

REVIEW

Organochlorine dynamics between zooplankton and their environment, a reassessment

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ABSTRACT: The level of organochlorine contamination in estuarine, coastal or oceanic waters and various depth strata, including surface films, is important in predicting bioaccumulation in zooplankton. Present and past levels of contamination in seawater are briefly reviewed and summarized. Most of the evidence indicates that little organochlorine is truly dissolved in seawater: most molecules are sorbed onto particulate material or sequestered into micellar structures. A first-order kinetic model is adequate to describe the accumulation of organochlorines by zooplankton from seawater because current laboratory techniques are not refined enough to distinguish pathways within the organism. The chemical nature of the compounds, such as chlorine content, influences the time taken to reach an equilibrium level in an organism and the bioaccumulation at equilibrium. Size of organism, and temperature and salinity of the environment affect organochlorine uptake rates; this is believed to be related to the weight-specific surface area of the zooplankter exposed per unit time. The lipid content influences the carrying capacity of the zooplankter or its bioconcentration factor from seawater. The same kinetic model can be expanded to include organochlorines assimilated through feeding on contaminated food. Experimental studies have shown that 'field' levels of contamination in zooplankton can be reached in the lab within days by accumulation from food. The transfer of organochlorines from generation to generation in the lipid-rich eggs of zooplankters is discussed and explored with the kinetic model. Juvenile stages are predicted to contain the highest organochlorine levels after yolk absorption and therefore should experience the greatest mortality. Finally, it is concluded, from consideration of present levels of contamination, that the water column is the present and probably ultimate repository of most organochlorine compounds in the marine environment and that zooplankton play a major role in distributing organochlorines from atmospheric 'fallout' throughout the ocean depths.

INTRODUCTION

The final repository for refractory anthropogenic compounds such as organochlorines has been hypothesized to be the marine sediments, in particular the abyssal plains (Woodwell et al. 1971); however recent studies suggest a more dynamic situation occurs, at least in shallow waters, with a continuing flux between sediments and seawater (Larsson 1985a, b, Baker et al. 1985). It has become obvious that although zooplankton contain a small fraction of the global chlorinated hydrocarbons in their bodies they may be responsible for important mass transfers throughout the ocean abyss (Burns et al. 1985). This

review evaluates what is known about the role of plankton in the dynamics of organochlorines in the ocean in an attempt to interpret their importance on a global scale.

Several groups of anthropogenic organochlorine compounds have been detected and reported frequently in the marine environment. These are DDT, cyclodiene, and HCH insecticides, industrial compounds such as polychlorinated biphenyls (PCBs) and hexachlorobenzene (HCB), and pentachlorophenol (PCP) both of which are produced as fungicides but may also be industrial by-products (Table 1). Chlorinated compounds such as polychlorinated terphenyls (PCTs), chlorobiphenyl derivatives, chlorinated paraf-

Table 1. Organochlorine compounds detected in the marine environment

| | |
|--------------------|---|
| <i>p,p'</i> -DDT | 2,2-bis-(<i>p</i> -chlorophenyl)-1,1,1-trichloroethane |
| <i>o,p'</i> -DDT | 2-(<i>o</i> -chlorophenyl)-2-(<i>p</i> -chlorophenyl)-1,1,1-trichloroethane |
| <i>p,p'</i> -DDE | 2,2-bis-(<i>p</i> -chlorophenyl)-1,1-dichloroethylene |
| <i>p,p'</i> -DDD | 2,2-bis-(<i>p</i> -chlorophenyl)-1,1-dichloroethane |
| Chlordane | Cis- and trans-isomers of 1,2,4,5,6,7,8,8-octachloro-3a,4,7,7a-tetrahydro-4,7-methanoindane |
| Dieldrin | 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8a-octahydro-1,4-endo-exo-5,8-dimethanonaphthalene |
| Endrin | 1,2,3,4,10,10-hexachloro-6,7-epoxy-1,4,4a,5,6,7,8,8a-octahydro-1,4-endo-endo-5,8-dimethanonaphthalene |
| Aldrin | 1,2,3,4,10,10-hexachloro-1,4,4a,5,8,8a-hexahydro-1,4-endo-exo-5,8-dimethanonaphthalene |
| Toxaphene | Chlorinated camphene (mixture, components resemble aldrin) |
| Heptachlor | 1,4,5,6,7,8,8-heptachloro-3a,4,7,7a-tetrahydro-4,7-methanoindene |
| Heptachlor epoxide | 1,4,5,6,7,8,8-heptachloro-2,3-epoxy-2,3,3a,4,7,7a-hexahydro-4,7-methanoindene |
| HCH | α -, β -, γ -, δ -isomers of 1,2,3,4,5,6-hexachlorocyclohexane |
| Lindane | γ -hexachlorocyclohexane |
| PCBs | Isomers of polychlorinated biphenyls (mixture) |

bins and many others have been produced for industrial use (Zitko 1976, Addison 1983), but have not yet been reported in the water, phytoplankton or zooplankton of the marine environment. The explanation may be that these substances, except chlorinated paraffins, have been recently introduced by industry and are expensive and time-consuming to assay.

All groups of organochlorines share common physical and chemical properties such as relative chemical stability, unreactivity, appreciable volatility at environmental temperatures (Mackay & Wolkoff 1973, Mackay & Leinonen 1975) and low solubility in seawater (0.1 to 2000 $\mu\text{g l}^{-1}$; Dexter & Pavlou 1978, Mackay et al. 1980) but with high lipid-water partition coefficients (Chiou et al. 1977, Mackay 1982). These properties account for their known environmental persistence, ubiquitous distribution throughout the world and accumulation in fat reserves of marine organisms.

Copepods can accumulate organochlorines directly by sorption from seawater and from particulate matter in the seawater or by feeding on contaminated prey. It is therefore desirable to first review the types and concentrations of organochlorines found in seawater and phytoplankton before considering their dynamics in zooplankton. At the same time we look for evidence of environmental gradients and temporal changes in organochlorine contamination.

COASTAL AND NEAR-SHORE SURFACE WATERS

The documented levels of organochlorines in seawater reported here were determined from bulk samples which included both living and inert particles. The implications of this crude measure of contamination in seawater are discussed more fully later when attempting to interpret marine organochlorine

dynamics. In the DDT family, *p,p'*-DDT, *p,p'*-DDE and *p,p'*-DDD are the most frequently reported forms in coastal surface waters with concentrations generally below 5 ng l^{-1} (pptr) (Table 2). Exceptions are the coastal waters off Marseille, France, in 1971 (Raybaud 1972) and Blanca Bay, Argentina, in 1980–81 (Sericano & Pucci 1984) where levels averaged more than 40 ng l^{-1} . The lowest values, 0.008 ng l^{-1} , were reported off Syowa Station, Antarctica. ΣDDT levels in the northern hemisphere do not appear to have changed over the period for which measurements are available (1970–1980), even though DDT usage declined in the 60's and was totally banned by the mid-70's in industrialized countries (Brooks 1977).

Dieldrin is the only cyclodiene pesticide that has been quantified at several locations; with concentrations of $13.8 \pm 12.4 \text{ ng l}^{-1}$ in Biscayne Bay, Florida (USA), 1968 (Seba & Corcoran 1969), 0.3 ± 0.1 in the German Bight, 1974 (Stadler & Ziebarth 1975), 0.2 ± 0.1 in the western Baltic Sea, 1974 (Stadler & Ziebarth 1976), 0.02 ± 0.06 around the British Isles, 1974 (Dawson & Riley 1977) and 0.05 ± 0.04 in Corpus Christi Bay, Texas (USA), 1980 (Ray et al. 1983a). Aldrin has been reported from Biscayne Bay, Florida, $17.8 \pm 9.4 \text{ ng l}^{-1}$, in 1968 (Seba & Corcoran 1969) and Blanca Bay, Argentina in 1980–81, $3.7 \pm 4.1 \text{ ng l}^{-1}$ (Sericano & Pucci 1984). Heptachlor has been reported only from Blanca Bay, Argentina, in 1980–81 at 40.8 ± 46.4 and chlordane only from Corpus Christi Bay, Texas, in 1980 at $0.18 \pm 0.31 \text{ ng l}^{-1}$ (Ray et al. 1983a). Toxaphene was quantified in amounts up to 10 ng l^{-1} in Mediterranean surface waters between 1981 and 1982 (Burns et al. 1985). The α - and γ -isomers are the more frequently encountered of the HCH insecticides with concentrations ranging between 1 and 10 ng l^{-1} in seawater (Table 3). As in the DDT group, highest values were measured near Marseille, France, in 1971 and Blanca Bay, Argentina, in 1980–81 (Table 3).

Polychlorinated biphenyls have been quantified in several inshore locations with most values lying between 1 and 10 ng l⁻¹ seawater (Table 4). High values of >100 ng l⁻¹ were measured in 1971 near Marseille, France (Raybaud 1972) and Narragansett Bay, Rhode Island (USA) (Duce et al. 1972). Lowest values, 0.05 ± 0.01 ng l⁻¹, were measured near Syowa Station,

Antarctica in 1981–82 (Tanabe et al. 1983). There was no trend in PCB levels in inshore waters between 1971 and 1980 (Table 4) in spite of greatly reduced usage during the 1970's (Addison 1983). Burns et al. (1985), however, have measured a 3-fold decline in PCB levels in Mediterranean seawater off Monaco between the mid-1970s and 1978–1982.

Table 2. DDT and metabolites in coastal and near-shore surface waters (ng l⁻¹)

| Year | Region | N | <i>p,p'</i> -DDT | <i>p,p'</i> -DDE | <i>p,p'</i> -DDD | ΣDDT | Source |
|--------------------------------|---|---------------------------|---|----------------------------|--------------------------|----------------------------------|---------------------------|
| 1969 | San Francisco Bay, California, USA | 24 | 5 ± 5 ^a (1–18) | 2 ± 1 (1–4) | 2 ± 2 (1–7) | 8 ± 5 (3–23) | Earnest & Benville (1971) |
| 1970 | Monterey Bay to San Diego, Calif., USA | 4 | + ^b | + | + | 2.4 ± 0.2 (2.4–2.7) | Cox (1971a) |
| | Vancouver, B.C., Canada to San Francisco Bay, USA | 4 | + | + | + | 4.0 ± 1.1 (3.0–5.6) | Cox (1971a) |
| 1971 | Hawaii Canal, Hawaii, USA | 6 | 9.0 (0.4–41.0) | 0.3 (0.1–0.6) | 3.6 (0.1–10.0) | | Bevenue et al. (1972) |
| 1971 | Near Marseille, France Mediterranean Sea | ? | (70–180) | (30–120) | | | Raybaud (1972) |
| 1972 | Off North Carolina, USA | 4 | nd ^c (<8.0) | 7.9 ± 6.9 (3.5–18.1) | nd (<0.8) | | Jonas & Pfaender (1976) |
| 1973 | California Coast, USA | ? | | | | (0.9–17.5) | Pavlou et al. (1974) |
| 1973 | Southern California Bight | 14 | 0.6 ± 0.3 (0.3–1.3) | 0.10 ± 0.03 (0.06–0.13) | | 0.7 ± 0.3 (0.34–1.43) | Risebrough et al. (1976) |
| 1974 | Off Monaco Mediterranean Sea | 3 | | | | 0.04 ± 0.01 (0.04–0.05) | Elder (1974) |
| 1974 | California Coast, USA | 7 | 0.5 ± 0.6 (<0.1–1.9) | 1.0 ± 1.6 (<0.1–4.6) | | | Scura & McClure (1975) |
| 1974 | Around British Isles | 31 | + | + | + | 0.05 ± 0.07 (<0.01–0.25) | Dawson & Riley (1977) |
| 1974 | German Bight, North Sea | 24 | 0.3 ± 0.2 (0.1–0.6) | ? | 1.5 ± 0.6 (0.5–2.6) | | Stadler & Ziebarth (1975) |
| 1974 | Western Baltic Sea | 21 | 0.2 ± 0.1 (0.1–0.3) | ? | 2.4 ± 0.6 (0.8–3.4) | | Stadler & Ziebarth (1976) |
| 1974–1975 | Santos Estuary, Brazil | 64 | | | | (nd–40) | Tommasi (1985) |
| 1975 | Hanö Bight, North Sea | 8 | 0.04 ± 0.08 (nd–0.21) | 1.7 ± 1.6 (0.8–5.2) | | | Osterroht (1977) |
| 1975 | German Bight, North Sea | 17 | 0.33 ± 0.15 (0.11–0.63) | 0.11 ± 0.06 (nd–0.21) | 0.20 ± 0.15 (nd–0.57) | | Stadler (1977) |
| 1975 | Western Baltic Sea | 18 | 0.2 ± 0.1 (nd–0.4) | | 0.1 ± 0.1 (nd–0.2) | | Stadler (1977) |
| 1975 | Southern California Bight, USA | 20 | (<0.05–0.8) | (<0.05–5.9) | (<0.03–2.2) | | Risebrough et al. (1976) |
| 1980 | Corpus Christi Bay, Texas, USA | 8 | | | | 1.32 ± 1.04 (0.2–3.1) | Ray et al. (1983a) |
| 1980–1981 | Blanca Bay, Argentina ^d | 15 | 37.9 ± 87.2 (nd–346.0) | | nd | | Sericano & Pucci (1984) |
| 1981 | Syowa Station, Antarctica ^e | 6 | + | + | | 0.008 ± 0.008 (0.001–0.021) | Tanabe et al. (1983) |
| ^a Mean ± sd (range) | ^b Present | ^c Not detected | ^d <i>o,p'</i> -DDT, 13.4 ± 24.5, (nd – 94.5) | | | ^e <i>o,p'</i> -DDT, + | |

Table 3. HCH isomers (BHC) in coastal and near-shore surface waters (ng l⁻¹)

| Year | Region | N | α-HCH | β-HCH | γ-HCH | δ-HCH | Σ HCH | Source |
|---------------|--|----|--------------------------------------|------------------------|-----------------------------|-------------------------|----------------------------|-------------------------|
| 1971 | Hawaii Canal, USA Pacific Ocean | 6 | | | 0.9 (0.3-2.0) | | | Bevenue et al. (1972) |
| 1971 | Near Marseille, France, Mediterranean Sea | ? | | | 70 (40-90) | | | Raybaud (1972) |
| 1974- 1975 | Santos Estuary, Brazil | 77 | | | | | (nd-1020) | Tommasi (1985) |
| 1975 | German Bight, North Sea | 17 | 6.5 ± 5.5 ^a (2.0-20.0) | | 5.6 ± 4.7 (1.9-16.0) | | | Stadler (1977) |
| 1975 | Western Baltic Sea | 18 | 6.8 ± 1.3 (4.7-8.7) | | 5.0 ± 1.0 (3.4-7.0) | | | Stadler (1977) |
| 1976 | Western Baltic Sea | 14 | 9.8 ± 4.1 (5.5-21.8) | 0.8 ± 0.2 (0.3-1.3) | 3.4 ± 0.8 (1.9-5.2) | | | Gaul & Ziebarth (1980) |
| 1978 | Western Baltic Sea | 13 | 7.5 ± 1.8 (4.1-11.0) | 0.4 ± 0.2 (0.2-0.7) | 2.9 ± 0.8 (1.8-4.5) | | | Gaul & Ziebarth (1980) |
| 1980 | Corpus Christi Bay, Texas, Gulf of Mexico | 8 | 0.27 ± 0.15 (<0.05-0.49) | | 0.07 ± 0.05 (<0.01-0.11) | | | Ray et al. (1983a) |
| 1980- 1981 | Blanca Bay, Argentina | 15 | 22.4 ± 16.4 (5.1-50.7) | | 27.8 ± 22.3 (5.4-65.1) | 6.6 ± 5.4 (0.5-17.5) | | Sericano & Pucci (1984) |
| 1981- 1982 | Syowa Station, Antarctica | 6 | | | | | 0.48 ± 0.27 (0.29-0.93) | Tanabe et al. (1983) |
| 1981- 1982 | Monaco, Mediterranean Sea | 4 | | | (nd-1.29) | | | Burns et al. (1985) |

nd: not detected ^a Mean ± SE

Hexachlorobenzene (HCB) was detected consistently in fjords off Norway although not quantified (Ofstad et al. 1979) and reported as 0.24 ± 0.25 ng l⁻¹ in 1980 in Corpus Christi Bay, Texas (USA) (Ray et al. 1983a). In the Mediterranean HCB is reported to range up to 1.29 ng l⁻¹ in unfiltered surface waters with highest concentrations occurring during the phytoplankton production season (Burns et al. 1985).

OCEANIC SURFACE WATERS

A better estimation of global contamination by organochlorines can be obtained by examining levels in the open ocean far from point sources. DDT compounds are generally around 1 ppb in open ocean waters (Table 5), and as expected, this is lower than that reported for coastal waters (Table 2). However, Jonas & Pfaender (1976) could find no trends in DDE distribution during a survey in 1972 extending from N. Carolina (USA) to the Sargasso Sea. Tanabe & Tatsukawa (1980) concluded that their measured levels of ΣDDT in the N.W. Pacific were higher than Atlantic values but this is not borne out by the measurements of Orlova (1983). The Atlantic values reported by Mikhaylov (1979), for 2 transects during 1973 and 1975

between Norway and Iceland, are anomalously high and probably represent a misprint. Surface water and atmospheric ΣDDT levels in the Pacific and Indo-Pacific region are higher in the southern hemisphere which probably reflects terrestrial usage (Tanabe & Tatsukawa 1980, Tanabe et al. 1982a). Finally, there is no obvious temporal trend in oceanic ΣDDT levels (Table 5).

Only one study reported finding cyclodiene pesticides in open ocean waters. Jonas & Pfaender (1976) measured 6.4 ± 5.6 ng l⁻¹ dieldrin and found traces of aldrin in surface waters of the western N. Atlantic.

HCH pesticides have been sought in most of the world oceans and seas (Table 6). Tanabe & Tatsukawa (1980) report γ-HCH (Lindane) levels between 1 and 10 ng l⁻¹ in Pacific surface waters, which is higher than the levels found in the Atlantic (Orlova 1983). They attribute this to the present heavy pesticide usage in Asia and this is also reflected in measured atmospheric levels (Tanabe et al. 1982a). ΣHCH residues in seawater were higher in the northern hemisphere of the Pacific region and α-HCH dominated the northern whereas γ-HCH dominated the southern hemisphere (Tanabe et al. 1982a).

In the early 1970s PCBs averaged 20 to 40 ng l⁻¹ in Atlantic surface waters but declined sharply in 1973 to

ca 1 ng l^{-1} (Table 7). Harvey et al. (1974) have attributed this decline to government restrictions in use and the resulting reduction in production. However, Risebrough et al. (1976) find Harvey's earlier measurements an order of magnitude too high based on indus-

trial sales and estimated release to the atmosphere from N. America. Williams & Robertson (1975) found only $2.6 \pm 1.4 \text{ ng l}^{-1}$ in the North Central Pacific Gyre in 1972, and Tanabe & Tatsukawa (1980) reported averages of 0.3 to 0.5 ng l^{-1} in the N.W. Pacific be-

Table 4. PCB's in coastal and near-shore surface waters (ng l^{-1})

| Year | Region | N | ng l^{-1} | Match | Source |
|---------------|--|----|-----------------------------------|----------------------------------|--|
| 1971 | Narragansett Bay Rhode Island, USA | ? | 150 ± 40^a | Aroclor 1254 | Duce et al. (1972) |
| 1971 | Near Marseille, France, Mediterranean Sea | ? | (100–210) ^b | ? | Raybaud (1972) |
| 1972 | Around Japan | ? | (0.3–13.9) | ? | Marine Safety Agency, Japan (1974) (cited in Stadler 1977) |
| 1973 | Around Japan | ? | (0–6.4) | ? | |
| 1973 | California (USA), near sewage outfall | ? | (5.4–16.3) | ? | Pavlou et al. (1974) |
| 1973 | Southern California Bight | 7 | 0.4–0.10 (0.3–0.5) | Aroclor 1254 | Risebrough et al. (1976) |
| 1974 | New England (USA), Continental Shelf | 6 | 0.8 | Aroclor 1260 | Harvey et al. (1974) |
| 1974 | Off California, USA | 7 | 12.7 ± 10.6 (3.0–35.6) | Aroclor 1254 | Scura & McClure (1975) |
| 1974 | Around British Isles | 31 | 0.4 ± 0.3 (< 0.15 –1.5) | Aroclors 1254 and 1260 | Dawson & Riley (1977) |
| 1974 | German Bight, North Sea | 22 | 3.1 ± 0.9 (2.0–4.7) | Clophen A 30 | Stadler & Ziebarth (1975) |
| 1974 | Western Baltic Sea | 21 | 2.9 ± 1.2 (1.1–5.9) | Clophen A 30 | Stadler & Ziebarth (1976) |
| 1975 | Hanö Bight, Baltic Sea | 8 | 0.9 ± 0.9 (0.3–3.0) | Clophen A 50 | Osterroht (1977) |
| 1975 | German Bight, North Sea | 17 | 2.1 ± 0.9 (0.8–3.6) | Clophen A 50 | Stadler (1977) |
| 1975 | Western Baltic Sea | 18 | 1.1 ± 0.8 (nd–3.9) | Clophen A 50 | Stadler (1977) |
| 1975 | France, Mediterranean coastline | 11 | 13.1 ± 12.3 (1.5–38.0) | Phenochlor DP-5 | Elder (1976) |
| 1975 | Southern California Bight, USA | 20 | (0.04–2.0) ^c | Aroclor 1254 | Risebrough et al. (1976) |
| 1976 | Western Baltic Sea | 14 | 7.2 ± 4.1 (1.1–15.6) | Clophen A 60 | Gaul & Ziebarth (1980) |
| 1976– 1977 | Oslofjord and Frierfjord, Norway | 3 | 5 ± 1 (4– < 10) | Aroclor 1254 and Clophen A 60 | Ofstad et al. (1979) |
| 1978 | Western Baltic | 12 | 5.7 ± 2.5 (3.5–11.9) | Clophen A 60 | Gaul & Ziebarth (1980) |
| 1980 | Corpus Christi Bay, Texas, USA | 8 | 4.8 ± 10.7 (0.1–31.0) | Aroclor 1260 | Ray et al. (1983a) |
| 1981– 1982 | Syowa Station, Antarctica | 6 | 0.05 ± 0.01 (0.03–0.07) | Kanechlor 300, 400 or 500 | Tanabe et al. (1983) |
| 1981– 1982 | Monaco, Mediterranean Sea | 4 | (0.2–1.2) | Aroclor 1254 | Burns et al. (1985) |

^a Mean \pm SE^b Range^c Pentachlorobiphenyls

Table 5. DDT and metabolites in oceanic surface waters (ng l⁻¹)

| Year | Region | N | <i>p,p'</i> -DDT | <i>o,p'</i> -DDT | <i>p,p'</i> -DDE | <i>p,p'</i> -DDD | ΣDDT | Source |
|-------------------|--------------------------------------|------------------------|-------------------------------------|------------------------------|------------------------------|------------------|----------------------------|--|
| 1972 | Mediterranean Sea | ? | (2–24) | | (0.5–2.0) | (0.7–2.0) | | Andryuschtschenko et al. (1975); (cited in Stadler 1977) |
| 1972 | North Central Pacific Gyre | 2 | <0.002 ^a | | <0.01 | | | Williams & Robertson (1975) |
| 1972 | Gulf Stream-Sargasso Sea | 6 | <8 nd ^b | | 2.9±2.7 (<0.35–7.5) | <0.8 nd | | Jonas & Pfaender (1976) |
| 1973 | Sargasso Sea | 9 | 0.2±0.1 ^c (<0.15–0.5) | <0.05 | | | | Bidleman & Olney (1974) |
| 1973 | NE Atlantic Ocean, Norway to Iceland | ? | + ^d | | + | + | 500±90 | Mikhaylov (1979) |
| 1975 | NE Atlantic Ocean, Norway to Iceland | ? | + | | + | + | 500±90 | Mikhaylov (1979) |
| 1975 | NW Pacific Ocean, | 13 | + | | + | + | 0.11±0.05 (0.06–0.23) | Tanabe & Tatsukawa (1980) |
| 1975 | Pacific Ocean, off Mexico | 23 | (<1–20) | | (<1–<10) | (<1–<80) | | Risebrough et al. (1976) |
| 1976 | NW Pacific Ocean | 8 | + | | + | + | 0.90±0.26 (0.52–1.35) | Tanabe & Tatsukawa (1980) |
| 1976 | Arabian Sea and Bay of Bengal | 6 | + | | + | + | 0.10±0.04 (0.06–0.16) | Tanabe & Tatsukawa (1980) |
| 1977 | Mediterranean Sea, | 3 | 1.2 | | 0.2=ΣDDE+DDD | | 1.4 | Orlova (1983) |
| | Subtropical Atlantic, | 15 | 0.7 | | 0.1=ΣDDE+DDD | | 0.8 | |
| | Subarctic Atlantic | 10 | 0.4 | | 0.5=ΣDDE+DDD | | 0.9 | |
| 1978 | Mediterranean Sea | 16 | 0.4 | | 0.2=ΣDDE+DDD | | 0.6 | Orlova (1983) |
| | Subtropical Atlantic, | 30 | 0.3 | | 0.2=ΣDDE+DDD | | 0.5 | |
| | Subarctic Atlantic | 73 | 0.2 | | 0.4=ΣDDE+DDD | | 0.6 | |
| 1977 | China Sea | 3 | + | | + | + | 0.08±0.02 | Tanabe & Tatsukawa (1980) |
| 1978 | NW Pacific Ocean | 6 | + | | + | + | 0.25±0.03 (0.22–0.27) | Tanabe & Tatsukawa (1980) |
| 1978 | Bering Sea | 7 | + | | + | + | 0.02±0.01 (0.01–0.04) | Tanabe & Tatsukawa (1980) |
| 1979 | NW Pacific Ocean | 8 | + | | + | + | 0.38–0.41 (0.02–1.17) | Tanabe & Tatsukawa (1980) |
| 1979 | Mediterranean Sea, | 32 | 0.7 | | 0.3=ΣDDE+DDD | | 1.0 | Orlova (1983) |
| | Tropical Atlantic, | 11 | 0.3 | | 0.4=ΣDDE+DDD | | 0.7 | |
| | Subtropical Atlantic, | 49 | 0.7 | | 0.1=ΣDDE+DDD | | 0.8 | |
| | Subarctic Atlantic | 54 | 0.4 | | 0.2=ΣDDE+DDD | | 0.6 | |
| 1980 | Mediterranean Sea, | 35 | 0.5 | | 0.5=ΣDDE+DDD | | 1.0 | Orlova (1983) |
| | Tropical Atlantic, | 34 | 0.4 | | 0.3=ΣDDE+DDD | | 0.7 | |
| | Subtropical Atlantic, | 39 | 0.5 | | 0.2=ΣDDE+DDD | | 0.7 | |
| | Subarctic Atlantic | 49 | 0.2 | | 0.1=ΣDDE+DDD | | 0.3 | |
| 1980–1981 | Indo-Pacific | 15 | 0.03±0.03 (0.005–0.091) | 0.01±0.01 (0.001–0.018) | 0.004±0.003 (0.001–0.013) | | 0.04±0.04 (0.007–0.13) | Tanabe et al. (1982a) |
| | Antarctic Ocean, below Australia | 12 | 0.01±0.01 (0.003–0.053) | 0.003±0.002 (0.001–0.006) | 0.002±0.001 (0.001–0.005) | | 0.02±0.01 (0.005–0.058) | Tanabe et al. (1982a) |
| ^a Mean | ^b Not detected | ^c Mean ± sd | ^d Present | | | | | |

tween 1975 and 1979. Open ocean levels are generally lower than those found in coastal waters (Table 4). Tanabe et al. (1982b) showed that coastal waters contained a higher proportion of penta, hexa and heptachlorobiphenyls over lower chlorinated PCBs. This finding was expected from their lower vapour pressures which would result in reduced atmospheric transport over long distances.

SURFACE FILMS

Surface films are primarily composed of fatty acids and alcohols of biological origin and are believed to

form a monomolecular layer between the air-sea interface (Garrett 1965, 1967). Organochlorine residues are now well-known to accumulate in sea-surface films because of their lipophilic properties (Seba & Corcoran 1969, Duce et al. 1972, Larsson et al. 1974, Ofstad et al. 1979). Sampling techniques for films are necessarily crude and include anywhere from 150 μm to 3.5 mm of the surface layer. I followed the example of Duce et al. (1972) to calculate the concentration factors, using published organochlorine levels in surface films, by conservatively estimating the film to be 5 molecular layers thick and assuming all organochlorines were associated with the lipids in this film. The concentration factors for all organochlorine compounds in sur-

Table 6. HCH isomers (BHC) in oceanic surface waters (ng l^{-1})

| Year | Region | N | α -HCH | β -HCH | γ -HCH ^a | Σ HCH | Source |
|---------------|---|----------------------|--------------------------------|---------------------------------|---|------------------------------|---|
| 1972 | NW Atlantic Ocean | 6 | | | 0.04 ± 0.07 (0.10–0.15) ^b | | Jonas & Pfaender (1976) |
| 1972 | Mediterranean Sea | ? | | | (2–9.5) | | Andryuschtschenko et al. (1975); (cited in Stadler 1977) |
| 1976 | NW Pacific Ocean | 8 | | | | 10.6 ± 5.4 (4.9–22.6) | Tanabe & Tatsukawa (1980) |
| 1976 | Arabian Sea and Bay of Bengal, Indian Ocean | 6 | | | | 1.1 ± 0.6 (0.2–1.9) | Tanabe & Tatsukawa (1980) |
| 1977 | China Sea | 3 | | | | 2.2 ± 1.1 (1.3–3.4) | Tanabe & Tatsukawa (1980) |
| 1977 | Mediterranean Sea, Subtropical Atlantic, Subarctic Atlantic | 3 15 10 | | | 1.3 1.4 0.5 | | Orlova (1983) |
| 1978 | Mediterranean Sea, Subtropical Atlantic, Subarctic Atlantic | 16 30 73 | | | 0.7 0.6 0.8 | | Orlova (1983) |
| 1978 | NW Pacific Ocean | 6 | | | | 1.3 ± 0.4 (0.8–1.9) | Tanabe & Tatsukawa (1980) |
| 1979 | Bering Sea | 7 | | | | 3.9 ± 0.4 (3.2–4.4) | Tanabe & Tatsukawa (1980) |
| 1979 | NW Pacific Ocean | 8 | | | | 7.3 ± 3.6 (3.7–14.2) | Tanabe & Tatsukawa (1980) |
| 1979 | Mediterranean Sea, Tropical Atlantic, Subtropical Atlantic, Subarctic Atlantic | 32 11 49 54 | | | 1.3 0 0.5 0.4 | | Orlova (1983) |
| 1980 | Mediterranean Sea, Tropical Atlantic, Subtropical Atlantic, Subarctic Atlantic | 35 34 39 49 | | | 1.5 0.1 0.8 0.3 | | Orlova (1983) |
| 1980– 1981 | Indo-Pacific Ocean | 15 | 0.9 ± 1.0 (0.1–3.4) | 0.2 ± 0.3 (0.02–1.0) | 1.4 ± 1.3 (0.16–3.7) | | Tanabe et al. (1982a) |
| 1980– 1981 | Antarctic Ocean, below Australia | 12 | 0.09 ± 0.07 (0.02–0.10) | 0.03 ± 0.02 (0.008–0.66) | 0.5 ± 0.3 (0.19–0.94) | | Tanabe et al. (1982a) |

^a Lindane (1,2,3,4,5,6,-hexachlorocyclohexane)

^b Mean \pm sd (range)

face films relative to subsurface waters (upper metre) ranged between 10^4 and $10^7 \times$, with 10^5 to $10^6 \times$ in Narragansett Bay, USA, German Bight, western Baltic Sea and the Sargasso Sea and 10^4 to $10^5 \times$ in Blanca Bay Argentina (Table 8). These extremely high factors are due to the lipophilic properties of organochlorines and are believed to be either a direct result of atmospheric 'fall-out' on the ocean surface and/or from subsurface biological origin, though the relative contribution of each pathway is presently unknown (Bidleman & Olney 1974, Larsson et al. 1974). Feeding schools of fish and whales could produce a surface slick by rupturing oily zooplankton, chiefly copepods, during consumption, with the wax esters escaping to the surface. Certainly extensive slicks have been observed where fish and whales are feeding on dense aggregations of zooplankton (Watkins & Schevill 1979). Wave-pro-

duced bubbles could also be a means by which 'dissolved' organics and associated organochlorines are concentrated and brought to the sea surface (Riley 1970, Johnson 1976).

DEPTH DISTRIBUTION

Three studies have dealt with organochlorine distribution in water columns of shallow environments. The information given by Sericano & Pucci (1984) on the water column of Blanca Bay, Argentina, reveals no significant differences (paired t-test) between *o,p'*-DDT, *p,p'*-DDT, aldrin, heptachlor, α -HCH, γ -HCH and δ -HCH concentrations between 1 m depth and bottom waters (7 to 12 m). The studies of Stadler (1977) show significantly higher concentrations of *p,p'*-DDD, α -HCH

Table 7. PCB's in oceanic surface waters (ng l^{-1})

| Year | Region | N | ng l^{-1} | Match | Source |
|---------------|--|----|----------------------------------|-------------------------------|------------------------------|
| 1971 | N Atlantic Ocean Iceland to Nova Scotia | 8 | $25 \pm ?$ | Aroclor 1260 | Harvey et al. (1974) |
| 1972 | North Central Pacific Gyre | 2 | 2.6 ± 1.4^a (1.6-3.6) | Aroclor 1254 | Williams & Robertson (1975) |
| 1972 | NE Atlantic Ocean 34°-63°N | 19 | 39 ± 36 (1-150) | Aroclor 1260 | Harvey et al. (1973) |
| 1972 | NW Atlantic Ocean Gulf Stream, Sargasso | 15 | 27 ± 24 (1-88) | Aroclor 1260 | Harvey et al. (1973) |
| 1973 | N Atlantic Ocean Azores to Barbados | 8 | $2 \pm ?$ | Aroclor 1260 | Harvey et al. (1974) |
| 1973 | N Atlantic Ocean, Sargasso to New York Bight, USA | 9 | $0.8 \pm ?$ | Aroclor 1260 | Harvey et al. (1974) |
| 1973 | Sargasso Sea | 9 | 1.4 ± 0.9 (<0.09 -3.6) | Aroclor 1254 | Bidleman & Olney (1974) |
| 1975 | Mediterranean Sea | 37 | 2.9 ± 3.6 (0.2-19.0) | Phenochlor DP-5 | Elder & Villeneuve (1977) |
| 1975 | NW Pacific Ocean | 13 | 0.41 ± 0.10 (0.25-0.56) | Kanechlor 300, 400 and 500 | Tanabe & Tatsukawa (1980) |
| 1975 | Pacific Ocean off Mexico | 23 | (<10 - <300) ^b | Aroclor 1254 | Risebrough et al. (1976) |
| 1976 | NW Pacific Ocean | 8 | 0.54 ± 0.29 (0.29-1.11) | Kanechlor 300, 400 and 500 | Tanabe & Tatsukawa (1980) |
| 1978 | NW Pacific Ocean | 6 | 0.35 ± 0.13 (0.23-0.59) | Kanechlor 300, 400 and 500 | Tanabe & Tatsukawa (1980) |
| 1979 | NW Pacific Ocean | 5 | 0.33 ± 0.05 (0.22-0.38) | Kanechlor 300, 400 and 500 | Tanabe & Tatsukawa (1980) |
| 1980- 1981 | Indo-Pacific Ocean | 18 | 0.12 ± 0.06 (0.04-0.25) | Kanechlor 300, 400 and 500 | Tanabe et al. (1982b) |
| 1980- 1981 | Antarctic, below Australia | 9 | 0.06 ± 0.01 (0.04-0.08) | Kanechlor 300, 400 and 500 | Tanabe et al. (1982b) |

^a Mean \pm sd (range) ^b Pentachlorobiphenyls

Table 8. Organochlorine concentration factors for the air-sea interface relative to water beneath the surface film

| Compound (s) | Concentration factor | Source |
|------------------|----------------------|---|
| PCB's | 10^4 – 10^7 | Duce et al. (1972), Bidleman & Olney (1974), Stadler (1977), Ofstad et al. (1979), Gaul & Ziebarth (1980) |
| <i>p,p'</i> -DDT | 10^4 – 10^6 | Bidleman & Olney (1974), Stadler (1977), Sericano & Pucci (1984) |
| <i>o,p'</i> -DDT | 10^4 | Bidleman & Olney (1974), Sericano & Pucci (1984) |
| <i>p,p'</i> -DDE | 10^6 | Stadler (1977) |
| <i>p,p'</i> -DDD | 10^6 | Stadler (1977) |
| α -HCH | 10^4 – 10^6 | Stadler (1977), Gaul & Ziebarth (1980), Sericano & Pucci (1984) |
| β -HCH | 10^5 – 10^6 | Gaul & Ziebarth (1980) |
| γ -HCH | 10^4 – 10^6 | Stadler (1977), Gaul & Ziebarth (1980), Sericano & Pucci (1984) |
| δ -HCH | 10^4 | Sericano & Pucci (1984) |
| Dieldrin | 10^6 | Stadler (1977) |
| Heptaclor | 10^4 | Sericano & Pucci (1984) |
| Aldrin | 10^5 | Sericano & Pucci (1984) |

and γ -HCH but not *p,p'*-DDT, *p,p'*-DDE, dieldrin and PCBs in the upper metre compared to bottom waters (10 to 44 m) of the German Bight, whereas in the western Baltic Sea only surface and bottom PCB concentrations were similar. The studies of Gaul & Ziebarth (1980) show significantly higher concentrations of β -HCH and γ -HCH but not α -HCH and PCBs in the western Baltic in both 1976 and 1978. However, both German studies sampled the surface waters with a bucket which would be contaminated by the surface film (Gordon et al. 1974).

Jonas & Pfaender (1976) found no difference in *p,p'*-DDE, dieldrin or γ -HCH in the upper 1000 m of the open N.W. Atlantic but they sampled with a Niskin bottle which would be contaminated as it passed through the surface film. Harvey et al. (1973) reported PCB surface concentrations in the N. Atlantic from bucket samples that were significantly greater than 200 m samples collected with a Bodman sampler. However, later work with closable samplers revealed no difference between 1 m and deeper water in the Atlantic (Harvey & Steinhauer 1976a), Mediterranean (Elder & Villeneuve 1977) and N.E. Pacific (Scura & McClure 1975). The studies of Tanabe & Tatsukawa (1983) in the W. Pacific to Antarctic region reveal no significant differences between surface and deep concentrations of PCBs, DDT compounds and isomers of HCH, except γ -HCH which was significantly higher in surface samples. A decreasing trend with depth was noted for all HCH isomer concentrations. However, their seawater sampler went through the surface film open (S. Tanabe pers. comm.). The definitive work on the depth distribution of organochlorines has yet to be done with sufficient replication for statistical treatment, and closable samplers to avoid surface contamination.

AVAILABILITY TO ZOOPLANKTON FROM SEAWATER

It is apparent that zooplankton inhabiting either estuarine, coastal or oceanic waters and various depth strata will each be subjected to a different level of organochlorine contamination. Neustonic coastal copepods are likely to be the most heavily contaminated if organochlorine compounds are not too tightly sequestered in lipid materials in the surface film.

The whole issue of the availability of organochlorines in seawater to zooplankton is far from clear because it is only recently that we have attempted to deal separately with the dissolved and suspended components in seawater. The availability of organochlorines depends on the quality, quantity and size of competing living and inert particles, because organochlorines are very hydrophobic and adsorb readily to other surfaces (Haque & Schmedding 1976, Picer et al. 1977, Hiraizumi et al. 1979). Many studies have shown that particulate matter high in organic content accumulates and retains greater concentrations of organochlorines than inorganic particles (Choi & Chen 1976, Picer et al. 1977, Hiraizumi et al. 1979, Nau-Ritter et al. 1982, O'Connors et al. 1982). The concentration of particles (vol/vol) in the marine environment, from bacteria to whales, has been found to be inversely proportional to particle diameter (Sheldon et al. 1972, 1973) which implies that most of the organochlorines in seawater should adsorb onto the bulk of the particulates which are less than 1 μ m in diameter. Organochlorines are also incorporated into colloidal micelles believed to be formed by humic substances in the presence of ionic species (Wershaw et al. 1969, Poirrier et al. 1972, Carter & Suffet 1982, Hassett & Milicic 1985). Humic substances may com-

prise as much as 90 % of the dissolved organic matter in seawater (Boehm & Quinn 1973, Skopintsev 1981). It follows that most organochlorines are probably bound to particulates or associated with the micellar structures present in seawater.

Operationally, 'solution' is defined in oceanography as passage through a 0.5 μm pore-size filter (Strickland & Parsons 1972). This definition would include organochlorines associated with micellar structures because filtration through a 0.45 μm membrane does not remove experimentally added organochlorines from humic solutions (Wershaw et al. 1969). However, membrane filters have been used to remove between 98 and 99 % of organochlorines added to double-distilled water (Kurtz 1977). Filtration is, therefore, of dubious value in separating the dissolved from the particulate unless all dissolved organochlorines in nature are in fact associated with micellar structures.

Organochlorine determinations on seawater have been made either by direct solvent extraction of bulk seawater, occasionally prefiltered through coarse mesh to remove larger organisms, or by concentration of the samples on resins such as Amberlite XAD-2, polyurethane foam, activated carbon or other absorbant (Sericano & Pucci 1984). The direct extraction approach combines organochlorines in 'solution' with those bound to both microorganisms and inert particles, whereas the sorbent methods allow a proportion of the organochlorines associated with particulates to escape detection (Bedford 1974).

Cox (1971a), after prefiltering seawater through 176 μm mesh to remove larger organisms, then concentrating particulates through centrifugation and filtration on GF/C filters, found that between 2 and 10 % of the DDT residues in seawater were associated with the filters. Tanabe & Tatsukawa (1983) reported 14 to 88 % of the ΣDDT , 7 to 58 % of the PCBs and 0.05 to 2.4 % of the ΣHCH residues in surface waters of the western Pacific to Antarctic region to be retained on GF-50 filters (0.5 μm). This increasing trend from DDT compounds to HCH isomers to pass through a glass-fiber filter is consistent with their known water solubilities (Gunther et al. 1968, Haque et al. 1974, Biggar & Riggs 1974). Similarly the more soluble lower chlorinated PCB isomers are proportionately more common in the seawater filtrate than on the filters (Tanabe & Tasukawa 1983). They also observed that the percentage of adsorbed to total organochlorines increased towards high latitudes and suggested that this was associated with increased standing stocks of phytoplankton.

However, a considerable amount of the auto- and heterotrophic organisms may pass through these filters, particularly in the more dilute conditions prevalent at low latitudes. Sheldon et al. (1972) have

hypothesized that particulate matter (vol/vol) [P] is distributed equally between logarithmic size intervals in the oceans; thus $[P] \propto D^{-1}$, where D is the equivalent spherical diameter of these particles. It follows that the particle surface area exposed per unit volume [S] in the oceans is proportional to D^{-2} . Given the affinity of organochlorines for surfaces, it would appear that little organochlorine is dissolved in seawater and that most of it is associated with minute particles.

The reported values of PCBs in seawater, therefore, vastly overestimate what is truly dissolved but these values may be realistic estimates of the contaminant 'pool' available for uptake by larger planktonic organisms. Harding & Phillips (1978) found that PCBs associated with natural particulates were transferred rapidly to phytoplankton, with an equilibrium occurring within hours. They speculated that either a 2-step process occurred via seawater or that PCBs were transferred directly during a transient association with particulate matter. The important point is that a dynamic equilibrium was established rapidly between particulate material and phytoplankton.

ACCUMULATION BY ZOOPLANKTON FROM THE SEAWATER MEDIUM

Many studies have described the accumulation of water-borne organochlorines by aquatic organisms but unfortunately these results are rarely intercomparable. The mathematical basis of first-order kinetic models was formulated many years ago for radionuclide research (Sheppard 1962, Ruzic 1972). Only recently have single compartment models been used to describe and predict the accumulation of organochlorines by aquatic organisms from their environment (Eberhardt et al. 1971, Sodergren & Svensson 1973, Branson et al. 1975, Harding & Vass 1977). These models assume that the rates of uptake and loss are proportional to the organochlorine concentration in seawater [W] and that in or on the organism [\emptyset], thus

$$\frac{d[\emptyset]}{dt} = k_i[W] - k_o[\emptyset] \quad (1)$$

where k_i and k_o = rate constants onto and/or into, and out of the organisms, respectively. Both coefficients incorporate a contaminant dilution factor which reflects the growth of the organism during experimental exposure, but this would be small in the zooplankton studies. Previous studies have established that the open-ocean concentrations of organochlorines in 'seawater' have been consistently in the low ng l^{-1} range during the past decade; thus the assumption that 'seawater' concentrations [W] are constant is justified.

Many experiments have shown independently that a negative exponential function best describes the clearance of organochlorines from aquatic organisms (Branston et al. 1975, Goerke & Ernst 1977, Fowler et al. 1978, Harding & Vass 1979, McManus et al. 1983). It appears that both the basic assumptions of the model are reasonable.

Few experimental studies have met the criteria of constant organochlorine levels in 'seawater' during the uptake phase and flow-through systems for determining the true clearance rate constants (k_i). It is easier to interpret the accumulation process if clearance rates are determined first under similar experimental conditions. The dynamics for p,p' -DDT in *Calanus finmarchicus* in contaminated filtered seawater have been described (Harding & Vass 1977) from concentrations comparable to the ppt levels found in the oceans (Tables 2, 5 & 8) to the maximum solubility in water (Biggar & Riggs 1974). The fit to the model is reasonable with $k_i = 0.048 \text{ d}^{-1}$, and k_r values between 1.0 and $2.5 \times 10^4 \text{ d}^{-1}$. This means equilibrium concentrations in *C. finmarchicus* can be predicted from water contamination alone.

In the previous section we discussed the availability of organochlorines to marine organisms from particulate matter. Peters & O'Connor (1982) exposed *Gammarus daiberi* and *Neomysis americana* to similar concentrations of ^{14}C -PCB (Aroclor 1254) in static experiments with filtered (0.45 μm) and unfiltered river water and found little difference in the equilibrium concentrations reached by these estuarine zooplankters. Wyman & O'Connors (1980) exposed *Acartia tonsa* to ^{14}C -PCB (Aroclor 1254) in static uptake experiments to initial concentrations of 10 $\mu\text{g PCB l}^{-1}$ filtered seawater (0.45 μm) and 1.6 $\mu\text{g PCB l}^{-1}$ seawater and illite clay particles, which resulted in equilibrium concentrations of 248 and 22 $\mu\text{g PCB g}^{-1}$ dry weight *A. tonsa*, respectively. Thus it does not appear to matter whether zooplankters are exposed to organochlorines on particles greater or less than 1 μm .

Several factors affect the accumulation process. The first is the chemical nature of the compounds. Zhang et al. (1983) have shown with *Daphnia magna* in static experiments that the higher the chlorine content, or the lower the water solubility of the PCB compounds used, the longer it takes to reach an equilibrium level. Furthermore, the bioaccumulation factor at equilibrium, $[\text{Ø}]/[\text{W}]$, increases with the chlorine content of the PCB isomers. Zhang et al. (1983) were able to demonstrate that the less soluble PCBs took a longer time to clear from *D. magna* even though a flow-through system was not used. It is reasonable to expect the rate-constants of DDT compounds and the more chlorinated PCB isomers to be similar from their water solubilities (Wallnofer et al. 1973, Biggar & Riggs 1974) but for the

same reason the cyclodiene and HCH pesticides should be radically different (Gunther et al. 1968). Recently many authors have successfully used octanol-water partition coefficients and aqueous solubility to predict bioaccumulation factors for a variety of chemicals (Veith et al. 1979, MacKay 1982).

In its simplest form the exchange equilibria theory of Hamelink et al. (1971) treats the biota as an immiscible liquid phase in an aqueous medium and assumes that purely physico-chemical processes control the equilibrium situation for each compound. Clayton et al. (1977) found zooplankton collected in Puget Sound, Washington (USA), contained 1 to 16 ppm PCBs on a lipid weight basis between 1973 and 1975. They believed these results could be explained solely by equilibrium partitioning between internal lipid pools of the biota and ambient water because bioconcentration factors, normalized for lipid, were the same at all locales and seasons sampled which covered a range of species assemblages. However, we are just beginning to appreciate that there are surface dynamics, such as particle-water-organism interactions (Harding & Phillips 1978, Nau-Ritter et al. 1982), and biological processes, such as feeding, egg laying, overwintering and death (Wyman & O'Connors 1980, Harding et al. 1981), which augment molecular partitioning. These factors will be discussed more fully later.

The size of an organism has long been recognized to affect the dynamics of organochlorine uptake and release. Cox (1971b) found that small *Euphausia pacifica* of 2 to 3 mg dry weight, initially accumulated $\approx 35 \text{ ng DDT g}^{-1}$ dry weight over a 2 h period compared to $\approx 9 \text{ ng DDT}$ for 10 mg specimens in static uptake experiments. Small *E. pacifica* reached an equilibrium of $\approx 80 \text{ ppb}$ by the third day. Unfortunately this study was not extended past 3 d but the large euphausiids were approaching the same level ($\approx 60 \text{ ppb}$) when the experiment was terminated. In flow-through clearance studies Cox (1971b) found large euphausiids to take longer to lose DDT, with a k_i of 0.046.

Harding & Vass (1979) studied the accumulation and loss of $^{14}\text{C}_{p,p}$ -DDT with *Thysanöessa raschii* to look for a possible organism-size difference in rate-constants compared to those previously measured with *Calanus finmarchicus*. The euphausiids used were 2 orders of magnitude larger than *C. finmarchicus* on a weight basis. The DDT accumulated by *T. raschii* (ng mg^{-1} dry weight), subjected to constant seawater contamination, did not change significantly with organism size and the derived k_i 's were of the same order of magnitude as those obtained for *C. finmarchicus*. Furthermore, in flow-through experiments with plenty of food, the clearance rate constant of 0.043 d^{-1} was no different from that previously measured in *C. finmarchicus*.

However, sickly euphausiids were observed to take up far less DDT than their healthy counterparts (Harding & Vass 1979). Previously Crosby & Tucker (1971) found heat-killed *Daphnia magna* accumulated half as much DDT as healthy individuals and Wildish & Zitko (1971) found that heat-killed *Gammarus oceanicus* did not take up measurable quantities of PCBs. Cox (1971b) exposed healthy and heat-killed euphausiids to pptr levels of DDT in seawater over a 2 h period and found no difference in DDT accumulated per unit weight. This discrepancy can be explained by the use of magnetic stirrers in those experiments, which would have kept the corpses in motion. It would appear that surface area exposed per unit time rather than surface area alone is an important factor controlling uptake. Why is it then that the uptake and clearance rates of *T. raschii*, with a calculated surface area of $\approx 10 \text{ mm}^2 \text{ mg}^{-1}$, are so similar to *C. finmarchicus* with $\approx 34 \text{ mm}^2 \text{ mg}^{-1}$ exposed? Hardy & Bainbridge (1954) measured the vertical swimming speed of both *C. finmarchicus* and euphausiids in an experimental plankton wheel and reported rates of 15 to 47 m h^{-1} and 40 to 128 m h^{-1} , respectively. Field estimates on vertically migrating populations are 36 to 108 m h^{-1} for *C. finmarchicus* and 180 to 216 m h^{-1} for euphausiids (Klyashtorin & Yarzhombek 1973). Thus, euphausiids swim about 2.7 times as fast as *C. finmarchicus*. This suggests that weight-specific surface area exposed per unit time should be about equal and explains why a size difference in uptake rates could not be found between *T. raschii* and *C. finmarchicus* (Harding & Vass 1979).

A previous collation of rate-constants, including both fish and aquatic invertebrates experimentally exposed to either DDT or PCBs isomers, showed that k_1 varied between 0.004 and 0.066 d^{-1} , whereas k_2 ranged over 3 orders of magnitude, 2.5×10^{-4} to 2.5×10^4 (Harding & Addison 1986). This variation cannot be explained by size of organism alone. Temperature affects the metabolism and activity of an organism and should also modify the exchange rates of organochlorines between plankton and their environment. Cox (1971b) found *Euphausia pacifica* accumulated slightly more DDT from seawater at 15° compared to 5°C over a 2 h exposure. However, Zhang et al. (1983) found that a 10°C rise in temperature doubled the loss of biphenyl, 2,2'-PCB and 2,4,6,2',4'-PCB from contaminated *Daphnia magna*. This corresponds to the 'Q₁₀' law whereby a rise of 10°C doubles metabolic rates (Prosser 1973) and presumably the respiratory surface area exposed per unit time. Fish also accumulate organochlorines at a faster rate at higher temperature, within the normal thermal range for the species, and in most cases a doubling of contaminant levels was found with a 10°C rise in temperature (Murphy & Murphy

1971, Reinert et al. 1974, Edgren et al. 1979, Veith et al. 1979). Edgren et al. (1979) found that perch cleared 2,3',4',5-PCB, 2,2',4,4',5,5'-PCB and *p,p*-DDT faster at 16° than at 8°C but the differences were not statistically significant. Reduced salinity also increases the uptake of organochlorines by fish from water (Murphy 1970, Tulp et al. 1979) and would be expected to apply to estuarine copepods as well. This too is probably related to the amount of water that comes in contact with the body surfaces due to increased osmoregulatory activity.

McManus et al. (1983) found that *Acartia tonsa* cleared ¹⁴C-PCB (Aroclor 1254) accumulated from either contaminated phytoplankton or seawater at about the same rate. Harding & Addison (1986) came to a similar conclusion from a review of the fish and aquatic invertebrate literature. However, McManus et al. (1983) determined that *A. tonsa* cleared PCB more rapidly in the presence of phytoplankton and attributed this to an increased metabolic rate and increased egg production. In further experiments they demonstrated that female *A. tonsa* cleared PCB twice as rapidly as males and were able to show that the difference was due to egg production.

Less is known about the transport of organochlorines within organisms. Organochlorines are sorbed externally (Crosby & Tucker 1971) or to gut surfaces, perhaps in micellular structures (Laher & Barrowman 1983), partitioned into the hemolymph or blood plasma lipoproteins (Dvorchik & Maren 1974) and transported to lipid-rich tissues, analogous to the experimental results following intravenous injection to invertebrates (Guarino et al. 1974) and fish (Darrow & Addison 1973, Pritchard et al. 1973, Zinck & Addison 1974). Several of these authors have reported blood plasma clearance curves of more than one phase, indicating the presence of more than one 'compartment' clearing at different rates. Harding & Vass (1977) attempted to examine the lipid as well as the body levels of *p,p*-DDT in *Calanus finmarchicus* during uptake and clearance experiments. However, the lipid compartment was found to contain a constant percentage of the total DDT which indicates either a rapid transferral to the lipid stores or contamination in the lipid extraction technique. Our current level of experimental sophistication has not yet warranted the use of multicompartment models (e.g. Ruzic 1972).

ORGANOCHLORINES ACQUIRED FROM CONTAMINATED PREY

Copepods can be herbivorous, detritivorous, omnivorous or carnivorous in feeding habits (Poulet 1983) and one species may change from one feeding

type to another either seasonally (Marshall 1924, Corner et al. 1976, Landry 1981) or as it develops through naupliar to copepodid stages (Lewis 1967, Poulet 1977, Uye & Kasahara 1983). Five studies have examined the transfer of organochlorines from prey to consuming zooplankton. Wyman & O'Connors (1980) described the accumulation of ^{14}C -PCB (Aroclor 1254) by *Acartia tonsa* through feeding on an algal mixture of *Skeletonema costatum* and *Thalassiosira pseudo*. *A. tonsa* fed labelled phytoplankton acquired $61 \mu\text{g PCB g}^{-1}$ dry wt. (ppm) within 5 h from an initial contamination of 22 ng PCB in the phytoplankton and $0.6 \mu\text{g l}^{-1}$ in the water. Once the phytoplankton was consumed *A. tonsa* body levels dropped to 28 ppm over the rest of the 2 d period whereas the water level declined to $0.25 \mu\text{g PCB l}^{-1}$. In a similar experiment *A. tonsa* was subjected to an initial 0.6 ppb in the water and a total of 80 ng PCB on illite clay particles (Wyman & O'Connors 1980). In this case maximum body levels were reached after 30 h at ≈ 22 ppm dry wt. It is obvious that the feeding process can triple the body contamination and in a much shorter period when water and particle contamination are roughly equal.

Peters & O'Connor (1982) introduced the estuarine amphipod *Gammarus daiberi* to ^{14}C -PCB (Aroclor 1254) in water in the presence of 2 food organisms, the rooted green plant *Myriophyllum spicatum* and the alga *Chroococcus* sp. They also exposed *G. daiberi* to a previously labelled *Chroococcus* sp. culture after it was first concentrated then diluted in fresh medium, $\approx 3 \times 10^5$ cells ml^{-1} . *G. daiberi* subjected to $1 \mu\text{g PCB l}^{-1}$ in either water, filtered water or water containing *M. spicatum* accumulated 5.5 ± 0.9 , 4.0 ± 0.4 , and $2.4 \pm 0.2 \mu\text{g PCB g}^{-1}$ dry wt (ppm), respectively, after 48 h. This indicates that feeding was less important in PCB uptake by *G. daiberi* than the reduction of PCBs available from the water, due to competition for organochlorines by particulates and the plant. *G. daiberi* accumulated less PCBs, 1.0 ± 0.2 and $0.2 \pm 0.1 \mu\text{g g}^{-1}$ dry wt., after 48 h exposures to either a labelled *Chroococcus* sp. culture or just the labelled algae in fresh medium, respectively. These results can only be explained by the low food intake of *G. daiberi*, together with the phytoplankton removing and binding PCBs from the water otherwise available for uptake. It is noteworthy that feeding by *G. daiberi* was never verified in this paper. In another study *G. tigrinus* accumulated maximal body burdens of $\approx 23 \mu\text{g }^{14}\text{C}$ -PCB g^{-1} dry weight in the first 9 h exposure to contaminated fungal food whereas control organisms took up 0.6 to 13.9 % of this level indirectly via the water over 24 h treatment periods (Pinkney et al. 1985). This clearly demonstrates the importance of the feeding pathway in estuarine amphipods.

Grazing has been studied with *Calanus finmar-*

chicus feeding on ^{14}C -*p,p'*-DDT contaminated phytoplankton cultures (Harding et al. 1981). In this case, 10 of the 13 experimental results are consistent with previous laboratory determinations of feeding rate. Two cell concentrations of *Thalassiosira weissflogii* were used to approximate average ($\approx 60 \mu\text{g C l}^{-1}$) and maximum ($\approx 600 \mu\text{g C l}^{-1}$) carbon levels encountered in nature. Calculated DDT assimilation efficiencies ranged from 5 to 94 %. However, an inverse linear relation exists between the percent DDT assimilation and food consumption rate. In other words, when the level of algal contamination is constant, experimentally held between 110 and $180 \mu\text{g DDT g}^{-1}$ algal dry wt., *C. finmarchicus* ingests more DDT and phytoplankton at the higher algal populations but retains proportionately less DDT.

Cox (1971b) measured the assimilation of ingested ^{14}C -*p,p'*-DDT by the euphausiid *Euphausia pacifica* through carnivorous feeding on *Artemia* sp. nauplii. The results of 5 experiments, each with 1 euphausiid feeding on contaminated *Artemia* nauplii, varied from 34 to 93 % assimilation after allowing for a 2 d gut clearance period. In a batch experiment with 12 euphausiids the assimilation efficiency for DDT ingested was 76 %. In these euphausiid studies there were not enough experiments to distinguish any dependence of assimilation efficiency on size of daily ration.

The exponential model (Eq. 1) used earlier to describe the flux of organochlorines between planktonic crustaceans and seawater can be expanded to include organochlorines assimilated through feeding on contaminated food

$$\frac{d[\emptyset]}{dt} = k_i[W] - k_e[\emptyset] + \alpha I[F] \quad (2)$$

where α = organochlorine assimilation ratio from food; I = weight-specific ingestion rate; $[F]$ = concentration of organochlorines in the food (Bruggeman et al. 1981, Harding et al. 1981). Experimentally determined α values for both invertebrates and fish are generally high but variable and there appears to be no relationship between the amount of organochlorine retained and the size of organism or trophic level (Harding & Addison 1986). McManus et al. (1983) found that PCB residues accumulated by *Acartia tonsa* feeding on contaminated phytoplankton are eliminated at a similar rate to those obtained from contaminated water alone. Thus, it is justified to retain the clearance rate proportional to the concentration in the organism.

This equation can be rewritten to describe organochlorine dynamics over the entire life span of the organism by incorporating the size dependency of ingestion rate

$$\frac{d[\emptyset]}{dt} = k_i[W] - k_j[\emptyset] + \alpha\beta t^{-\gamma}[F] \quad (3)$$

where t = time, β and γ = proportionality constants which depend on environmental temperatures (Harding et al. 1981). These constants were obtained for *Calanus finmarchicus* by combining the experimentally determined size and ingestion rate relationship of Paffenhöfer (1971) with the size and stage-duration curve of Mullin & Brooks (1970). Several variants on this modelling approach to simulate organochlorine residue dynamics in aquatic food webs have recently been described (Norstrom et al. 1976, Thomann 1981, Brown et al. 1982).

Predator-prey bioconcentration factors $\{[\emptyset]/[F]\}$ simulated with this model (Eq. 3) indicate that organochlorine retention from food exceeds clearance early in life. This reverses at some point due to the decreasing specific ingestion-rate as a copepod grows to adult size. The maximum bioconcentration derived from food is directly proportional to the organochlorine assimilation efficiency. The exact timing of this peak is governed by environmental temperature, with higher temperatures causing maximum bioconcentration to occur earlier in developmental time. However, the predicted ultimate bioconcentration factor at maturity does not differ substantially for *Calanus finmarchicus*

Table 9. Records of organochlorine levels in prey: phytoplankton and microplankton

| Year | Region | Mesh size | N | DDT (ng g ⁻¹) | | | PCB (ng g ⁻¹) | | | Dieldrin (ng/g) | | | Source |
|--------------------|--|---------------------|----------|---------------------------|-------------------------|----------|---------------------------|----------|------------------|-----------------|----------|----------------------|-------------------------------|
| | | | | Wet wt | Dry wt | Lipid wt | Wet wt | Dry wt | Lipid wt | Wet wt | Dry wt | Lipid wt | |
| 1965–1966 | Off Northumberland, UK, coastal water | ? | 1 | 30 | | | | | | 20 | | | Robinson et al. (1967) |
| 1971–1972 | Off Stockholm, Sweden, estuarine and coastal waters | 100 µm | 25 | | | | | | 300–3500 | | | Jensen et al. (1972) | |
| 1972 | Gulf of St. Lawrence, Canada, PEI, coastal water | 73 µm | 9 | | | | 90–3050 | | | | | | Ware & Addison (1973) |
| 1972–1973 | Turku archipelago, Baltic Sea, Finland, coastal water | 150 µm | 134 | | | | 40–750 | | | 4000–77 000 | | | Linko et al. (1974) |
| 1973 | Osaku Bay, Japan, coastal water | 93 µm | 21 | | 400–25 000 ^a | | | | | | | | Hiraizumi et al. (1975) |
| 1973 | North Sea, off Holland, coastal ^c water | 50 µm | 2 | 0.23 ^b | | 540 | 3.5 | | 8.4 | 0.3 | 690 | | Ten Berge & Hillebrand (1974) |
| 1974 | NW coast Mediterranean, coastal water | 60 µm | 6 | 40–76 | | | | | 232–741 (66 420) | | | | Marchand et al. (1974) |
| 1974 | Turku archipelago, Baltic Sea, Finland, coastal water | 150 µm | 67 | | | | 30–3300 | | | 26 000–340 000 | | | Linko et al. (1979) |
| 1974 | Mediterranean Sea, Villefranche, France, coastal water | 76 µm | 1 | | nd | | | | 4500 | | | | Fowler & Elder (1978) |
| 1974–1975 | Mediterranean Sea, Villefranche, France, coastal water | 76 µm | 3 | | | | 168–420 | | | 1800–4500 | | | Elder & Fowler (1977) |
| 1975 | Turku archipelago, Baltic Sea, Finland, coastal water | 150 µm | 47 | | | | 40–720 | | | 4000–66 000 | | | Linko et al. (1979) |
| 1976 | Gulf of St. Lawrence, NS, coastal water | 66–125 µm | 9 | | | | 1.0–6.4 | 6.1–74.4 | 86–2237 | | | | Harding et al. (1978) |
| | | 125–250 µm | 10 | | | | 0.6–7.0 | 5.8–60.8 | 104–2899 | | | | |
| 1977 | Gulf of St. Lawrence, NS, Canada, coastal water | 25–66 µm | 4 | | | | 0.1–0.9 | 1.4–4.7 | 173–232 | | | | Harding et al. (unpubl.) |
| | | 66–125 µm | 15 | | | | 0.2–3.3 | 1.5–31.6 | 152–440 | | | | |
| | | 125–250 µm | 15 | | | | 0.2–2.7 | 1.9–19.3 | 78–469 | | | | |
| ^a Range | | ^c Endrin | | Pentachlorobenzene | | α-HCH | | β-HCH | | HCB | | | |
| ^b Mean | | Wet wt | Lipid wt | Wet wt | Lipid wt | Wet wt | Lipid wt | Wet wt | Lipid wt | Wet wt | Lipid wt | Wet wt | Lipid wt |
| | | 0.12 | 0.27 | 0.05 | 0.14 | 0.09 | 0.22 | 0.21 | 0.49 | 0.06 | 0.13 | | |

Table 10. Records of organochlorine levels in marine plankton Crustacea

| Year | Region | Organism type Mesh size | N | DDT (ng g ⁻¹) | | | PCB (ng g ⁻¹) | | | Dieldrin (ng g ⁻¹) | | | Source |
|---------------|--|--------------------------------------|----|---------------------------|--------------------------|-------------|---------------------------|------------------|-----------------|--------------------------------|--------------|-------|----------------------------------|
| | | | | Wet | Dry | Lipid | Wet | Dry | Lipid | Wet | Dry | Lipid | |
| 1965– 1966 | Off Northumberland, UK, coastal water | Macrozooplankton | 1 | 160 ^a | | | | | | 160 | | | Robinson et al. (1967) |
| 1966 | Long Island, NY, USA, estuarine water | Bulk plankton >239 µm | 1 | 40 | | | | | | | | | Woodwell et al. (1967) |
| 1969 | Firth of Clyde, UK, estuarine water | Bulk plankton >300 µm | 1 | | | | 30 | | | | | | Holden (1970) |
| 1969 | NW Atlantic Ocean, shelf water | bulk plankton >239 µm | 14 | | 2.5– 110 ^b | 42– 7000 | | 71– 3000 | 320– 260 000 | | | | Risebrough et al. (1972) |
| 1970– 1971 | N Atlantic Ocean, oceanic water | Bulk plankton >333 µm | 4 | <0.01– 9.5 | 120– 1250 | 4– 450 | | 925– 19 300 | | | | | Grice et al. (1972) |
| 1970 | N Atlantic Ocean | Bulk plankton >239 µm | 2 | <0.01 | | | 300– 450 | | | | | | Risebrough et al. (1972) |
| 1971 | S Atlantic Ocean, oceanic water | Bulk plankton >239 µm | 4 | 0.3–2.6 | 90– 625 | 18– 638 | | 7300– 124 000 | | | | | Risebrough et al. (1972) |
| 1971 | Gulf of Mexico, shelf and oceanic waters | Bulk plankton >300 µm | 29 | tr. ^c –34 | | | <3– 1055 | | | | | | Giam et al. (1973b) |
| 1971– 1972 | Firth of Clyde, UK, coastal water | Bulk plankton >300 µm | 12 | 21–107 | 30– 460 | 80– 2200 | | 1100– 17 100 | 20– 230 | | 430– 2880 | | Williams & Holden (1973) |
| | North Channel, UK, shelf water | | 4 | <4–50 | <90– 750 | 10– 920 | | 200– 2500 | 2–15 | | 60– 260 | | Williams & Holden (1973) |
| | N Atlantic UK, oceanic water | | 10 | 2–16 | 20– 8000 | 10– 120 | | 100– 5500 | 1–2 | | 20– 1000 | | Williams & Holden (1973) |
| 1972 | Antarctic, Ross Sea, oceanic water | Bulk plankton >300 µm | 1 | <0.8 | <20 | <3 | | <76 | | | | | Giam et al. (1973a) |
| 1972 | Gulf of St. Lawrence, PEI, coastal water | Bulk plankton >239 µm | 9 | | | | tr.– 1860 | | | | | | Ware & Addison (1973) |
| 1973 | Gulf of Mexico, USA, estuarine to shelf waters | Bulk plankton >333 µm | 5 | 2–3 | 258– 3681 | 40– 157 | | 3370– 112 359 | <1–61 | | <1– 6742 | | Baird et al. (1975) |
| 1973 | North Sea, off Holland, coastal water | Bulk plankton >300 µm | 2 | 1.3 | 560 | 20 | | 10 300 | 1.5 | | 650 | | Ten Berge & Hillebrand (1974) |
| 1974 | St. Margaret's Bay NS, coastal water | copepods | 1 | <1 | | | | | | | | | Darrow & Harding (1975) |
| 1974 | NW Mediterranean Sea, coastal water | Bulk plankton >300 µm | 9 | | 35– 135 | | | 122– 74,500 | | | | | Marchand et al. (1974) |
| 1974 | Mediterranean Sea, Villefranche, France, coastal water | <i>Meganyctiphanes norvegica</i> | 1 | | 26 | | | 620 | | | | | Fowler & Elder (1978) |
| | | <i>Sergestes arcticus</i> | 1 | | 15 | | | 470 | | | | | |
| | | <i>Pasiphaea sivado</i> | 1 | | 5 | | | 210 | | | | | |
| 1973– 1975 | Puget Sound, Wash., USA, coastal water | Bulk plankton >333 µm | 37 | | | | 0– 200 | 0– 1600 | | | | | Clayton et al (1977) |
| 1974 | N Pacific Ocean, oceanic water | Copepods | 3 | | | | 100– 190 | 3260– 14 390 | | | | | Takagi et al. (1975) |
| 1976 | Gulf of St. Lawrence, Canada, coastal water | Bulk plankton >230 µm | 40 | | | | 0.2– 9.7 | 2.6– 39.0 | 40– 742 | | | | Harding et al. (1978) |
| 1977 | Spain, Atlantic Ocean, coastal and shelf waters | Bulk plankton >200 µm | 37 | | | | 3.7– 16 | 27.2– 231 | 230– 4812 | | | | Franco et al. (1981) |
| 1977 | Mediterranean Sea | Bulk plankton >280 µm | 7 | 1–11 | 12–85 | | 2–25 | 25– 230 | | | | | Fowler & Elder (1980–81) |

Table 10. continued

| Year | Region | Organism type Mesh size | N | DDT (ng g ⁻¹) | | | PCB (ng g ⁻¹) | | | Dieldrin (ng g ⁻¹) | | | Source |
|-------------------|---|----------------------------|---|---|-----|-------|---------------------------|-------------|-------------|--------------------------------|-----|-------|-----------------------------|
| | | | | Wet | Dry | Lipid | Wet | Dry | Lipid | Wet | Dry | Lipid | |
| 1977 | Gulf of St. Lawrence, NS, Canada, coastal water | Bulk plankton >250 µm | 60 | | | | 0.1- 21 | 0.5- 147 | 101- 820 | | | | Harding et al. (unpubl.) |
| 1978 | Mediterranean Sea ^e | Copepods | 3 | | | | | 180- 620 | | | | | Burns et al. (1985) |
| 1979 | | Pteropods | 1 | | | | | 190 | | | | | |
| | | Copepods | 7 | | | | | 68- 220 | | | | | |
| 1981 | | Copepods | 2 | 2.7- 20.4 | | | | 110- 490 | | | | | |
| 1982 | | Copepods | 1 | 21.6 | | | | 190 | | | | | |
| 1979 | NW Pacific Ocean ^d | Bulk plankton >300 µm | 1 | 1.7 | | | | 1.8 | | | | | Tanabe et al. (1984) |
| ^a Mean | ^b Range | ^c Trace | ^d 0.26 ng g ⁻¹ ΣHCH | ^e 0.24-7.20 ng g ⁻¹ dry γ-HCH; 0.30-1.40 ng g ⁻¹ dry HCB | | | | | | | | | |

grown at 10 and 15 °C, at least through the interaction of temperature with ingestion rate.

It is informative to compare the bioconcentration factors found in nature with the model results. Organochlorine levels in natural phytoplankton assemblages have not been determined, because of the impracticality of separating phytoplankton from microzooplankton in field samples. As much as 50 % of the 25 to 66 µm plankton size fraction can be present as animal species, mainly copepod nauplii (Harding et al. 1980). However, this fraction can still be representative of all prey available for ingestion because most copepods, including herbivores such as *Calanus finmarchicus*, can feed omnivorously on phytoplankton and microzooplankton (Corner et al. 1976). Published reports of organochlorine levels in bulk plankton collected in fine and coarse mesh nets are listed in Tables 9 and 10, respectively. Fewer compounds have been reported in plankton compared to seawater but this is due to either a lack of searching by the investigator or to masking of certain compounds by large quantities of another, e.g. PCBs obscuring the DDE GC peak (Harding et al. 1978). It is noteworthy that the ΣDDT, PCBs and dieldrin levels found in phyto- and zooplankton, on a wet weight basis, are at least 3 orders of magnitude greater than that found in seawater with its associated particulates.

There has been an enduring controversy in the literature over the relative importance of diet versus direct sorption from water in the accumulation of organochlorines by aquatic organisms (Harding et al. 1981). Eight studies exist from which it has been possible to calculate bioconcentration factors for zooplankton, in the copepod size range, relative to the concentration in their prey (Table 11). The results of Ware & Addison (1973) are exceptional in that the phytoplankton has

one to 2 orders of magnitude higher concentration of PCBs than the zooplankton. They attributed their observations to recent atmospheric input through precipitation, followed by sorption to the greatest surface area available. Fowler & Elder (1978) measured PCBs and DDTs in a microzooplankton-euphausiid-shrimp food chain and found no evidence for trophic magnification in the Mediterranean (Table 11). Most predator-prey bioconcentration values are close to or greater than unity, indicating higher concentrations of organochlorine compounds in the copepod size fractions than associated with their prey (Table 11). This observation can be explained if the input through feeding maintains organochlorine levels in copepods above a purely sorption equilibrium. There is experimental evidence to demonstrate that this can occur in the short term (Wyman & O'Connors 1980). It could also be argued that copepods accumulate higher concentrations because of their longer generation spans with the resultant increased exposure to particle-bound organochlorines other than food. However, laboratory studies of copepod feeding on contaminated prey show rapid bioaccumulation on a time scale of hours (Wyman & O'Connors 1980, Harding et al. 1981) which is sufficient to explain the organochlorine levels found in nature.

ORGANOCHLORINES TRANSFERRED FROM GENERATION TO GENERATION

A final process by which copepod populations could maintain substantial amounts of organochlorines is through the transfer of residues accumulated over a lifetime from mother to offspring. A number of studies document high organochlorine levels in eggs and

Table 11. Bioconcentration factors ($\{[\text{O}]/[\text{F}]\}$) for zooplankters over their prey

| Year | Region | N | ΣDDT | PCB | Dieldrin | Endrin | ΣHCH | HCB | 6-CB | Source |
|---------------|--|----|--------------------|----------|----------|--------|--------------------|-----|------|----------------------------------|
| 1965– 1966 | Off Northumberland, UK, coastal water | 1 | 2.0 | | 3.0 | | | | | Robinson et al. (1967) |
| 1970 | Monterey Bay, Calif., USA, coastal water | 1 | 1.0 | | | | | | | Cox (1970, 1971b) |
| 1972 | Gulf of St. Lawrence, PEI, Canada, coastal water | 7 | | 0.01–0.2 | | | | | | Ware & Addison (1973) |
| 1973 | North Sea, Holland coastal water | 2 | 2.1 | 2.1 | 1.9 | 1.9 | 1.5 | 1.3 | 0.9 | Ten Berge & Hillebrand (1974) |
| 1974 | NW Mediterranean Sea, France, coastal water | 6 | 0.9–2.6 | 0.4–1.6 | | | | | | Marchand et al. (1974) |
| 1974 | Mediterranean Sea, Villefranche, France, coastal water | 1 | 0.2–26 | 0.1–0.8 | | | | | | Fowler & Elder (1978) |
| 1976 | Gulf of St. Lawrence NS, Canada, coastal water | 11 | | 0.1–1.9 | | | | | | Harding et al. (1978) |
| 1977 | Gulf of St. Lawrence, NS, Canada, coastal water | 15 | | 0.8–9.2 | | | | | | Harding et al. (unpubl.) |

embryos of invertebrates and fish, which is not surprising considering the affinity these compounds have for lipids (Harding & Addison 1986).

McManus et al. (1983) found that adult female *Acartia tonsa* cleared ^{14}C -PCB at a significantly faster rate than males over a 36 h depuration study. They also discovered that the concentrations in the eggs, $407.5 \mu\text{g g}^{-1}$ dry wt., was considerably higher than the original concentration in the body of the mother, $95 \mu\text{g PCB g}^{-1}$ dry wt. McManus and coworkers then measured PCB concentrations in shed eggs and fecal pellets between 3 time periods to evaluate their importance in clearance studies. The weight-specific PCB concentration in the eggs was always higher than the corresponding concentration in the adult females, which is probably due to the higher lipid content of the eggs. There was no difference between the sexes in the amount of PCBs cleared in fecal pellets. A PCB budget of egg and fecal pellet extrusion and clearance through other routes illustrated that egg production accounted for the observation difference of PCB clearance in male and female *A. tonsa*.

It is instructive to use model Eq. (3) to predict the transfer of organochlorines from copepod generation to generation. Initially, any carry-over of organochlorines in the egg would have a large effect on the concentration in the earlier developmental stages but this is greatly reduced by the time maturity is reached (Harding et al. 1981). In a continuously cool environment with plentiful prey, a constant assimilation efficiency,

and a low seawater contamination, an equilibrium situation can be established after 4 generations. The observed higher contaminant concentrations in copepod eggs (McManus et al. 1983) is faithfully reproduced by the model. Increasing the environmental temperature has the effect of shortening the generation length but increases the number of generations needed for the copepod population to reach an equilibrium. In temperature latitudes with a marked seasonality of environmental variables, cold and warm seasons should induce an alternating contaminant equilibrium between seasons for both the bioconcentration in eggs and adult females. However, the reduction of copepod feeding during periods of food scarcity could not be incorporated into the model because of a lack of experimental information. Reduced feeding would lower the organochlorine content in successive clutches and obscure any alternating equilibrium between generations in nature.

EFFECT OF TOXICITY ON ORGANOCHLORINE-ORGANISM DYNAMICS AND SPECIES SURVIVAL

The toxic effect of various organochlorines on marine zooplankton rarely has been studied. More important, the level at which a contaminant burden reduces the long-term survival of a species is unknown. There is a distinct possibility that the

experimentally determined organochlorine rate-constants and assimilation efficiencies for copepods have been modified by the effects of the chemical on the physiology of the organism. This is apparent from the results of the organochlorine uptake studies performed on *Acartia tonsa* (Wyman & O'Connors 1980) and *Calanus finmarchicus* at high levels of contamination in the seawater (Harding & Vass 1977). The solution is to experiment at concentrations well below the acute level.

A few generalizations about invertebrate toxicity have been proposed, though a review of the more extensive fish literature (Cairns et al. 1975, Houston 1982) indicates that these may be premature. It appears that the lower the water solubility of individual organochlorine compounds the greater their toxicity to freshwater cladocerans (Crosby et al. 1966, Gunther et al. 1968). It is tentatively established that many organochlorines are more toxic to invertebrates at lower temperatures (Crosby et al. 1966, Boothe 1978).

The most vulnerable period for planktonic copepods must be in the non-feeding naupliar stages. As we have seen, organochlorines are partitioned into the lipid reserves of the organism which results in high levels in copepod eggs (McManus et al. 1983). Now, these lipids are rapidly utilized during early copepod development before active feeding takes place (Lee et al. 1974). This indicates that the early naupliar stages would be the first to experience the toxic effects of critical body burdens of organochlorines.

McManus et al. (1983) have shown that the first laid eggs of *Acartia tonsa* contain the highest level of contamination. High mortality of first laid eggs could serve as a mechanism for clearing the copepod population of organochlorines. However, it could equally well endanger the species since females produce only a few clutches before removal from the population by predators (Peterson 1979) and the first clutch is demographically the most important (Slobodkin 1961).

It was inadvertently found during uptake experiments that *p,p'*-DDT was acutely lethal to *Calanus finmarchicus* at body concentrations greater than 80 ng mg⁻¹ dry weight (Harding & Vass 1977) and to *Thysanøessa raschii* at greater than 20 ng mg⁻¹ dry weight (Harding & Vass 1979). Wyman & O'Connors (1980) found that PCBs were toxic to *Acartia tonsa* at body concentrations around 20 ng mg⁻¹ dry weight. The highest values found in nature to date approach this toxic level (Table 10).

ULTIMATE REPOSITORY

It is fitting to conclude this review with a discussion of the role played by zooplankton in the distribution of

organochlorines in the oceans. To achieve this, I have used PCBs as a representative group of compounds and divided the world oceans, because of broad differences in contamination levels, into that above the continental shelves and that above the ocean basins. The amount of PCBs in surface film, seawater, zooplankton, sediments, macrobenthos and fish were estimated separately (Table 12). As previously discussed, the seawater compartment includes particulate matter and phytoplankton and surprisingly contains most of the PCBs in the world oceans. Similarly, meiobenthos is included in the sediment compartment because large organisms (macrobenthos) are usually removed before organochlorine analysis. We have not included microplankton in the zooplankton compartment which would raise biomass concentrations by approximately one-third (Sheldon et al. 1977). The scarcity of observations of organochlorine determinations in abyssal fauna and deep-sea sediments is presently the weakest part of this tabulation (Table 12).

The physical rates of vertical mixing and advection in the oceans are too slow to explain the quantities of PCBs that have found their way into abyssal water, sediments and organisms. Conservative properties such as bomb-³H, originating in the mid-50s and -60s, indicate that vertical mixing in the ocean is largely confined to the surface layer above the thermocline (Broecker 1981). Surface formation of bottom water occurs in the North Atlantic and off the Antarctic continent. The most recently formed North Atlantic Bottom Water has an age of ≈80 yr whereas deep-sea bottom water in general has a residence time of ≈1000 yr. Away from areas of deep-water formation the vertical penetration of bomb-¹⁴C in seawater is more than twice that of ³H (Broecker et al. 1980). This suggests a biological transport which can best be explained by the known nocturnal migrations of mesopelagic organisms (Roe 1972, Roe et al. 1984) into the surface layer at mid-latitudes. This nocturnal feeding migration also helps to explain why similar levels of organochlorines have been found in epipelagic and mesopelagic communities (Baird et al. 1975, Barber & Warlen 1979). However, there is presently little support for Vinogradov's (1962) hypothesis of deep-sea food transfer by migrating zooplankters below 1500 m. Few species have been found to migrate daily through the 1000 m depth level (Foxton 1970, Roe 1972). Ontogenic migrations of high latitude copepods, to as deep as 2000 m annually (Østvedt 1955, Vinogradov 1968, Sekiguchi 1975), provide a similar mechanism on a different time scale for transporting PCBs to the ocean depths.

The oldest mechanism proposed for transporting organics, and therefore organochlorines, to the deep sea is the so-called 'rain' of detritus (Agassiz 1888). At cool latitudes mass settlements of phytoplankton

Table 12. PCB's in world oceans

| Continental shelf waters | Oceanic waters |
|---|---|
| $26.4 \times 10^{12} \text{ m}^2$ $1.8 \times 10^{15} \text{ m}^3$ | $334.6 \times 10^{12} \text{ m}^2$ area (Groves & Hunt 1980) $1.8 \times 10^{18} \text{ m}^3$ seawater (Groves & Hunt 1980) |
| <i>Surface film (150 μm)</i> | |
| $5 \times 10^5 \mu\text{g PCB m}^{-3}$ (Table 8) $1.9 \times 10^3 \text{ mt PCB}$ (9.5 %) | $0.5 \times 10^5 \mu\text{g PCB m}^{-3}$ (Table 8) $2.5 \times 10^3 \text{ mt PCB}$ (0.3 %) |
| <i>Seawater and particulates</i> | |
| $5 \mu\text{g PCB m}^{-3}$ (Table 4) $9.2 \times 10^3 \text{ mt PCB}$ (46.1 %) | $0.5 \mu\text{g PCB m}^{-3}$ (Table 7) $880.1 \times 10^3 \text{ mt PCB}$ (98.5 %) |
| <i>Zooplankton</i> | |
| $20.1 \text{ g wet m}^{-2}$ (Vinogradov 1968) $10 \text{ ng PCB g}^{-1} \text{ wet}$ (Table 10) $0.005 \times 10^3 \text{ mt PCB}$ (<0.1 %) | 160 g wet m^{-2} (Vinogradov 1968) $10 \text{ ng PCB g}^{-1} \text{ wet}$ (Table 10) $0.5 \times 10^3 \text{ mt PCB}$ (0.1 %) |
| <i>Sediments, micro- and meiobenthos</i> | |
| $327 \times 10^{-6} \text{ g PCB m}^{-2}$ (Addison 1980) | $30 \times 10^{-6} \text{ g PCB m}^{-2}$ (Eder et al. 1976, Harvey & Steinhauer 1976b) |
| $8.7 \times 10^3 \text{ mt PCB}$ (43.6 %) | $10 \times 10^3 \text{ mt PCB}$ (1.1 %) |
| <i>Macrobenthos</i> | |
| 100 g wet m^{-2} (Nesis 1965, Mills 1980) | 0.5 g wet m^{-2} (Nesis 1965, Rowe et al. 1974, Haedrich & Rowe 1977) |
| $50 \text{ ng PCB g}^{-1} \text{ wet}$ (Moller et al. 1983, Ray et al. 1983b, Weber 1983, Greig & Sennefelder 1985) | $25 \text{ ng PCB g}^{-1} \text{ wet}$ (Eder et al. 1976) |
| $0.1 \times 10^3 \text{ mt PCB}$ (0.5 %) | $0.004 \times 10^3 \text{ mt PCB}$ (<0.1 %) |
| <i>Pelagic and demersal fish</i> | |
| 8 g wet m^{-2} (Mills & Fournier 1979) | 6 g wet m^{-2} (Haedrich & Rowe 1977, Mills & Fournier 1979, Gjosaeter & Kawaguchi 1980) |
| $200 \text{ ng PCB g}^{-1} \text{ wet}$ (Schaefer et al. 1976, van den Broek 1979, Schneider 1981, Eisenberg & Topping 1985) | $200 \text{ ng PCB g}^{-1} \text{ wet}$ (Baird et al. 1975, Eder et al. 1976) |
| $0.04 \times 10^3 \text{ mt PCB}$ (0.2 %) | $0.4 \times 10^3 \text{ mt PCB}$ (<0.1 %) |
| <i>Total</i> | |
| $19.9 \times 10^3 \text{ mt PCB}$ (100 %) | $893.5 \times 10^3 \text{ mt PCB}$ (100 %) |

remains have been observed to reach abyssal depths (4100 m) within weeks of the demise of plankton blooms in overlying surface water (Billett et al. 1983). However, in oligotrophic subtropical seas almost all of the organic matter produced is recycled in the warm upper layer (Riley 1970, Knauer & Martin 1981). Nevertheless, a certain degree of seasonality exists even in oligotrophic waters, such as the Sargasso Sea, where winter sedimentation at 3200 m depth is twice the summer rate (Deuser et al. 1981). Larger particles such as carcasses and molts (Wheeler 1967), fecal pellets (Urrère & Knauer 1981) and 'marine snow' (Shanks & Trent 1980, Silver & Alldredge 1981) make up most of the organic material settling through the thermocline in strongly stratified regions. Elder & Fowler (1977) and Fowler et al. (1979) have shown that euphausiid and copepod fecal pellets can transport substantial amounts of PCBs to the deep-sea.

A final possibility, inferred by Harding (1974), is that deep-sea heterotrophs draw on the large 'pool' of dis-

solved and particulate organic matter and that they in turn are consumed by copepods. Urrère & Knauer (1981) found a higher rate of fecal pellet sedimentation at 1500 m than at 500 m in the northeast Pacific and concluded that detrital feeders were repackaging organics into pellets at various depths. Karl & Knauer (1984) also observed a subsurface increase in the number of fecal pellets settling, and presumably produced, between 600 and 1100 m depth. This increase in pellets at mid-depths is either the result of migrating mesopelagic organisms defecating at depth or repackaging of heterotrophs or detritus by deep-sea filter feeders. Fecal pellets produced below the thermocline are more likely to reach greater depths because turbulence is less and the cooler waters slow the decomposition process. Zooplankton carcasses and molts produced below the surface layer will also decompose more slowly at the lower temperatures (Harding 1973). Migrating zooplankton and nekton are therefore indirectly important in augmenting downward detrital and

organochlorine transport below 1000 m.

Knap et al. (1986) measured $4.4 \pm 3.3 \text{ ng PCB m}^{-2} \text{ d}^{-1}$ in material settling into traps moored 1000 m above bottom in a 4200 m water column in the Sargasso Sea. This value is in reasonable agreement with the calculated average PCB accumulation on the deep sea floor (Table 12). Coastal measurements of PCBs settling into traps with particulate matter are an order of magnitude greater than those for the Sargasso which is consistent with other nearshore estimates of input from sinking fecal pellets and flux calculations into bottom sediments (Osterroht & Smetacek 1980, Burns et al. 1985).

It is no longer certain that the sediments are the final resting place for organochlorines. Larsson (1985a,b) has shown experimentally that there is a seasonal cycle whereby PCBs are released from bottom sediments in shallow waters during the summer, mediated by bioturbation as well as temperature. Resuspension of particulates into the nepheloid layer (Baker et al. 1985) could also be important in maintaining a dynamic equilibrium of organochlorines between sediments and the water column overlying the continental shelf and canyons.

In conclusion, the importance of zooplankton in the dynamics of PCBs in the world oceans is misleading if one simply takes into account the amount bound to their biomass (Table 12). The dispersion rates have not been quantified but it appears certain that zooplankton play a major role in distributing organochlorines throughout the ocean depths by increasing the deposition rates of particulate matter. At the present time it appears that the abyssal sediments may not be the ultimate repository of organochlorines, as previously thought (Woodwell et al. 1971), but rather the water column with its biota is retaining or recycling these compounds.

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