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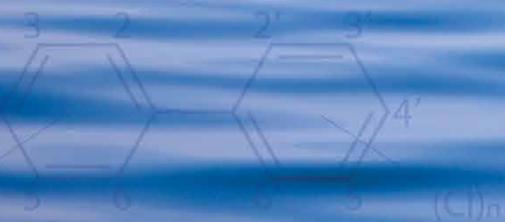
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# Persistent Organic Pollutants (POPs), Polycyclic Aromatic Hydrocarbons (PAHs), and Plastics

Examples of the Status, Trend, and Cycling of  
Organic Chemicals of Environmental Concern in the Ocean

BY JOHN W. FARRINGTON AND HIDESHIGE TAKADA



**ABSTRACT.** Four decades of research have provided a reasonable understanding of the outline of the biogeochemical cycles of persistent organic pollutants (POPs) and polycyclic aromatic hydrocarbons (PAHs) in coastal ocean and surface ocean ecosystems, including atmospheric transport to the sea, air-sea exchange processes, and the role of particulate matter in removing these chemicals from surface waters. It is clear that deep ocean fish are contaminated with POPs. However, despite available sampling and analytical capabilities, deep ocean ecosystems are much less sampled and understood. A multidecade assessment of POPs and PAHs in US coastal waters using bivalve sentinel organisms documents high concentrations near urban areas and also some stations where concentrations have begun to decline. The results are consistent with coastal sediments near urban areas being a leaky sink for POPs and PAHs, and sources from land continuing to contribute these contaminants to the sea. Other studies document coastal and continental margin surface sediments as a sink, albeit a potentially leaky sink, for POPs and PAHs. Floating plastic debris, including small pellets, has reemerged as an oceanic environmental concern. A “Pellet Watch” assessing plastic pellets and associated POPs and PAHs is underway. Enhanced studies of deep-ocean ecosystems are recommended. The findings are also relevant to biogeochemical cycles for emerging organic pollutants.

## INTRODUCTION

Organic chemicals of environmental concern (OCEC) have been detected in oceanic ecosystems, including polar region food webs, abyssal ocean sediments and biota, and open ocean surface mixed layer waters, as well as in a few deep-sea water samples (e.g., WHO, 1976, 1979; Fowler, 1990; Schulz-Bull et al., 1998; Bidleman et al., 2005, and references therein). Globally, OCEC have polluted many coastal ecosystems, mainly near urban areas (UNEP, 1984). This statement may seem trivial, given that such findings were to be expected based on the pioneering pollution studies of the early 1970s. However, having actual data regarding the extent and severity of chemical contamination, and the basis for longer-term assessments, provides a means to determine how concentrations of chemicals of environmental concern respond

spatially and temporally to human actions, be they deliberate or inadvertent inputs, or regulatory actions to curtail or eliminate inputs.

There are two main categories of OCEC: (1) natural organic chemicals, such as fossil fuel hydrocarbons mobilized by modern human activities and introduced to the marine environment in amounts and at rates in excess of those of a few centuries ago, and (2) organic chemicals that are new to nature and synthesized in the modern era, such as polychlorinated biphenyls (PCBs), that are of potential concern when deliberately or inadvertently released to the environment. These latter compounds (category 2) are often termed xenobiotic chemicals. The very presence of

detectable xenobiotic chemicals in samples from ocean ecosystems is indicative of a change in ocean chemistry.

National, regional, and United Nations regulatory actions have been taken to eliminate or curtail production and release of some OCEC, such as PCBs and several chlorinated pesticides, including DDT and hexachlorocyclohexanes (Table 1). Nevertheless, these chemicals are still moving and being redistributed through the environment. They are often termed legacy pollutants. More recently, other OCEC produced for reasons that were thought to benefit society, such as the flame retardant polybrominated diphenyl ethers, have entered the environment and become OCEC. These chemicals, and others such as some pharmaceuticals and homecare products, are often referred to as emerging pollutants, although some have now been around for over two decades (see Box 1).

This raises a point of terminology for the words “contaminant” and “pollutant.” For most scientific considerations, contaminants are chemicals present at concentrations above natural background for category 1 chemicals or level of detection for category 2 chemicals. Pollutants are chemicals present at concentrations that cause harm to organisms, ecosystems, or humans. Our discussion here does not revise the words of authors we cite but rather accepts the terms as they have used them.

Our task in this paper is to use a few examples to provide a glimpse, more like

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**John W. Farrington** ([jfarrington@whoi.edu](mailto:jfarrington@whoi.edu)) is Dean Emeritus, Woods Hole Oceanographic Institution, Woods Hole, MA, USA. **Hideshige Takada** is Professor, Laboratory of Organic Geochemistry, Tokyo University of Agriculture and Technology, Fuchu, Tokyo, Japan.

a survey, for ocean scientists as a whole of current knowledge of OCEC in the marine environment, changes over time, and biogeochemical cycles in the marine environment. We acknowledge that this overview does not do justice to the extensive efforts of many scientists.

Chemical measurements by themselves are a small part of the picture. Lohman et al. (2007, and references therein) document the need for a more complete understanding of biogeochemical cycles of OCEC. They note the importance of having reasonably accurate assessments of production and inputs of OCEC to the environment and review example efforts. We recognize that this is not easy to accomplish for a global inventory and do not attempt such an effort in this paper. In addition, we recognize that biogeochemical cycles (including bioavailability) need to be coupled with biological effects and potential human health effects of chemicals transferred through the marine food web to humans.

Given the time scales of decades to millennia for the cycling of some chemicals in seawater and surface sediments of the deep ocean, it is important to be able to detect very low concentrations

of such chemicals and to understand their marine biogeochemical cycles. This enables an early warning of their presence and pathways of movement to allow action to be taken to prevent their buildup to the point of causing adverse effects.

### ASSESSMENTS OF OCEC IN THE MARINE ENVIRONMENT

Polychlorinated biphenyls (PCBs), a few chlorinated pesticides such as DDT and its main environmental degradation products DDE and DDD (most often noted herein as ΣDDT), and a few examples for polycyclic aromatic hydrocarbons (PAHs) are chosen as examples to illustrate the processes acting on many of the OCEC and the resulting distribution in the environment. PAHs are not in the Stockholm Convention POPs list, but are of environmental concern, (e.g., they are specified in the United Nations/Economic Council for Europe Aarhus Protocol [Lohmann et al, 2007]). These examples were chosen because there is some, but by no means adequate, geographical coverage for concentrations in water, sediment, or selected biota, for which there are also some time series data. (See Box 2 for a brief

discussion of chemical structures and shorthand notations.)

We also provide a brief assessment of contamination in certain areas of the surface ocean containing small size floating debris because of the recent reemergence of interest in this topic.

### Framing the Challenge

Assessments of contaminant and pollutant concentrations can be classified as follows:

1. Integrative (e.g., concentrations in a top predator as a means to assess food web and ecosystem contamination/pollution)
2. Baseline or benchmark (the first pass at determining spatial distribution and concentration)
3. Temporal (changing concentrations over time at a given location)
4. Temporal and spatial distributions (concentrations over time at several locations)

For (2) through (4), the usual sample types are available: water (dissolved and particulate matter concentrations, including large and small sinking particles), air immediately over the water surface, surface slicks, sediment,

Table 1. Persistent organic pollutants listed in the Stockholm Convention of 2001 plus Amendments of 2009 and 2011 (United Nations, 2013b).

Chemical Annex A Elimination <sup>a</sup>		Chemical Annex B Restricted <sup>b</sup>
Aldrin	Hexachlorobenzene	DDT
Alpha hexachlorocyclohexane (α HCH)	Lindane	Perfluorooctane sulfonic acid and its salts
Beta hexachlorocyclohexane (β HCH)	Mirex	Perfluorooctane sulfonyl fluoride
Chlordane	Pentachlorobenzene	
Chlordecone	Polychlorinated biphenyls (PCB)	
Dieldrin	Tetrabromodiphenyl ether	
Endrin	Pentabromodiphenyl ether	
Heptachlor	Toxaphene	
Hexabromobiphenylether	Technical enodsulfan and its related isomers	
Heptabromodiphenylether		

<sup>a</sup>Elimination – Signatories will undertake measures to eliminate production and use of the chemicals listed. See Convention for detailed legal explanations.

<sup>b</sup>Restricted – Signatories will produce and use the listed chemicals only for restricted uses specified according to the Convention and Annex B. See Convention for detailed legal explanations.

and myriad organisms.

Most importantly, we need to have an understanding of the biogeochemical cycles of OCEC to be able to estimate the routes of input and movement through marine ecosystems, including both chemical and biochemical reactions (e.g., biodegradation, detoxification, metabolism),

as well as reservoirs of temporary (days to decades) and long-term (centuries or more) accumulation. Figure 1 shows most of the essential features of a biogeochemical cycle. For a complete picture, the processes depicted should be coupled with regional and global atmospheric and ocean circulation processes.

### OCEC in Marine Organisms: Widespread Contamination, Temporal Trends

Numerous additional and more detailed studies conducted since the 1970s have documented OCEC contamination in deep-sea fish, marine mammals, and marine birds, such as the North Pacific

## BOX 1. BRIEF HISTORICAL PERSPECTIVE

In a manner understood by the public, Rachel Carson (1962) documented harm to organisms exposed to chlorinated pesticides (e.g., DDT). Thus began the saga of organic chemicals of environmental concern (OCEC). Although sparse, studies of OCEC from the 1950s to about 1970 (Blumer et al., 1970; SCEP, 1970; NRC, 1971; Goldberg, 1972) were of considerable concern. When brought to the attention of the United Nations Conference on the Human Environment held in June 1972 in Stockholm, Sweden, along with data for several trace metals such as lead and mercury, these studies informed Principle 7 of the conference's declaration (United Nations, 2013a):

*States should take all possible steps to prevent pollution of the seas by substances that are liable to create hazards to human health, to harm living resources and marine life, to damage amenities or to interfere with other legitimate uses of the sea.*

OCEC, mainly those with nonpolar (lipophilic) properties, can bioaccumulate at 1,000 to 100,000 times from seawater to fatty tissues of marine organisms. This means that concentrations of  $10^{-8}$  g L<sup>-1</sup> seawater of a nonpolar organic chemical has the potential to bioaccumulate to  $10^{-6}$  g g<sup>-1</sup> tissue dry weight (sometimes noted as 1 ppm) and also be transferred through the food web, accompanied by various adverse biological effects on some species (NRC, 1975, 1979; WHO, 1976, 1979). It is also a potential pathway back to human consumers of seafood, with accompanying health concerns (e.g., Dawe and Stegeman, 1991).

Research studies and environmental assessments from the 1970s through the 1990s led to The Stockholm Convention of May 22, 2001, regarding persistent organic pollutants (POPs; United Nations, 2013b)<sup>1</sup>. POPs (Table 1) have common properties: they are persistent, bioaccumulative, and toxic (PBT) in one way or another to organisms (Muir and Howard, 2006). This knowledge led to screening procedures for all organic chemicals and, along with analytical chemical assessments of environmental distributions, led to significant efforts to identify what are now designated as "emerging contaminants" (Field et al., 2006a).

An early effort to identify emerging chemicals of environmental concern (NRC, 1975) was led by marine geochemist Edward D. Goldberg of Scripps Institution of Oceanography, a pioneer in identifying chemical contaminants in the marine environment. Following this lead, various registries of chemicals have been assembled over the years. On the order of 30,000 organic chemicals are widely used in quantities greater than one ton per year (Muir and Howard, 2006), mostly with the intent of providing for needs of modern civilization. As of October 21, 2013, there were 308,000 substances on the CHEMLIST™ (Regulated Chemicals Listing; CAS, 2013), which includes listings made under the US Toxic Substances Control Act (TSCA)<sup>2</sup> and other similar listings from Europe and China. More than 50 new items that require screening for marine environmental concerns are added to these listings each week (CAS, 2013).

Even though only a small percentage of all organic chemicals in use are of environmental concern, a small percent of a large number can still be a large number. Fortunately, research and environmental assessments have begun to catch up with needs, and a substantive understanding of the fundamentals of the aquatic chemistry of organic chemicals has evolved and is admirably summarized and explained in *Environmental Organic Chemistry* (Schwarzenbach et al., 2003). This body of knowledge, coupled with a growing understanding of toxicology and ecotoxicology, structural chemistry, and a few key chemical properties, provides an initial indication as to which organic chemicals may indeed prove to be serious pollutants if they enter aquatic ecosystems in appreciable quantities.

Progress in understanding the biogeochemical cycles of OCEC in the marine environment has been part of progress in the past few decades for more general knowledge of marine organic chemistry or the biogeochemistry of organic matter and chemicals in the marine environment as documented in several review-type publications (e.g., Duursma and Dawson, 1981; Farrington, 1992; Hansell and Carlson, 2002; Lee et al., 2004; Hansell et al., 2009; Bianchi and Canuel, 2011). We acknowledge the importance of natural organic matter and chemicals as they interact with OCEC in the marine environment.

<sup>1</sup> The Stockholm Convention came into force May 17, 2004, and was amended in 2009 and 2011.

<sup>2</sup> The objective of the Toxic Substances Control Act (TSCA) is to allow EPA to regulate new commercial chemicals before they enter the market, to regulate existing chemicals (1976) when they pose an unreasonable risk to health or to the environment, and to regulate their distribution and use (from <http://www.epa.gov/oecaagct/lsc.html>).

A complete presentation of known detailed chemical structures of organic chemicals of environmental concern would probably require the entire number of pages allotted to this special issue of *Oceanography* because of the large number of chemicals involved (see Box 1. Brief Historical Overview). Here, we use a few representative chemical structures to illustrate this point.

There can be 209 possible molecular structures in the entire set of mixtures of polychlorinated biphenyls (PCBs). Each individual chlorinated biphenyl molecule is designated as a congener of all other chlorinated biphenyls, meaning that they have either the same number of chlorines substituted on the biphenyl molecule (in which case they are also homologs, and a subset of these homologs are isomers) or have different numbers of chlorines substituted on the biphenyl molecule. Figure B2-1 gives examples of chlorobiphenyl congeners along with the numbering system of the biphenyl ring carbons to be able to correctly identify the chlorobiphenyl according to the nomenclature of the International Union of Pure and Applied Chemistry (IUPAC). In order to avoid the time-consuming repeat of the formal chemical name for each chlorobiphenyl in publications (also minimizing naming mistakes), IUPAC has assigned a congener number to each congener using a shorthand notation for polychlorinated biphenyl (PCB) or chlorobiphenyl (CB). Figure B2-1 shows CB 28 and CB 138 as examples of this notation. Erickson (1997), among others, provides a detailed discussion of PCBs.

Toxaphene, a chlorinated pesticide, is an even more complicated mixture of individual compounds of at least 670 possible chlorinated bicyclic terpenes that have a wide range of molecular weights, with polychlorinated camphenes being the major constituents. DDT is a mixture of two molecules, *p, p'* dichlorodiphenyltrichloroethane (*p, p'* DDT) and *o, p'* dichlorodiphenyltrichloroethane (*o, p'* DDT). The differentiation of the two chemicals is the position of substitution of a chlorine atom on the carbon atoms of the diphenylethane rings. Hexachlorocyclohexane has three configurations, alpha, beta, or gamma hexachlorocyclohexane, and again, the differentiation is the positioning of chlorine atoms on carbon atoms of the cyclohexane ring.

Figure B2-2 depicts representative structures of polycyclic aromatic hydrocarbons (PAHs). PAHs enter the ocean from a variety of sources, including forest and grass fires, wood combustion, fossil fuels (natural gas, coal, and oil), oil spills, and chronic inputs of petroleum (Lima et al., 2005). The representative structures in Figure B2-2 are only a few of the hundreds of PAHs potentially present in samples from the marine environment.

The exact chemical structures control the fate and effects of the chemicals in the environment, for example, solubility (even though they may have low solubility compared to, for example, NaCl, the chemicals do have some solubility in seawater); partitioning among the dissolved phase, colloidal phase, and particulate matter; bioaccumulation; differential metabolism by fish; and differential biodegradation by microbes and photochemical reactions (Schwarzenbach et al., 2003).

### Polychlorinated Biphenyls

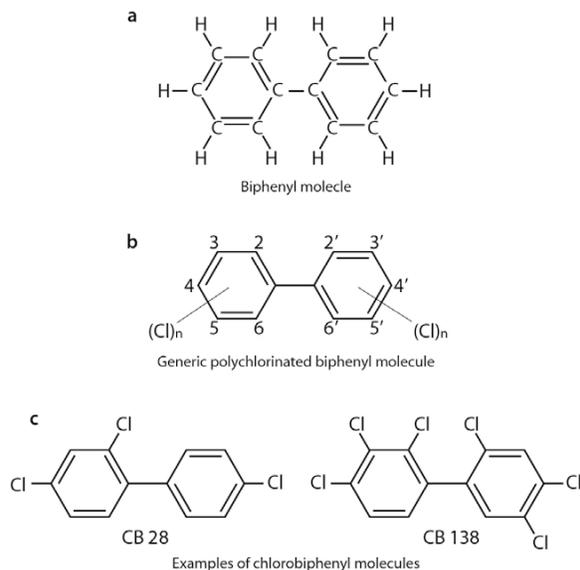


Figure B2-1. (a) Chemical structure of biphenyl, with carbon atoms, hydrogen atoms, and carbon-carbon bonds with aromaticity indicated by the carbon-carbon double bonds. (b) Generic structure of polychlorinated biphenyls in “short-hand” type notation with hydrogen atoms not shown and the carbon atom backbone depicted by two connected six-sided rings. (c) Two examples of individual chlorobiphenyls using the IUPAC shorthand number notation. The chlorine atoms have replaced the hydrogen atoms bonded to the carbon atom at the position shown. CB 28 is 2, 4, 4' trichlorobiphenyl and CB 138 is 2, 2', 3, 4, 4', 5' hexachlorobiphenyl.

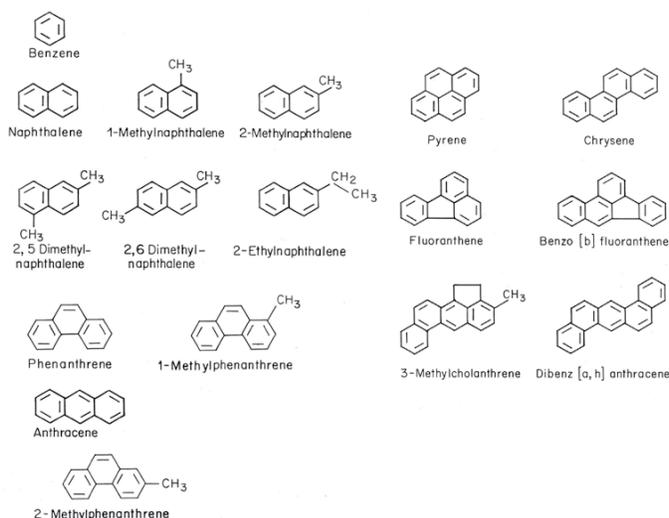


Figure B2-2. Example chemical structures using shorthand notation for aromatic rings where carbon atoms and hydrogen atoms are not shown and hexagon rings represent the carbon backbone. Benzene, with one cyclic ring, is not a polycyclic aromatic hydrocarbon, but is included for comparison. Some examples of alkyl (e.g., CH<sub>3</sub> or methyl and CH<sub>2</sub>CH<sub>3</sub> or ethyl) groups substituted on the ring structure are shown. Alkyl groups can be substituted on one, two, or several of the carbons in the cyclic rings. There can be various carbon chain lengths for the alkyl substituents.

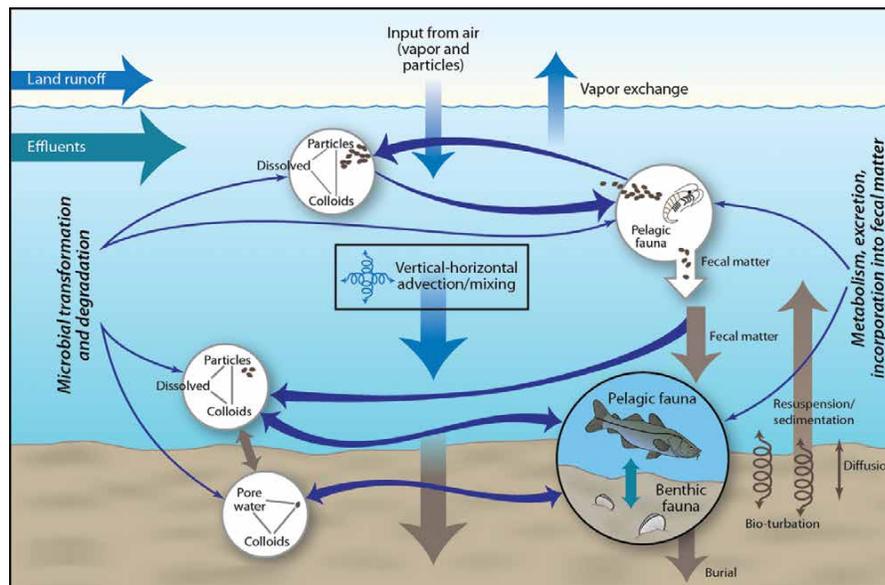


Figure 1. Schematic of the biogeochemical cycle of organic chemicals of environmental concern (OCEC) in the marine environment.

albatross (e.g., Stegeman et al., 1986; Fowler, 1990; Mössner and Ballschmiter, 1997; Froeschis et al., 2000; Yamashita et al., 2007; Howard et al., 2011).

There are sufficient quality analyses in some locations for assessments of temporal trends. For example, medical grade cod liver oil collected from a processing plant in Gdynia, Poland, from 1971 to 1989 was analyzed for PCBs and chlorinated pesticides (Kannan et al., 1992). DDT concentrations decreased from  $25 \times 10^{-6} \text{ g g}^{-1}$  fat (lipids) in 1974 to  $3.1 \times 10^{-6} \text{ g g}^{-1}$  fat (lipids) in 1989, and PCB concentrations decreased at a slower rate from  $18 \times 10^{-6} \text{ g g}^{-1}$  fat (lipids) in 1974 to  $7.0 \times 10^{-6} \text{ g g}^{-1}$  fat (lipids) in 1989. Pikkarainen and Parmanne (2006) report PCB (sum of seven chlorobiphenyls [CBs]) and organochlorine pesticide concentration data for herring from 1985 to 2002 in the northern Baltic Sea and also review other data from OCEC monitoring using fish tissue from the Baltic. The sum of the seven CBs decreased from

9–16 to  $2\text{--}6 \times 10^{-6} \text{ g kg}^{-1}$  wet weight of tissue from two-year-old female herring, and the sum of DDT concentrations decreased from 8–15 to  $1\text{--}5 \times 10^{-6} \text{ g kg}^{-1}$  wet weight tissue between 1985 and 2002.

Rigét et al. (2010) assemble and review multiple sets of data for legacy POPs for Arctic fish, bivalves, mammals, and birds. Most legacy POPs showed decreasing concentrations over the two to three decades preceding 2010 at the same time as there were decreasing trends for many of the same legacy POPs in Arctic air. An interesting exception was the chlorinated pesticide  $\beta$ -HCH, whose concentrations increased in the Canadian Arctic. Rigét et al. (2010) suggest this increase might be due to long-range oceanic transport to the Arctic rather than long-range atmospheric transport. This conclusion is consistent with earlier findings that atmosphere-seawater partitioning differences between the two isomers,  $\alpha$ -HCH and  $\beta$ -HCH, and accompanying transport pathway differences explain divergence in the concentrations of the two isomers in biota (Li and MacDonald, 2005).

Bivalve Organisms as Sentinels of OCEC in the Coastal Marine Environment: “Mussel Watch” Pioneering efforts that examined OCEC in land and marine organisms (e.g., Butler 1973) resulted in a call for a bivalve sentinel organism program of repeated collection and analysis over several annual cycles that led to establishment of the “Mussel Watch” program (Goldberg, 1975). A prototype US Environmental Protection Agency (EPA) Mussel Watch program from 1976 to 1978 that included PCBs, DDT family compounds, and PAHs, among the analytes, proved the feasibility of a large nationwide program (Goldberg et al., 1978; Farrington et al., 1983). Consistent background concentration benchmarks for several OCEC were established at rural coastal sites, and three-year consistent elevated concentrations were documented for several urban sites (Farrington et al., 1983). Mussel Watch programs have also been pursued elsewhere, including the United Kingdom, France, the Mediterranean

region, South America, Central America, the Caribbean region, Mexico, and the Asian Pacific region (e.g., Topping, 1983; Cantillo, 1991; Sericano et al., 1995; Villeneuve et al., 1999; Monirith et al., 2003; Isobe et al., 2007).

The US EPA Mussel Watch program evolved into the US National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) Program launched in 1985, which is ongoing. The program samples and analyzes both bivalves and surface sediments (not discussed here) on an annual basis. The collected data are available in an online database (<http://ccma.nos.noaa.gov/about/coast/nsandt/download.aspx>). A two-decade report that provides an overview and summary of data for 1986–2004/2005 is

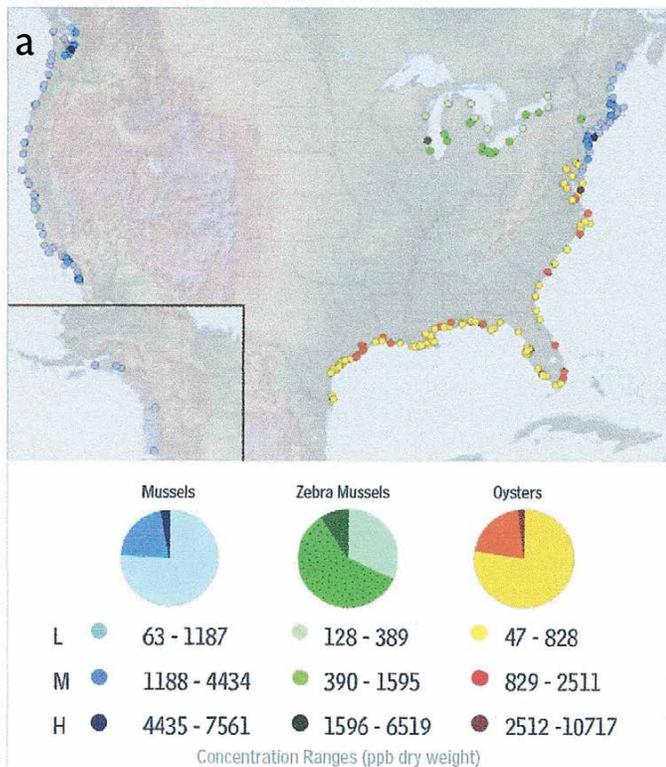
available online (Kimbrough et al., 2008). Figures 2 and 3 show OCEC concentrations found in the database for PAHs (Figures 2a and 3a) and PCBs (Figures 2b and 3b).

Ongoing sources of PAH to the coastal environment include oil inputs, fossil fuel and wood combustion, and forest and grass fires (Lima et al., 2005). Elevated concentrations near urban areas result from legacy PAHs in sediments and soils (Figure 2a). Kimbrough et al. (2008) report 33 sites with significant decreasing PAH concentrations and two sites with significant increasing concentrations (Figure 3a). Continuing elevated concentrations of PCBs (Figure 2b) are associated with legacy pollution; concentrations in coastal areas are controlled by continuing input from upstream

riverine areas with legacy PCB pollution or by flux from PCB-contaminated sediments to the water column and other components of the ecosystem. Forty-six sites have decreasing concentrations of PCBs for 1986–2004/2005, and one station shows an increase in concentration (Figure 3b).

There are limitations to the use of bivalves as sentinel organisms. Perhaps the most serious one is the lack of adequate populations in specific geographic locations where OCEC assessment is needed. One mechanism for overcoming this limitation is to transplant bivalves from nearby locations and suspend them on moorings (e.g., Widdows et al., 1981; McDowell-Capuzzo et al., 1989; Tanabe and Subramanian, 2006; Dabrowska et al., 2013).

### REGIONAL SPECIES CHARACTERIZATION



### REGIONAL SPECIES CHARACTERIZATION

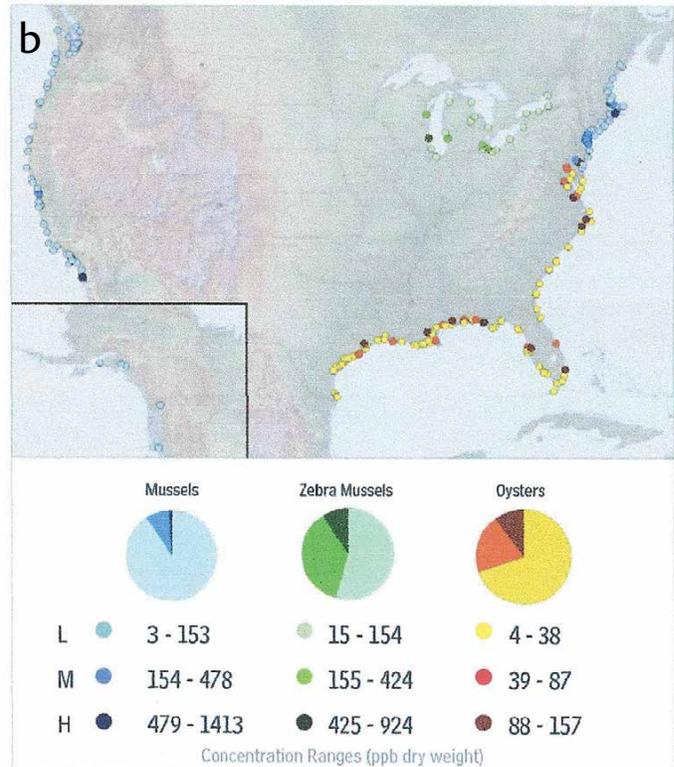


Figure 2. Concentrations of polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs) in bivalve sentinel organisms at stations along the US coast as reported in the US National Oceanic and Atmospheric Administration (NOAA) National Status and Trends (NS&T) program. (a) PAHs. (b) PCBs. From Kimbrough et al. (2008)

## Passive Samplers for OCEC Assessment

Passive samplers are another approach to overcoming the limitations of using bivalves as sentinel organisms. Examples are semipermeable membrane devices (SPMDs) of a pure lipid, such as triolein, within a permeable membrane, and solid phase devices such as polyethylene that can be moored at desired places (e.g., Prest et al., 1995; Adams et al., 2007; Morgan and Lohmann, 2008; Bourgeault and Courlay-France, 2013, and references therein). The one significant difference is that bivalves can accumulate OCEC associated with particulate matter due to filter feeding (e.g., Richardson et al., 2001, 2003). Thus, there is something to be gained by using both bivalves and passive samplers in locations where it is feasible (Richardson et al., 2001, 2003).

Lohmann (2012) critically reviews low-density polyethylene use for passive sampling, and details its utility and

underlying science. Lohmann notes that available literature concerning pressure effects on polyethylene suggests that there will be only a small effect of pressure to as great as 5,000 m depth, thus opening the potential utility of polyethylene for sampling in the deep ocean.

## OCEC in Ocean Atmosphere and Ocean Water (Dissolved and Particulate)

Understanding the details of the biogeochemical cycle of OCEC in the world and especially in the ocean—sources to sinks—requires measuring concentrations in the atmosphere over the ocean and in the water column, including particulate matter and dissolved phases (e.g., Jurado et al., 2004; Lohmann et al., 2006). Results of a recent survey of OCEC in the sea surface microlayer and in surface slick samples remind us of the importance of these habitats for understanding the fate and effects of OCEC in the marine environment (Menzies et al.,

2013, and references therein).

Iwata et al. (1993) report on their analyses of air and surface seawater samples collected at the same stations in 1989/1990. They note more uniform concentrations for  $\Sigma$ PCBs when comparing results from mid-latitude and equatorial regions for 1989/1990 compared to earlier measurements in the same places. One potential explanation they offer is a reduction of highly contaminated areas in developed countries and increased use in developing nations. They compare their data with limited data sets for atmospheric  $\Sigma$ PCB concentrations over the ocean for the 1980s and found little decrease. The result for  $\Sigma$ DDT compounds for the same stations demonstrates elevated concentrations for both air samples and surface seawater (Figure 4a,b) in both South and Southeast Asian areas compared to all other regions, consistent with reports of continued use of DDT for antimalarial purposes and crop pest control.

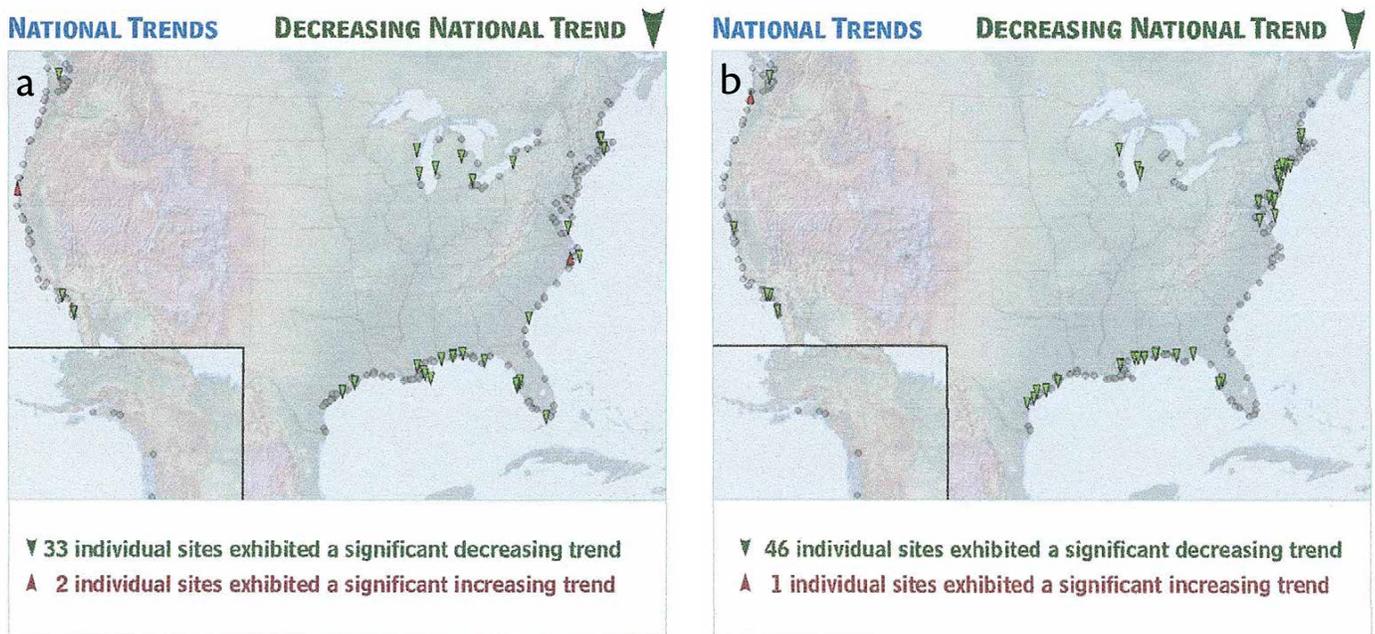


Figure 3. Decreasing or increasing trends for PAH and PCB concentrations in bivalve sentinel organisms for the US coast as reported in the US NOAA NS&T program from 1986 to 2005. (a) PAHs. (b) PCBs. From Kimbrough et al. (2008)

There were significant debates during the 1970s into the early 1980s about the efficacy of obtaining open-ocean and deepwater samples not contaminated by the sampling operation (discussed briefly by Farrington, 2014, in this issue). Schulz-Bull et al. (1991, 1995) proved the possibilities by using a system that

pumped water while underway to obtain up to 1,100 L volume samples about 1 m under the hull of R/V *Poseidon*, transmitted the water through a clean hose to a clean laboratory arrangement, and passed it through a sorbent XAD-2 resin column after filtration to obtain samples for column chromatographic separation

and individual CB analyses by multi-dimensional gas chromatography with electron capture detector. The sums of 31 measured CBs present in significant amounts were  $13.7\text{--}415 \times 10^{-12} \text{ g L}^{-1}$ , with lowest concentrations measured in the northeastern part of the North Sea and the English Channel and highest concentrations in the coastal waters of the German Bight.

The Kiel in situ Pump (KISP) was developed and deployed to sample large volumes of water by in situ filtration of particles and in situ sorption of dissolved OCEC at up to 5,000 m water depth and sampling 2,000 L (Petrick et al., 1996). Sampling during a May 1992 test cruise at three depths in the North Atlantic ( $50^\circ\text{N}$ ,  $21^\circ\text{W}$ ) and in the Skagerrak (north of Denmark between Norway and Sweden) in February 1993 yielded what are arguably the highest quality data obtained until that time for deep waters.

Multiple KISPs were deployed during a cruise to sample northern North Atlantic water masses around Iceland in August 1993 to obtain data for particulate and dissolved CBs (23 individual CBs were quantified) and PAHs (six individual PAHs were quantified; Schulz-Bull et al., 1998). The  $\Sigma\text{CBs}$  concentrations were  $286\text{--}11,241 \times 10^{-15} \text{ g L}^{-1}$ . There appeared to be a consistent decrease from shallower samples to deeper samples. Concentrations in the dissolved phase ranged between  $< 2$  and  $126 \times 10^{-15} \text{ g L}^{-1}$  for individual CBs. The deepest samples at each station had the lowest concentrations. The  $\Sigma\text{CBs}$  concentrations ranged between 10 and  $1,048 \times 10^{-15} \text{ g L}^{-1}$ , and the composition of the CBs in the dissolved phase were similar for all samples. In contrast to findings elsewhere in the Atlantic by these researchers, concentrations of CBs associated with particulate

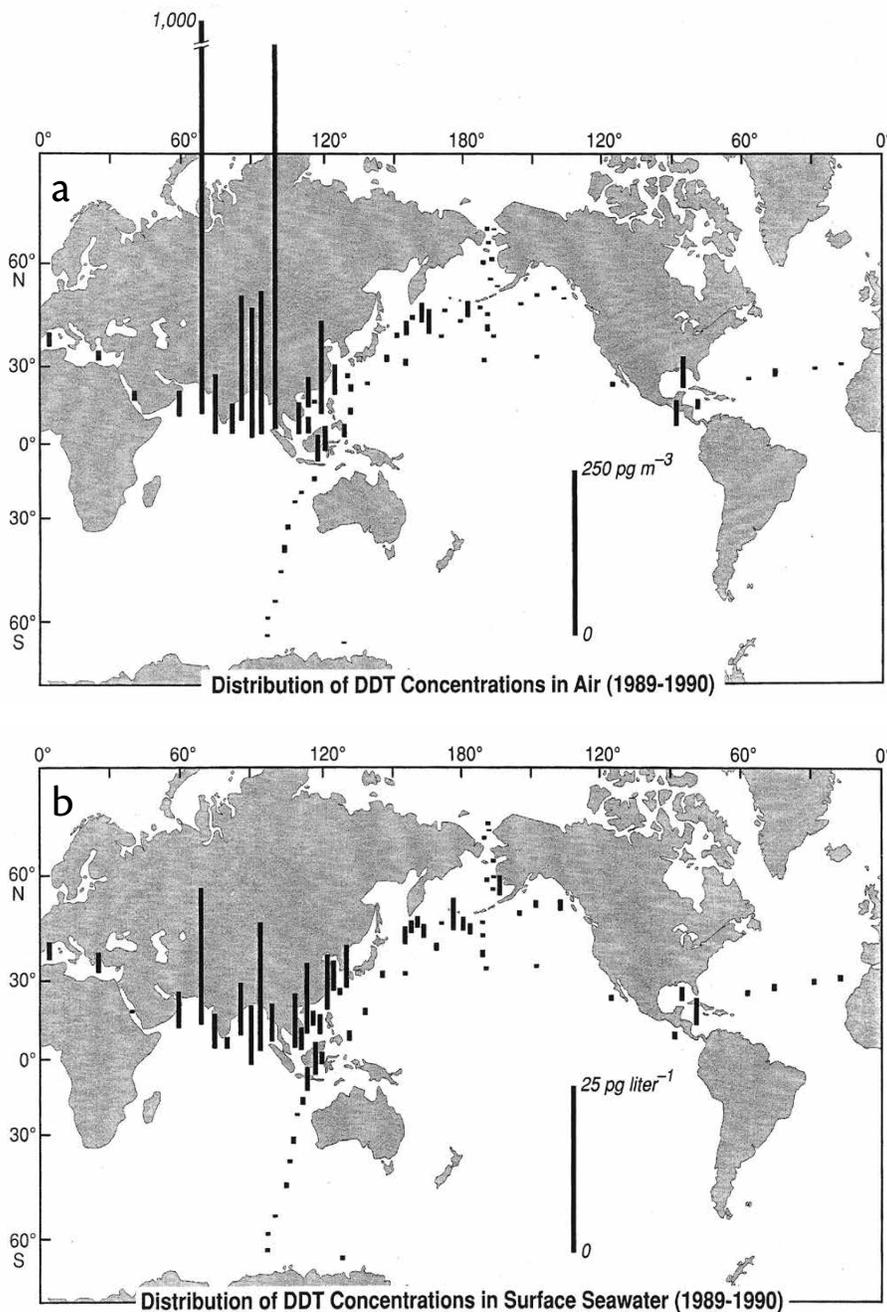


Figure 4. (a) DDT concentrations in air at indicated sampling locations in 1989/1990. (b) DDT concentrations in surface seawater at indicated sampling locations in 1989/1990. From Iwata et al. (1993). Reprinted with permission of the American Chemical Society

matter exceeded concentrations in solution, similar to results for the North and Mediterranean Seas; this is most likely a function of relatively high particulate matter concentrations in northern North Atlantic waters (Schulz-Bull et al., 1998). However, comparison of CB composition in solution and particulate CB composition documented that CBs with lower degrees of chlorination and therefore lower molecular weight (e.g., tri- and tetra-chlorobiphenyls) composed more of the CB mixture in solution compared to the CB mixture associated with particulate matter, consistent with expected partitioning of CBs between dissolved and particulate matter (Schulz-Bull et al., 1998). Regrettably, to our knowledge, these are the few deployments of the KISP (or any other similar device) made to sample the ocean's deeper waters below the mixed layer for determining both dissolved and particulate OCEC concentrations.

#### The Ocean as a Reservoir/Sink for OCEC: Biogeochemical Cycles

The majority of measurements connected with biogeochemical cycles away from the immediate coastal ocean focus on air-sea exchanges or particle fluxes collected in sediment traps or inferred from particulate matter OCEC filtered from seawater samples. An example of an early assessment is that of Burns and Villeneuve (1987), who estimated mass balances for a portion of the Mediterranean ecosystem off the Gulf of Lions. Three important conclusions of their study were: (1) detecting changes in PCB inventories over a reasonable time scale of years would require high-precision sampling and analysis for seawater samples; (2) trends are better assessed by improving estimates of fluxes through ocean systems, especially

air-sea exchange, riverine discharges, particulate fluxes through the water column, and PCB accumulation rates in sediments; and (3) the concentrations of PCBs in the layer of flocculant material at the sediment-water interface were consistent with large particle fluxes as an important removal mechanism from the overlying water column.

Gustaffson et al. (1997) studied the removal of PCBs from surface waters at seven stations in the northwestern Atlantic by measuring CBs concentrations in particulate matter and in cores of surface sediments. The particulate matter concentrations were in the fM (femtomolar) range, which is equivalent to approximately  $0.2\text{--}0.4 \times 10^{-12} \text{ g L}^{-1}$ , and is in the concentration range reported for particulate matter in Baltic Sea (Schulz-Bull et al., 1995) and the North Atlantic Ocean (Schulz et al., 1988). Particle mass fluxes out of the mixed layer were calculated using  $^{238}\text{U}\text{--}^{234}\text{Th}$  disequilibria and then multiplying them by concentrations of CBs associated with particulate matter. Export fluxes from the surface ocean decreased with increasing distance offshore for all CB congeners measured, with the flux of the more particle-reactive, more chlorinated CBs decreasing more rapidly than the flux for the less chlorinated CBs. The cited authors note that although the data set is limited, it indicates that the largest fraction of each CB is removed to the deep open ocean. They estimate that of the 14,000 moles per year of one CB congener (no. 52 – a tetrachlorobiphenyl; see Box 2 for an overview of the CB numbering system) for 1993/1994 sequestered in the northwestern Atlantic Ocean, 75% of the flux was in the pelagic sector, with 20% on the continental shelf and 5% in the 20 km wide coastal

region closest to the continental source (Gustaffson et al., 1997). By back extrapolation through time, they suggest that a significant fraction of PCBs released in the United States may have found their way into the North Atlantic Ocean.

Dachs et al. (1997a) report a budget of organochlorine compounds in the Western Mediterranean. Table 2 summarizes some of their data. They note that residence times estimated from a simple box model of PCBs in open sea surface waters were longer than in continental shelf waters and suggested that this was due to the rapid sinking of particles associated with coastal runoff. The biogeochemical budgets for PCBs and DDTs led the authors to conclude that atmospheric deposition is the primary mechanism for PCB transport to the region, while DDTs come mostly from continental runoff and river inputs. Of importance, Dachs et al. (1997a) note subsurface concentration maxima of PCBs and DDTs in zones of phytoplankton and zooplankton biomass.

A study of PAHs for the Western Mediterranean (Dachs et al., 1997b) found PAH enrichments in deeper samples for profiles collected near submarine canyons and near a sewage disposal zone that they attributed to horizontal advection of particles. PAH residence times were estimated to be 0.95 years for the continental shelf region and 6.5 years for the open sea region. Bouloubasi et al. (2012) report that the shelf sediments in the Gulf of Lions region act as a key sink for PAH inputs that originate mainly from the Rhone River. However, they also report that the shelf is a leaky sink, with transport away from the shelf via submarine canyons toward the slope and open sea amounting to about 21% of the total input to the region. A transport mechanism of this type may result

in sufficient transport of OCEC into mid-depth and deepwater regions of the Mediterranean, which would explain the observation of Marti et al. (2001) that the Mediterranean outflow may be a source of OCEC input to the mid-depths of the northeastern Atlantic. More detailed sampling is required to connect the two observations and test this hypothesis. There is no doubt that there are numerous submarine canyons in the continental margins of the world and that they are active in various types of resuspension and sediment transport from shallow to deep-sea regions (Puig et al., 2014). Deep advective transport of particulate matter has been documented in some areas of the ocean (e.g., Honjo et al., 1982).

The role of particulate matter fluxes as a mechanism for removing POPs from surface waters to deep waters and to surface sediments is well established for OCEC using various example POPs and PAH (e.g., for PAH, see Burns and Villeneuve, 1983; Deyme et al., 2011; Theodosi et al., 2013, and references

therein). Modeling of the PCB concentration in the surface ocean by Dachs et al. (2002) and Jurado et al. (2004) incorporated phytoplankton and mixed layer depth in addition to air-sea exchange from previous models. The results of the modeling's assimilation of some key field data demonstrate the importance of coupling air to water exchange, partitioning of PCBs to organic matter, and removal of PCBs to deeper waters by organic matter flux to the deep ocean. While this may seem of obvious importance to chemical oceanographers studying biogeochemical cycles of other chemicals (see articles in this special issue), prior studies focused mainly on the role of air-sea exchange processes, as noted by Dachs et al. (2002) and Jurado et al. (2004). The modeling suggests complex latitudinal and seasonal variations in both air-sea exchange and removal of PCBs from the surface mixed layer. The importance of phytoplankton blooms to the fate of PAHs was noted by Witt (2002) in a large-scale

study in the Baltic.

Lohmann et al. (2006) addressed the role of oceanic deepwater formation as a sink for POPs. By employing box models and using selected CBs, they determined that deepwater formation removed 420 kg  $\Sigma$ CBs  $\text{yr}^{-1}$  for the Norwegian Sea and 140–160  $\Sigma$ CBs  $\text{kg yr}^{-1}$  for each of the Labrador, Ross, and Weddell Seas. Combining the totals,  $\sim 870 \text{ kg yr}^{-1}$  were removed by deepwater formation, compared to a total  $\sim 320 \text{ kg } \Sigma\text{CBs yr}^{-1}$  removed from surface waters of these deepwater formation regions by CBs associated with sinking particles. Previous studies for the entire Atlantic basin provide an estimate of 22,000 kg  $\Sigma$ CBs  $\text{yr}^{-1}$  of  $\Sigma$ CBs removed from surface ocean waters associated with partitioning to particulate organic matter and sinking of that particulate matter out of the mixed layer (Jurado et al., 2004, 2005). Deepwater formation as a removal mechanism is relatively more important for the more-soluble OCEC (e.g., CB 52 versus CB 180) because they would be removed less efficiently by the particulate organic matter removal process due to less partitioning to the organic matter (Lohmann et al., 2006). In a separate study, Lohmann et al. (2009) measured concentrations of several POPs in the North Atlantic Ocean and Arctic Ocean air-sea boundary layer and surface waters. Among the interesting results was the observation that HCH and HCB are being exported from the Arctic into the North Atlantic Ocean via the East Greenland Current.

### Sediments as Sinks or Leaky Sinks for OCEC

With data collected in estuarine, coastal, and continental shelf areas, decades of research document that sediments are temporary and

Table 2. Budget of organochlorine compounds in the western Mediterranean Sea circa 1993–1994.\*

		Continental Shelf (0–200 m)	Open Sea (0–200 m)	Open Sea (200–1,900 m)
Surface (km <sup>2</sup> )		1.3 x 10 <sup>5</sup>	7.2 x 10 <sup>5</sup>	7.2 x 10 <sup>5</sup>
Volume (km <sup>3</sup> )		2.6 x 10 <sup>4</sup>	1.44 x 10 <sup>5</sup>	1.22 x 10 <sup>6</sup>
<b>Concentrations (10<sup>-12</sup> g L<sup>-1</sup>)</b>				
PCB	Particulate matter	13	6	6
	Dissolved	60	36	6
DDT	Particulate matter	1	0.3	0.09
	Dissolved	4	1.2	0.36
<b>Budget (kg)</b>				
PCB	Particulate matter	338	864	1,224
	Dissolved	1,560	5,184	7,344
DDT	Particulate matter	26	43.2	110
	Dissolved	104	172.8	440

\*Reformatted from Dachs et al. (1997a)

long-term sinks for OCEC (Figure 1), for example, PAH data summarized by Saha et al. (2009; Figure 5). PAH concentrations for 174 locations in India, Indonesia, Malaysia, Thailand, Vietnam, Cambodia, and Laos were high in or near urban areas (e.g.,  $\Sigma$ PAHs  $\sim 1,000\text{--}10,000 \times 10^{-9} \text{ g g}^{-1}$  dry weight), contrasted with much lower concentrations (e.g.,  $\Sigma$ PAHs  $\sim 10\text{--}100 \times 10^{-9} \text{ g g}^{-1}$  dry weight) near rural coastal locations.

Compilation and interpretation of data from 4,214 distinct continental shelf sediments around the globe by Jönsson et al. (2003) provide an estimate of global inventories and also of inventories divided among 18 different continental shelf areas for eight CB congeners common to all samples in the inventory. Table 3 provides a compilation of estimated burial flux for four CBs. About 80% of estimated historical heptachlorobiphenyl CB 180 global emissions are deposited in continental shelf sediments. Less than 10% of CB 28 trichlorobiphenyl emissions and less than 50% of tetrachlorobiphenyl emissions are deposited and accumulated in continental shelf sediments.

The presence of OCEC in sediment, as determined by extraction and chemical analyses, does not mean that they are either sequestered forever in the sediments or that they are biologically available. Resuspension and transport to other locations is one mechanism for moving the contaminated sediments and the associated OCEC, as noted earlier in the discussion about transport to deeper waters via submarine canyons. In addition, OCEC can partition from the solid phase to dissolved and colloidal phases of interstitial water in sediments, with the potential for release to the overlying water or biological uptake by benthic infauna and

epifauna, as shown in Figure 1 (see Schwarzenbach et al., 2003).

A large deposit of DDT and its transformation product pollutants (mainly DDE) accumulated in the continental margin area of California's Palos Verdes shelf as a result mainly of DDT manufacturing wastes discharged to a large municipal sewer system that, in turn, discharged to the coastal ocean. Studies of this site demonstrate in a first-order

manner how legacy chemicals deposited in sediments can be continuing sources of contamination or pollution for higher trophic levels such as birds and sea lions (see papers in Lee and Wiberg, 2002). Collectively, the authors of these papers modeled, on a quantitative or semi-quantitative basis, processes depicted in Figure 1 for DDT and DDE.

There is an issue that needs resolution by further field campaigns. Continental

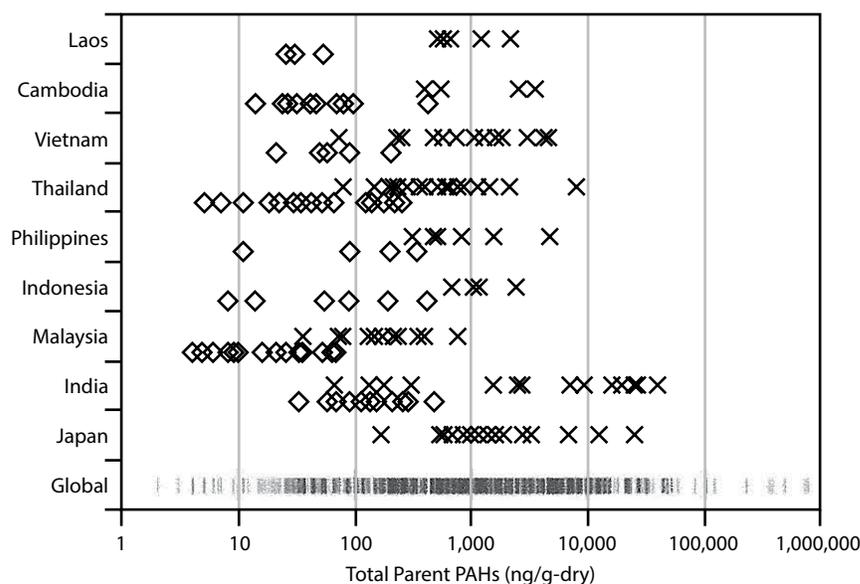


Figure 5. Compilation of concentrations of parent PAHs in surface sediments sampled in South Asia and Southeast Asia compared to a worldwide compilation. Cross = urban locations. Diamond = rural locations. Short vertical line = global locations. (Boonyatumanond et al., 2006). Summed PAHs are phenanthrene, anthracene, fluoranthene, pyrene, benzo[a]anthracene, chrysene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[e]pyrene, benzo[a]pyrene, indeno[1,2,3-cd]pyrene, benzo[ghi]perylene, and coronene. From Saha et al. (2009), reprinted with permission

Table 3. Global inventories and burial fluxes of selected CBs in continental shelf sediments (from Jönsson et al., 2003). Reprinted with permission of the American Chemical Society

	CB 28	CB 52	CB 153	CB 180
Number of Chlorines	3	4	6	7
Global Inventory (ton)	460 (290–910)*	700 (420–1,500)*	1,200 (720–2,100)*	760 (380–2,220)*
Global Burial Flux (ton yr <sup>-1</sup> )	9.6 (3.9–20)*	14 (5.1–31)*	24 (8.8–47)*	15 (4.7–46)*

\* Lower and upper 95% confidence limits

shelf and continental margin sediments where high OCEC concentrations are present may be resuspended and then transported and deposited in deep waters where OCEC concentrations are low compared to the shelf and upper margin waters. OCEC could then desorb from the sediments and enter the dissolved phase in the mid-depth or deep-water regions. The mid-depth and deep waters in turn advect to the continental shelf and boundary regions of lower OCEC concentrations where sediment resuspension may result in sorption of the dissolved phase OCEC and incorporation into shelf and slope sediment in a process akin to the chemical scavenging process described by Bacon et al. (1976). There are insufficient data at this time to resolve this issue because of the lack of sufficient deepwater concentration profiles of particulate and dissolved phase OCEC.

#### Metabolism and Biodegradation of OCEC

Several of the OCEC, while persistent relative to many other organic chemicals such as proteins, amino acids, and lipids, can be degraded slowly by microorganisms and also metabolized by biota such as fish, crustacea, and marine mammals. Individual chemicals in PCBs and PAHs can exhibit differential rates of degradation and metabolism depending on their exact chemical structure. These processes transform the PCBs and PAHs entering the ocean to different mixtures depending on the types and locations of samples, for example, fish compared to shellfish compared to sediment. Further discussion is beyond the scope of this paper, and the reader is referred to Dawe and Stegeman (1991), Leahy and Colwell (1990), and Abramowicz (1990), among others, for beginning reading.

#### FLOATING DEBRIS: PLASTICS

Floating debris, including relatively large pieces of plastic, discarded fishing nets, and garbage that are mostly made up of organic chemicals, have been recognized in the surface waters of the North Atlantic and Pacific open ocean gyres and along beaches for decades (Carpenter and Smith, 1972; Venrick et al., 1973; NRC, 2008). The aesthetic impacts on the sea surface, beaches, and shorelines are real. Teuten et al. (2009) note concern about plastics as a mechanism for transporting hydrophobic contaminants (e.g., OCEC) and transferring them to marine life, including birds. Derraik (2002) reviews other concerns in terms of impact on marine organisms, such as entanglement in discarded fishing nets and ingestion of plastics, and, more generally, the broad distribution of plastics on the ocean surface. Recent studies indicate that marine plastics bring OCEC such as PBDEs (polybrominated diphenyl ethers) directly to higher-trophic level organisms where they are found, for example, in the tissues of oceanic seabirds (Tanaka et al., 2013). Renewed interest in floating debris in open ocean gyres was stimulated by quantitative observations of plastics in the North Pacific Gyre (e.g., Moore et al., 2001) and spawned the term “ocean garbage patch” (Kaiser, 2010). The explosion of debris in the North Pacific Ocean resulting from the March 2011 Tōhoku tsunami reinforced current interest in the origins and fates of floating debris of all sizes (Bagulayan et al., 2012).

The early studies by Carpenter and Smith (1972) and Venrick et al. (1973) note the presence of small plastic spherules collected in neuston nets. Recently, there has been a resurgence of research on and assessment of the presence of

plastic debris, including plastic spherules or “pellets” in the ocean (e.g., Morét-Ferguson et al., 2010; Doyle et al., 2011). The Morét-Ferguson et al. data on plastic fragments of various sizes were collected on the same expeditions as those for floating tar reported by Peters and Suida (2014, in this issue). The largely polypropylene and polyethylene particles were mostly less than 10 mm in size, with densities ranging from 0.808 to 1.24 g ml<sup>-1</sup>. The densities of the pelagic plastic particles varied considerably from densities reported for beached plastic particles or pellets. A “Pellet Watch” has been launched (see Box 3) to further enhance this work.

#### SUMMARY AND NEXT STEPS

The research to date and assessments of POPs and PAH provide a reasonable basis for developing further understanding of the inputs, cycling, and fates of OCEC, including both legacy and emerging contaminant chemicals. The coastal ocean and interactions at the atmosphere/surface ocean interface are much better understood than are continental margin waters and the open ocean below the mixed layer.

The response time of the nearshore coastal ocean to legacy pollutants such as PCBs and DDT as assessed by bivalve sentinel organism programs and measurements in sediments confirms fears and predictions of the late 1960s and the 1970s that these POPs would persist in such ecosystems for many decades, if not a century or more. Ross et al. (2009) provide an example of the growing concerns regarding emerging contaminants (or perhaps they should now be designated emerged contaminants/pollutants) for polybrominated diphenyl ethers in the Canadian environment.

There is continuing and intensifying



concern about the atmospheric and oceanic processes that transport POPs and possibly other OCEC to places far removed from sources of input, such as the polar regions, especially the Arctic. We have provided only brief reference to this topic and refer the reader to Bidleman et al. (2005) for a compilation of papers on the subject.

Several emerging or emerged OCEC are not as hydrophobic as the POPs and PAH, and their possible differences from known rates of OCEC solubility in seawater and partitioning between dissolved and particulate phases should be taken into account in studies of biogeochemical cycles in the marine environment (see Field et al., 2006b).

While the technology exists to obtain high-quality water samples for dissolved and particulate OCEC measurements in the deep sea, very few measurements have been made even though the deep ocean appears to be a large sink for OCEC on time scales of centuries or longer. This is especially true if we consider deposition and accumulation in sediments and interaction with overlying deep waters. Vertical transport processes involving large and small particulate matter remove OCEC from the surface waters to the deep ocean. There are indications that horizontal transport from continental shelf and slope areas, especially via submarine canyons, may transport OCEC to the deep ocean. The quantitative importance of such processes for regional- and ocean-scale biogeochemical balances of OCEC remains poorly understood.

The lack of detailed hydrographic profiles for these POPs and other OCEC compared to those obtained for elements as reported elsewhere in this special issue needs to be rectified. Surely, if bright minds can develop devices to

sample and measure toxins and microbes in the field (Scholin et al., 2009), and in situ mass spectrometers can be deployed on autonomous underwater vehicles to detect aromatic hydrocarbons such as benzene and naphthalene (Camilli et al., 2010), then the development and deployment of new field sampling and measurement devices and sensors for OCEC is also feasible and in order.

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