Marine Environmental Research **28** (1989) 259–264

The Relationship between Lipid Composition and Seasonal Differences in the Distribution of PCBs in *Mytilus edulis* L.

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ABSTRACT

The concentrations of individual chlorobiphenyl congeners were measured in the mussel Mytilus edulis *transplanted to several stations in Buzzards Bay and Nantucket Sound, MA (USA). Individual stations represented a gradient of chemical contamination and the sampling period extended over a complete annual cycle. Fluctuations in concentrations of some chlorobiphenyl congeners were apparent at all stations during the late spring and early summer with a marked decline occurring during autumn; this pattern was correlated with the seasonal cycle of gametogenesis and spawning activity. Relative redistribution and release of individual chlorobiphenyl congeners associated with spawning is not consistent, suggesting differential partitioning of specific congeners in different tissues or lipid pools. These patterns are consistent with our general view of the bioconcentration of organic contaminants in marine organisms. The major factors controlling the distribution of PCBs in mussels appear to be the relative concentrations of individual contaminants in ambient waters, modified to some extent by differences in partitioning between organisms and water (as indicated by differences in* K_{ow}*), and seasonal variations in lipid content.*

Bioconcentration of lipophilic organic contaminants may be influenced by physico-chemical properties, such as molecular configuration or stearic properties that influence biotransformation and membrane transfer

Marine Environ. Res. 0141-1136/90/$03·50
Printed in Great Britain

kinetics,[1,2] and biological factors, such as the partitioning between storage lipids and structural lipids[3,4] and differential distribution of contaminants among different tissues.[5] Steady-state pharmacokinetic models, generally used to describe the kinetics of uptake and release of lipophilic organic contaminants, consider that partitioning between the environment and the organism is controlled by steady-state uptake and depuration and the organism is considered to be a single homogeneous unit.[6,7] Empirical data on the relationship of bioconcentration factors (K_B) to octanol–water partition coefficients (K_{OW}), however, often deviate from the predicted values derived from single-compartment models, suggesting the interaction of several factors in determining bioconcentration with the potential for major seasonal differences to occur. Seasonal variation in bioconcentration may be related to dependency on specific metabolic processes involved in the storage and mobilization of lipid reserves during reproductive events.

Ongoing studies in New Bedford Harbor and adjacent Buzzards Bay, Massachusetts (USA) provide an excellent opportunity to investigate the bioavailability, bioconcentration, and biological effects of lipophilic organic contaminants in marine bivalve molluscs. The history of contaminant inputs to New Bedford Harbor is a carefully documented series of disturbances. Inputs of polychlorinated biphenyls (PCBs) and heavy metals are well-described and the fates of these contaminants have been examined in regards to chemical modifications, distributions, biogeochemical cycling and physical transport of contaminants within the Acushnet River Estuary and in the outer regions of Buzzards Bay.[1]

Specimens of *Mytilus edulis* L. were transferred to several stations in Buzzards Bay and Nantucket Sound, Massachusetts (USA), sampled bi-weekly and analyzed for concentrations of 20 chlorobiphenyl congeners or pairs of congeners in addition to changes in condition index and the distribution of neutral and polar lipid classes to examine differences in seasonal distribution of lipophilic organic contaminants. Individual stations represented a gradient of chemical contamination and the sampling period extended over a complete annual cycle (November 1984 to November 1985). Mussels were collected from a reference station in Sandwich, Massachusetts (USA), acclimated to transfer conditions, and transplanted in cages to specified stations. An array of six cages was deployed at each station and the array was suspended 1 m above the bottom. During bi-weekly sampling, 24 mussels were taken from each of the cages at random and subsampled for bioenergetic, biochemical, histological and chemical analyses. Three replicates were taken for lipid analyses and total lipids were determined by gravimetric analysis, followed by identification of specific lipid classes.[8] A pool of eight animals were prepared for organic contaminant analysis by

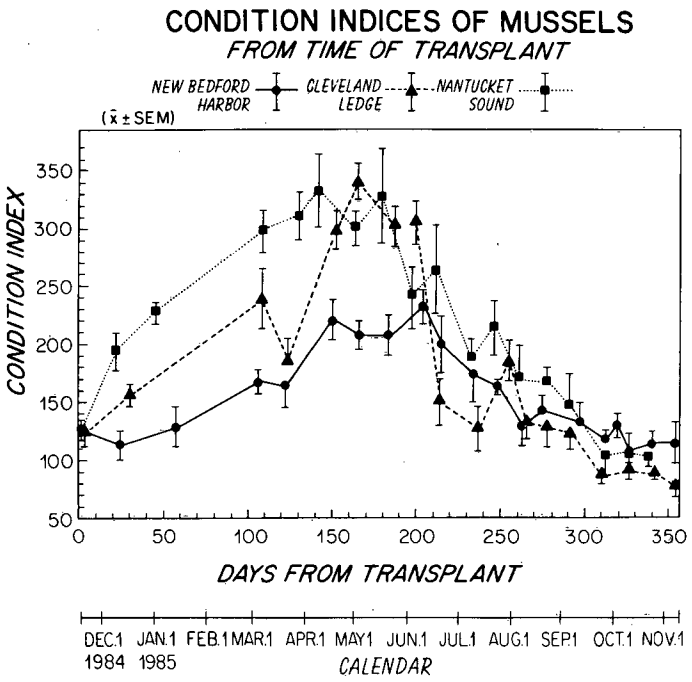

Fig. 1. Condition indices of *Mytilus edulis* from New Bedford Harbor, Nantucket Sound and Cleveland Ledge; mean of eight replicate measurements ± 1 SE.

alkaline digestion and column chromatographic isolation of specific classes of compounds; duplicate samples were analyzed for PCBs by glass capillary gas chromatography and gas chromatography–mass spectrometry.[1] Eight replicate samples were assayed for condition index, measured as the ratio of dry weight to shell volume.

Condition indices for mussels at New Bedford Harbor are lower than values for mussels at other stations during the pre-spawning period (Fig. 1). Values for mussels from the Cleveland Ledge, Massachusetts (USA) station are lower than values for mussels from the Nantucket Sound station during the first few months of the study, but are equal during the period of gametogenesis. Accumulation and utilization of lipid reserves (primarily triacylglycerols) correlate with changes in condition index and significant differences are evident between mussels from New Bedford Harbor and the other two stations. Maximum lipid accumulation among mussels from New Bedford Harbor is only 57·2% of values obtained at Nantucket Sound. As spawning proceeds, condition indices and lipid content are reduced in all populations.

Mussel transplants in New Bedford Harbor show uptake of chlorobiphenyl congeners to exceedingly high concentrations (10^{-6} g/g dry wt

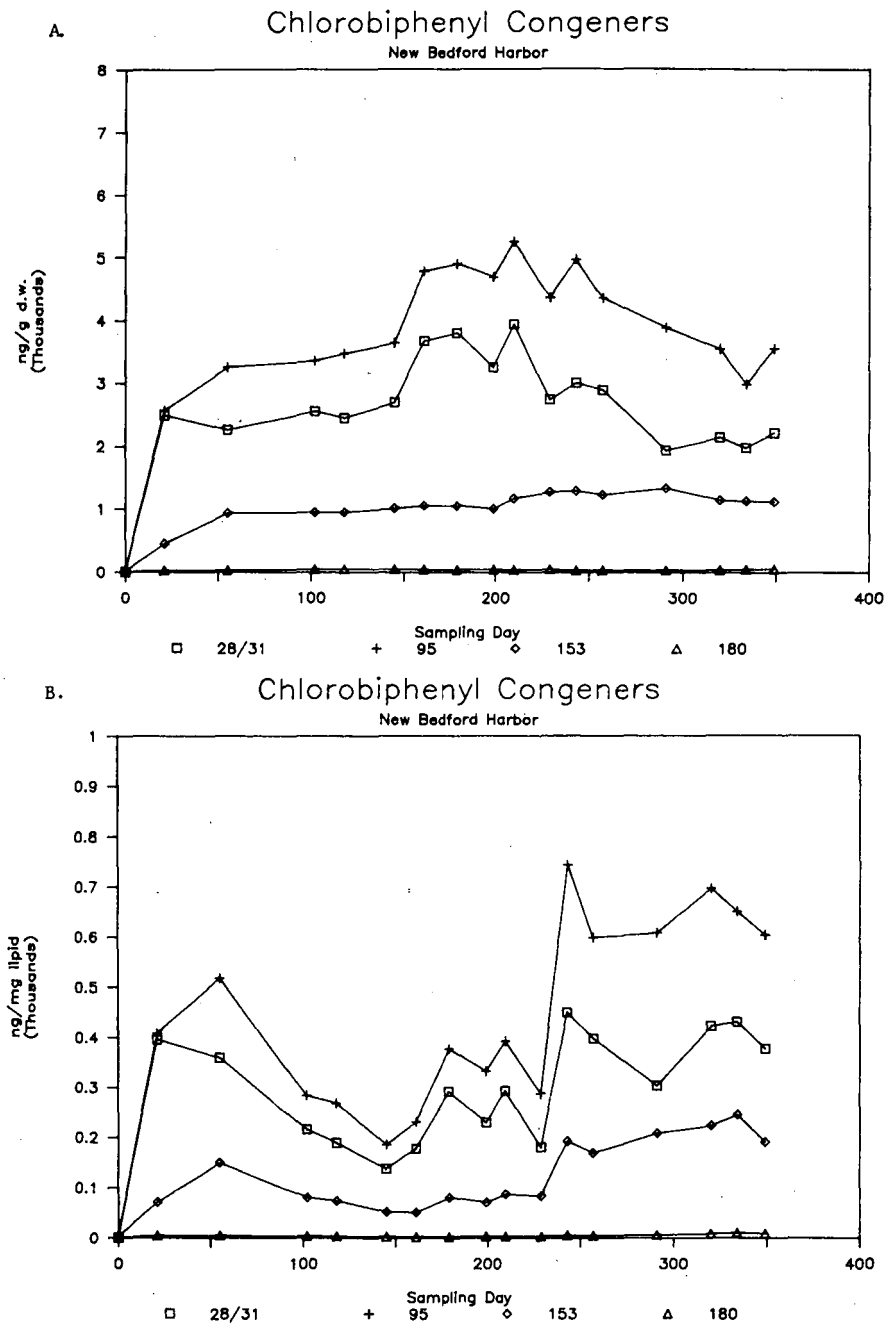

Fig. 2. Chlorobiphenyl congener distribution in *Mytilus edulis* from New Bedford Harbor. (A) Plotted on a dry weight basis, (B) plotted on a lipid weight basis. Numbers refer to IUPAC numbers for specific chlorobiphenyl congeners.

range). Fluctuation in the concentration of some chlorobiphenyl congeners (IUPAC No. 28/31 and 95) is apparent during the late spring and early summer (from day 150 onward) with a marked decline occurring during autumn; this pattern was correlated with gametogenesis and spawning activity (Fig. 2A). Relative redistribution and release of individual chlorobiphenyl congeners associated with spawning is not consistent, suggesting differential partitioning of specific congeners in different tissues or different lipid pools. Plotting these data on a lipid weight basis further supports the differential distribution hypothesis (Fig. 2B). Similar seasonal patterns in the fluctuation of individual chlorobiphenyl congeners were observed among mussels at other stations, although concentrations at each of these stations are at least 1–2 orders of magnitude lower than those observed at New Bedford Harbor. Differential uptake and loss of lighter chlorinated chlorobiphenyls (di- and trichlorobiphenyls) and heavier chlorinated chlorobiphenyls (hexa- and heptachlorobiphenyls) have also been noted for the green-lipped mussel, *Perna viridis.*[9]

The relative distribution of specific chlorobiphenyls in mussels transplanted to New Bedford Harbor is markedly different from the distributions observed in mussels from other sites. For example, IUPAC 28/31 (a pair of trichlorobiphenyls) and IUPAC 95 (a pentachlorobiphenyl) are present in greater relative abundance than other chlorobiphenyls in mussels transplanted to New Bedford Harbor, whereas these compounds are at relatively low to intermediate concentrations at the other two sites. Although IUPAC 28/31 have lower K_{ow} values than other chlorobiphenyls measured, their relatively high concentrations in harbor waters influence their rate of uptake.

These patterns are consistent with our general view of the bioconcentration of organic contaminants in marine organisms. The major factors controlling the distribution of PCBs in mussels appear to be the relative concentration of individual contaminants in ambient waters, modified to some extent by differences in partitioning between organisms and water (as indicated by differences in K_{ow}), and seasonal variations in lipid content. Unlike previous laboratory experiments[10] in which steady-state concentrations of PCBs were achieved in *Mytilus* within 20 days, the present study suggests that under actual field conditions only quasi-steady-state conditions can be achieved given the high seasonal variation in lipid content associated with the annual reproductive cycle of *Mytilus* and the potential for toxic effects on the synthesis of lipid reserves and the allocation of lipids to reproductive and energetic demands. Therefore, simple lipid (K_{ow}) partitioning phenomena cannot account for some of the second-order fluctuations apparent in contaminant distribution and models to describe selective partitioning within specific tissues are needed.

ACKNOWLEDGEMENTS

This research was supported by NOAA, Office of Sea Grant award NA 86-AA-D-S6090 (RP/22 and RP/17) to J.M.C. and J.W.F. with matching funds from the Coastal Research Center, Woods Hole Oceanographic Institution, and US Environmental Protection Agency CR-814895-01-1 to J.M.C. We thank Carl Johnson for assistance with GC/MS analyses and Bruce Tripp for assistance in the laboratory. Contribution No. 7076 from the Woods Hole Oceanographic Institution.

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