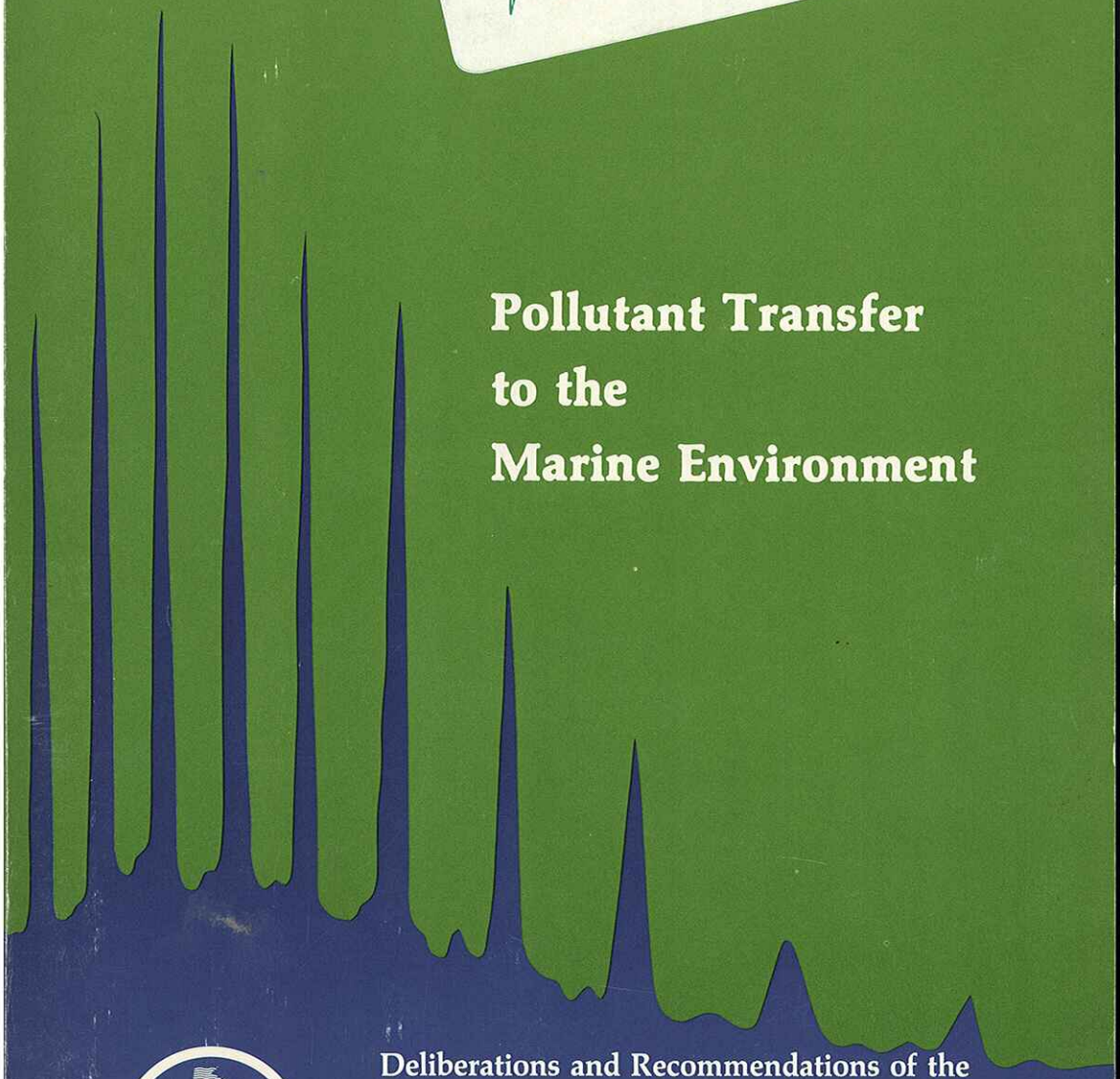
Pollutant Transfer to the Marine Environment



Deliberations and Recommendations of the
National Science Foundation,
International Decade of Ocean Exploration
Pollutant Transfer Workshop
January 11-12, 1974

Pollutant Transfer to the Marine Environment

Deliberations and Recommendations of the NSF/IDOE Pollutant Transfer Workshop held in Port Aransas, Texas January 11-12, 1974

Robert A. Duce, convener and editor
Graduate School of Oceanography
University of Rhode Island, Kingston

Patrick L. Parker, host and co-editor
Marine Science Institute
University of Texas, Port Aransas

C. S. Giam
Department of Chemistry
Texas A&M University, College Station
(on leave of absence to NSF/IDOE as
Program Manager, Environment Quality)

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Foreword

"Preserve the ocean environment by accelerating scientific observations of the natural state of the ocean and its interactions with the coastal margin—to provide a basis for (a.) assessing and predicting man-induced and natural modifications of the character of the oceans; (b.) identifying damaging or irreversible effects of waste disposal at sea; and (c.) comprehending the interaction of various levels of marine life to permit steps to prevent depletion or extinction of valuable species as a result of man's activities."

The above is the first of the six prescribed goals of the National Science Foundation's Office for the International Decade of Ocean Exploration. In pursuit of this goal, IDOE, in 1971, carried out a one-year Baseline Data Acquisition Program and in the following year initiated the Pollutant Transfer Program. The Baseline Program indicated the levels of contamination by trace metals and petroleum and chlorinated hydrocarbons in water, sediment and biota in the oceans. However, it did not provide information on the pathways or mechanisms controlling the rate of pollutant transfer from the source to and within the ocean, the information so necessary for predicting the distribution of pollutants, for assessing whether the oceans are becoming measurably polluted, and ultimately for indicating rates at which pollutants may be released safely to the environment.

The goals of the research projects in the Pollutant Transfer Program were determined to be: (1.) to identify important transfer pathways and mechanisms; (2.) to evaluate the major environmental factors which affect transfer processes and (3.) to develop principles governing transfer of pollutants.

To attain these ends, a research program was designed around a matrix consisting of the pollutants of concern, especially those reported in the Baseline Program, and the major processes by which they are transported in the environment (atmospheric, marine-outfalls, chemical, biological and geological). This report is an assessment of the first two-year (1972-74) effort of the investigators of the Transfer Program.

The publication of this volume has resulted from the dedicated efforts of the staff of IDOE including (visiting) Program Managers, Dr. George Grice, Woods Hole Oceanographic Institution; Dr. Patrick L. Parker, University of Texas, Marine Science Institute; and Dr. C. S. Giam, Texas A & M University, and Program Secretary, Ms. S. Robinson. I am grateful to the conveners, participants and special guests who participated in the meeting in Port Aransas, Texas, especially, the IDOE grantees whose data resulted in the publication of this report. I would also like to thank Dr. Parker for the hospitality extended to the group at Port Aransas.

Feenan D. Jennings, *Head*
Office for the International Decade of Ocean Exploration

Preface

Scientists in the two-year-old Pollutant Transfer Program of the National Science Foundation, International Decade of Ocean Exploration met in a workshop at Port Aransas, Texas, on January 11-12, 1974. This report summarizes their presentations as well as their discussions of and recommendations for future research. Due to time and space limitations, this volume does not include data accumulated by these investigators nor a review of the literature. The reader may write to individual investigators for detailed scientific publications.

During the first two years, primary research emphasis in the Pollutant Transfer Program has been on the transport of trace metals, and petroleum and chlorinated hydrocarbons to the ocean via the atmosphere, rivers, sewage and industrial outfalls, and dumping. Research also was begun on the chemical form and degradation of these pollutants in the marine environment. Biological studies were designed to investigate the mechanisms of pollutant uptake by organisms, to verify transfer through the food web and to quantify the role of organisms in pollutant dispersion. In less than two years, the investigators have made significant progress in all of these areas.

The accelerated development of natural resources and the increased use of the ocean as a dump have made studies of marine environmental quality a necessity. Fortunately NSF/IDOE funds have allowed marine scientists to make such studies. Thanks are also due the University of Texas Marine Science Institute, host for this Workshop. Herbert Windom was chairman of the Workshop's trace metal section and James Butler and Robert Risebrough were co-chairmen of the petroleum and chlorinated hydrocarbon section. Janice Stearns, Linda Anderson and Laurence Pearce provided invaluable technical and editing assistance in preparing this report.

Robert A. Duce

Introduction and Recommendations

The objectives of the Pollutant Transfer Program of the National Science Foundation, International Decade of Ocean Exploration are to determine the mechanisms which control the rate of pollutant transfer, both to the ocean and within the ocean, and to identify major environmental factors affecting transfer processes. Such information is necessary to predict pollutant distribution in the marine environment and to assess future ocean pollution. The importance of and rationale for studying transfer processes in the marine environment are adequately described in *Marine Environmental Quality* (NAS, 1971).

The following report summarizes the research accomplishments during the first two years of the Pollutant Transfer Program. The report also includes general and specific recommendations by scientists working in this program concerning future research in this area.

The discussion of scientific accomplishments within the Pollutant Transfer Program has been separated into two chapters: trace metals, and chlorinated and petroleum hydrocarbons. Within these chapters are descriptions of intercalibration and standardization programs presently underway and planned. Also included are discussions of research on transfer processes to the marine environment (e.g., atmospheric, river, sewage and estuarine transfer) and transfer processes within the marine environment (e.g., physical and chemical transfer in the water column and sediments, and biological transfer) for trace metals, and chlorinated and petroleum hydrocarbons. For details of any of the research programs described in this report, the reader may contact the appropriate principal investigator listed in Appendix 2. Scientific publications resulting from research in this program are given in Appendix 3.

Recommendations

The following general and specific recommendations are not meant to describe or outline a complete program for studying pollutant transfer to and within the marine environment. This was accomplished at the Durham (New Hampshire) Conference (NAS, 1971), and much of the program outlined at that time is underway in the NSF/IDOE Pollutant Transfer Program as presently constituted. The recommendations below were made by the participants at the Port Aransas Workshop in identifying areas, either missing from or requiring more intensive study in the present program, where additional emphasis might be placed in the future.

General Recommendations

1. Establish an Executive Committee for the Pollutant Transfer Program composed of several participants in the program as well as several outside experts. The Committee would have primary responsibility for developing the long-range objectives of the Program, coordinating and channeling the various individual programs toward the overall goals, and identifying areas of weakness in the Program. It also would coordinate the Pollutant Transfer Program with other major components of the IDOE Environmental Quality Program and would be responsible for initiating intercalibration and standardization programs as needed—a critical part of this and any other study of chemical substances in the environment. Appendix 1 gives further details.

2. Develop and apply methods for determining the chemical speciation and stability of the various pollutants during their transfer to and within the marine environment. This critical gap in present knowledge must be filled before we can adequately determine the mechanisms of biological and physical transfer processes.

3. Design field programs to determine numerical fluxes of the various pollutants by the different pathways to (e.g., atmosphere, rivers, etc.) and within (e.g., sedimentation, biological uptake, etc.) the marine environment. For those substances which occur naturally as well as a result of man's activities, methods for distinguishing between natural and pollutant fluxes should be developed.

4. Formulate predictive models, wherever possible, for determining the rate of pollutant transfer to and within the various reservoirs of the marine environment. While some of these models will be quite specific for particular pollutants and/or transfer paths, more generalized models will be important in predicting transfer processes for pollutants which may be of concern in the future.

5. Consider the ultimate fate of pollutants in the marine environment. Such considerations should include not only the final resting place of the non-degradable pollutants, but details of the degradation of organic pollutants through various types of chemical reactions. Much more information is needed on the identification of metabolic derivatives, degradative pathways, and partitioning between the various physical phases in the sea.

Specific Recommendations

1. Develop methods to distinguish more precisely between the biogenic and petroleum hydrocarbons in the marine environment and to determine which compounds are common to both sources. In order to accomplish this, continuing research is essential on the biogenesis of hydrocarbons by marine organisms, and the transfer of biogenic hydrocarbons through food webs, their metabolism and their degradation.

2. Continue investigations of food-web magnification of organic and metal pollutants in order to resolve present uncertainties about this controversial subject. These investigations should include both field studies involving known food chains and closely controlled laboratory studies.

3. Investigate potentially damaging organic pollutants such as low-molecular-weight hydrocarbons, low-molecular-weight chlorinated hydrocarbons, hexachlorobenzene, and heterocyclic petroleum components. These are members of a large group of synthetic organic substances which are presently receiving little or no attention in terms of their potential effect on the marine environment.

4. Determine the contribution of point sources of pollutants, particularly ocean dumping, and assess their significance.

5. Investigate the distribution of atmospheric pollutants, including organic and inorganic substances, between the vapor phase and particles in the marine atmosphere. Knowledge of gas-particle reactions is also important in understanding the atmospheric transport of many of these substances to the marine environment.

6. Determine the association of trace metals with organic substances and/or mineral matter in particles in the sea. More information on the physical transfer of metals, especially in the particulate phase, and more accurate residence times should be obtained.

7. Measure the concentrations of metals, and of chlorinated and petroleum hydrocarbons in the appropriate organs and tissues of marine organisms. This will provide valuable information on transport paths and mechanisms within organisms.

8. Evaluate the transfer of all pollutants to sediments and remobilization of these pollutants into the marine environment after post-deposition transformation. Largely ignored in the present program, transfer processes across the water-sediment boundary in both directions must be elucidated.

Program Accomplishments: Trace Metals

The identification of heavy metal pollution in the marine environment is difficult. Natural input may equal or exceed man's, but lacking baseline data prior to the Industrial Revolution, it is difficult to assess man's impact. Even if the concentration of a metal is not presently elevated due to anthropogenic sources, the continued studies are critical in predicting the impact of man's future input as technology evolves.

In contrast to synthetic contaminants, such as chlorinated hydrocarbons, where studies of the variations in concentration levels are important, heavy metal pollution of the marine environment may best be understood by the study of transfer processes. For example, metal pollution has definitely been identified in the coastal regions of oceans and seas throughout the world. In some cases such pollution has led to financial loss to shellfish and other commercial fisheries, and, in isolated cases, it has been hazardous to human health. A greater understanding of the long-range impact of these ever increasing instances on the total marine ecosystem requires that we understand metal transfer processes in the marine environment. Problems related to lead pollution in the open ocean also remain unresolved. The reason for elevated levels of mercury in open ocean fish, such as tuna and swordfish, continues to be a subject of controversy. The solution to these and related problems will come only through an understanding of heavy metal transfer processes.

Intercalibration and Standardization

Intercalibration studies are a critical part of any environmental quality program; results obtained by the different laboratories must be compared with some confidence. An initial trace-metal intercalibration program was part of the IDOE Baseline Studies Program, when standardized substances such as NBS orchard leaves, tuna meal, and bovine liver were analyzed by the participating laboratories. This initial study pointed out many weaknesses and errors in early analytical procedures of the participating laboratories, and significant improvement in reliability resulted. For the variety of sample matrices being analyzed in the Pollutant Transfer Program (i.e., organisms, sediments, seawater, atmospheric particles, etc.), one or two types of intercalibration samples generally would not satisfy all of the Program's aspects. Thus, a continuing plan of intercalibration and standardization, designed to meet the particular requirements of various components of the Pollutant Transfer Program, is a very important part of further Program development.

For 18 months all laboratories participated in a Lead in Seawater Workshop as a first step in the quality control of data coming from heavy metal transfer studies (Patterson, 1974). A crucial requirement in the distribution of samples of seawater containing standardized concentrations of lead at very low levels is that the shipping containers not contaminate the sample. A reliable method for cleaning FEP teflon shipping containers was developed by the group from the California Institute of Technology for this study. The cleaning operations, carried out in a low-contamination-level laboratory, used successive treatments with hot concentrated nitric acid for periods totaling six to ten days, followed by soaking in ultra-pure, acidified water for another three to five days. All handling was with stainless steel tongs and talc-free gloves. If the seawater samples were adjusted to pH one with a small amount of ultra-pure hydrochloric acid before they were shipped to the participant analysts, problems associated with the partitioning of lead between dissolved and particulate phases and with adsorption on container walls were largely eliminated. Four samples in which lead concentrations were standardized by isotope-dilution analytical techniques were circulated among workshop participants over a six-month period. Participants used atomic absorption and anodic stripping voltammetry techniques to analyze samples. Results obtained by these methods did not correspond to isotope dilution-mass spectrometry analysis; the principal source of error appeared to be lead contamination during analysis in the atomic absorption and anodic stripping laboratories.

The analytical results obtained in this program were evaluated by all the participants at a meeting held at the California Institute of Technology in the fall of 1973. The participants agreed upon modifications of their procedures to significantly improve the accuracy of their lead analyses. They also agreed that the Workshop would be continued until some researchers had demonstrated an ability to determine lead in seawater reliably, using atomic absorption and/or anodic stripping voltammetry techniques. The lead intercalibration program will continue, and other trace metals also will be similarly evaluated in the future.

Transfer Processes to the Marine Environment

Atmospheric Transfer

The NSF/IDOE Durham Workshop on Marine Environmental Quality (NAS, 1971) identified the atmosphere as a potentially significant, but largely

unstudied, path for the transfer of both metal and organic pollutants to open ocean areas. As part of the Pollutant Transfer Program, a study was initiated at the University of Rhode Island to investigate the atmospheric transport of these substances to open ocean areas over the North Atlantic and their deposition on the ocean surface.

A critical part of the study of atmospheric trace substances in remote areas is developing the capability to collect samples free of local contamination, and a sophisticated procedure for contamination-free atmospheric sampling was developed by the Rhode Island group (Duce et al., 1974). The primary atmospheric sampling site for this study is a 20-meter-high walk-up tower located on the southwest coast of Bermuda. Collection of samples at the tower is controlled automatically by wind direction and by the number of condensation nuclei (which may be indicative of local pollution) in the air to avoid the collection of contamination produced on the island. Sample collection occurs only when the wind is off the water and condensation nuclei counts are near background for marine air, 2 to 300 per cubic centimeter. Under all other conditions the collection equipment automatically shuts down and protective doors close over the collectors to prevent local contamination.

Additional atmospheric samples have been collected by the University of Rhode Island research vessel *Trident* on twelve cruises covering wide areas of the North Atlantic (figure 1). Atmospheric sampling from R/V *Trident* takes place at the top of an eight-meter-high A-frame tower installed on the bow. Wind directional control of the sampling, similar to that in Bermuda, is used on the ship tower. In a joint program with the University of Maryland, a fully automated atmospheric sampling station has also been set up at the geographic South Pole (Zoller et al., 1974).

Results of atmospheric trace metal studies to date suggest that, except for the seawater-derived alkali-alkaline earth metals (Hoffman, Hoffman, and Duce, 1974), most of the atmospheric trace metals measured over the open ocean and in Antarctica are from normal crustal weathering. However, several trace metals are found in concentrations far in excess of that predicted from a crustal source. An enrichment factor (EF) for each element in the atmosphere relative to the crust may be calculated using aluminum, which is present in high concentrations in the crust, as a reference element:

$$EF = (X/Al)_{atm}/(X/Al)_{crust} \qquad (1)$$

If EF is close to unity for any element, X, a crustal weathering source is indicated for that element. Geometric mean values of the calculated enrichment

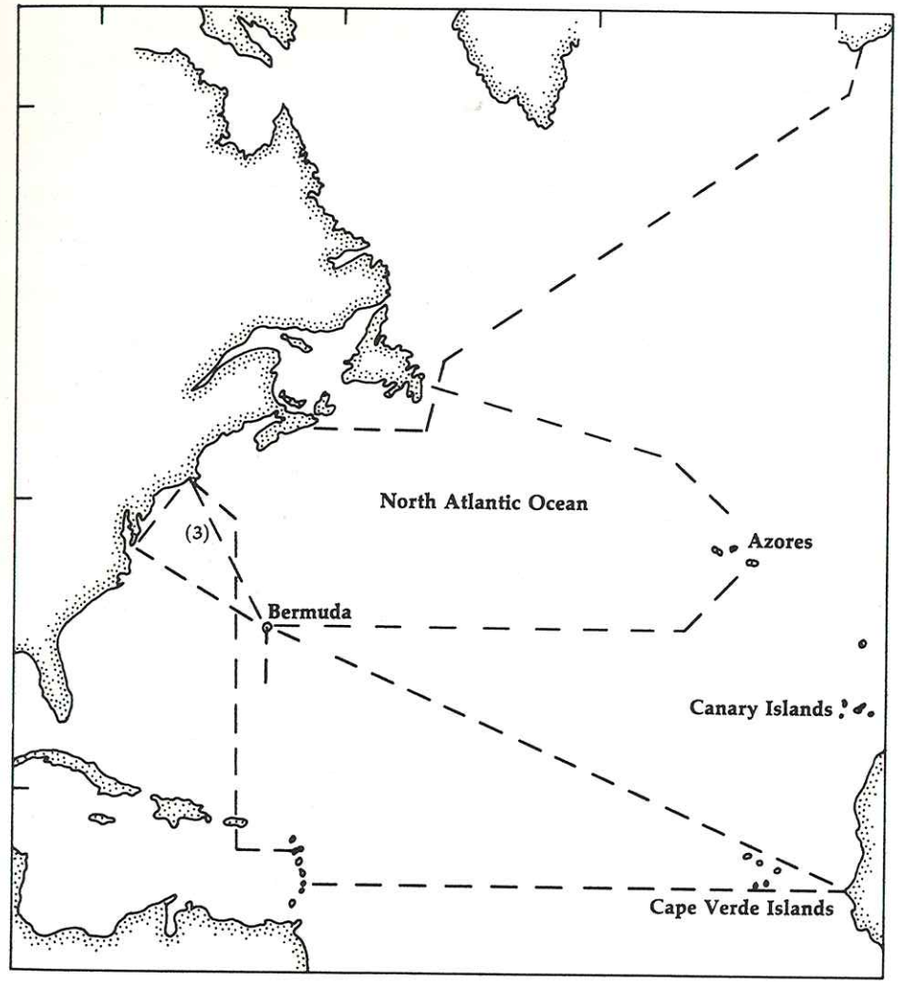

Figure 1. Atmospheric sampling areas covered by R/V *Trident* on 12 cruises from May, 1970, to October, 1973.

factors for samples collected from the westerlies (north of 30°N) over the North Atlantic and from Antarctica are presented in table 1. The EF values suggest that most of these trace elements, i.e. scandium, thorium, samarium, manganese, europium, iron, lanthanum, cobalt, and of course, aluminum, are from normal crustal erosion. The high vanadium enrichment found in the

Table 1. Enrichment of heavy metals in atmospheric particles relative to the earth's crust.

Element	Enrichment Factor (EF)	
	South Pole	*North Atlantic Westerlies*
Scandium	0.8	1.1
Thorium	0.9	—
Aluminum	1.0	1.0
Samarium	1.3	—
Manganese	1.4	1.9
Europium	1.9	—
Iron	2.1	1.1
Lanthanum	2.2	—
Cobalt	4.7	3.5
Vanadium	1.4	13
Zinc	69	40
Copper	93	84
Cadmium	—	300
Antimony	1300	3000
Lead	2500	2300
Selenium	18000	16000

After Zoller et al. (1974) and Duce et al. (1974).

North Atlantic, but not in Antarctica, is likely due to vanadium produced by the burning of heavy fuel oil, containing high concentrations of vanadium porphyrin complexes, along the east coast of North America. However, the elements zinc, copper, antimony, lead, and selenium (as well as cadmium over the North Atlantic) are significantly enriched in the atmosphere. The enrichment factors for these elements are also quite similar in both locations. Due to the short residence time of aerosols (a few weeks at most), the long tropospheric mixing times between the northern and southern hemispheres (approximately one year), and the fact that most of the anthropogenic sources for these elements are in the northern hemisphere, it is concluded that many of the elements which have high enrichment factors are likely of natural rather than anthropogenic origin. It should also be noted that, with the possible exception of copper, these enriched elements are relatively volatile in their inorganic forms compared to the other metals and are among the elements most likely to form stable carbon-metal bonds. Possible natural sources for the high atmospheric concentrations of some of these elements might be some high temperature process such as volcanism, or biological mobilization of the elements by processes such as methylation.

It should be pointed out that the conclusions above may not apply to lead, and the close agreement of the lead enrichment values at the two locations may be coincidental. Recent measurements using the improved collection techniques at the Bermuda tower suggest the background atmospheric lead concentration over marine areas may be significantly lower than previously measured by the Rhode Island group and others due to contamination during sample collection and analysis. Atmospheric lead concentrations of less than 0.1 nanogram per cubic meter have been measured recently at this location (Duce and Hoffman, unpublished data).

An important part of the atmospheric transport program at Rhode Island is the evaluation of the chemical composition and concentration of particulate matter in the oceanic surface microlayer relative to the concentrations in atmospheric particulate matter (Duce et al., 1972). The top 100 to 300 micrometers of the ocean surface are sampled using the screen technique of Garrett (1965). All particulate heavy metals investigated in the surface microlayer to date (lead, aluminum, iron, vanadium, copper, cadmium, chromium, and manganese) are significantly enriched compared to water approximately 20 to 30 centimeters below the surface. Typical surface microlayer and subsurface concentrations of these particulate trace metals found on a *Trident* cruise from Dakar to Bermuda are shown in figure 2. It appears, however, that only in regions with extremely high atmospheric particle loadings, e.g., the Sahara dust plume in the northeast trades off West Africa, does the direct deposition of atmospheric particles affect the surface microlayer particulate enrichment for most trace metals (Hoffman et al., 1974). Other areas, e.g., between Bermuda and the Azores, have rather similar enrichments in the surface microlayer, but atmospheric particle loadings are several orders of magnitude less. It also appears that the enrichments of many particulate trace metals found in the surface microlayer may be derived from a bubble flotation process (Wallace and Duce, 1974) in areas of the ocean which are not dominated by large fluxes of atmospheric continental material. The Rhode Island group has also calculated that the lifetime of directly deposited atmospheric dust in the oceanic surface microlayer is relatively short (i.e., a few seconds at most) under winds of five to ten meters per second (Hoffman et al., 1974). In an associated study, a group at the University of Connecticut has found no enrichment of total mercury or of any dissolved mercury species in the oceanic surface microlayer. Particulate mercury alone was not determined (Fitzgerald and Hunt, 1974).

As part of a comprehensive study of lead in the marine environment at the California Institute of Technology, atmospheric particle input has been

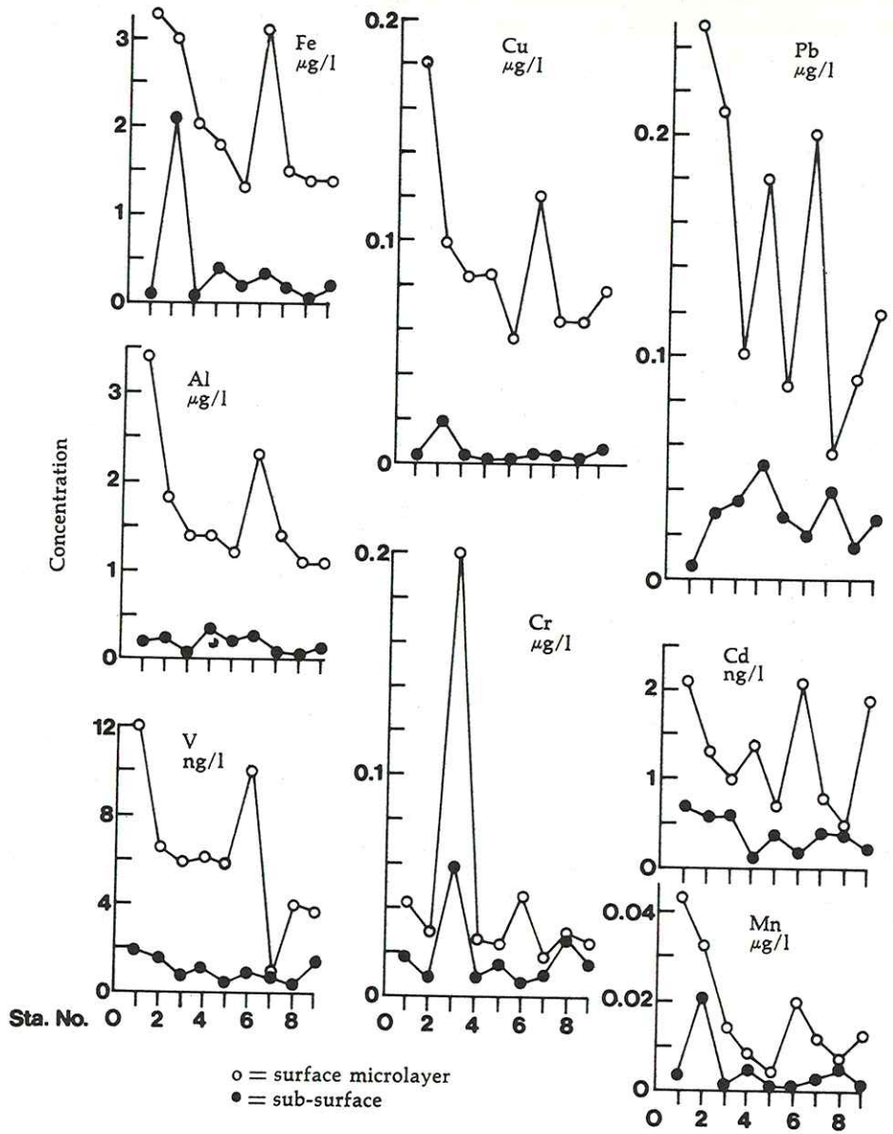

Figure 2. Oceanic particulate trace metal concentrations between Dakar (left) and Bermuda (right). Data obtained from a cruise of R/V *Trident* (February, 1972). Open circles represent surface microlayer concentrations; filled circles represent subsurface (20 to 30 centimeters) concentrations (Hoffman and Duce, unpublished data).

found to be a major source of industrial lead to the Southern California Bight, comparable in magnitude to the input of lead from storm runoff, rain, and sewage reported by other investigators (Patterson and Settle, 1974). The atmospheric lead originates from automobiles burning leaded gasoline. A mass balance of lead to the Southern California Bight is shown in table 2.

The California Institute of Technology group has also found that the isotopic composition of atmospheric lead may be important in determining the source area for this lead. They found significant differences in the $^{206}Pb/^{207}Pb$ ratio in atmospheric samples collected in the southern California basin, northern California basin, and in marine air off the southern California coast.

Riverine, Sewage, and Estuarine Transfer

The major routes for pollutant transfer to coastal areas are rivers and sewage and industrial outfalls. These subject estuarine ecosystems to the most severe anthropogenic stresses in the marine environment.

At the Skidaway Institute of Oceanography researchers are studying the fluxes of iron, manganese, copper, cadmium, and mercury through the salt marsh estuaries of the southeastern Atlantic coast to the adjacent continental shelf (Windom, 1974). The flux of heavy metals through salt marsh estuaries is controlled by processes which occur at river-estuary and salt marsh-sediment boundaries. These include adsorption-desorption reactions, flocculation, precipitation, and sedimentation. Along with estuarine circulation, these processes effectively control the residence time of metals in estuaries. Some biological processes, such as uptake by marsh vegetation, may serve as efficient mechanisms to recycle metals otherwise lost to estuarine sediments.

Assuming the southeastern salt marsh estuarine environment is in steady-state, where input of metals is equal to loss, the flux of metals can be expressed by the following equation:

$$K_i = K_d + K_p = K_s + K_f \qquad (2)$$

where K_i is the rate of total metal input by rivers; K_d and K_p are the rates of dissolved and particulate metal input by rivers, respectively; K_s is the rate of metal loss in salt marsh sediments, and K_f is the net flux of metals through the estuarine system. The total input of metals to the system was determined by sampling the major rivers, and the sedimentation loss was calculated by determining the rate of accumulation of sediments in salt marshes and metal concentration in the sediments. Using (2), the net flux of metals through the

Table 2. Mass balance of lead to the Southern California Bight.

Input	Tons Pb/yr/12000 km²	Reference
Dry aerosol deposition	290	Patterson, unpublished data
Rain	20	Patterson, unpublished data
Waste water	200	SCCWRP (1972)
Storm runoff and dry flow	170	SCCWRP (1972)
Industrial river dissolved lead	20	Chow and Patterson (1962)
Prehistoric natural river clay lead	40	Chow and Patterson (1962)
Prehistoric natural river dissolved lead	1	Chow and Patterson (1962)
Ambient seawater lead entering Bight by advection	700	Patterson, unpublished data
Output		
Prehistoric natural sediment clay lead	30	Bruland et al. (1974)
Industrial sediment lead	160	Bruland et al. (1974)
Ambient seawater lead leaving Bight by advection	700	Patterson, unpublished data
Addition of pollutant lead to ambient seawater lead leaving Bight	540	Patterson, unpublished data
Addition of natural river clay lead to ambient seawater lead leaving Bight	10	Chow and Patterson (1962)

system was calculated, as shown in table 3. Processes occurring in the estuarine environment influence the amount, and possibly the form, of metals entering continental shelf waters. Upon entry into estuaries in river runoff, a large portion of the dissolved iron and manganese precipitates and accumulates in sediments along with particulate fractions of these metals. Most cadmium and copper in solution are transported through salt marsh estuaries while the particulate fractions are apparently lost to the sediments. All dissolved mercury and, apparently, a desorbable fraction of that in particulate matter are transported through the salt marsh system. Uptake by salt marsh vegetation is a relatively important transfer mechanism for mercury, as indicated in table 4 (Rahn, 1973).

A project group at Oregon State University is investigating the particulate-dissolved distribution of riverborne metals and changes in this distribution which occur when river water mixes with seawater. Studies in the Columbia River system at its peak discharge indicate that the soluble, extractable copper, zinc, and manganese are not conservative during mixing with seawater (Cutshall, unpublished data). A linear mixing model predicts that soluble metal concentrations should fall on a straight line drawn between the mean metal concentration in zero-salinity river water and that in seawater

Table 3. Budget for the annual flux of metals through southeastern Atlantic salt marsh estuaries.

	Iron (10^6Kg)	Manganese (10^3Kg)	Cadmium (10^3Kg)	Copper (10^3Kg)	Mercury (10^3Kg)
Total Input (K_i)	62	1870	52	304	3.6
Sedimentation Loss (K_s)	210	1200	9	66	0.4
Sedimentation Loss (%)	> 100	64	17	22	11
Net Flux through Estuary (K_f)	0	670	43	238	3.2

Total discharge is approximately 6 × 10^{13} liters annually from the major southeastern Atlantic rivers. Salt marsh area is approximately 10^6 acres. After Windom (1974).

Table 4. Average concentration in and annual uptake of metals by *Spartina alterniflora* (leaves and stalks).

	Iron	Manganese	Cadmium	Copper	Mercury
	μg/g dry weight				
Mean	750	50	0.5	3.7	0.20
Maximum	2100	300	1.9	7.8	0.37
Minimum	100	17	0.1	0.5	0.07
Average Annual Uptake (10^3Kg)	2.1 × 10^3	140	1.4	10	0.6
Percent of Total Input	3	7	3	3	17

Annual production of Spartina alterniflora *is approximately 700 g/m². After Windom (1974).*

(salinity greater than 30 parts per thousand). However, these three metals show a positive deviation from this line, with a maximum deviation at approximately seven percent salinity. These results are in agreement with those of Evans and Cutshall (1973) for ^{54}Mn and ^{65}Zn in the Columbia River.

The group at the California Institute of Technology is beginning an extensive investigation of the input of lead to the Southern California Bight from storm runoff and sewage outfalls. They have found that less than one percent of the waste effluent lead is in dissolved form while the rest is particulate; these findings are significantly different from previous investigations. The isotopic composition of the dissolved lead was also considerably different from particulate lead, suggesting different sources for these forms of this metal. Extensive sampling of dissolved and particulate lead in the ocean in the vicinity of sewage outfalls as well as in rain and storm runoff is presently underway (Patterson, unpublished data).

Transfer Processes within the Marine Environment

Physical and Chemical Transfer

Once heavy metal pollutants have reached the marine environment, it is critical to understand their physical distribution and dispersion in the water column and their accumulation at various interfaces. Knowledge of the chemical speciation of metals and the reactions they undergo as they migrate through the marine environment is also needed.

As mentioned previously, it appears that particulate trace metals may be enriched in the sea surface microlayer by the transport to that region on the surface of bubbles. A program is underway at the University of Rhode Island to evaluate the flux of particulate organic material and trace metals to the sea surface by bubbles. Laboratory studies using fresh Narragansett Bay water have shown that copper, zinc, nickel, lead, iron, manganese, chromium, cadmium, and vanadium present on particles in the water column are transported to the water surface in significant quantities by bubbles. Similar shipboard studies are underway in the North Atlantic. Results to date suggest that in most open ocean areas, the upward flux of many of these trace metals to the sea surface on particles associated with bubbles is at least of the same order of magnitude and perhaps considerably higher than the downward flux of the same metals to the sea surface from the atmosphere (Wallace and Duce, 1974). This is in agreement with the suggestions made previously (Hoffman et al., 1974, see page 9).

The Rhode Island group has also developed a profiling system for the top meter, incorporating temperature and pH sensors, which is capable of depth resolution of $\pm$ 2 centimeters. Using this system along with the screen technique for surface microlayer sampling, both reactive and organic forms of phosphorus were found to be enriched in the oceanic surface microlayer compared to their concentration in subsurface water. Using the top meter profiler, a phosphorus concentration gradient was noted down to depths ranging from 15 to 40 centimeters below the surface. In the microlayer reactive phosphorus showed the greater enrichment in open ocean, low nutrient areas, while organic phosphorus was relatively more enriched in Narragansett Bay (Kester and Pilson, unpublished data).

The California Institute of Technology group has been studying lead speciation in seawater as part of their pollutant lead transport program. While they have found variable amounts of dissolved and particulate lead in the open ocean, they believe that the majority of the particulate lead was not incorporated within biological or inorganic solids in the Southern California Bight area, but was adsorbed on the surface of the particles (Patterson, unpublished data). This will be discussed in more detail in a later section.

The isotopic composition of total lead in seawater was also measured for samples collected in several areas of the Southern California Bight and was found to vary considerably with collection location (table 5). In the absence of appreciable amounts of industrial lead, ambient seawater lead within the Southern California Bight should be isotopically homogeneous. The demonstrated existence of a heterogeneity in the leads of the Southern California Bight indicate the existence of different mixtures of industrial pollutant lead with open ocean lead. The variety of leads of different isotopic compositions which was observed in these and other samples suggests that it is feasible to determine the extent and source of industrial lead pollution in this area. The isotopic compositions of leads older than 1910 in sediments of the Southern California Bight showed a $^{206}Pb/^{207}Pb$ ratio of about 1.21. After this date the ratio changed to smaller values ranging from 1.176 to 1.152. The sediments were dated with ^{210}Pb. The youngest sediment in this investigation was dated at 1965 and its isotopic composition was similar to that of industrial lead in Los Angeles Basin aerosols determined in 1964. At the present time, however, the isotopic composition of industrial lead in aerosols, snow, rain and storm runoff averages about 1.19. This recent change has likely occurred as a result of the incorporation of a greater proportion of lead from Missouri, which has a $^{206}Pb/^{207}Pb$ ratio of approximately 1.6, into the pool of less radiogenic lead previously used to manufacture lead alkyls (Patterson, unpublished data).

The Connecticut group is investigating the distribution of total, inorganic, and organic mercury in coastal and adjacent river waters of Long Island Sound as well as in open ocean areas of the North Atlantic. (Organic mercury is defined as the difference between the measured concentrations of total and inorganic mercury.) They have found that as much as 50 to 60 percent of the mercury present in the coastal and river waters may exist either as organic compounds or in association with organic matter (Fitzgerald and Lyons, 1973). Total mercury in open ocean waters was less than ten nanograms per liter, a factor of five to eight less than that for coastal waters.

Biological Transfer

There are several investigations in the Pollutant Transfer Program which are directed largely toward determining the concentrations, distribution

Table 5. Isotopic composition of total seawater lead.

Type of Sample	$^{206}Pb/^{207}Pb$
Open north Pacific, Spring 1971	1.176 ± 0.002
La Jolla, California coast, Nov. 1, 1972 (taken during strong upwelling)	1.195 ± 0.002
La Jolla, California coast, March 31, 1973	1.188 ± 0.002
La Jolla, California coast, April 10, 1973	1.188 ± 0.002
Los Angeles, California coast, April 9, 1973	1.194 ± 0.002

Patterson, unpublished data.

and transfer of heavy metals such as lead, mercury, cadmium, copper, and zinc in various marine biological communities. The Skidaway group has been investigating the uptake of mercury by *Spartina alterniflora*, and has found that this plant very efficiently takes up mercury through its roots. This uptake is followed by transfer of the mercury to the leaves, where it is subsequently released to estuarine waters. The root system apparently concentrates inorganic mercury, while the leaves concentrate methylmercury. With the exception of mercury, no correlation was found between the heavy metal (iron, manganese, cadmium, copper) concentrations found in *Spartina* tissue and concentrations measured in the marsh sediment, river, or estuarine waters of seven major river systems of the southeastern coast of the United States. However, the ranges of metal concentrations in *Spartina* tissue are narrow in comparison with the ranges measured in the sediment and water; it appears that either metal concentrations in the plants from the southeastern marshes are "saturated" or that the concentration of metals in the plant are controlled by the plant. Thus, present evidence suggests that the changes in heavy metal concentrations which might be caused by dredging and by increased effluents in rivers would not seem to be critical to *Spartina*. The mean ratio of metal in the plant to metal in the marsh sediment ranges from less than one for iron to approximately one for most of the other metals (Dunstan and Windom, 1974).

The Skidaway group has also identified several important food chains of the southeastern innercontinental food web (Stickney et al., 1974) and is investigating the transfer of metals in this food web. The natural variations in the concentration of cadmium, copper, lead, mercury, and zinc in members of these food chains have been determined as a basis for laboratory transfer studies. Many metal variations observed between species appear to be due to trophic level relationships. Initial studies regarding the form of metals in the different organisms have been limited to methylmercury. These data, while

not yet fully synthesized, generally indicate that a relatively large quantity (about 70 percent) of the mercury in vertebrate muscle occurs in the methyl form. The percentage of methylmercury (compared to total mercury) is less in invertebrate omnivores, which depend to a great extent on primary producers as a food source (table 6). This suggests that the form of metal in the diet may be important in determining the forms subsequently found in organisms. (Only a small portion, less than 20 percent, of the mercury in *Spartina* leaves and stems is methylated.) Other possibilities are that higher trophic level organisms may assimilate more methylmercury from the water through their gill systems or may metabolically convert mercury to the methyl form. In contrast to the muscle tissues of carnivorous fish, samples of liver and spleen from sharks contained negligible quantities of methyl relative to total mercury, which was present in high concentrations ranging from five to eight parts per million. More information is needed to determine how the modes of uptake, metabolism, storage and excretion of heavy metals influence their transport in marine ecosystems (Windom, unpublished data).

An understanding of the interaction of industrial pollutant lead with marine food chains is critically dependent upon our knowledge of the forms in which lead occurs in seawater. A major advance in this area has been the discovery by the California Institute of Technology group that much of the lead in seawater may be adsorbed on the mucilage of algae. In the previous studies of the occurrence of heavy metals in plankton, investigators have ignored the distinction between metabolized metals contained within the organisms and adsorbed metals on their outside surfaces. The above finding, together with the discovery that excessive amounts of lead collect on the epidermal mucus of fish (Chow, Patterson, and Settle, 1974), shows that the biologically active fraction of the total lead in marine organisms may be quite small, especially at the lowest trophic levels of the food chain. The profuse occurrence of biologically inactive adsorbed lead had almost completely obscured our understanding of the distribution of biologically active lead in the marine food chain.

In tank studies of a number of adult sculpin *(Scopaena guttata)* which were subjected to lead acetate concentrations about 60,000 times greater than ambient seawater levels, lead was found to accumulate continuously over a period of three months in the mucosal covering of the epidermis. This suggests that the residence time of lead adsorbed on skin mucus is longer than a few weeks. Analyses of muscle reveal that lead does not increase with time when fish are exposed to these lead concentrations over a period of three months (figure 3). However, analyses of the fish kidneys indicate the fish are me-

Table 6. Average percent of methylmercury in muscles of estuarine organisms.

Types of Organisms	Number of Animals Examined	Percent of Mercury in Methyl Form		
		Average	*Maximum*	*Minimum*
Carnivorous fish	61	73	> 100	10
Crabs	5	52	> 100	28
Shrimp	22	26	82	< 4
Clams	3	14	25	< 6

Data from Gardner et al. (1974).

tabolizing the increased amounts of lead they were exposed to during the three-month period. Some of this metabolized lead is accumulating in the bone, as revealed by analyses of this material. These data suggest that both kidney tissue and epidermal mucus could serve as indicators of different levels of lead exposure (Patterson, unpublished data).

A group at the University of Alaska is just beginning an investigation of the budget of heavy metals within coastal areas dominated by seagrass communities, e.g., *Zostera marina* (eelgrass). The unique feature of the seagrass system is that the plants are theoretically capable of absorbing trace metals from the sediments or from the water column, i.e., metals that would be lost to the sediments in most systems can be returned to the food web by rooted plants. Very preliminary results suggest that the roots and rhizomes of both shallow (tide pool) and deep (sub-tidal) plants are more enriched with zinc and copper than are their leaves, while cadmium exhibits a reverse trend. Deep plant leaves, roots, and rhizomes contain less zinc and copper but more cadmium than do shallow plant parts.

Preliminary results also suggest that an oxidized microlayer exists around the roots of seagrasses and this, although not yet measured, may facilitate the incorporation of metals into the seagrass from the sediments that would otherwise be bound by the reducing conditions normally found there. In this way metals that would otherwise be lost in the sediments are returned to the food chain and the water column (McRoy and Burrell, unpublished data).

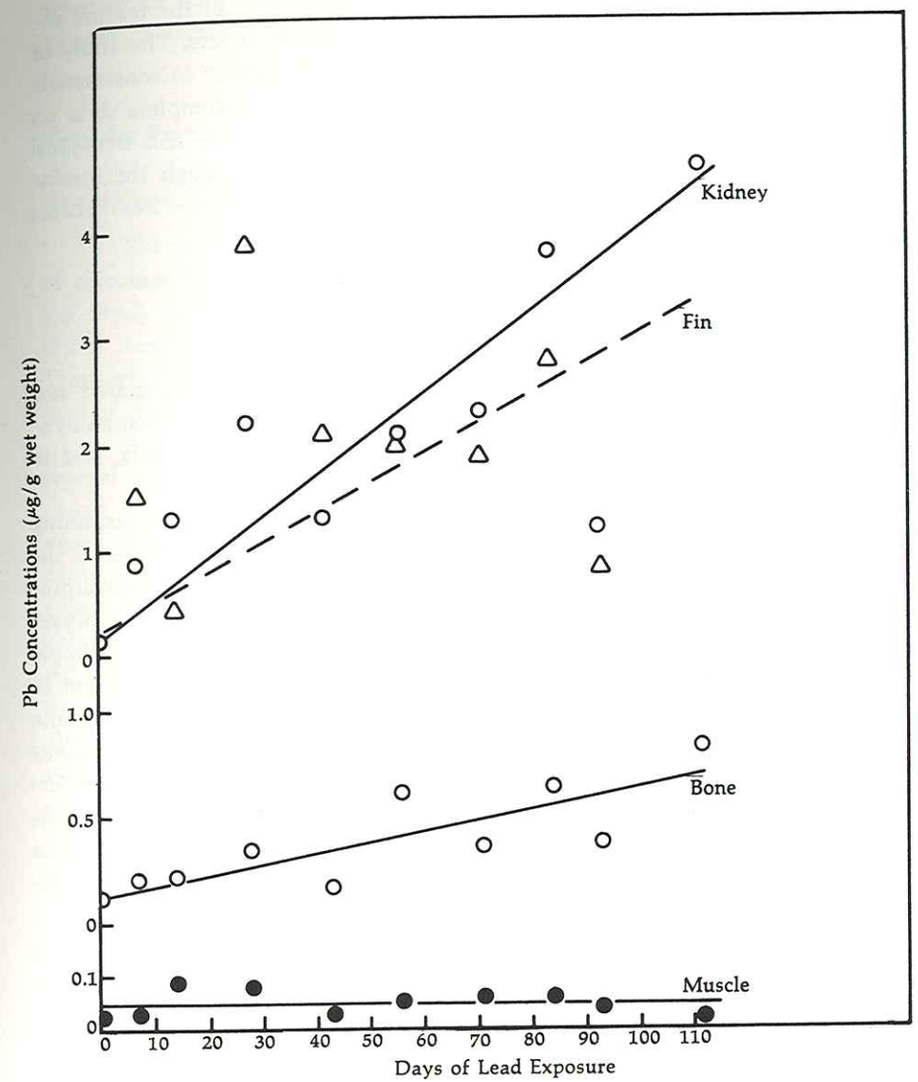

Figure 3. Analysis of sculpin tissue exposed to increased lead concentrations (Patterson, unpublished data).

Program Accomplishments: Chlorinated and Petroleum Hydrocarbons

Among the many organic compounds entering the oceans as a result of man's activities, the chlorinated and petroleum hydrocarbons are currently of greatest concern. This concern stems from the relatively large quantities of these substances added to the oceans, their persistence in the marine environment and their potential for detrimental biological effects. The study of these substances in the marine environment has been subject to considerable controversy during the past few years, in part due to incomplete data on which to base our understanding of the physical, chemical, and biological processes involved in the transport of these substances through the marine environment. Explaining many aspects of these transport paths has become a major goal of the IDOE Pollutant Transfer Program.

Intercalibration and Standardization

An important prerequisite to meaningful studies of chlorinated and petroleum hydrocarbons in the marine environment is to establish that analytical methods used by different investigators give comparable results, and to determine the magnitude of the error to be expected.

At present no adequate standardized methods exist for determining the levels of petroleum contamination in marine samples. Thus three of the laboratories involved in the IDOE Program have conducted an intercalibration exercise to determine the accuracy and precision of hydrocarbon analyses as a first step toward routine intercalibration. The intercalibration sample consisted of cod liver oil spiked with approximately 372 parts per million of a distillate cut (boiling range, from n-C_{16} to n-C_{28} alkanes) of a Louisiana crude oil. The mean concentration found by the three laboratories was 406 $\pm$ 26 parts per million (Farrington et al., 1973). While continued intercalibration of this type will be of great importance, determination and standardization of methods which will allow the analyst to distinguish between biogenic and petroleum hydrocarbons are perhaps even more critical to future programs.

Chlorinated hydrocarbon determinations were intercalibrated among IDOE participants and with the international monitoring program of the Organization for Economic Cooperation and Development (OECD), in which 26 laboratories of 12 countries participate (Holden, 1973). Initially, researchers obtained a sample of corn oil, spiked with several chlorinated hydrocarbon pesticides and used for intercalibration in the OECD program. Subsequently, a liver of a silky shark was homogenized, ground with sodium sulfate and

divided into aliquots for distribution among IDOE participants. Agreement among the laboratories was surprisingly good, and in general was comparable to the reproducibility obtained within one laboratory. The PCB values, reported on a lipid basis by six laboratories were, respectively, 13, 13, 6, 16, 13, and 13 parts per million (Harvey et al., 1974).

Transfer Processes to the Marine Environment

Atmospheric Transfer

The potential importance of the atmosphere as a route for the transfer of chlorinated and petroleum hydrocarbons to the ocean was emphasized by the Study on Critical Environmental Problems (SCEP, 1970) and the National Academy of Sciences (1971). Thus, several groups in the Pollutant Transfer Program are investigating various aspects of the atmospheric transport of these substances.

A group at the Scripps Institute of Oceanography is investigating several halogenated hydrocarbons of low molecular weight which are produced globally at levels of megatons per year and which eventually are nearly completely released to the environment. Included are the chlorofluorocarbons used primarily as aerosol propellants and the low-molecular-weight chlorinated aliphatics, such as trichloroethane, perchloroethylene and trichloroethylene, which are used as dry-cleaning fluids, solvents and degreasers. All are stable in the environment and have been detected both in air and natural waters. Levels in seawater are in the nanogram-per-liter range while atmospheric concentrations are approximately 10^{-9} parts per part of air by volume (Su and Goldberg, 1973). Compressed laboratory gases, terrestrial waters and organisms now contain measurable quantities of these materials. Residence times in the atmosphere appear to be on the order of a decade or more.

Trichlorofluoromethane, dichlorodifluoromethane, trichloroethylene, perchloroethylene, trichloroethane, carbon tetrachloride, chloroform and methyl iodide have all been detected in seawater, and it is believed that many of these substances are transported to the ocean via the atmosphere. The latter three also may be produced naturally.

Several programs are investigating the atmospheric transport of the heavier chlorinated hydrocarbons to marine areas. Previous studies have been concerned with the transport of these substances to marine areas on atmos-

pheric particles or aerosols. Harvey and Steinhauer (1974) at the Woods Hole Oceanographic Institution and Bidleman and Olney (1974) at the University of Rhode Island independently developed new collection techniques which allow them to determine both the particulate and gas phase concentrations of these substances. The concentration ranges they have observed for total (gas and particulate) PCBs, DDT and chlordane in the atmosphere at several locations over the North Atlantic are presented in table 7. There is a nearly exponential decrease in atmospheric concentrations of PCBs from near shore to open ocean regions. Both Bidleman and Olney (1974) and Harvey and Steinhauer (1974) found that more than 90 percent of these chlorinated hydrocarbons passed through glass fiber filters, which have an efficiency greater than or equal to 98 percent for particles with radii $\geqq$ 0.015 micrometers, suggesting that the bulk of these substances may be present in the vapor phase over the ocean. Bidleman and Olney (1974) also found that the PCBs in the atmospheric samples matched Aroclor 1242 or 1248 while those in water from the Sargasso Sea more closely resembled the heavier Aroclors 1254 or 1260. The total DDT levels found in Bermuda were approximately 100 times the particulate DDT values reported in Barbados by Risebrough et al. (1968) and Seba and Prospero (1971), supporting the concept of a predominantly vapor phase for this pesticide over the North Atlantic Ocean. In open ocean areas where both substances were measured, PCB concentrations were at least a factor of ten greater than those of DDT.

Bidleman and Olney (1974) have calculated a maximum atmospheric residence time of 40 days for PCBs over the Sargasso Sea area of the North Atlantic. This calculation assumed a uniform PCB distribution in the top 100 meters of the ocean and in the troposphere above the ocean, a PCB residence time in the oceanic mixed layer of four years, and the only source of oceanic PCBs to be the atmosphere. These authors point out that a considerably shorter atmospheric residence time would result if the atmospheric concentration decreases with height, if the PCB residence time in the oceanic mixed layer is less than four years (as may be suggested by recent seawater data, see below) or if PCB removal mechanisms, e.g., photochemical degradation, exist within the atmosphere. In any event, it appears that either the residence time of PCBs in the atmosphere is much shorter than the estimated DDT residence time of 3.3 years proposed by Woodwell et al. (1971), or there is significant PCB input into the North Atlantic water from sources other than atmospheric deposition.

A group at the Scripps Institute of Oceanography and the La Jolla Laboratory of the National Marine Fisheries Service are investigating atmos-

Table 7. Atmospheric chlorinated hydrocarbon concentrations over the North Atlantic.

Location	Concentration Range (ng/m³)			
	PCB	*DDT*	*Chlordane*	*Reference*
Bermuda (30°20'N, 64°40'W)	0.2-0.6	0.02-0.06	0.01	Bidleman and Olney (1974); Harvey and Steinhauer (1974)
Georges Banks (41°40'N, 67°30'W)	0.6-1.6	—	—	Harvey and Steinhauer (1974)
Grand Banks (45°16'N, 52°68'W)	0.05-0.16	—	—	Harvey and Steinhauer (1974)
Cruise, Bermuda-Rhode Island	0.7-1.6	0.03-0.10	—	Bidleman and Olney (1974)
Vineyard Sound	4-5	—	—	Harvey and Steinhauer (1974)
Near Narragansett Bay	2-6	—	—	Bidleman and Olney (1974)

pheric PCB transport in the southern California area. They have shown that transport of *particulate* chlorinated hydrocarbons is a meso-scale (characteristic distances of 100 kilometers) phenomenon rather than a global phenomenon, with the majority of the residues being deposited within a radius of 100 kilometers of the source area (McClure and LaGrange, unpublished data). This is in agreement with the Atlantic data described above. These authors also have shown that the source of aerosol PCB is located within the Los Angeles metropolitan area.

Studies at the University of California, Berkeley, give indirect evidence for the atmospheric transport of PCB and DDT compounds from their observed distribution in the biota of the more remote marine environment (Risebrough, unpublished data). Both types of compounds are universally present in Arctic wildlife, both terrestrial and marine, in areas where transport via other pathways is highly unlikely. The use of marine birds, principally fish-eating species such as cormorants and pelicans or surface feeders such as petrels and shearwaters, has permitted an assessment of the present contamination patterns in the Pacific. Highest DDT residues occur in the Southern California Bight, the result of input from a single point land source. Elsewhere, however, aerial dispersal and removal appear to account for the majority of the residues. In the vicinity of Amchitka in the Aleutians, PCB concentrations are relatively high in birds, and PCB exceeds DDT. The strongest winds are westerly from the vicinity of Japan, the most likely source of the PCB. South of the equator, residues of both DDT and PCB compounds drop by an order of magnitude in birds, a reflection of the lower environmental

input into the southern hemisphere. DDE/PCB ratios are approximately unity. PCB was detected in some, but not all, specimens of Antarctic marine birds, whereas DDT residues were present in all samples. It was concluded that the atmosphere was the most likely pathway to the Antarctic (Risebrough, unpublished data). A comparison of levels of organochlorine compounds in the vicinity of Bermuda, using the Bermuda petrel and white-tailed tropic bird as indicator species, has shown the area to be less contaminated than the California coastal current (Wingate, Mendola, and Risebrough, unpublished data).

The Rhode Island group is also investigating the atmospheric transport of petroleum hydrocarbons over the North Atlantic. Measurements from the Bermuda tower and R/V *Trident* indicate that approximately 95 percent of the heavier hydrocarbons (boiling range, n-C_{14} to n-C_{32}) in the vicinity of Bermuda are not retained on glass fiber filters, but are retained on polyurethane plugs behind the filters (Quinn and Wade, unpublished data). Based on gas chromatographic analysis, the major source of these hydrocarbons appears to be petroleum products. The hydrocarbons retained on the plug are operationally defined as "gaseous" and those retained on the filter as "particulate." The ranges of concentration found during 1973 in these two phases are presented in table 8.

The Rhode Island group also found a significant variation in the percentage of heavier hydrocarbons in the "gas" phase with distance from Rhode Island on a cruise between Bermuda and Rhode Island. Although the total concentration remained essentially constant, there was a regular progression from approximately 60 percent "gas"-40 percent "particulate" on the continent to approximately 95 percent "gas" at Bermuda. This change may be due in part to the shorter residence time of the particles relative to the gas and, perhaps, in part to a conversion of particulate to gaseous hydrocarbons by volatilization as the particles age and move away from the continent.

Riverine, Sewage, and Estuarine Transfer

Few systematic studies have been made of either the transfer of chlorinated or petroleum hydrocarbons to the ocean via rivers, sewage outfalls and other runoff sources, or of the transport of these materials in estuaries. This is an important gap in our present knowledge, since the principal ecological effects of these substances are thought to occur in estuarine and coastal regions.

Table 8. Atmospheric hydrocarbons, Bermuda (boiling range, n-C_{14} to n-C_{32}).

Date (1973)	"Gas" Concentration Range (ng/m^3)	"Particulate" Concentration (ng/m^3)	Number of Samples
May	220-340	< 35	4
June	60-600	< 43	5
July	460	< 20	1
August	210-270	< 18	2
October	120-260	< 16	2
November	130-160	< 14	2
December	60	< 21	1

Quinn and Wade, unpublished data.

Measurements of the chlorinated hydrocarbons entering the Southern California Bight from rivers and major municipal waste water treatment plants have been made since 1971 by the group at the University of California, Berkeley. Estimates of total input from these sources indicate that the surface runoff from rivers has contributed only a relatively small amount of PCB and DDT compounds to the total residue burden. The White's Point outfall of the Los Angeles County Sanitation Districts has accounted for the majority of the DDT residues in the Bight and has also been, with other waste water treatment plants, the major source of PCB. Input of DDT and PCB compounds into the Bight from sewage has been declining since 1971 (Young, Risebrough, and de Lappe, unpublished data).

Data obtained at the Scripps Institution of Oceanography and the La Jolla Laboratory of the National Marine Fisheries Service on preserved fishes and plankton also show that DDT in the California Current has for the past 25 years come chiefly from the Los Angeles sewer system into the Southern California Bight. A 1969 survey (McClure and LaGrange, unpublished data) showed that the highest concentration of DDT in plankton of the California Current occurs around Los Angeles; it diminishes in plankton found north, south, or west of Los Angeles. Myctophid fishes, resident in the waters around the White's Point sewer outfall, almost precisely reflected the continual output of a major DDT manufacturer who apparently dumped DDT into that sewer system from 1949 to 1970 (MacGregor, 1974, in press).

A survey of hydrocarbons in a few samples from sewage treatment plants has been made by the Woods Hole and Rhode Island groups (Farrington and Quinn, 1973a). Effluent from the Field's Point Sewage Plant, which serves the cities of North Providence, Providence, and some industries in

Cranston, Rhode Island, was found to contain from 2.4 to 16.2 milligrams per liter total hydrocarbons on the three days it was sampled. The lowest concentration detectable by the methods employed was 0.5 milligram per liter. Gas chromatographic analysis of the hydrocarbons indicates that the bulk have a petroleum source.

From the discharge rate and the concentrations of hydrocarbons in the effluent, it was estimated that the Field's Point plant discharged between 130 and 655 metric tons of hydrocarbons per year, if the concentrations found are representative of concentrations throughout the year. Approximately 30 billion liters of municipal wastes are discharged to the coastal waters of the United States each day (U.S. Dept. of Interior, 1969). If the concentrations of hydrocarbons in these effluents are similar to those of the Field's Point effluent, then 28,000 to 140,000 metric tons of petroleum hydrocarbons are discharged each year to United States coastal waters by wastewater effluents. Treatment plants servicing small towns and cities may have lower concentrations of petroleum hydrocarbons, however. For example, the West Warwick, Rhode Island, sewage plant effluent contained no detectable hydrocarbons. On the other hand, treatment plants servicing large cities and plants providing only primary treatment may have higher concentrations of petroleum hydrocarbons than those found at Field's Point.

Based on available data (U.S. Coast Guard, 1971) it is calculated that approximately 28,000 metric tons of oil were spilled into United States coastal waters during 1970. Thus, the estimated amount of petroleum discharged by wastewater effluents may be in the same range as the reported amount spilled in coastal areas. Other work by this group (Farrington and Quinn, 1973b) has indicated that sewage effluents were one of the major sources of petroleum hydrocarbon input to Narragansett Bay at the time of the study. It is clear that transport of petroleum hydrocarbons from waste oil disposal and other urban sources such as street runoff which pass through sewage systems is an active process and a significant source of petroleum hydrocarbons in some coastal areas.

Dumping

No studies have so far been carried out on the effects of dumping chlorinated organic wastes in the United States coastal environment. These wastes include the EDC tars, byproducts from the oxychlorination of ethylene in the manufacture of ethylene chloride, used in the production of vinyl chloride. Compounds present in the EDC tars include a variety of chlorinated hydrocarbons of low molecular weight (Jensen et al., 1970). Amounts discharged into United States coastal waters can be obtained from Environmental Protection Agency permit applications. Thus, the Shell Chemical Company is currently permitted to dump 1.7×10^6 kilograms per month of a low-molecular-weight chlorinated hydrocarbon mixture into the Gulf of Mexico (Environmental Protection Agency, permit application number 730D0084). Low-molecular-weight chlorinated hydrocarbons detected at stations in the North Atlantic, the Barents Sea and the North Sea are believed to derive in part from the dumping of waste products of vinyl chloride manufacture in the North Sea (Jensen et al., 1970). Also significant is the deliberate or accidental introduction of petroleum and petrochemical wastes into the open ocean as a result of shipping operations or of dumping to dispose of unwanted byproduct material. It has been estimated that approximately two million metric tons per year of petroleum wastes are introduced to the open ocean from tankers, particularly those not using the load-on-top technique, as well as from the bilges of tankers and other ships (Porricelli et al., 1971).

Investigators at the Bermuda Biological Station presently are studying the distribution and composition of pelagic tar lumps in the North Atlantic Ocean. Over 250 samples from near Bermuda, Bermuda beaches, and other parts of the North Atlantic, have been analyzed using gas chromatographic techniques. Although many distinctive patterns are found, nearly all show normal paraffin profiles typical of the weathered waxy residue from a paraffinic crude oil (figure 4). In some tar lumps, inclusions of almost pure paraffinic wax (n-C_{25} to n-C_{33}) have been found.

It has been inferred from these observations that the principal source of pelagic tar in the North Atlantic is the normal operation of tankers which do not use the load-on-top method. Anecdotal evidence seems to indicate a substantial increase in pelagic tar near Bermuda following the closing of the Suez Canal and the alteration of tanker routes to pass around South Africa (Butler, Morris, and Sass, 1973). Because of this, and the much smaller amount of tar found on the North American continental shelf, it may be inferred that much of the tar in the Sargasso Sea comes from tanker washings dumped off the west coast of Africa (Morris and Butler, 1973).

Transfer Processes within the Marine Environment

Physical and Chemical Transfer

Water Column. As with other pollutants, the transfer of chlorinated and petroleum hydrocarbons in the ocean depends upon their physical distri-

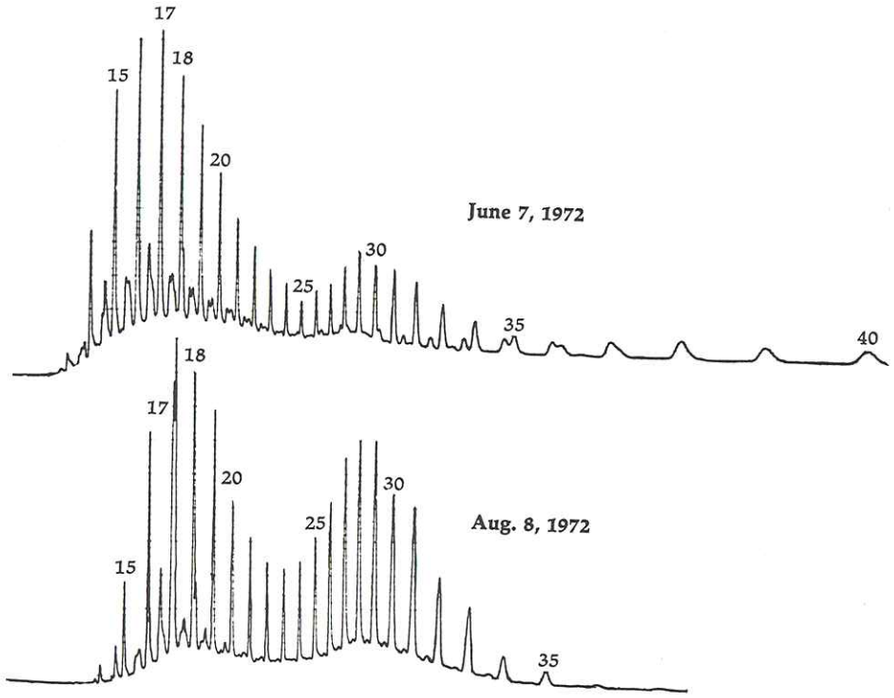

Figure 4. Gas chromatograms of two tar lumps from station S near Bermuda, showing the bimodal paraffin distribution characteristic of crude oil sludges. After Butler, Morris, and Sass (1973).

bution between dissolved and particulate phases, their concentration at phase interfaces, and their chemical speciation and reactivity. Unfortunately little was known until recently about these parameters for the chlorinated or petroleum hydrocarbons, and a significant fraction of our recent knowledge has come from the Pollutant Transfer Program. For example, only two years ago there were virtually no reliable data available on the PCB concentration of open ocean water. In the summer of 1972, a total of 51 surface and 200-meter water samples from the open North Atlantic were analyzed for PCBs by the Woods Hole group. The average concentration was 35 nanograms per liter at the surface and 10 nanograms per liter at 200 meters (Harvey et al., 1973). It was estimated that the mixed layer of the North Atlantic contained about 20,000 tons of PCB. During the summer of 1973, the average surface

water PCB concentrations in the Sargasso Sea were found to be 0.6 to 1.0 nanogram per liter (Harvey, unpublished data). In general, PCB depth profiles followed the main features of the water column and showed a slight inflection at 200 to 300 meters, the base of the thermocline. Also subsurface, high salinity layers generated in the Sargasso Sea showed minimum PCB concentrations. PCB was detectable (0.5 nanogram per liter $\pm$ 0.3) to at least 3000 meters. The 1/e concentration depth was about 100 meters in 1972 and 1973. Apparently 90 percent of the PCB was in the dissolved phase (Harvey, unpublished data).

Harvey (private communication) believes the apparent tenfold decrease in surface water concentration of PCB in the North Atlantic between 1972 and 1973 may have been due to the use and containment controls for PCB instituted in the United States in 1970-1971 (*Chemical and Engineering News*, 6 Dec. 1971, p. 15). This rapid rate of decrease of PCB in the entire mixed layer would require adsorptive scavenging by particulates with a fairly high settling rate, e.g., 100 to 300 meters per year, and would imply a very short PCB residence time in the mixed layer. Clearly much further work is needed in this critical area.

Determinations of chlorinated hydrocarbons in seawater along the California coast have shown that concentrations of PCB and DDT compounds are on the order of a nanogram per liter and occasionally lower, except in the vicinity of the White's Point sewage outfall. Aroclor 1242 components of the PCB have been present in many of the samples and the chlorinated hydrocarbon composition has been found to vary, depending on the station site (de Lappe and Risebrough, unpublished data).

The few studies which have been made of the composition of the top 100 to 300 micrometers of the open ocean surface (collected using the screen technique of Garrett (1965)) have indicated that, with the possible exception of pelagic tar lumps, this is not a major reservoir for either chlorinated or petroleum hydrocarbons, although elevated concentrations are present. Twelve samples from this surface microlayer in the Sargasso Sea area collected by the Rhode Island group contained approximately ten nanograms per liter of PCB, ten times the subsurface (20 to 30 centimeters) concentration, and approximately 0.5 nanogram per liter of DDT, at least twice the subsurface value (Bidleman and Olney, 1974). Similar enrichments for PCBs have been found in the surface microlayer of Narragansett Bay, Rhode Island, although the actual concentrations in both surface and sub-surface water are considerably higher (Duce et al., 1972). The mean concentration of heavier hydrocarbons (boiling range n-C_{14} to n-C_{32}) in 20 unfiltered surface

microlayer samples from the Sargasso Sea was approximately 150 micrograms per liter, generally about twice that in samples taken simultaneously at 20 to 30 centimeters depth (Quinn and Wade, unpublished data).

The quantities of pelagic tar (petroleum residue) in the Bermuda and Sargasso Sea area from 1969 through 1972 were reported by Morris and Butler (1973). Figure 5 shows the geographic area covered in this study and emphasizes the much higher amounts of pelagic tar in the Sargasso Sea than on the Scotian Shelf. Other surveys have suggested that the amount of tar found off the east coast of the United States is similar to the amount found on the Scotian Shelf (Attaway et al., 1974; Sherman et al., 1973; Sleeter et al., 1974). Figure 6 shows that, on the average, tar is about five percent of the wet weight of *Sargassum* and four times the wet weight of daytime surface zooplankton. Data taken over a two-year period from station S near Bermuda show that the amount of tar collected at one station at one time is not representative, except for order of magitude, of the average amount at that location over a long time period (Butler, Morris, and Sass, 1973). Approximately one-third, 30 out of 110, of the pelagic tar chromatograms show a double maximum in the envelope of normal paraffin peaks (Butler, Morris, and Sass, 1973). This has previously been suggested as evidence of crude-oil sludge (Miller, 1973; Brunnock et al., 1968).

Additional pelagic tar samples were collected during 1973 on cruises of the R.V. *Eastward* and R.V. *Atlantis II* (Sleeter et al., 1974). Tar quantities found in the southern Sargasso Sea were higher than expected (up to 90 milligrams per square meter) but amounts found in the vicinity of the Azores were somewhat smaller and comparable to the amounts found near Bermuda (see figure 7).

The Rhode Island group has found that suspended hydrocarbon material obtained by filtration of surface microlayer and subsurface water (20 to 30 centimeters deep) samples in the Sargasso Sea appears to be primarily particles of weathered pelagic tar less than one millimeter diameter with a considerable fraction probably less than 300 micrometers diameter. The estimated concentration of these particles in the subsurface samples is 100 to 300 micrograms per liter (Quinn and Wade, unpublished data). These particles probably account for most of the hydrocarbons observed in unfiltered surface and subsurface samples. Similar particles have been observed in water collected near Bermuda by Morris (unpublished data), and the mass of hydrocarbons calculated from the number and size of particles corresponds approximately to the mass concentration of hydrocarbons obtained by solvent extraction of the seawater followed by gas chromatographic analysis (Morris

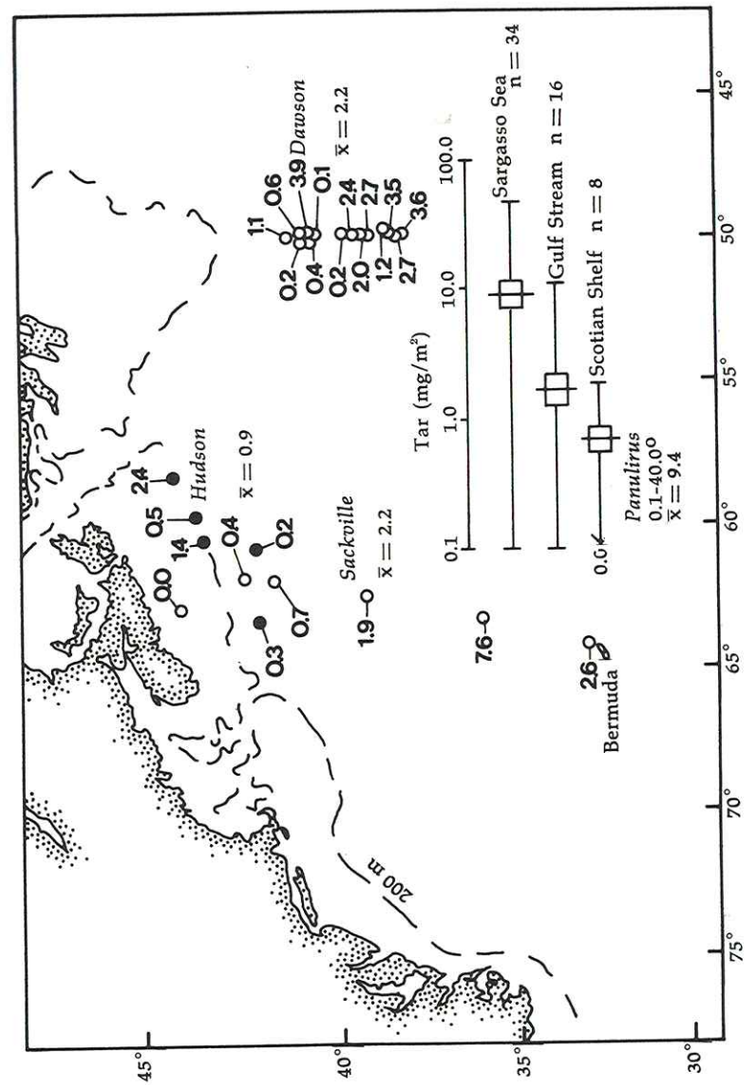

Figure 5. Distribution of tar in the northwest Atlantic and Sargasso Sea. Numbers by each station represent the quantity of tar found, in milligrams per square meter of ocean surface. Data obtained from cruises of the *Panulirus* (May, July, December, 1970; March, 1971 to November, 1972); *Sackville* (October, 1971); *Hudson* (September, 1969); and *Dawson* (June-July, 1970). After Butler, Morris, and Sass (1973). Also represented are the mean (vertical bar), standard deviation of the mean (white area) and range for each geographical region.

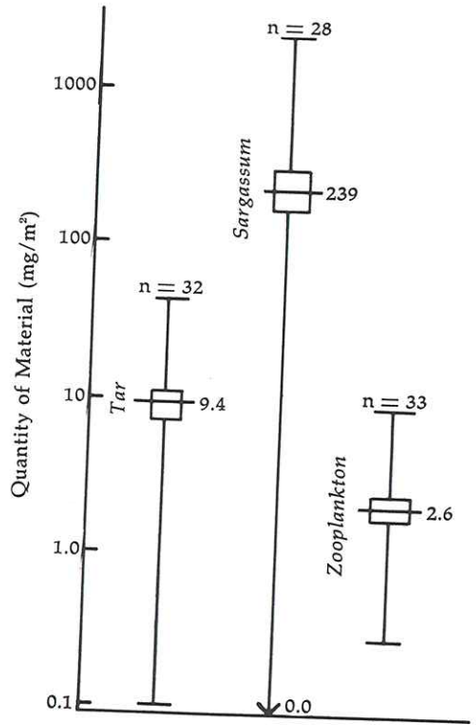

Figure 6. Comparison of the quantities of tar, *Sargassum*, and daytime surface zooplankton at station S, near Bermuda. All samples for 1970-72 included. After Butler, Morris, and Sass (1973).

and Gordon, unpublished data). Attempts were made to estimate the possible lifetime and fate of pelagic tar (Morris and Butler, 1973). Although no firm conclusions have yet been drawn, certain facts are of significance:

1. Tar lumps of various types (as indicated by the normal paraffin profile) are well-mixed. In the same net numerous distinct types have been found.

2. The outside surface of the lumps does not show substantial degradation compared to the interior. However, many lumps carry epiphytic algae, barnacles, and polychaete worms which may be several months old at the time of collection. Inhomogeneities appear to be associated with waxy inclusions, not with a concentric weathering pattern.

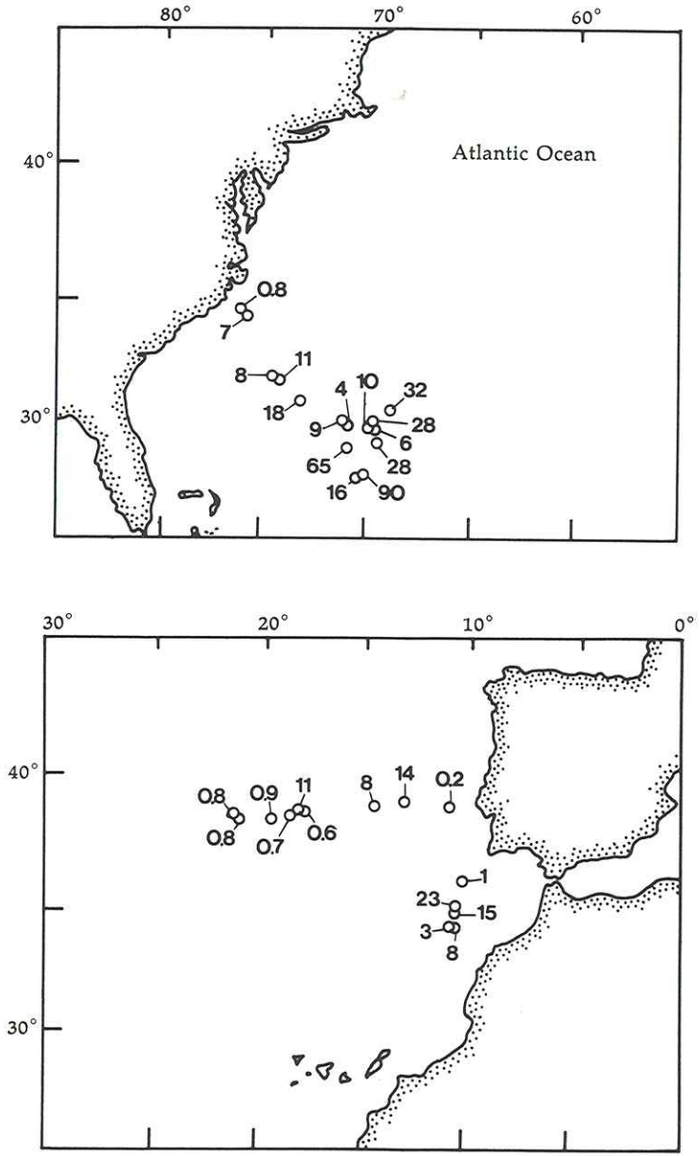

Figure 7. Distribution of tar in the southern Sargasso Sea and east Atlantic. Numbers by each station represent the quantity of tar found, in milligrams per square meter of ocean surface. Data obtained from cruises of the *Eastward* (June-July, 1973) and *Atlantis II* (July, 1973). After Sleeter, Morris, and Butler (1974).

3. Lumps of a wide size range show similar internal compositions.

4. Particles down to the mesh size of the net (0.3 millimeter) are found in the samples, and filtration of seawater from shallow depths near areas where pelagic tar is abundant also reveals black particles which may be the product of disintegrating tar lumps.

A very approximate estimate of the total standing stock of pelagic tar in the world's oceans (0.3 to 1.0 million metric ton), when combined with the input of petroleum to the high seas from all sources (three to four million metric tons per year) and an assumption of the fraction of petroleum remaining as tar (10 to 20 percent) lead to an estimated residence time of the order of one year (Butler, Morris, and Sass, 1973).

The current hypothesis is that waxy tanker wastes degrade initially by evaporation and dissolution, and that the residue from this short-term process constitutes the bulk of pelagic tar. Tar lumps degrade more slowly by becoming brittle at the outer surface and disintegrating to form fine particles of approximately neutral buoyancy. These particles are then mixed by convection in the upper layers of the ocean, and may contribute a substantial fraction of the hydrocarbons analyzed by solvent extraction of unfiltered seawater (Quinn and Wade, unpublished data). The degradation of these particles may be extremely slow because of the low concentrations of nutrient and hydrocarbon-oxidizing bacteria in the open ocean, and they may have lifetimes of many decades. At present, however, this hypothesis is largely speculative (Butler, private communication).

Sediments. The ultimate resting place for a significant fraction of the petroleum and chlorinated hydrocarbons is marine sediments. Harvey (unpublished data) has found that the surface layers (top two centimeters) of North Atlantic sediments appear to have accumulated significant amounts of PCB. The concentrations found are proportional to the depth of the overlying water, as shown in table 9.

Analysis of dated oceanic sediments from the Santa Barbara Basin of the Southern California Bight by the Berkeley group has shown that the deposition of PCB began about 1945 and DDE first appeared in sediments deposited about 1952. Concentrations of both substances show a progressive increase until 1967, with no indication that the rates of deposition were beginning to decline in that year. Estimated deposition rates of DDE and PCB in 1967 were 1.9×10^{-4} grams per square meter per year and 1.2×10^{-4} grams per square meter per year, respectively (Hom, Risebrough, Soutar, and Young, 1974).

Table 9. PCBs in sediments.

Location	Water Depth (meters)	PCBs (ng/g sediment, dry wt.)
Buzzards Bay, Massachusetts	3	500
Hudson Canyon	137	2.2
Hudson Canyon	2626	1.7
Hudson Canyon	3785	1.0
Hatteras Plain	5465	0.3

Harvey, unpublished data.

Surface sediments (one to ten centimeters deep) from several locations on a transect from Bermuda to the New York Bight to Woods Hole, Massachusetts, have been analyzed for hydrocarbons (including alkanes, alkenes, and two- to six-ring aromatics) by the Woods Hole group (Farrington, unpublished data). The concentrations of hydrocarbons from three-column chromatography fractions isolated from the sediments are presented in figure 8.

The sediments at stations 7, 8, 9, and 10 contain hydrocarbon concentrations distinctly greater than sediments at other stations. There are many sources for hydrocarbons in the New York Bight region. Thus, the higher concentrations of hydrocarbons in the sediments in and adjacent to this region are expected. Sediments at station 11 are near the acid-iron waste dump area. Sediments at station 10 are very near both the waste disposal area for spoils from harbor dredging and the sewage sludge disposal area (Gross, 1972). Sediments at station 9 are 12.5 kilometers down the Hudson Shelf trough from the sewage sludge and dredge spills disposal areas. It may be that transport of sediments from these disposal areas to station 9 accounts for the higher concentrations of hydrocarbons at this location. It is interesting to note that the concentrations of hydrocarbons in sediments at station 12, off the southern Massachusetts coast, are approximately the same as those of the continental slope and abyssal plain sediments at stations 1 to 5, but are much lower than at other inner continental shelf stations which are in or near the New York Bight region.

Biological Transfer

One of the more controversial aspects of the pollution of the marine environment by chlorinated and petroleum hydrocarbons has been the premise that these substances may be trophically concentrated as one proceeds up

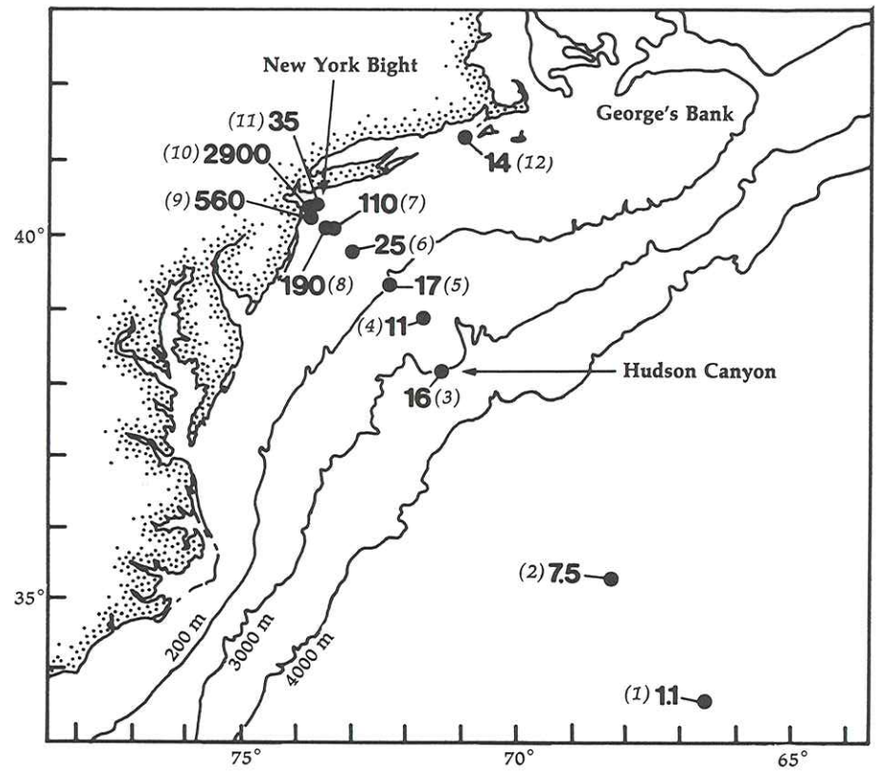

Figure 8. Hydrocarbon concentrations in sediments (micrograms per gram dry weight), including alkanes, alkenes, and two- to six-ring aromatic hydrocarbons. Station numbers are in parentheses. (Farrington, unpublished data.)

the food chain. Food-web magnification of non-metabolized chlorinated hydrocarbon pollutants has generally been an accepted premise in the past. Several studies, however, including some in the IDOE Pollutant Transfer Program, have obtained results suggesting that this concept should be modified in order to explain some of the observed pollutant distributions.

High concentrations of DDT and PCB compounds in marine birds and mammals support the trophic concentration theory when these chlorinated hydrocarbons are transferred from aquatic organisms to birds and mammals. Among the aquatic members of food webs, however, the available evidence

suggests that trophic accumulation of chlorinated hydrocarbons is but one of the factors affecting the concentrations of these compounds in marine organisms. Other factors are the lipid content, permitting increased storage; partition coefficients of individual compounds, which determine the degree of accumulation in the lipids of organisms over concentrations in the ambient water, and characteristics of integument, cell surfaces, excretion pathways, etc.

Chlorinated hydrocarbons have been found in a variety of marine organisms in the North and South Atlantic Ocean by the Woods Hole group (Harvey et al., 1974). The results of 53 analyses for PCBs in plankton are graphically presented in figure 9. The mean concentration of PCBs in both North and South Atlantic plankton was 200 micrograms per kilogram wet weight. The four samples containing more than 1000 micrograms per kilogram were all very rich in phytoplankton. The rest were mainly zooplankton. The PCB/DDT ratio was always greater than 30. Lower concentrations of PCBs in planktonic material have been reported in the Pacific, Gulf of Mexico, and Antarctic Ocean (Giam et al., 1973a & b; Claeys, 1972). The similar PCB concentrations observed in the North and South Atlantic samples by Harvey et al. (1974) cannot be explained at this time but obviously have important implications for global PCB transport.

Harvey et al. (1974) found that vertically migrating mesopelagic organisms in the Atlantic Ocean, which inhabit the 300 to 1000 meter depths, contain 2 to 200 micrograms per kilogram PCB and 1 to 20 micrograms per kilogram DDT wet weight. The relative concentrations among the six families and eleven genera on a wet- or lipid-weight basis reveal no correlation with lipid content, age, location, depth of habitat, size or trophic level. Commercial groundfish, e.g., cod, haddock, pollock, etc., caught on the New England fishing grounds contain about ten times more PCB and DDT than similar fish caught in the Denmark Strait.

The transfer of parts-per-trillion quantities (10^{-12} grams per gram) of PCBs and DDT in laboratory food chains is being studied at the Scripps Institution of Oceanography and the National Marine Fisheries Service to simulate typical trophic processes in the plankton of the ocean. Evidence suggests that unicellular algae scavenge these small quantities of PCB and DDT by as much as ten-thousandfold. In contrast, animals feeding on the algae accumulate these chemicals only two- to fivefold and a similar enrichment is found in fish larvae feeding on these herbivorous forms. The implication of these results for food-web magnification theories is under further investigation (MacGregor, 1974).

The mussel, *Mytilus californianus*, was used as an indicator species to determine the distribution of DDT and PCB compounds in southern Califor-

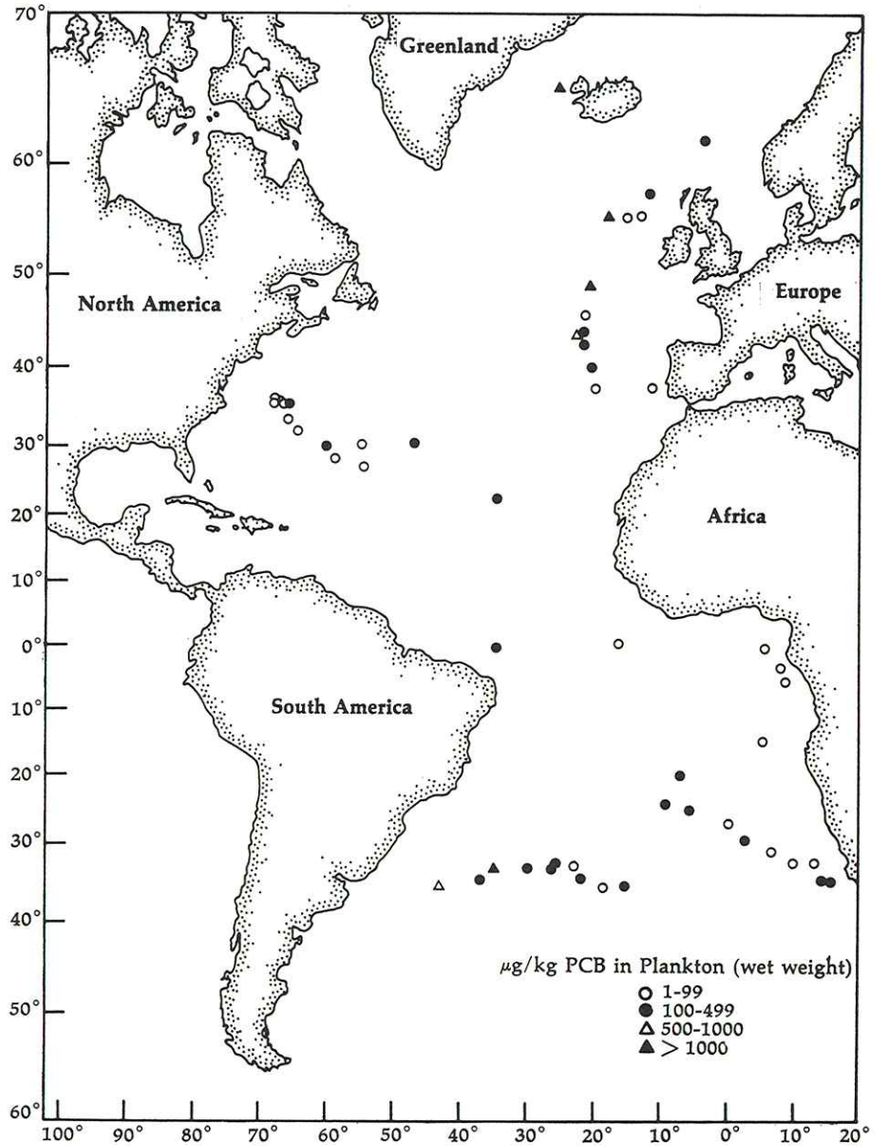

Figure 9. PCB concentrations in plankton in the Atlantic Ocean. Data obtained from cruises of *Chain, Atlantis II, Gosnold, Knorr,* and *Bjarni Saemundsson* from December, 1970, to August, 1972. After Harvey et al. (1974).

nia coastal waters by the Berkeley group. Three distinct zones of DDT contamination are recognized in this area: (1.) an exceptionally contaminated area in the vicinity of the White's Point outfall of the water treatment plant of the Los Angeles County Sanitation Districts on the Palos Verdes Peninsula; (2.) a highly polluted adjacent area extending from Oxnard in the north southward to Oceanside and seaward to the vicinity of Santa Barbara Island, into which DDT compounds are assumed to have dispersed from the White's Point region; and (3.) the remaining coastal areas and the offshore Channel Islands. Contamination levels in these latter areas are considerably lower and are assumed to derive principally from aerial fallout. The distribution of the PCB compounds is approximately equivalent to that of the DDT group, with concentration gradients extending from the Los Angeles area. Residues appear to derive from several input sources, however, in contrast to the dominant point source of input of the DDT compounds (Risebrough, unpublished data).

Similar results were found off the California coast by the Scripps group. The concentrations of PCB and DDT in zooplankton lipid (250 samples in an area of 500 × 3000 kilometers) indicate that DDT emanates from a point source near Los Angeles while PCB shows a more even distribution which is not inconsistent with a diffuse aerosol source. Evidence for advection in the California Current southward from the Los Angeles Bight appears in the PCB distribution (McClure, Talkington, and LaGrange, unpublished data).

Mussels obtained at Royal Palms on the Palos Verdes Peninsula by the Berkeley group show a downward trend in both DDT and PCB concentrations from 1971 to 1973. DDE and PCB concentrations were found not to be dependent upon the size of the mussels. Mussels brought from the Royal Palms site to Bodega Bay, where concentrations of chlorinated hydrocarbons in local mussels were one to two orders of magnitude lower, were found to lose 90 percent of the DDE and 75 percent of the PCB in five months (deLappe, Risebrough, and Young, unpublished data).

As part of their study of petroleum residues the Bermuda Biological Station group have obtained over 300 distinct samples of *Sargassum* communities in the western North Atlantic over the past two to three years. These samples include nearly 200 in a time series at station S, 18 miles southeast of Bermuda, extending over more than two years, as well as samples from most other parts of the Sargasso Sea. The relative amount of pelagic tar associated with the communities collected varies by a factor of over 5000. Most of the 300 samples of *Sargassum* communities have already been separated into major taxa in preparation for a statistical study of the species composition and organization. Identification and counts of the motile macrofauna are

nearly complete. The motile microfauna (mainly copepods) are still to be identified and counted.

Investigation of the food web of the *Sargassum* community has been directed towards discerning the primary food sources of each of the common species within the community. Analyses of the gut contents of preserved specimens were combined with direct observations of feeding behavior of live specimens in the laboratory and examination of their gut and fecal contents. The *Sargassum* community is supported by four major food sources: (1.) the plankton and particulate organic matter in the surrounding water, (2.) the *Sargassum* weed itself, (3.) the epiphytic algae, including diatoms, attached to the *Sargassum*, and (4.) the detritus produced within the community. The animals of the first trophic level include primarily the planktivores (the filter-feeding bryozoans, serpulid polychaetes, the barnacles, and the hydroids), the omnivores feeding on *Sargassum* and epiphytic algae (the amphipods, polychaetes, and other organisms with major alternative food sources) and the detritus-feeders (harpacticoid copepods, amphipods, and possibly flatworms). Trophic pathways overlap even at the first trophic level, and above this level the distinction between trophic levels is blurred. For the higher level omnivores and carnivores there are alternative food sources, and it is apparent that the limits of food availability within the *Sargassum* have imposed a necessity for the larger organisms (such as crabs, shrimp, and fish) to have the greatest number of alternative food sources available to them. The common Sargassum crab has been observed to readily eat *Sargassum* weed and even tar lumps in the absence of other food sources (Morris and Gieselman, unpublished data).

Samples of clean *Sargassum natans* collected 16 miles south of Bermuda contained from three to twelve micrograms per gram wet weight (12 to 60 micrograms per gram dry weight) hydrocarbons. This compares well with the results of Burns and Teal (1973) on material collected on the open ocean, which contained one to five micrograms per gram of petroleum hydrocarbons in the C_{14}-C_{30} range. Samples of *Sargassum natans* from the Bermuda area which had visible tar lumps and bryozoans (indicating old colonies) associated with them contained much higher hydrocarbon concentrations. Gas chromatograms of *Sargassum* material were similar to those of the tar, even though efforts were made to physically remove all the visible tar particles before extraction. These samples contained 20 to 250 micrograms per gram wet weight (77 to 970 micrograms per gram dry weight) hydrocarbons, but it is not yet certain that all of these hydrocarbons were incorporated in the algal tissues (Butler and Baum, unpublished data).

Summary

It is clear that investigations under the NSF/IDOE Pollutant Transfer Program have resulted in significant advances in our knowledge and understanding of pollutant transport to the marine environment. However, it is equally clear that the program is still in the early stages of attaining the objectives outlined in the Introduction. Detailed information is being obtained on the transport of trace metals, PCBs, DDT and, to a certain extent, petroleum hydrocarbons to some regions of the marine environment. Other pollutants; however, particularly organic substances such as hexachlorobenzene, heterocyclic compounds, and a number of chlorinated and non-chlorinated hydrocarbons of low molecular weight, have received little or no attention. Studies designed to determine quantitative fluxes of the three major pollutant classes—trace metals, petroleum, and chlorinated hydrocarbons—to the marine environment via the atmosphere, rivers, and industrial and sewage outfalls are underway, but should be expanded. There has been virtually no work on the contribution and significance of ocean dumping for these substances. The possible importance of particulates in the ocean as a mechanism for removal of pollutants to the sediments has only recently been realized, and much more work is needed on the pollutant scavenging properties of various types of marine particles, their residence time and interaction with the biota in the water column and their flux to the ocean floor. The sediments have received the least attention in this program, but they are the potential ultimate resting place for many, if not most, of the pollutants being investigated. Thus, the transport of pollutants to the sediments and their possible remobilization into the water column are most important.

Our understanding of the transfer of pollutants within the marine food web is limited at best. Recently some theories of food web magnification of pollutants have been questioned in some areas, partially as a result of investigations made in this program. Continued research on pollutant transfer within the marine biological community is certainly needed, as the results of these studies will be the cornerstone of any conclusions reached on the effect of these pollutants on the most important part of the marine environment—the biota.

Also of fundamental importance in investigating the transport of trace metals and petroleum-like hydrocarbons is the ability to distinguish between natural and anthropogenic sources for these substances found in the marine environment. This does not pose such a problem for the chlorinated hydrocarbons, since no natural sources are known for such substances as DDT and PCBs. While some progress has been made in distinguishing between naturally and anthropogenically-produced trace metals and petroleum-like hy-

drocarbons, the criteria used to date are largely empirical and, at best, subject to doubt in many cases.

Finally, we must strive to develop predictive models which will allow us to determine what, how much, how fast, and with what effect pollutant substances can be transported to and through the various reservoirs in the marine environment. To date, the Pollutant Transport Program has largely been concentrating on determining just what the primary pathways of transport are and obtaining some initial information on fluxes via these pathways. This information is, of course, critical in the development of such predictive models and much more information is also required about the specific mechanisms of transfer within the various reservoirs before adequate predictive models can be derived.

In summary, the first two years of the IDOE Pollutant Transfer Program have resulted in a number of significant new insights into pollutant transport to and within the marine environment. Perhaps more important, however, is that the work accomplished to date has brought the overall problem of pollutant transport into sharper focus as it has become apparent what additional types of information are needed and in what areas our basic understanding of transfer processes is lacking. Thus, these first two years will serve as a strong base for obtaining a more quantitative description of pollutant transfer to and within the marine environment during the remaining years of the International Decade of Ocean Exploration.

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Appendices

Appendix 1: NSF/IDOE Pollutant Transfer Program Executive Committee

One of the primary objectives of the IDOE Pollutant Transfer Workshop at Port Aransas, Texas, was to evaluate the scientific management of the Pollutant Transfer Program. Of particular importance was the consideration of the formation of an IDOE Pollutant Transfer Program Executive Committee, made up largely of individuals actively involved in this program.

The Pollutant Transfer Program as presently constituted consists of ten separate and distinct research grants investigating a wide variety of problems associated with pollutant transfer to the marine environment. There has not been a concerted effort by the participants in this program to develop a comprehensive plan for investigating transfer processes to and within the marine environment. Ultimately, the Pollutant Transfer Program should be able to provide details for a framework, based on theoretical, laboratory, and field work, from which one can determine the important transfer mechanisms, identify the major factors that affect the transfer process, and develop the basic principles governing pollutant transfer for any pollutant, present and future.

In addition to the need for a comprehensive master plan for pollutant transfer research, the participants felt that there are many obvious areas where cooperative and joint efforts, both in the laboratory and in the field, would be highly desirable and likely quite productive within the present program.

Thus, the participants at the Port Aransas Workshop recommended strongly that an IDOE Pollutant Transfer Program Executive Committee be set up as rapidly as possible. The primary functions and responsibilities of this Executive Committee would be:

1. To provide scientific guidance to the NSF/IDOE Office with respect to the Pollutant Transfer Program, with particular attention to the development of comprehensive long-range plans for accomplishing the objectives of this program and the identification of weaknesses as they develop in the existing program.

2. To initiate and coordinate intercalibration, sample exchange, and joint sampling programs among the Transfer Program participants.

3. To organize summer institutes, special workshops, the production of special standards and other similar cooperative efforts to further the goals of the Pollutant Transfer Program.

4. To cooperate with the NSF/IDOE Environmental Quality Program

Manager in the preparation and presentation of the annual oral report of the Pollutant Transfer Program to the National Science Foundation.

5. To stimulate and solicit strong research proposals relevant to the long-range plan and goals of the Pollutant Transfer Program and to *comment on the applicability* of submitted proposals to the long-range plan and goals.

6. To establish panels to assist in the development of programs in any particular areas of need, such as modelling, intercalibration, sampling techniques, data processing, etc.

7. To establish an effective system of communication of scientific results among Transfer Program participants, the scientific community in general, and the public.

It is further recommended that the Executive Committee be composed of three people actively involved in the IDOE Pollutant Transfer Program and one person, appointed by the IDOE office, actively involved in similar research but not a part of this program. The three members from the Pollutant Transfer Program should be elected by the principal investigators of active grants. All members should be appointed for two-year terms, except two of the initial four members should be appointed to three-year terms. The Program Manager for Environmental Quality at IDOE and his immediate predecessor should both be ex-officio members of the Executive Committee. One member of the Committee should be elected chairman by the Committee members.

The Executive Committee should have an administrative system to provide travel, communication, secretarial assistance, etc. for various functions outlined above. The administrative function should be located with the Chairman of the Executive Committee.

Appendix 2: NSF/IDOE Pollutant Transfer Program Active Grants, 1972-74

Principal Investigator	Organization	Project Title
David C. Burrell and Peter McRoy	University of Alaska	Heavy Metal Dynamics in Seagrass Ecosystems: Processes and Oceanic Interaction
James N. Butler and Byron Morris	Harvard University and Bermuda Biological Station	Transfer of Persistent Pollutants in *Sargassum* Communities
Norman Cutshall	Oregon State University	Effects of Ocean Water on the Physico-Chemical Form of Heavy Metals
Robert A. Duce	University of Rhode Island and University of Connecticut	Atmospheric Pollutant Transport and Deposition on the Sea Surface
Edward D. Goldberg	University of California, Scripps Institution of Oceanography	The Fluxes of Synthetic Organics in the Marine Environment
and		and
Reuben Lasker		Exchange Rates of Chlorinated Hydrocarbons and Similar Chemicals in Marine Food Chains
George R. Harvey	Woods Hole Oceanographic Institution	Uptake and Transfer of Chlorinated Hydrocarbons in the Atlantic Ocean
Clair C. Patterson	California Institute of Technology	Determination of the Input and Transport of Pollutant Lead in Marine Environments Using Isotope Tracers
Robert W. Risebrough	University of California, Berkeley	Formulation of Mass Balance Equations for Polychlorinated Biphenyls in Marine Ecosystems
Herbert L. Windom	University of Georgia, Skidaway Institute of Oceanography	The Transfer of Heavy Metals Through the Inner Continental Shelf to the Open Ocean

Appendix 3: Publications Resulting from the NSF/IDOE Pollutant Transfer Program

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Appendix 4: NSF/IDOE Pollutant Transfer Program Workshop Participants

University of Texas Marine Science Institute, Port Aransas, Texas, January 11-12, 1974

Terry Bidleman
Department of Food and Resource Chemistry
University of Rhode Island
Kingston, Rhode Island 02881

James Butler
Division of Engineering and Applied Physics
Pierce Hall
Harvard University
Cambridge, Massachusetts 02138

Norman Cutshall
School of Oceanography
Oregon State University
Corvallis, Oregon 97331

Robert Duce
Graduate School of Oceanography
University of Rhode Island
Kingston, Rhode Island 02881

John Farrington
Woods Hole Oceanographic Institution
Woods Hole, Massachusetts 02543

William Forster
Ecological Sciences Branch
Division of Biomedical and Environmental Research
U.S. Atomic Energy Commission
Washington, D. C. 20545

C. S. Giam
Office of the International Decade of Ocean Exploration
National Science Foundation
Washington, D. C. 20550

Edward Goldberg
Scripps Institution of Oceanography
University of California
P.O. Box 109
La Jolla, California 92037

Grant Gross
Oceanography Section
National Science Foundation
Washington, D. C. 20550

George Harvey
Woods Hole Oceanographic Institution
Woods Hole, Massachusetts 02543

John Hem
Water Resources Division
U.S. Geological Survey
345 Middlefield Road
Menlo Park, California 94025

Gerald Hoffman
Graduate School of Oceanography
University of Rhode Island
Kingston, Rhode Island 02881

Feenan Jennings
Office of the International Decade of Ocean Exploration
National Science Foundation
Washington, D. C. 20550

Reuben Lasker
National Marine Fisheries Service
Fishery Oceanography Center
La Jolla, California 92037

John Martin
Moss Landing Marine Laboratory
Moss Landing, California 95039

Vance McClure
Tiburon Fisheries Laboratory
P.O. Box 98
Tiburon, California 94920

Peter McRoy
Institute of Marine Science
University of Alaska
College, Alaska 99701

Patrick Parker
Marine Science Institute
University of Texas
Port Aransas, Texas 78373

Bob Presley
Department of Oceanography
Texas A & M University
College Station, Texas 77843

James Quinn
Graduate School of Oceanography
University of Rhode Island
Kingston, Rhode Island 02881

Reinhold Rasmussen
Air Pollution Research Section
College of Engineering
Washington State University
Pullman, Washington 99163

Robert Risebrough
Bodega Marine Laboratory
University of California
P.O. Box 247
Bodega Bay, California 94923

William Sackett
Department of Oceanography
Texas A & M University
College Station, Texas 77843

Dorothy Settle
Division of Geological and Planetary Sciences
California Institute of Technology
Pasadena, California 91109

David Stallings
Fish Pesticide Research Laboratory
Route 1
Columbia, Missouri 65201

Robert Stickney
Skidaway Institute of Oceanography
55 West Bluff Road
Savannah, Georgia 31406

Herbert Windom
Skidaway Institute of Oceanography
55 West Bluff Road
Savannah, Georgia 31406

Kenneth Winters
Marine Science Institute
University of Texas
Port Aransas, Texas 78373

William Zoller
Department of Chemistry
University of Maryland
College Park, Maryland 20742