
- Tanabe, S., Watanabe, S., Kan, H. and Tatsukawa, R. (1988) Capacity and mode of PCB metabolism in small cetaceans. *Marine Mammal Science* **4**, 103–124.
- Tanabe, S., Iwata, H. and Tatsukawa, R. (1994) Global contamination by persistent organochlorines and their ecotoxicological impact on marine mammals. *The Science of the Total Environment* **154**, 163–177.
- Tatton, J. O'G. and Ruzicka, J. H. A. (1967) Organochlorine pesticides in Antarctica. *Nature* **215**, 346–348.

Wagemann, R. and Muir, D. C. G. (1984) Concentrations of heavy metals and organochlorines in marine mammals of northern waters: overview and evaluation. *Canadian Technical Report of Fisheries and Aquatic Sciences* **1279**.

Wania, F. and Mackay, D. (1993) Global fractionation and cold condensation of low volatility organochlorine compounds in polar regions. *Ambio* **22**, 10–18.
