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Temporal trends and spatial distribution of DDT in bivalves from the coastal marine environments of the continental United States, 1986–2009

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ABSTRACT

Analysis of DDT isomers and breakdown products, DDD and DDE, in over 3500 bivalve samples collected from more than 300 locations along the continental United States indicates that concentrations are decreasing. Overall average concentrations for the East (45.8 ± 24.6 ng/g dw), Gulf (42.4 ± 21.1 ng/g dw), and West (90.9 ± 43.3 ng/g dw) coasts are declining with an environmental half-life between 10 and 14 years and are predicted to decrease below 10% of today's concentrations by 2050. Geographically, areas with high and low levels are well identified. Bivalves yielding the highest concentrations were collected in areas linked to areas of DDT production or heavy usage. These areas are clustered in the southern California and San Francisco area, on the West coast; Delaware and Hudson/Raritan Estuary, on the East coast; and in Alabama and northwestern Florida, on the Gulf of Mexico. Statistically significant decreasing trends in ΣDDT concentrations are apparent at most of these locations.

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1. Introduction

DDT (1,1,1-trichloro-2,2′bis(*p*-chlorophenyl)ethane or dichlorodiphenyl-trichloroethane) was one of the most widely used pesticides in the United States and worldwide for many years. Originally synthesized by Austrian chemist Othmar Zeiler in 1874; its insecticidal properties were not discovered until 1939 by Swiss chemist Paul H. Muller, which resulted in his award of the 1948 Nobel Prize in Medicine. Its attractiveness was largely due to its reasonable production cost, effectiveness, persistence, and versatility. In the United States, DDT production steadily increased after 1945 and peaked at 80,000 metric tons in 1963 (Cheremisinoff and Rosenfeld, 2011). Increasing pest resistance and the development of alternative pesticides, paired with growing public concern regarding adverse side effects [e.g., bird egg shell thinning (Vos et al., 2000)] and government restrictions to its use, resulted in the decline in the popularity and usage of DDT. DDT production decreased to just 5500 metric tons in early 1970s. It was the chemical stability and persistence in the environment, the basis of public concerns, which led to the final prohibition, effective December 31st, 1972, of all applications of DDT in the United States (U.S. EPA, 1975). The ban imposed onto the use of DDT in the United States did not affect its export and international application. Similarly, and in spite of the regulatory actions that led to the ban of DDT, exemptions for its application in U.S. crops were granted in many instances when deemed as economical emergencies in subsequent years (U.S. EPA, 1975). After 1972, the use of DDT in other developed countries also declined gradually but routine use continued in developing countries, mainly for malaria control. In 2001, the Stockholm Convention included DDT as one of 12 persistent organic pollutants to be banned worldwide to reduce the risks to human health and the environment (UNEP, 2001). Consequently, Parties to the Convention can only use DDT for "disease vector control" under strict guidelines for use and in accordance with the World Health Organization (WHO) recommendations (UNEP, 2001). In 2009, the Conference of the Parties to the Stockholm Convention ratified this action and concluded that "countries that are currently using DDT for disease vector control may need to continue such use until locally appropriate and cost-effective alternatives are available for sustainable transition away from DDT" (UNEP, 2009). At least, 15 countries in Asia, Pacific and Africa currently use DDT for vector control (UNEP, 2009). A few others are reevaluating or are in the

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process of policy change on the application of DDT (Longnecker, 2005). Until recently, only India, China, and DPR Korea were producing DDT (UNEP, 2009).

In 1984, the National Oceanic and Atmospheric Administration (NOAA) created the National Status and Trends (NS&T) Program to assess the status and long term trends of selected contaminants, including isomers of DDT and breakdown products DDD and DDE, in coastal marine environments. This was done in response to public and scientific concerns regarding the quality of these environments and the lack of systematic monitoring programs in the U.S. In 1986, under the umbrella of the NS&T Program, the "Mussel Watch" Project initiated a yearly nationwide sampling effort to collect bivalves from pre-established sites along the East, Gulf, and West coasts to describe the spatial distribution of selected organic and inorganic contaminant concentrations and their temporal changes at both regional and national scales. During the following years, the NS&T Program added sampling locations in Alaska, Hawaii, Puerto Rico and the Great Lakes. The approach of the "Mussel Watch" Project echoed the idea, introduced by Professor Edward Goldberg, of proposing a mussel watch monitoring program using *Mytilus edulis*, or similar species, as sentinel organism (Goldberg, 1975; Goldberg et al., 1978). The rationale behind Goldberg's idea has been extensively discussed by many authors (e.g., Phillips and Rainbow, 1993; O'Connor et al., 1994; de Kock and Kramer, 1994; Sericano, 2000) and successfully implemented in many monitoring programs worldwide (e.g., Besada et al., 2011; Boonyatumanond et al., 2002; Monirith et al., 2003; O'Connor, 2002; Sericano et al., 1995; Sericano, 1993; Sukasem and Tabucanon, 1993). Presently, the NOAA's "Mussel Watch" project is the longest, continuous coastal monitoring program that is national in scope. This paper provides (1) an overview of the concentrations of DDT isomers and their metabolites, DDD and DDE, measured in bivalves collected from the continental United States coastal regions between 1986 and 2009, (2) a discussion of observed concentration trends, and (3) a prediction for a 90% reduction of today concentrations.

2. Methods

2.1. Sampling

Detailed descriptions of "Mussel Watch" Project sampling locations and rationale for their selection have been provided in several NOAA's publication (Lauenstein and O'Connor, 1988; Lauenstein and Cantillo, 1993; Kimbrough et al., 2008); therefore, a brief summary follows. The overall sampling scheme contemplates the annual collection of bivalves from indigenous populations, within a target size range, at the same location and similar annual timeframe. These sampling locations, roughly separated by 10–20 km in partially enclosed coastal areas and by 70–100 km in open coastlines, were selected to provide samples representative of contaminant accumulation from nearby or surrounding areas outside of known contaminant point sources. Sampling efforts were generally conducted from December to February when bivalves are less reproductively active. The sampling scheme experienced a few changes over the years. Originally, 20–30 (oysters or mussels, respectively) bivalves were collected from three stations, approximately 100–1000 m apart (at intertidal sites), within each selected site and composites from each station analyzed individually; since 1992, only one composited sample from each location has been collected and analyzed. The initial 145 locations, sampled in 1986 and 1987, on the East, Gulf, and West coasts, were augmented with new sites that were added during the subsequent years to either fill in large spatial gaps or to gather data closer to urban centers. Similarly, locations in Alaska, Great lakes, Puerto Rico and Hawaii were also added. From 1986 to 2009, over 300 different sites have been sampled to present, with the majority of them located along the East, Gulf, and West coasts (Fig. 1). After 1995, not all the sites were sampled every year; most sites were collected in alternate years. Because of the large area covered by the "Mussel Watch" Project, latitude plays a very important role in the distribution of species; thus, different bivalve species have been collected along the East (*M. edulis*, from Maine to Cape May, New Jersey and *Crassostrea virginica* from Delaware Bay southward), Gulf (*C. virginica*), West (*Mytilus galloprovincialis*, *Mytilus trossulus*, and *Mytilus californianus*), Alaska (*Mytilus species*), and Great Lakes (*Dreissena species*) coasts (Fig. 2). In spite of species differences, the analysis of different bivalve species that co-exist at some locations revealed comparable concentrations for all organic compounds (O'Connor, 2002).

2.2. Analytical procedures

The analytical procedure used by the Geochemical and Environmental Research Group (GERG), at Texas A&M University, and TDI Brooks for the extraction, fractionation and cleanup of DDT, DDD, and DDE isomers in bivalve samples followed, with some minor modifications, a method developed for the "Mussel Watch" Project (MacLeod et al., 1985) and described in more detail elsewhere (e.g., Sericano et al., 1990a,b; Kimbrough and Lauenstein, 2006; Kimbrough et al., 2006). GERG used approximately 15 g of wet tissue that were spiked with 4,4′ dibromooctafluorobiphenyl (DBOFB), PCB 103 and PCB 198, as surrogates standards, extracted with methylene chloride after the addition of anhydrous Na_2SO_4,

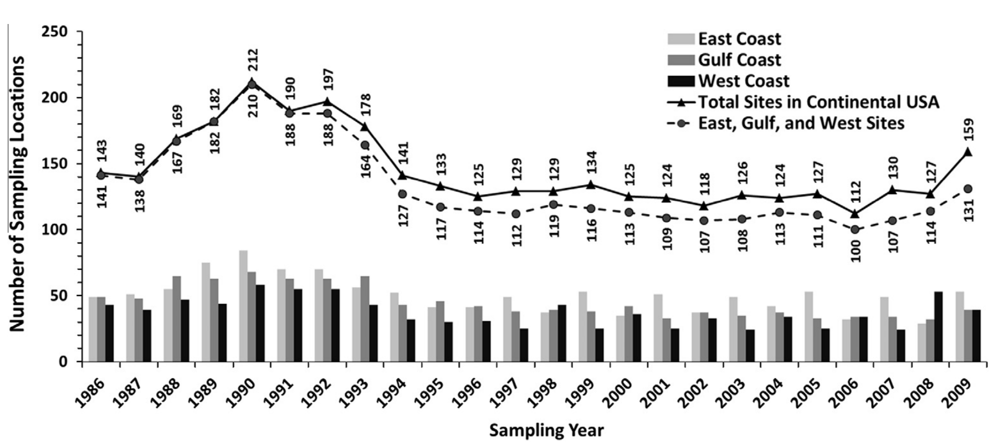

Fig. 1. Samples collected annually from 1896 to 2009 during the NOAA's national status and trends program.

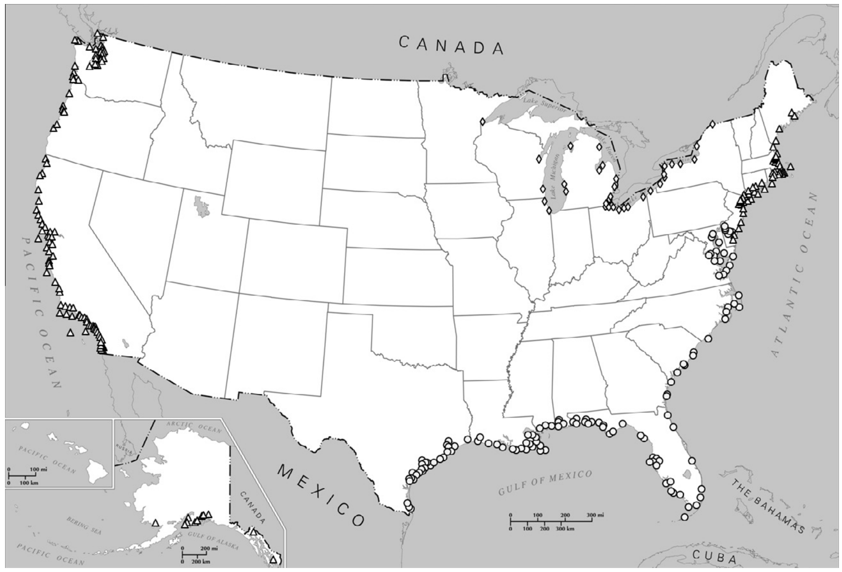

Fig. 2. Distribution of oysters (○, *Crassostrea virginica*) mussels (Δ, *Mytilus species*), and zebra mussels (◇, *Dreissena species*) collected and measured as part of the mussel watch program.

and homogenized. An automated extraction apparatus (Dionex ASE200 Accelerated Solvent Extractor) was used at TDI Brooks to extract organic contaminants from 0.5 to 15 g of wet tissue sample mixed with Hydromatrix, or an equivalent mass of dry tissue, after spiking with the same surrogate standards. The extractions were performed using 100% dichloromethane inside stainless-steel extraction cells held at elevated temperature and pressure. The extracted compounds dissolved in the solvent were collected in 60-mL glass vials. Extracts are concentrated to a volume of 1–3 mL, using an evaporative solvent reduction apparatus (Zymark TurboVap II or water bath). In both laboratories, an aliquot was removed for gravimetrical determination of the percent lipid and the remaining portion of the concentrated extracts purified. The extracts were initially cleaned using column chromatography (partially deactivated silica gel:alumina column chromatography using a 1:1 mixture of pentane and methylene chloride as eluent) and further purified by gel permeation chromatography to remove compounds that would interfere with the gas chromatography in the determinative step of DDT, DDD, and DDE isomers along with several other chlorinated pesticides. DDT, DDD, and DDE isomers present in a final 1 mL sample extract were quantitatively analyzed by gas chromatography with electron capture detection (GC-ECD, ^{63}Ni) using a Hewlett Packard 5890 Series II Gas Chromatograph. The sample extracts were injected in the splitless mode into a 30 m × 0.25 mm i.d. (0.25 μm film thickness) DB-5 fused silica capillary column (J&W Scientific, Inc.), or similar, at an initial temperature of 100 °C, held for 1 min and ramped to 140, 250, and 300 °C at 5, 1.5, and 10 °C min^{-1}, respectively. The oven was held at the higher temperature for 5 min for a total run time of 90.33 min. Injector temperature was maintained at 275 °C. Prior to or during each analytical run, the instrument was calibrated by injection of standard mixtures at four different concentrations. 2,4,5,6-tetrachloro-*m*-xylene (TCMX) was used as internal standard. Calibration solutions were prepared at five concentrations ranging from 5 to 200 (GERG) and 5 to 500 ng/mL (TDI Brooks) by diluting commercially available solutions containing the analytes of interest. Calibration curves are established by analyzing each of 5 calibration standards. A typical analytical set consisted of standards, samples, and quality control samples which included some or all of the following: method blank, duplicate, matrix spike, matrix spike duplicate or blank spike, blank spike duplicate, and standard reference material. As part of the NS&T program, both analytical laboratories (i.e., GERG and TDI Brooks) have participated in several laboratory intercalibration exercises and maintain strict Quality Assurance/Quality Control (QA/QC) activities to ensure that the data produced are reproducible, accurate, and comparable.

2.3. Data analysis

The lack of differences in the concentrations of organic contaminants between bivalve species exposed to the same environmental contaminant levels allows for data comparison without distinctions regarding the sampled species. A simple linear regression was used to examine the relationship between dependent and independent variables and to determine if the probability that the observed correlation coefficient occurred by chance. The significance of the slope of the regression line was determined from the *t*-statistic test at the 0.05 and 0.10 critical alpha levels. Half-lives discussed in the text were calculated from the exponential decay in concentration following the single compartment model (Sericano and Wade, 2011) in which a transformation from a linear to logarithmic scale is necessary to calculate the time required for a decaying quantity to fall to one half of its initial value.

3. Results and discussion

Since 1986, and for the next 23 years, *o*-*p*′ and *p*-*p*′ isomers of DDD, DDE, and DDT were analyzed in over 3500 bivalve samples collected from over 300 different locations along the East, Gulf, West, including Alaska, Hawaii, Puerto Rico and Great Lakes coastlines. For the purpose of this discussion, Alaskan and West coast data are kept separated. Data from Hawaii and Puerto Rico locations are not included in this discussion. Tables 1–3 summarize the number of sites sampled and analyzed each year since 1985 as well as average and median concentrations and ranges for each target analyte. In spite of the ban on the use of DDT in the United States in 1972, DDTs isomers and related metabolites are still ubiquitous contaminants in bivalves from coastal areas and they have been detected in virtually every sample collected since the beginning of the MW project.

Table 1
DDD, DDE, and DDT isomer concentrations (ng/g dw) in East coast bivalves.

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
1986 (n = 49)				*1995 (n = 41)*			
o,p′ DDD	2.23	22.4 ± 63.0	<MDL–407	*o,p′* DDD	3.80	5.22 ± 5.47	0.60–22.6
p,p′ DDD	23.7	36.5 ± 50.8	<MDL–297	*p,p′* DDD	7.75	11.7 ± 12.4	1.44–55.6
o,p′ DDE	2.97	10.9 ± 17.5	<MDL–90.7	*o,p′* DDE	0.69	1.73 ± 2.61	<MDL–11.0
p,p′ DDE	21.3	27.9 ± 35.2	<MDL–220	*p,p′* DDE	17.3	21.4 ± 16.8	3.99–71.3
o,p′ DDT	<MDL	2.89 ± 6.25	<MDL–29.0	*o,p′* DDT	1.09	1.74 ± 1.72	<MDL–8.05
p,p DDT	<MDL	5.46 ± 12.9	<MDL–66.7	*p,p* DDT	0.89	1.82 ± 2.22	<MDL–8.98
ΣDDTs	65.5	106 ± 164	2.76–1110	ΣDDTs	34.0	43.6 ± 39.1	9.14–175
1987 (n = 51)				*1996 (n = 41)*			
o,p′ DDD	1.27	4.96 ± 8.60	<MDL–38.7	*o,p′* DDD	3.13	5.03 ± 5.25	<MDL–27.7
p,p′ DDD	24.7	46.9 ± 49.1	<MDL–250	*p,p′* DDD	5.45	8.13 ± 8.21	<MDL–34.9
o,p′ DDE	<MDL	5.18 ± 12.4	<MDL–56.7	*o,p′* DDE	1.00	1.69 ± 2.40	<MDL–11.4
p,p′ DDE	35.3	46.5 ± 46.1	6.30–253	*p,p′* DDE	12.6	17.5 ± 16.0	<MDL–63.4
o,p′ DDT	<MDL	1.65 ± 4.23	<MDL–23.3	*o,p′* DDT	1.67	2.67 ± 4.49	<MDL–27.3
p,p DDT	6.43	10.3 ± 14.1	<MDL–55.0	*p,p* DDT	1.36	1.98 ± 2.09	<MDL–7.76
ΣDDTs	83.4	116 ± 109	9.70–548	ΣDDTs	26.2	37.0 ± 33.5	2.42–139
1988 (n = 55)				*1997 (n = 49)*			
o,p′ DDD	3.87	6.33 ± 7.77	<MDL–36.3	*o,p′* DDD	2.31	7.73 ± 12.5	<MDL–63.7
p,p′ DDD	9.43	21.8 ± 32.7	<MDL–170	*p,p′* DDD	5.40	13.8 ± 21.8	<MDL–104
o,p′ DDE	<MDL	1.81 ± 3.74	<MDL–16.67	*o,p′* DDE	<MDL	1.53 ± 4.00	<MDL–19.1
p,p′ DDE	17.7	28.9 ± 27.5	2.63–141	*p,p′* DDE	11.0	21.2 ± 23.3	2.06–111
o,p′ DDT	<MDL	1.13 ± 2.46	<MDL–10.2	*o,p′* DDT	0.70	1.96 ± 2.98	<MDL–11.6
p,p DDT	2.73	3.65 ± 5.12	<MDL–26.3	*p,p* DDT	2.79	4.28 ± 4.96	<MDL–23.0
ΣDDTs	43.0	63.6 ± 73.2	5.36–383	ΣDDTs	23.0	50.5 ± 66.5	5.72–319
1989 (n = 75)				*1998*			
o,p′ DDD	1.27	3.61 ± 4.06	<MDL–13.3	*o,p′* DDD	3.07	4.59 ± 4.49	<MDL–19.6
p,p′ DDD	12.3	23.6 ± 45.6	<MDL–347	*p,p′* DDD	5.54	8.21 ± 7.52	<MDL–37.1
o,p′ DDE	<MDL	3.80 ± 6.96	<MDL–29.7	*o,p′* DDE	1.13	1.50 ± 2.09	<MDL–10.7
p,p′ DDE	27.7	39.3 ± 51.0	5.46–310	*p,p′* DDE	13.4	23.2 ± 32.1	<MDL–194
o,p′ DDT	2.47	5.86 ± 17.0	<MDL–143	*o,p′* DDT	1.84	2.30 ± 2.57	<MDL–13.0
p,p DDT	<MDL	2.89 ± 4.61	<MDL–21.3	*p,p* DDT	1.18	1.86 ± 3.21	<MDL–19.1
ΣDDTs	46.8	79.0 ± 108	6.76–722	ΣDDTs	30.8	41.7 ± 44.7	0.83–264
1990 (n = 84)				*1999*			
o,p′ DDD	1.04	2.63 ± 3.82	<MDL–19.2	*o,p′* DDD	2.01	3.29 ± 4.66	<MDL–28.3
p,p′ DDD	9.49	18.7 ± 25.4	<MDL–136	*p,p′* DDD	3.22	6.48 ± 11.4	<MDL–74.4
o,p′ DDE	1.26	3.02 ± 5.01	<MDL–24.6	*o,p′* DDE	0.98	1.38 ± 1.78	<MDL–9.63
p,p′ DDE	21.3	33.4 ± 34.9	4.35–212	*p,p′* DDE	11.5	13.7 ± 11.6	1.83–59.6
o,p′ DDT	3.50	5.44 ± 5.78	<MDL–34.8	*o,p′* DDT	<MDL	0.93 ± 1.06	<MDL–3.88
p,p DDT	2.16	3.19 ± 4.51	<MDL–28.4	*p,p* DDT	0.86	1.14 ± 1.04	<MDL–4.11
ΣDDTs	40.3	66.5 ± 74.5	4.35–456	ΣDDTs	20.2	27.0 ± 29.2	2.83–175
1991 (n = 70)				*2000*			
o,p′ DDD	<MDL	1.13 ± 2.51	<MDL–13.0	*o,p′* DDD	2.40	3.91 ± 4.18	<MDL–19.8
p,p′ DDD	3.99	8.46 ± 14.7	<MDL–94.7	*p,p′* DDD	3.57	5.25 ± 5.33	<MDL–23.6
o,p′ DDE	<MDL	1.56 ± 3.13	<MDL–15.5	*o,p′* DDE	0.55	0.99 ± 1.09	<MDL–4.40
p,p′ DDE	12.2	19.7 ± 23.2	<MDL–115	*p,p′* DDE	7.66	11.9 ± 10.4	<MDL–41.2
o,p′ DDT	<MDL	1.31 ± 2.80	<MDL–17.0	*o,p′* DDT	0.86	1.46 ± 2.03	<MDL–9.51
p,p DDT	<MDL	1.54 ± 4.03	<MDL–24.4	*p,p* DDT	<MDL	0.80 ± 0.98	<MDL–3.82
ΣDDTs	19.2	33.7 ± 45.1	2.46–248	ΣDDTs	15.6	24.3 ± 21.5	<MDL–86.4
1992 (n = 70)				*2001*			
o,p′ DDD	2.07	4.02 ± 5.08	<MDL–21.8	*o,p′* DDD	2.05	3.96 ± 5.65	<MDL–27.4
p,p′ DDD	4.62	8.71 ± 10.7	<MDL–54.0	*p,p′* DDD	3.06	7.10 ± 11.7	<MDL–56.2
o,p′ DDE	0.80	2.02 ± 6.45	<MDL–52.1	*o,p′* DDE	<MDL	1.19 ± 2.57	<MDL–12.7
p,p′ DDE	15.5	20.4 ± 17.5	<MDL–74.6	*p,p′* DDE	8.15	11.0 ± 11.2	<MDL–49.6
o,p′ DDT	0.85	1.83 ± 3.02	<MDL–19.8	*o,p′* DDT	0.69	1.31 ± 1.78	<MDL–9.48
p,p DDT	<MDL	1.37 ± 3.37	<MDL–19.6	*p,p* DDT	0.51	1.24 ± 1.83	<MDL–8.35
ΣDDTs	26.3	38.3 ± 36.0	0.51–170	ΣDDTs	14.5	25.9 ± 33.1	0.92–156
1993 (n = 56)				*2002*			
o,p′ DDD	3.66	5.45 ± 6.09	<MDL–32.3	*o,p′* DDD	2.04	3.82 ± 5.91	<MDL–29.6
p,p′ DDD	7.14	10.3 ± 10.2	<MDL–44.2	*p,p′* DDD	2.87	4.81 ± 6.81	<MDL–37.9
o,p′ DDE	1.26	2.15 ± 4.44	<MDL–32.3	*o,p′* DDE	<MDL	0.95 ± 1.40	<MDL–6.39
p,p′ DDE	15.6	21.4 ± 19.4	2.50–93.2	*p,p′* DDE	8.88	10.6 ± 9.95	<MDL–47.6
o,p′ DDT	3.04	4.99 ± 6.03	<MDL–37.2	*o,p′* DDT	0.94	2.08 ± 4.27	<MDL–21.3
p,p DDT	1.66	3.48 ± 5.77	<MDL–40.8	*p,p* DDT	0.52	0.79 ± 1.18	<MDL–6.76
ΣDDTs	34.9	47.7 ± 41.0	2.50–178	ΣDDTs	16.7	23.0 ± 24.9	0.70–117
1994 (n = 52)				*2003*			
o,p′ DDD	3.30	5.83 ± 6.26	<MDL–28.3	*o,p′* DDD	2.30	4.08 ± 6.27	<MDL–32.4
p,p′ DDD	7.37	12.4 ± 15.7	<MDL–83.5	*p,p′* DDD	2.41	7.22 ± 15.4	<MDL–91.4
o,p′ DDE	0.98	1.60 ± 2.37	<MDL–12.6	*o,p′* DDE	<MDL	0.96 ± 2.20	<MDL–11.3
p,p′ DDE	19.2	26.4 ± 27.7	<MDL–146	*p,p′* DDE	9.58	15.0 ± 25.7	0.51–173
o,p′ DDT	<MDL	1.05 ± 3.53	<MDL–21.0	*o,p′* DDT	1.24	1.95 ± 2.49	<MDL–12.0
p,p DDT	0.72	3.12 ± 6.24	<MDL–30.2	*p,p* DDT	1.21	2.35 ± 3.87	<MDL–22.2
ΣDDTs	32.2	50.4 ± 52.3	1.66–235	ΣDDTs	19.0	31.5 ± 46.7	1.30–241

Table 1 (*continued*)

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
2004				*2007*			
o,p′ DDD	3.41	5.63 ± 6.85	<MDL–39.7	*o,p′* DDD	1.81	4.75 ± 11.3	<MDL–64.4
p,p′ DDD	3.02	7.04 ± 7.86	<MDL–32.4	*p,p′* DDD	3.67	10.8 ± 25.7	<MDL–131
o,p′ DDE	0.67	1.15 ± 1.29	<MDL–4.22	*o,p′* DDE	<MDL	1.77 ± 4.36	<MDL–24.4
p,p′ DDE	9.85	13.8 ± 11.7	<MDL–42.8	*p,p′* DDE	13.4	19.0 ± 23.3	1.30–152
o,p′ DDT	1.40	2.28 ± 3.29	<MDL–18.7	*o,p′* DDT	0.83	1.18 ± 1.21	<MDL–6.32
p,p DDT	0.66	0.98 ± 1.29	<MDL–7.71	*p,p* DDT	0.73	1.49 ± 1.87	<MDL–9.44
ΣDDTs	18.6	30.9 ± 28.0	2.11–125	ΣDDTs	20.8	39.0 ± 64.5	2.04–372
2005				*2008*			
o,p′ DDD	2.13	4.61 ± 5.85	<MDL–27.1	*o,p′* DDD	2.27	4.24 ± 5.33	<MDL–27.0
p,p′ DDD	3.56	8.00 ± 9.70	<MDL–38.2	*p,p′* DDD	4.02	5.91 ± 6.27	<MDL–24.2
o,p′ DDE	<MDL	1.61 ± 3.42	<MDL–18.2	*o,p′* DDE	0.55	0.86 ± 1.09	<MDL–5.26
p,p′ DDE	13.0	19.6 ± 22.3	1.54–131	*p,p′* DDE	8.41	14.7 ± 14.2	1.80–56.2
o,p′ DDT	0.71	1.26 ± 1.68	<MDL–10.3	*o,p′* DDT	<MDL	0.75 ± 1.25	<MDL–6.06
p,p DDT	0.64	1.72 ± 5.63	<MDL–41.3	*p,p* DDT	0.63	1.04 ± 1.14	<MDL–5.10
ΣDDTs	24.6	36.8 ± 43.4	2.44–219	ΣDDTs	17.1	27.6 ± 26.7	2.46–97.6
2006				*2009*			
o,p′ DDD	2.57	4.30 ± 5.10	<MDL–22.7	*o,p′* DDD	1.28	3.27 ± 5.04	<MDL–25.6
p,p′ DDD	5.24	7.75 ± 9.45	<MDL–43.0	*p,p′* DDD	2.43	5.99 ± 9.69	<MDL–59.0
o,p′ DDE	1.13	1.80 ± 2.79	<MDL–14.0	*o,p′* DDE	<MDL	0.82 ± 1.40	<MDL–7.74
p,p′ DDE	14.1	19.7 ± 22.0	<MDL–111	*p,p′* DDE	7.42	11.5 ± 11.9	0.62–50.9
o,p′ DDT	1.40	2.14 ± 2.21	<MDL–7.85	*o,p′* DDT	<MDL	0.61 ± 0.87	<MDL–3.55
p,p DDT	0.66	1.08 ± 1.28	<MDL–5.36	*p,p* DDT	0.54	1.16 ± 1.84	<MDL–9.83
ΣDDTs	31.0	36.8 ± 40.1	<MDL–196	ΣDDTs	12.2	23.4 ± 28.9	1.34–148

The highest overall average concentration of ΣDDTs, defined as the sum of *o-p′*- and *p,p′*- isomers of DDD, DDE, and DDT, was detected in bivalves from the West coast (90.9 ± 43.3 ng/g dw), twice the concentration detected in oysters and mussels from the Gulf (42.4 ± 21.1 ng/g dw) and East (45.8 ± 24.6 ng/g dw) coasts. The DDT breakdown products, DDEs, as the sum of o,p′ and p,p′ isomers, was the most abundant DDT-related compound averaging 52.4 ± 5.73%, 62.8 ± 10.68%, and 74.2 ± 11.9% of the total average concentrations encountered in bivalves from the East, Gulf, and West coasts, respectively. DDEs were particularly dominant in the West coast. DDDs, on the other hand, had comparable contributions to ΣDDTs in bivalves from the East coast and the Gulf of Mexico (37.7 ± 5.71% and 30.4 ± 10.5%, respectively) while average percentage in bivalves from the West coast was less than half (14.2 ± 5.15%). DDTs accounted for a small proportion of the total DDT detected. On average, DDTs represented 9.90 ± 2.66%, 6.76 ± 3.02%, and 11.6 ± 8.62% in bivalves from the East, Gulf of Mexico, and West coasts, respectively, with apparent spikes in the proportion of DDT detected in 1997 (27.9%), 1998 (29.0%), and 2001 (31.9%). The proportional profiles encountered for DDT and related compounds along the three coasts of the USA are different from the generally reported composition of the technical DDT mixture containing 75% p,p′-DDT, 15% o,p′-DDT, <0.5% p-p′-DDD, <0.5% o,p′-DDD, 5% p,p′DDE, <0.5% o,p′-DDE, and <5% unidentified compounds (WHO, 1979). As reported earlier for the Gulf of Mexico oysters (Sericano et al., 1990a,b), approximately 75–80% of the total DDT concentrations measured in bivalves from the three coasts corresponded to the sum of *p,p′*-isomers in good agreement with the 80% reported for technical DDT (WHO, 1979).

Evaluating ΣDDT instead of just DDT parent compounds reflects better the potential contribution of degradation products to environmental exposure to DDT (Schenker et al., 2007). In general, while the levels of ΣDDT declined over the years in all three coastal environments, these decreases are more pronounced during the earlier years of this study. This can be inferred from Tables 1–3 by the decreasing average and median concentrations, and tighter, lower concentration ranges. The temporal decrease of ΣDDT concentrations is similar in West and East coastal environments but slightly faster along the northern Gulf of Mexico coast (Fig. 3). Bivalves are good surrogates for monitoring environmental trends because they adjust to environmental changes within 30–60 days for most organic contaminants with little metabolic transformation (Sericano, 1993). The environmental half-lives of ΣDDTs, reflected by native bivalves from the East, Gulf, and West coasts and calculated as indicated in Sericano and Wade (2011), are 13.3, 10.5, and 13.9 years, respectively. Under the present scenario, one would expect that ΣDDT concentrations would decrease more than 90% in approximately 45, 35, and 45 years for the East, Gulf and Gulf coasts, respectively.

These environmental half-life values are in good agreement with previous reports. Woodwell et al. (1971) estimated the residues half-life in a range upward to 20 years in the environment and closer to 10 years within the biosphere as a whole (Woodwell, 1969). The approximately 10 year estimation made by Woodwell was tested when historical DDT measured in bivalves from the three coasts of the United States was compared to the levels encountered during the first three years of the MW project (Sericano et al., 1990a,b). That estimation is now confirmed in this study and is in concordance with the generally reported half-life of between 2 and 15 years (U.S. EPA, 1989). It must be noted that ΣDDT losses in the marine environment are due to a combination of physical, chemical and biological processes that have different kinetics. Some of these processes are fast and initially predominant while others are slow and became more important with time. DDT loss is a nonlinear process, once a half-life is calculated; it does not necessarily mean that the losses for the remaining 50% will occur in the same amount of time since DDT degradation is a nonlinear process following the transition from the first mechanisms to the second ones.

DDT enters the atmosphere mainly as a result of spraying operations in areas where its use is permitted under specific health emergencies (e.g., malaria outbreak) and from volatilization of residual DDT and related compounds from soil and water from past use. Once in the atmosphere, these chemicals can be transported globally by air mass movement. Levels of DDD, DDE, and DDT isomers can be reduced by environmental processes (e.g., hydrolysis, oxidation/reduction, microbial degradation, volatility, and photolysis). The relative significance of several of these processes

Table 2
DDD, DDE, and DDT isomer concentrations (ng/g dw) in Gulf of Mexico bivalves.

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
1986 (n = 49)				*1995 (n = 46)*			
o,p′-DDD	2.32	5.68 ± 11.0	<MDL–67.1	*o,p′*-DDD	<MDL	1.72 ± 4.64	<MDL–30.9
p,p′-DDD	9.25	17.2 ± 26.5	<MDL–137	*p,p′*-DDD	3.49	10.3 ± 24.0	<MDL–154
o,p′-DDE	<MDL	1.44 ± 7.07	<MDL–49.7	*o,p′*-DDE	<MDL	1.26 ± 3.15	<MDL–18.0
p,p′-DDE	12.4	17.2 ± 15.2	2.68–64.7	*p,p′*-DDE	12.7	27.9 ± 52.1	2.38–323
o,p′-DDT	<MDL	0.97 ± 2.42	<MDL–14.5	*o,p′*-DDT	0.54	4.46 ± 15.1	<MDL–97.3
p,p′-DDT	0.66	1.69 ± 3.73	<MDL–23.6	*p,p′*-DDT	<MDL	3.32 ± 11.7	<MDL–77.7
ΣDDTs	26.5	44.2 ± 54.8	2.82–302	ΣDDTs	20.5	49.0 ± 89.0	2.38–500
1987 (n = 48)				*1996 (n = 42)*			
o,p′-DDD	1.42	9.93 ± 47.0	<MDL–326	*o,p′*-DDD	1.41	3.98 ± 6.69	<MDL–33.0
p,p′-DDD	9.41	25.1 ± 64.0	<MDL–443	*p,p′*-DDD	2.74	11.3 ± 24.3	<MDL–133
o,p′-DDE	<MDL	1.14 ± 6.03	<MDL–41.9	*o,p′*-DDE	1.94	6.59 ± 15.8	<MDL–84.6
p,p′-DDE	13.0	29.3 ± 71.2	1.14–495	*p,p′*-DDE	13.0	31.7 ± 55.2	0.92–310
o,p′-DDT	<MDL	0.51 ± 1.47	<MDL–8.12	*o,p′*-DDT	<MDL	1.24 ± 2.73	<MDL–12.6
p,p′-DDT	<MDL	1.40 ± 2.10	<MDL–9.38	*p,p′*-DDT	0.78	2.03 ± 3.66	<MDL–16.5
ΣDDTs	30.3	67.4 ± 188	3.13–1320	ΣDDTs	22.0	56.8 ± 96.3	3.30–507
1988 (n = 65)				*1997 (n = 38)*			
o,p′-DDD	3.27	9.55 ± 24.5	<MDL–157	*o,p′*-DDD	1.43	4.91 ± 11.8	<MDL–70.4
p,p′-DDD	9.38	28.1 ± 62.8	<MDL–474	*p,p′*-DDD	2.15	15.0 ± 54.6	<MDL–335
o,p′-DDE	<MDL	6.17 ± 28.3	<MDL–213	*o,p′*-DDE	0.59	1.16 ± 1.76	<MDL–9.43
p,p′-DDE	18.6	30.0 ± 37.6	<MDL–213	*p,p′*-DDE	11.1	27.3 ± 58.1	1.99–339
o,p′-DDT	1.09	1.99 ± 2.86	<MDL–15.5	*o,p′*-DDT	<MDL	1.17 ± 3.25	<MDL–19.5
p,p′-DDT	0.89	2.24 ± 4.84	<MDL–35.1	*p,p′*-DDT	0.81	3.98 ± 13.4	<MDL–82.5
ΣDDTs	38.0	78.0 ± 131	3.07–818	ΣDDTs	17.7	53.5 ± 139	3.12–851
1989 (n = 63)				*1998 (n = 39)*			
o,p′-DDD	5.83	9.21 ± 18.1	<MDL–131	*o,p′*-DDD	0.94	3.78 ± 8.14	<MDL–47.3
p,p′-DDD	6.06	27.3 ± 74.0	<MDL–510	*p,p′*-DDD	1.86	12.8 ± 45.1	<MDL–282
o,p′-DDE	1.67	8.53 ± 40.8	<MDL–317	*o,p′*-DDE	<MDL	4.74 ± 16.7	<MDL–83.2
p,p′-DDE	20.0	46.8 ± 86.5	3.09–522	*p,p′*-DDE	9.47	24.7 ± 47.0	1.00–236
o,p′-DDT	1.16	2.23 ± 4.40	<MDL–32.6	*o,p′*-DDT	<MDL	0.69 ± 2.10	<MDL–9.75
p,p′-DDT	1.65	3.69 ± 6.56	<MDL–37.2	*p,p′*-DDT	<MDL	2.82 ± 9.11	<MDL–56.4
ΣDDTs	35.0	97.8 ± 201	11.3–1150	ΣDDTs	11.1	49.3 ± 113	1.00–629
1990 (n = 68)				*1999 (n = 38)*			
o,p′-DDD	3.82	5.69 ± 9.36	<MDL–62.4	*o,p′*-DDD	0.98	1.50 ± 1.74	<MDL–7.67
p,p′-DDD	4.19	19.4 ± 56.8	<MDL–390	*p,p′*-DDD	2.66	4.67 ± 5.35	<MDL–24.0
o,p′-DDE	1.05	5.97 ± 23.1	<MDL–167	*o,p′*-DDE	0.97	1.38 ± 1.80	<MDL–9.35
p,p′-DDE	9.92	25.4 ± 48.7	<MDL–286	*p,p′*-DDE	11.5	18.4 ± 28.3	1.61–171
o,p′-DDT	0.73	2.26 ± 6.03	<MDL–45.0	*o,p′*-DDT	0.78	1.08 ± 1.19	<MDL–5.27
p,p′-DDT	3.10	4.83 ± 7.45	<MDL–45.6	*p,p′*-DDT	<MDL	1.25 ± 1.74	<MDL–6.41
ΣDDTs	24.5	63.6 ± 130	3.16–769	ΣDDTs	16.0	28.3 ± 33.6	3.51–197
1991 (n = 63)				*2000 (n = 42)*			
o,p′-DDD	<MDL	2.07 ± 5.35	<MDL–33.2	*o,p′*-DDD	0.54	2.36 ± 8.05	<MDL–52.2
p,p′-DDD	2.15	15.3 ± 42.5	<MDL–258	*p,p′*-DDD	0.96	9.58 ± 42.1	<MDL–273
o,p′-DDE	0.56	2.73 ± 9.69	<MDL–71.3	*o,p′*-DDE	<MDL	2.50 ± 12.4	<MDL–80.3
p,p′-DDE	11.9	29.1 ± 50.0	1.38–302	*p,p′*-DDE	4.28	15.4 ± 37.9	0.67–217
o,p′-DDT	<MDL	0.75 ± 2.45	<MDL–16.8	*o,p′*-DDT	<MDL	0.50 ± 0.77	<MDL–3.41
p,p′-DDT	0.86	2.73 ± 6.52	<MDL–40.8	*p,p′*-DDT	<MDL	<MDL	<MDL–7.36
ΣDDTs	18.2	52.6 ± 104	2.18–548	ΣDDTs	7.10	30.8 ± 90.6	1.11–556
1992 (n = 63)				*2001 (n = 33)*			
o,p′-DDD	<MDL	1.18 ± 2.01	<MDL–11.1	*o,p′*-DDD	0.85	1.77 ± 2.51	<MDL–11.8
p,p′-DDD	3.84	10.9 ± 16.4	<MDL–73.8	*p,p′*-DDD	0.84	3.55 ± 6.68	<MDL–32.6
o,p′-DDE	0.99	2.62 ± 11.1	<MDL–88.3	*o,p′*-DDE	<MDL	<MDL	<MDL–1.73
p,p′-DDE	14.6	32.1 ± 41.1	2.29–220	*p,p′*-DDE	4.87	15.9 ± 24.7	0.66–103
o,p′-DDT	0.54	1.18 ± 1.69	<MDL–8.79	*o,p′*-DDT	<MDL	0.91 ± 1.60	<MDL–7.38
p,p′-DDT	1.37	3.00 ± 4.50	<MDL–27.0	*p,p′*-DDT	<MDL	1.24 ± 2.85	<MDL–15.7
ΣDDTs	22.2	51.0 ± 60.4	3.87–276	ΣDDTs	6.46	23.7 ± 34.0	1.21–115
1993 (n = 65)				*2002 (n = 37)*			
o,p′-DDD	<MDL	1.50 ± 4.41	<MDL–34.1	*o,p′*-DDD	0.75	2.19 ± 3.56	<MDL–14.8
p,p′-DDD	2.67	10.5 ± 35.4	<MDL–283	*p,p′*-DDD	1.17	4.36 ± 8.78	<MDL–38.4
o,p′-DDE	<MDL	1.48 ± 6.49	<MDL–38.3	*o,p′*-DDE	0.62	5.60 ± 18.6	<MDL–90.0
p,p′-DDE	12.5	26.4 ± 50.0	1.61–294	*p,p′*-DDE	5.33	16.0 ± 31.2	0.90–141
o,p′-DDT	<MDL	0.96 ± 2.03	<MDL–14.7	*o,p′*-DDT	<MDL	0.72 ± 0.96	<MDL–4.34
p,p′-DDT	0.88	1.87 ± 5.08	<MDL–40.0	*p,p′*-DDT	<MDL	0.74 ± 1.32	<MDL–5.13
ΣDDTs	17.0	42.7 ± 90.8	1.72–670	ΣDDTs	9.55	29.6 ± 61.3	1.99–280
1994 (n = 43)				*2003 (n = 35)*			
o,p′-DDD	0.80	3.61 ± 7.99	<MDL–48.2	*o,p′*-DDD	1.57	2.34 ± 2.29	<MDL–9.30
p,p′-DDD	1.96	12.3 ± 34.2	<MDL–212	*p,p′*-DDD	1.38	4.13 ± 6.31	<MDL–25.4
o,p′-DDE	1.06	3.51 ± 8.15	<MDL–49.6	*o,p′*-DDE	<MDL	<MDL	<MDL–3.16
p,p′-DDE	9.39	30.2 ± 77.1	1.43–505	*p,p′*-DDE	8.63	20.1 ± 29.0	1.28–157
o,p′-DDT	<MDL	0.57 ± 1.01	<MDL–6.32	*o,p′*-DDT	0.72	1.27 ± 1.82	<MDL–9.89
p,p′-DDT	<MDL	1.35 ± 3.61	<MDL–23.5	*p,p′*-DDT	<MDL	1.52 ± 2.73	<MDL–13.0
ΣDDTs	18.1	51.5 ± 123	3.34–795	ΣDDTs	12.9	29.7 ± 36.6	1.93–173

Table 2 (*continued*)

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
2004 (n = 37)				*2007 (n = 34)*			
o,p′-DDD	0.98	1.59 ± 2.12	<MDL–8.61	*o,p′*-DDD	<MDL	1.00 ± 1.60	<MDL–7.40
p,p′-DDD	0.80	4.04 ± 9.00	<MDL–37.6	*p,p′*-DDD	1.02	2.57 ± 4.34	<MDL–16.2
o,p′-DDE	<MDL	2.07 ± 9.31	<MDL–55.6	*o,p′*-DDE	<MDL	<MDL	<MDL–1.90
p,p′-DDE	5.55	14.9 ± 25.0	0.60–107	*p,p′*-DDE	7.15	11.4 ± 13.8	1.49–63.3
o,p′-DDT	0.64	1.02 ± 1.20	<MDL–4.93	*o,p′*-DDT	<MDL	<MDL	<MDL–4.77
p,p′-DDT	<MDL	0.73 ± 1.47	<MDL–6.89	*p,p′*-DDT	<MDL	0.53 ± 1.58	<MDL–8.82
ΣDDTs	8.07	24.4 ± 43.6	0.88–202	ΣDDTs	9.70	16.1 ± 19.0	1.49–72.8
2005 (n = 33)				*2008 (n = 32)*			
o,p′-DDD	<MDL	1.20 ± 1.90	<MDL–6.48	*o,p′*-DDD	MDL	0.61 ± 0.95	<MDL–3.66
p,p′-DDD	1.24	2.98 ± 4.48	<MDL–16.2	*p,p′*-DDD	0.64	1.95 ± 3.88	<MDL–16.9
o,p′-DDE	<MDL	<MDL	<MDL–2.50	*o,p′*-DDE	<MDL	3.46 ± 15.8	<MDL–88.2
p,p′-DDE	5.84	18.0 ± 31.6	1.09–172	*p,p′*-DDE	5.70	11.4 ± 18.3	0.67–98.9
o,p′-DDT	<MDL	<MDL	<MDL–2.07	*o,p′*-DDT	<MDL	<MDL	<MDL–0.71
p,p′-DDT	<MDL	0.59 ± 0.90	<MDL–3.85	*p,p′*-DDT	<MDL	<MDL	<MDL–1.79
ΣDDTs	7.74	23.5 ± 35.5	1.65–182	ΣDDTs	8.33	17.7 ± 37.3	0.94–209
2006 (n = 34)				*2009 (n = 39)*			
o,p′-DDD	0.63	1.26 ± 1.63	<MDL–5.54	*o,p′*-DDD	<MDL	1.47 ± 3.64	<MDL–16.0
p,p′-DDD	1.18	3.37 ± 6.67	<MDL–34.8	*p,p′*-DDD	0.72	4.43 ± 11.3	<MDL–53.9
o,p′-DDE	<MDL	2.33 ± 9.54	<MDL–53.5	*o,p′*-DDE	<MDL	<MDL	<MDL–1.46
p,p′-DDE	6.41	12.5 ± 17.6	0.78–72.0	*p,p′*-DDE	3.46	8.31 ± 14.0	<MDL–77.0
o,p′-DDT	<MDL	0.53 ± 0.78	<MDL–3.05	*o,p′*-DDT	<MDL	<MDL	<MDL–4.06
p,p′-DDT	<MDL	0.61 ± 0.87	<MDL–2.71	*p,p′*-DDT	<MDL	1.08 ± 4.09	<MDL–24.8
ΣDDTs	9.69	20.6 ± 32.3	0.78–152	ΣDDTs	5.87	15.8 ± 24.0	<MDL–81.7

depends of prevailing environmental conditions (e.g., ambient temperature). DDT isomers and derivatives are reported to have significantly lower, by a factor of 2–20, half-lives in soils of tropical areas compared to more temperate regions (e.g., Racke et al., 1997; Cheremisinoff and Rosenfeld, 2011). Unfortunately, there is no readily available, if any, comparable information on the half-life of ΣDDT in tropical marine waters or coastal zones. Comparison of a warmer area within this study (i.e., northern Gulf of Mexico) with a colder area (i.e., Alaska) reveals that the half-life of ΣDDT calculated for Alaska (14.4 years) is nearly 40% higher than the half-life calculated for the northern Gulf of Mexico (10.5 years) and slightly higher than the values estimated for the East (13.3 years) and West (13.9 years) coasts. In addition to a temperature factor, the longer times estimated for coastal United States at higher latitudes might also be a consequence of a more significant deposition of DDD, DDE and/or DDT due to long-range atmospheric transport from lower latitudes (Shen et al., 2005; Gioia et al., 2005; Goel et al., 2010). The influence of this transport/deposition on the half-life of ΣDDT would be proportionally more significant in areas with the lowest concentrations. Thus, the faster decrease observed in the northern Gulf of Mexico compared to Alaska might be related to a faster mobilization by any or all the mentioned environmental processes, a lower atmospheric input or a combination of both. Similar findings were reported when comparing *p,p′*-DDE concentrations in the airshed of Chesapeake and Delaware Bays with levels in the Great Lakes region (Goel et al. (2010)). Half-life values, estimated for Chesapeake and Delaware Bays, were, in general, two to three times lower than the half-life estimated for the Great Lakes. Goel et al. (2010) indicate that this may be related to the sandier soils and warmer temperatures of the Chesapeake and Delaware regions that facilitate degradation and volatilization of legacy pesticides and predict regional concentrations would likely be below 10% of present levels in 30–40 years. This prediction agrees well with the half-lives estimated for each of the three coasts of the United States.

The observed decrease in the average concentrations of ΣDDT is also detected in most, but not all, sampling locations. The linear regression used to examine the significance of the relationship between dependent and independent variables permitted the distinction among locations experiencing significant, at two different levels of confidence, or no change. Fig. 4 shows typical results of this analysis. Including only locations sampled 5 or more times, 75% (170) of the sites showed a statistically significant decrease in ΣDDT concentrations at the 95% level of confidence, 8% (19) additional sites revealed a significant change, mostly decreases, at the 90% level of confidence, while the remaining 38 sites (17%) did not show any statistically significant trend at either level. Most of the significant decreases were observed in Gulf of Mexico bivalves where 85%, 8%, and 8% of the locations revealed a significant decrease ($\alpha = 0.05$), a less certain decline ($\alpha = 0.10$) and no trend at all, respectively. Similar percentages are calculated for the East coast (76%, 6%, and 18%, respectively). In contrast, bivalves from the West coast were more resilient to show changes with 60%, 13%, and 27%, respectively. Although it is not clear why the ΣDDT concentrations in some locations are still increasing nearly forty years after the ban imposed to all applications of DDT in the United States, it seems that this is mostly related to past inputs of manufacture wastes or runoff from historically contaminated agricultural lands as discussed below. It is noteworthy to mention that these increases, when detected, are only slightly significant ($\alpha = 0.10$) such as is the case of Long Island Sound, Housatonic River (LIHR) site on the East coast (Fig. 4c).

Previously reported data, published after the first few years of the NS&T program, commenced to define the distribution of contaminants in the U.S. coastal areas (e.g., Sericano et al., 1990a,b; O'Connor, 2002; O'Connor and Lauenstein, 2006). The present data set confirms some of those observations and integrates all three coasts of the U.S. to redefine, in a global context, areas of highest (above the national 85th percentile, Table 4) and lowest (below the national 15th percentile, Table 5) concentrations averaged over time (Fig. 5). The highest ΣDDT concentrations are mainly concentrated in southern California from Tyler Bight in San Miguel Island (SANM) to the Tijuana River estuary (TJRE) in the National Estuarine Research Reserve (Table 4). With the exception of a single sample collected in 2008 from Point Mugu Lagoon (MULG), the highest concentrations within this group correspond to bivalves

Table 3
DDD, DDE, and DDT isomer concentrations (ng/g dw) in West Coast bivalves.

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
1986 (n = 43)				*1995 (n = 30)*			
o,p′ DDD	3.13	7.45 ± 11.0	<MDL–45.0	*o,p′* DDD	3.92	4.99 ± 4.17	0.60–16.1
p,p′ DDD	6.20	24.9 ± 43.2	<MDL–186	*p,p′* DDD	5.25	11.0 ± 12.9	0.79–52.6
o,p′ DDE	1.38	11.7 ± 32.2	<MDL–167	*o,p′* DDE	1.04	3.54 ± 8.19	<MDL–42.0
p,p′ DDE	17.0	63.9 ± 146	1.53–697	*p,p′* DDE	41.5	58.4 ± 77.4	2.85–375
o,p′ DDT	2.38	3.39 ± 4.18	<MDL–21.3	*o,p′* DDT	1.31	4.13 ± 6.34	<MDL–27.7
p,p′ DDT	3.53	6.04 ± 7.55	<MDL–31.0	*p,p′* DDT	2.48	9.05 ± 15.7	<MDL–64.1
ΣDDTs	33.9	117 ± 229	5.43–1080	ΣDDTs	54.3	91.1 ± 104	5.22–459
1987 (n = 39)				*1996 (n = 31)*			
o,p′ DDD	<MDL	4.10 ± 10.6	<MDL–51.0	*o,p′* DDD	1.69	4.63 ± 6.66	<MDL–27.5
p,p′ DDD	6.93	38.3 ± 62.3	<MDL–240	*p,p′* DDD	2.67	6.53 ± 8.81	0.60–37.8
o,p′ DDE	<MDL	18.0 ± 44.0	<MDL–170	*o,p′* DDE	1.04	6.56 ± 15.5	<MDL–78.6
p,p′ DDE	27.3	117 ± 228	3.66–1180	*p,p′* DDE	12.8	55.2 ± 95.8	2.51–424
o,p′ DDT	<MDL	1.38 ± 5.32	<MDL–32.0	*o,p′* DDT	0.63	0.96 ± 1.00	<MDL–4.19
p,p′ DDT	<MDL	10.9 ± 19.3	<MDL–73.7	*p,p′* DDT	2.57	3.24 ± 3.07	<MDL–13.6
ΣDDTs	36.0	190 ± 344	5.10–1650	ΣDDTs	28.0	77.1 ± 122	5.20–565
1988 (n = 47)				*1997 (n = 25)*			
o,p′ DDD	<MDL	3.56 ± 6.44	<MDL–27.3	*o,p′* DDD	1.56	10.3 ± 19.3	<MDL–88.0
p,p′ DDD	2.43	9.69 ± 16.0	<MDL–72.7	*p,p′* DDD	3.15	21.4 ± 34.7	<MDL–134
o,p′ DDE	<MDL	6.53 ± 20.3	<MDL–109	*o,p′* DDE	0.72	2.88 ± 4.98	<MDL–21.8
p,p′ DDE	23.9	106 ± 223	3.96–1010	*p,p′* DDE	18.0	68.0 ± 92.7	2.38–374
o,p′ DDT	<MDL	3.34 ± 13.2	<MDL–86.7	*o,p′* DDT	1.16	11.5 ± 20.8	<MDL–70.7
p,p′ DDT	0.60	6.08 ± 18.4	<MDL–113	*p,p′* DDT	3.86	28.3 ± 53.9	<MDL–203
ΣDDTs	32.6	136 ± 264	5.16–1170	ΣDDTs	28.2	142 ± 217	3.34–847
1989 (n = 44)				1998 (n = 43)			
o,p′ DDD	<MDL	3.87 ± 6.25	<MDL–30.0	*o,p′* DDD	1.68	9.31 ± 27.5	<MDL–168
p,p′ DDD	3.42	8.76 ± 12.2	<MDL–58.3	*p,p′* DDD	2.62	17.1 ± 45.6	<MDL–287
o,p′ DDE	<MDL	7.74 ± 31.2	<MDL–197	*o,p′* DDE	<MDL	8.35 ± 19.7	<MDL–108
p,p′ DDE	23.5	68.3 ± 146	<MDL–753	*p,p′* DDE	16.6	95.2 ± 149	2.88–567
o,p′ DDT	0.53	1.91 ± 3.24	<MDL–16.0	*o,p′* DDT	0.70	18.8 ± 90.5	<MDL–589
p,p′ DDT	1.27	2.30 ± 3.38	<MDL–13.4	*p,p′* DDT	2.15	34.3 ± 133	<MDL–836
ΣDDTs	32.1	92.8 ± 191	<MDL–1040	ΣDDTs	23.7	183 ± 376	3.92–2120
1990 (n = 58)				1999 (n = 25)			
o,p′ DDD	<MDL	1.47 ± 2.38	<MDL–11.0	*o,p′* DDD	1.17	4.09 ± 5.68	<MDL–18.0
p,p′ DDD	3.63	14.0 ± 24.5	<MDL–136	*p,p′* DDD	5.20	8.50 ± 9.56	<MDL–36.5
o,p′ DDE	1.04	9.02 ± 25.8	<MDL–148	*o,p′* DDE	3.62	3.60 ± 2.36	<MDL–9.68
p,p′ DDE	22.4	105 ± 211	2.81–1010	*p,p′* DDE	24.7	47.0 ± 57.9	2.77–182
o,p′ DDT	2.70	4.46 ± 7.67	<MDL–54.5	*o,p′* DDT	4.06	4.61 ± 3.33	<MDL–15.0
p,p′ DDT	3.22	6.69 ± 20.4	<MDL–156	*p,p′* DDT	1.01	5.54 ± 10.9	<MDL–36.5
ΣDDTs	31.7	141 ± 261	3.36–1220	ΣDDTs	36.9	73.4 ± 82.2	10.1–284
1991 (n = 55)				2000 (n = 36)			
o,p′ DDD	<MDL	0.51 ± 1.13	<MDL–5.46	*o,p′* DDD	1.39	3.10 ± 3.84	<MDL–18.1
p,p′ DDD	1.84	6.00 ± 9.69	<MDL–51.7	*p,p′* DDD	1.94	7.33 ± 11.6	<MDL–57.2
o,p′ DDE	0.81	6.65 ± 20.5	<MDL–141	*o,p′* DDE	0.97	5.87 ± 12.0	<MDL–51.6
p,p′ DDE	12.4	61.0 ± 123	3.70–589	*p,p′* DDE	9.89	73.3 ± 125	1.57–494
o,p′ DDT	<MDL	1.19 ± 1.82	<MDL–7.44	*o,p′* DDT	0.70	1.98 ± 4.17	<MDL–17.9
p,p′ DDT	0.64	1.65 ± 2.18	<MDL–10.5	*p,p′* DDT	1.25	6.35 ± 16.5	<MDL–78.8
ΣDDTs	17.9	77.1 ± 149	3.90–689	ΣDDTs	16.4	97.9 ± 161	3.02–673
1992 (n = 55)				2001 (n = 25)			
o,p′ DDD	<MDL	2.71 ± 4.54	<MDL–18.0	*o,p′* DDD	0.57	3.37 ± 6.20	<MDL–28.9
p,p′ DDD	2.34	8.51 ± 13.7	<MDL–63.5	*p,p′* DDD	1.63	6.36 ± 13.0	<MDL–62.6
o,p′ DDE	0.98	8.42 ± 27.3	<MDL–183	*o,p′* DDE	0.60	1.12 ± 1.61	<MDL–7.50
p,p′ DDE	20.3	93.7 ± 216	2.33–1250	*p,p′* DDE	12.0	26.9 ± 35.2	1.15–146
o,p′ DDT	<MDL	1.30 ± 2.60	<MDL–13.2	*o,p′* DDT	<MDL	7.54 ± 30.2	<MDL–152
p,p′ DDT	<MDL	2.32 ± 5.36	<MDL–31.5	*p,p′* DDT	0.72	10.1 ± 38.5	<MDL–193
ΣDDTs	24.0	117 ± 253	2.36–1410	ΣDDTs	17.4	55.4 ± 103	2.01–491
1993 (n = 43)				2002 (n = 33)			
o,p′ DDD	2.30	4.26 ± 5.35	<MDL–27.5	*o,p′* DDD	0.75	1.33 ± 2.21	<MDL–11.0
p,p′ DDD	4.21	8.48 ± 9.83	<MDL–36.0	*p,p′* DDD	1.57	3.66 ± 5.23	<MDL–20.9
o,p′ DDE	1.34	4.38 ± 8.05	<MDL–44.0	*o,p′* DDE	<MDL	5.77 ± 14.6	<MDL–73.7
p,p′ DDE	11.4	65.6 ± 127	<MDL–700	*p,p′* DDE	5.85	60.2 ± 130	1.63–642
o,p′ DDT	2.19	4.79 ± 6.79	<MDL–31.5	*o,p′* DDT	0.62	0.92 ± 0.96	<MDL–3.53
p,p′ DDT	2.10	6.98 ± 13.8	<MDL–67.6	*p,p′* DDT	0.63	1.57 ± 2.32	<MDL–12.4
ΣDDTs	32.1	94.6 ± 153	<MDL–799	ΣDDTs	10.8	73.5 ± 152	2.81–753
1994 (n = 32)				2003 (n = 24)			
o,p′ DDD	1.09	2.22 ± 3.44	<MDL–18.4	*o,p′* DDD	0.89	2.70 ± 3.41	<MDL–11.5
p,p′ DDD	2.05	6.02 ± 13.4	0.54–74.5	*p,p′* DDD	0.89	3.35 ± 5.06	<MDL–22.4
o,p′ DDE	0.80	7.78 ± 25.2	<MDL–133	*o,p′* DDE	<MDL	0.66 ± 0.93	<MDL–4.04
p,p′ DDE	12.9	42.2 ± 65.5	1.48–263	*p,p′* DDE	11.9	26.7 ± 37.9	0.65–167
o,p′ DDT	<MDL	0.59 ± 1.21	<MDL–5.90	*o,p′* DDT	<MDL	3.12 ± 5.20	<MDL–19.8
p,p′ DDT	1.34	2.24 ± 3.50	<MDL–14.7	*p,p′* DDT	<MDL	4.48 ± 10.6	<MDL–46.9
ΣDDTs	21.7	61.1 ± 103	3.57–459	ΣDDTs	15.4	41.0 ± 59.5	1.43–266

Table 3 (*continued*)

	Median	Avg ± Std	Min–Max		Median	Avg ± Std	Min–Max
2004 (n = 34)				2007 (n = 24)			
o,p′ DDD	1.07	2.03 ± 2.78	<MDL–15.0	*o,p′* DDD	0.73	2.10 ± 2.92	<MDL–9.89
p,p′ DDD	1.07	3.30 ± 5.26	<MDL–22.7	*p,p′* DDD	1.15	3.98 ± 5.03	<MDL–18.7
o,p′ DDE	<MDL	3.74 ± 7.88	<MDL–29.8	*o,p′* DDE	<MDL	0.78 ± 0.96	<MDL–3.17
p,p′ DDE	7.56	49.4 ± 93.2	1.02–410	*p,p′* DDE	20.5	32.8 ± 42.8	2.46–193
o,p′ DDT	0.64	1.87 ± 5.42	<MDL–32.1	*o,p′* DDT	<MDL	1.18 ± 1.89	<MDL–6.53
p,p′ DDT	0.66	3.20 ± 11.3	<MDL–66.8	*p,p′* DDT	0.61	1.78 ± 3.38	<MDL–15.5
ΣDDTs	14.1	63.5 ± 117	1.59–462	ΣDDTs	24.4	42.6 ± 53.9	3.17–241
2005 (n = 25)				2008 (n = 53)			
o,p′ DDD	0.63	3.37 ± 4.89	<MDL–18.9	*o,p′* DDD	0.70	2.26 ± 4.33	<MDL–25.7
p,p′ DDD	0.95	6.12 ± 11.0	<MDL–52.6	*p,p′* DDD	1.50	5.82 ± 12.2	<MDL–73.1
o,p′ DDE	<MDL	1.05 ± 1.91	<MDL–8.36	*o,p′* DDE	0.50	3.25 ± 6.40	<MDL–33.9
p,p′ DDE	11.8	41.2 ± 69.3	1.68–317	*p,p′* DDE	13.1	59.9 ± 115	1.47–703
o,p′ DDT	0.52	3.32 ± 6.52	<MDL–30.0	*o,p′* DDT	<MDL	1.84 ± 7.94	<MDL–57.0
p,p′ DDT	1.13	7.69 ± 20.0	<MDL–93.2	*p,p′* DDT	0.96	4.68 ± 21.0	<MDL–152
ΣDDTs	14.4	62.8 ± 111	1.99–520	ΣDDTs	16.3	77.7 ± 158	2.85–1030
2006 (n = 34)				2009 (n = 39)			
o,p′ DDD	0.73	1.00 ± 1.10	<MDL–4.09	*o,p′* DDD	0.58	1.46 ± 3.71	<MDL–23.0
p,p′ DDD	1.26	2.90 ± 3.56	<MDL–13.6	*p,p′* DDD	<MDL	2.66 ± 8.81	<MDL–54.5
o,p′ DDE	0.68	3.39 ± 6.43	<MDL–23.8	*o,p′* DDE	<MDL	<MDL	<MDL–2.50
p,p′ DDE	7.73	41.1 ± 72.2	1.33–278	*p,p′* DDE	5.16	12.1 ± 19.2	0.62–99.3
o,p′ DDT	0.64	0.99 ± 1.40	<MDL–7.53	*o,p′* DDT	<MDL	1.36 ± 6.04	<MDL–37.8
p,p′ DDT	0.98	1.36 ± 1.75	<MDL–8.35	*p,p′* DDT	<MDL	3.44 ± 16.0	<MDL–98.3
ΣDDTs	12.0	50.7 ± 83.1	1.97–321	ΣDDTs	7.00	21.5 ± 49.0	0.99–283

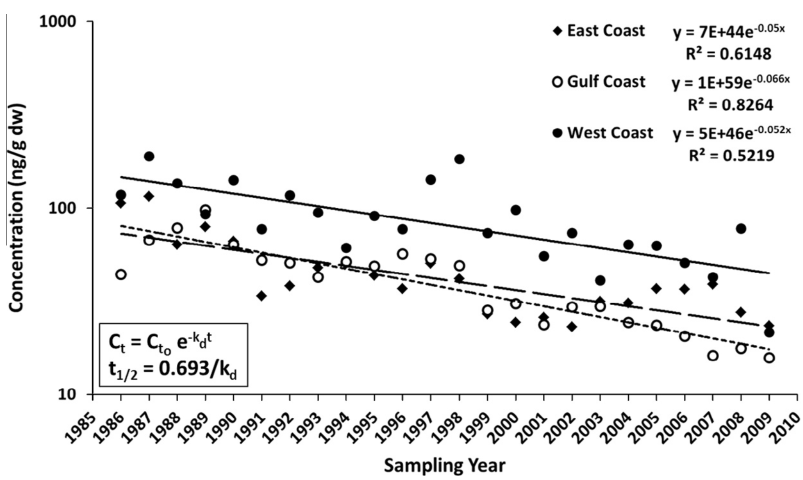

Fig. 3. Environmental dissipation trends of ΣDDTs reflected by bivalves from the East, Gulf, and West environmental coasts. Markers represent annual averages (Tables 1–3); standard deviations are omitted for clarity.

collected from the Southern California Bight area [San Pedro Harbor (SPFP), Palos Verdes (PVRP), Newport Bay (NHPB), Anaheim Bay (ABWJ), and Long Beach (LBBW)]. The high levels of ΣDDT detected in these bivalves, most of them listed at the top of the locations ranked above the national 85th percentile, corresponded well with concentrations previously reported in sediment samples from this area (e.g., Lee et al., 2002; Venkatesan et al., 2010; Chen et al., 2012). The Southern California Bight is known to be the recipient of over 2400 metric tons of DDT manufacture wastes, among other contaminants, in process water discharged between 1940 and the early 1970s through the White Point sewage outfalls (Lee et al., 2002; Chen et al., 2012). Even after 40 years, the DDT that has accumulated in the coastal sediments is being constantly remobilized, either as DDT of any of its metabolites, to the overlaying seawater and the biota therein (Eganhouse et al., 2000; Zeng and Tran, 2002; Blasius and Goodmanlowe, 2008). Table 4 shows the trends observed in locations with 5 or more samplings for those sites listed above the national 85th percentile. In spite of the lack of enough data points for some of the sites within the Southern California Bight or the lack of a clear trend evidenced at others, the fact that ΣDDT concentrations are decreasing at most of these geographically contiguous sites, suggest the likelihood of a downward trend for the entire area. Without new inputs, these decreasing trends are anticipated to continue at sampling sites located to the northwest of the Whites Point sewage outfalls, near the Palos Verdes site (PVRP), as the area will remain depositional for the next 40 years (Sherwood et al., 2002). Thus, the existing inventory of DDT and related compounds will remain buried, deeper with time, and practically isolated from the overlaying seawater. Unfortunately, the same model suggests that erosion is likely to occur to

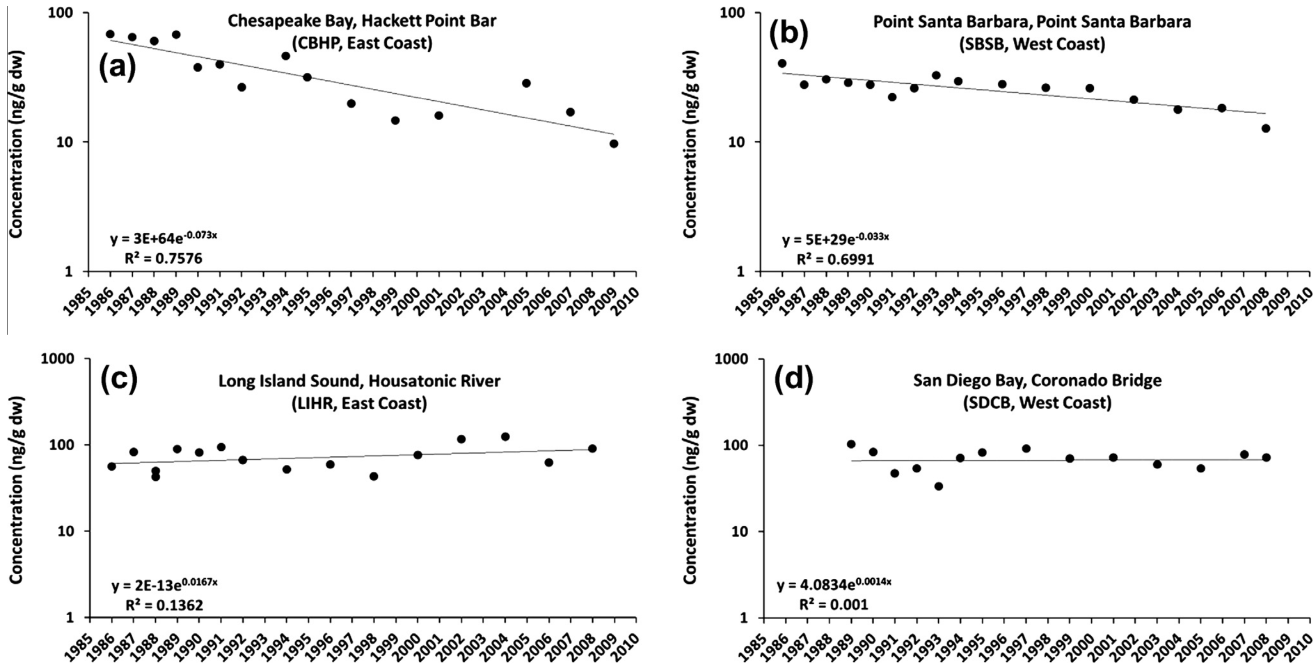

Fig. 4. Examples of trends observed in locations with 5 or more sampling episodes showing significant decreases at the 95% level of confidence (a and b), a significant increase at the 90% level of confidence (c), and a no statistically significant trend at either level (d).

the southeast of the outfalls resulting in reintroduction of DDE, DDD, and DDT isomers to the upper sediment layer and subsequent diffusion to the overlying seawater. This release of contaminants will likely be reflected in bivalves from nearby locations such as SPFP, LARM, LBBM, ABWJ, NHPB, and NBWJ.

Other sites with high ΣDDT concentrations on the West coast are concentrated in San Francisco [Emeryville (SFEM), Dumbarton Bridge (SFDB), and San Mateo Bridge (SFSM)] and Monterey [Elkhorn Slough (MBES) and Moss Landing (MBML)] Bays. DDT and metabolites, from a large residual mass deposited in historically contaminated adjacent soils/sediments, enter San Francisco and Monterey Bays from a variety of sources but primarily from runoff from agricultural lands and local watersheds (Connor et al., 2007; Hartwell, 2008). Concentrations in bivalves from two of the sites in San Francisco Bay listed in the above the national 85th percentile (Dumbarton Bridge and San Mateo Bridge) are decreasing while the third one (Emeryville) revealed no trend. Connor et al. (2007) predicted that, even without new inputs, DDT as well as other legacy pesticides will decline very slowly and their concentrations in water and superficial sediment will take from nearly one to three decades to reach risk-reduction goals. Under this scenario, the half-life of ΣDDT was estimated in about 4 years. Under a scenario of continued inputs, however, the half-life of ΣDDT in San Francisco mussels was projected to about 16 years, more in line with the estimation given above for the West coast. In addition to an expected decrease to inputs into the Bay, environmental degradation, outflow through the Golden Gate, and volatilization (in that order) are cited as mechanisms of removal of chemicals from the Bay (Connor et al., 2007). A similar situation is described for Monterey Bay where excess of DDT appears to be delivered to the deep ocean via the Canyons (Hartwell, 2008) although trends in the concentrations detected in bivalves from Elkhorn Slough and Moss Landing were not evident.

Sites having bivalves with the highest ΣDDT concentrations on the East coast are mainly concentrated on the northeast, particularly in Delaware Bay [Hope Creek (DBHC), Ben Davis Pt. Shoal (DBBD), Arnolds Point Shoal (DBAP), Kelly Island (DBKI), and Woodland Beach (DBWB)] and Hudson/Raritan Estuary [Lower Bay (HRLB), Upper Bay (HRUB), Raritan Bay (HRRB), and Battery Park (HRBP)]. Three of the locations in Delaware Bay (Ben Davis Pt. Shoal, Arnolds Point Shoal and Kelly Island) revealed decreasing trends in ΣDDT concentrations while not enough data have been collected in the remaining two sites (Hope Creek and Woodland Beach) to define a trend. These high concentrations are not surprising since a DDT manufacturing plant was located just south of Philadelphia on the Delaware River (Gioia et al., 2005). The two locations showing the highest ΣDDT concentrations in the Hudson/Raritan Estuary area (Lower Bay and Upper Bay) revealed decreasing trends. A third station (Raritan Bay) did not show a significant trend while the fourth site (Battery Park), located near the Upper Bay site, was sampled only once, after the attack on the World Trade Center. A few other isolated sites with high ΣDDT concentrations on the East coast include Throgs Neck in Long Island Sound (LITN), Dorchester Bay in Boston Harbor (BHDB), and Angelica Rock in Buzzards Bay (BBAR). Concentrations at all three locations showed significant decreasing trends in ΣDDT concentrations.

Three Locations on the Great Lakes [Eighteenmile Creek (LOEC), Milwaukee Bay (LMMB), and Holland Breakwater (LMHB)] also revealed ΣDDT concentrations above the national 85th percentile. Several studies have demonstrated that atmosphere plays an important role in the transport of contaminants over long distances and concluded that atmospheric transport/deposition is a significant input pathway for persistent organic pollutants into the lakes (Shen et al., 2005; Sun et al., 2006; Gewurtz et al., 2008). This pathway to the Great Lakes alone, however, cannot fully explain the patchy distribution of ΣDDT concentrations observed with less than 10% (3) of the sampled locations ($n = 39$) above the national 85th percentile and 20% (8) of the sites below the national 15th percentile. Thus, local "point sources" of DDTs and breakdown compounds (e.g., runoff from manufacturing sites, wastewater treatment facilities, and agricultural lands) that outweigh atmospheric inputs must still exist at Eighteenmile Creek, Milwaukee Bay, and Holland Breakwater locations. Only the analysis of bivalves from Holland Breakwater indicated that concentrations are significantly decreasing.

Table 4
Concentrations (ng/g dw) of DDD, DDE, and DDT isomers in bivalves from locations above the national 85th percentile.

Site	General location	Specific location	State	Lat_DD	Lon_DD	n	*o,p′*-DDD ± Std	*o,p′*-DDE ± Std	*o,p′*-DDT ± Std	*p,p′*-DDD ± Std	*p,p′*-DDE ± Std	*p,p′*-DDT ± Std	ΣDDTs ± Std	TREND*
MULG	Point Mugu Lagoon	Point Mugu Lagoon	CA	34.10230	−119.10390	1	25.7	15.2	57.0	73.1	703	152	1030	?
SPFP	San Pedro Harbor	Fishing Pier	CA	33.70667	−118.27417	15	16.9 ± 11.7	109 ± 59.1	4.73 ± 7.87	53.1 ± 45.3	542 ± 262	9.25 ± 17.1	736 ± 324	D
PVRP	Palos Verdes	Royal Palms State Pk.	CA	33.71700	−118.32267	16	12.9 ± 14.5	61.0 ± 42.0	0.90 ± 1.27	57.2 ± 65.0	593 ± 336	5.96 ± 5.57	731 ± 433	D
CBPP	Choctawhatchee Bay	Postil Point	FL	30.48233	−86.47930	17	54.4 ± 77.0	7.65 ± 11.3	4.53 ± 5.49	214 ± 176	195 ± 151	21.9 ± 24.0	498 ± 396	D
MBES	Monterey Bay	Elkhorn Slough	CA	36.80983	−121.78517	10	16.5 ± 12.3	8.75 ± 6.71	27.9 ± 31.4	49.9 ± 40.1	254 ± 172	84.2 ± 90.4	441 ± 329	nt
SFEM	San Francisco Bay	Emeryville	CA	37.82050	−122.33000	12	34.9 ± 47.3	3.20 ± 2.28	84.0 ± 164	71.9 ± 72.0	88.6 ± 57.8	135 ± 230	417 ± 558	nt
NHPB	Newport Bay	PCH Bridge	CA	33.61660	−117.90485	1	7.37	7.86	<MDL	39.4	345	7.80	407	?
ABWJ	Anaheim Bay	West Jetty	CA	33.73350	−118.10100	16	7.39 ± 6.99	33.4 ± 33.3	2.27 ± 2.17	40.6 ± 57.8	309 ± 189	7.50 ± 17.7	400 ± 266	D
LBBW	Long Beach	Breakwater	CA	33.72317	−118.17350	12	3.77 ± 2.02	33.7 ± 17.7	2.04 ± 2.30	16.7 ± 9.67	317 ± 152	5.82 ± 4.77	380 ± 175	D
MBHI	Mobile Bay	Hollingers Is. Chan.	AL	30.56333	−88.07500	10	40.9 ± 55.2	112 ± 86.6	10.9 ± 9.44	63.1 ± 64.7	131 ± 71.2	5.43 ± 4.06	362 ± 270	d
HRLB	Hudson/Raritan Estuary	Lower Bay	NY	40.56600	−74.05083	16	43.3 ± 97.4	18.8 ± 22.5	7.82 ± 7.22	99.6 ± 105	98.9 ± 78.8	14.8 ± 18.2	283 ± 285	D
RBMJ	Redondo Beach	Municipal Jetty	CA	33.83200	−118.39283	12	3.98 ± 2.96	19.8 ± 10.4	1.12 ± 1.38	12.6 ± 9.26	234 ± 163	1.87 ± 1.26	274 ± 182	d
PCLO	Panama City	Little Oyster Bar	FL	30.25133	−85.68100	12	13.3 ± 14.8	2.82 ± 1.72	1.83 ± 1.46	89.8 ± 97.9	145 ± 157	11.9 ± 12.3	265 ± 275	D
CBBL	Choctawhatchee Bay	Ben's Lake	FL	30.45317	−86.54100	3	15.3 ± 15.8	3.76 ± 2.54	3.65 ± 3.80	82.8 ± 45.1	150 ± 139	6.99 ± 8.24	262 ± 213	?
MBDR	Mobile Bay	Dog River	FL	30.59167	−88.03980	8	12.1 ± 9.71	79.5 ± 39.4	9.86 ± 14.8	28.8 ± 25.1	102 ± 23.2	3.74 ± 3.70	236 ± 101	D
MBML	Monterey Bay	Moss Landing	CA	36.80117	−121.78967	15	6.78 ± 5.85	4.28 ± 2.79	14.2 ± 12.7	19.8 ± 16.5	127 ± 57.2	38.9 ± 36.6	211 ± 123	nt
MDSJ	Marina Del Rey	South Jetty	CA	33.96183	−118.45800	16	6.95 ± 6.79	10.5 ± 9.47	9.29 ± 21.1	31.2 ± 34.1	132 ± 77.2	16.3 ± 29.8	207 ± 151	D
HRUB	Hudson/Raritan Estuary	Upper Bay	NY	40.68933	−74.04317	15	17.9 ± 11.7	8.42 ± 8.55	8.12 ± 8.5	67.9 ± 50.3	82.1 ± 52.6	14.7 ± 9.45	199 ± 121	D
LOEC	Lake Ontario	Eighteenmile Creek	NY	43.33870	−78.18783	1	16.6	<MDL	5.31	38.7	125	11.7	197	
LARM	Los Angeles	River mouth	CA	33.75525	−118.19498	1	9.46	9.35	<MDL	35.7	136	3.11	193	?
SANM	San Miguel Island	Tyler Bight	CA	34.02800	−120.41933	1	<MDL	1.66	<MDL	<MDL	187	<MDL	188	?
LMMB	Lake Michigan	Milwaukee Bay	WI	43.03217	−87.89517	9	18.1 ± 5.51	9.08 ± 16.5	10.2 ± 4.57	56.4 ± 23.4	79.3 ± 24.1	8.01 ± 4.07	181 ± 63.0	
BRFS	Brazos River	Freeport Surfside	TX	28.92117	−95.33950	14	3.39 ± 2.42	1.39 ± 0.97	1.82 ± 1.80	6.25 ± 4.08	166 ± 76.0	2.09 ± 1.89	181 ± 80.4	D
DBHC	Delaware Bay	Hope Creek	NJ	39.42667	−75.49333	2	16.2 ± 15.3	23.9 ± 8.08	7.23 ± 2.50	29.6 ± 7.97	92.5 ± 54.9	0.59 ± 0.83	170 ± 68.5	?
HRRB	Hudson/Raritan Estuary	Raritan Bay	NY	40.51900	−74.18450	11	29.5 ± 18.3	9.88 ± 4.96	4.38 ± 3.72	66.5 ± 32.6	49.7 ± 22.9	9.36 ± 12.2	169 ± 76.5	nt
NYSH	New York Bight	Sandy Hook	NJ	40.48750	−74.03333	15	15.6 ± 10.5	12.6 ± 13.8	4.17 ± 4.94	60.4 ± 41.3	66.2 ± 42.5	6.00 ± 5.61	165 ± 90.8	D
IBNJ	Imperial Beach	North Jetty	CA	32.58767	−117.13350	16	6.28 ± 7.27	2.17 ± 1.66	2.16 ± 2.09	18.7 ± 25.6	122 ± 84.3	5.95 ± 7.61	158 ± 108	D
LMHB	Lake Michigan	Holland Breakwater	MI	42.77317	−86.21500	8	5.42 ± 4.45	3.55 ± 5.27	7.68 ± 4.23	20.2 ± 14.5	93.7 ± 88.6	14.5 ± 11.3	145 ± 120	
CBBB	Choctawhatchee Bay	Boggy Bayou	FL	30.50400	−86.49400	3	8.65 ± 6.70	1.71 ± 0.45	1.71 ± 0.23	45.4 ± 16.8	82.2 ± 6.97	3.88 ± 1.14	144 ± 30.9	?
MUOS	Point Mugu	Old Stairs	CA	34.06618	−118.99823	1	4.75	6.62	9.47	9.62	88.6	24.2	143	?
DBBD	Delaware Bay	Ben Davis Pt. Shoal	NJ	39.25233	−75.30283	14	14.2 ± 8.14	14.2 ± 13.9	2.06 ± 1.45	30.3 ± 26.0	77.5 ± 53.7	1.19 ± 1.78	139 ± 101	D
PDPD	Point Dume	Point Dume	CA	34.00100	−118.80883	15	2.68 ± 3.62	11.8 ± 17.2	1.16 ± 1.05	10.3 ± 18.0	108 ± 94.3	2.61 ± 3.63	137 ± 133	D
SFDB	San Francisco Bay	Dumbarton Bridge	CA	37.50267	−122.12133	17	9.52 ± 9.06	2.83 ± 4.44	6.03 ± 6.51	33.1 ± 40.0	74.3 ± 42.5	6.83 ± 9.17	133 ± 91.3	D
DBAP	Delaware Bay	Arnolds Point Shoal	NJ	39.38333	−75.45000	14	15.7 ± 6.69	11.4 ± 5.82	1.96 ± 1.62	32.2 ± 18.8	70.1 ± 34.4	0.90 ± 1.53	132 ± 58.9	d
NBWJ	Newport Beach	West Jetty	CA	33.59100	−117.89000	15	2.22 ± 2.68	7.56 ± 3.13	1.10 ± 1.28	9.53 ± 7.47	106 ± 50.0	5.35 ± 5.84	131 ± 61.2	D
CDRF	Cardiff Reef	Cardiff Reef	CA	32.99988	−117.27867	1	3.15	1.83	<MDL	8.37	112	4.97	131	?
DBKI	Delaware Bay	Kelly Island	DE	39.20317	−75.35900	14	13.2 ± 10.5	11.9 ± 8.88	1.48 ± 1.76	25.2 ± 13.7	75.2 ± 50.1	1.07 ± 2.56	128 ± 70.3	D
TJRE	Tijuana River	Estuary	CA	32.56982	−117.12693	1	3.90	2.09	0.86	5.85	114	1.12	128	?
DBWB	Delaware Bay	Woodland Beach	DE	39.33200	−75.45700	1	5.30	21.7	7.00	27.0	66.7	<MDL	128	?
LITN	Long Island Sound	Throgs Neck	NY	40.81667	−73.79833	16	8.65 ± 5.36	2.12 ± 2.51	3.77 ± 4.30	54.3 ± 54.5	55.0 ± 28.1	3.37 ± 2.39	127 ± 73.4	D
MRPL	Mississippi River	Pass A Loutre	LA	29.08950	−89.07480	9	6.74 ± 4.51	0.88 ± 0.79	4.42 ± 4.36	46.2 ± 37.4	54.6 ± 40.1	13.5 ± 15.0	126 ± 97.5	D
OSBJ	Oceanside	Municipal Beach Jetty	CA	33.20167	−117.39367	16	3.47 ± 3.62	3.90 ± 3.02	5.92 ± 7.76	11.6 ± 10.3	84.5 ± 79.7	13.4 ± 14.5	123 ± 114	nt
GBSC	Galveston Bay	Ship Channel	TX	29.70450	−94.99300	11	9.76 ± 6.16	3.46 ± 6.04	5.25 ± 4.52	42.8 ± 22.9	45.7 ± 24.5	4.20 ± 4.74	111 ± 55.4	D
SAWB	St. Andrew Bay	Watson Bayou	FL	30.14250	−85.63220	16	9.43 ± 6.89	1.45 ± 1.31	3.38 ± 2.17	35.0 ± 26.6	54.9 ± 22.7	6.73 ± 4.41	111 ± 50.4	D
TBSM	Las Tunas Beach	Santa Monica Bay	CA	34.03900	−118.59717	12	1.86 ± 1.69	7.69 ± 3.56	1.34 ± 1.53	7.02 ± 5.16	89.4 ± 66.0	2.56 ± 2.37	110 ± 75.6	D
BBAR	Buzzards Bay	Angelica Rock	MA	41.57967	−70.85900	14	22.3 ± 39.7	3.77 ± 12.6	24.6 ± 35.2	16.1 ± 13.1	38.1 ± 66.5	2.26 ± 2.49	107 ± 121	D
HRBP	Hudson/Raritan Estuary	Battery Park	NY	40.70456	−74.01832	1	20.0	3.48	<MDL	26.4	50.9	6.19	107	?
SFSM	San Francisco Bay	San Mateo Bridge	CA	37.57800	−122.25367	17	7.59 ± 4.50	1.53 ± 1.45	5.81 ± 5.68	24.5 ± 24.9	60.0 ± 32.9	7.23 ± 8.81	107 ± 63.4	D
MBCP	Mobile Bay	Cedar Point Reef	FL	30.31550	−88.13380	16	11.6 ± 13.5	27.2 ± 21.6	2.42 ± 3.63	20.9 ± 21.8	41.9 ± 20.2	1.97 ± 2.83	106 ± 70.0	D
BHDB	Boston Harbor	Dorchester Bay	MA	42.30217	−71.03633	14	9.09 ± 5.72	1.57 ± 3.07	6.07 ± 6.72	36.2 ± 31.6	39.3 ± 25.2	9.06 ± 12.3	101 ± 69.6	D
SDHI	San Diego Bay	Harbor Island	CA	32.72467	−117.19467	16	9.48 ± 3.96	3.24 ± 7.53	7.25 ± 4.19	21.9 ± 22.1	54.7 ± 19.0	2.48 ± 1.74	99.1 ± 42.3	D

* Trends are shown as significant decrease (D or d at the 95 or 90% level of confidence, respectively), no significant trend (nt), or not enough data point to analyze (?).

Table 5
Concentrations (ng/g dw) of DDD, DDE, and DDT isomers in bivalves from locations below the national 15th percentile.

Site	General location	Specific location	State	Lat_DD	Lon_DD	*n*	*o,p′*-DDD ± Std	*o,p′*-DDE ± Std	*o,p′*-DDT ± Std	*p,p′*-DDD ± Std	*p,p′*-DDE ± Std	*p,p′*-DDT ± Std	ΣDDTs ± Std	TREND[*]
SSSI	Sapelo Sound	Sapelo Island	GA	31.39283	−81.28800	15	<MDL[**]	0.28 ± 0.47	0.27 ± 0.60	1.45 ± 3.03	4.18 ± 2.67	0.50 ± 1.14	7.18 ± 5.31	D
ARWI	Altamaha River	Wolfe Island	GA	31.32417	−81.31083	12	0.61 ± 0.72	0.30 ± 0.61	0.68 ± 1.06	0.90 ± 0.94	4.07 ± 2.10	0.47 ± 1.11	7.05 ± 4.38	D
PPJB	Partington Point	Julia P. Burns ASBS	CA	36.17500	−121.94000	1	0.72	0.54	<MDL	0.20	5.54	<MDL	7.00	?
LMPI	Lower Laguna Madre	Port Isabel	TX	26.07483	−97.19950	9	1.15 ± 0.84	<MDL	0.35 ± 0.33	1.62 ± 3.07	3.61 ± 2.79	0.18 ± 0.14	6.95 ± 6.30	D
DRSE	Detroit River	South End	MI	42.10690	−83.13550	1	1.13	0.20	<MDL	1.64	3.47	0.47	6.91	?
CBRP	Coos Bay	Russell Point	OR	43.43133	−124.22117	16	0.26 ± 0.39	0.22 ± 0.43	0.50 ± 0.52	1.30 ± 1.44	4.25 ± 3.97	0.31 ± 0.33	6.87 ± 5.07	D
LSAB	Lake St. Clair	Anchor Bay	MI	42.64917	−82.71100	6	0.23 ± 0.40	0.80 ± 0.95	0.23 ± 0.49	0.78 ± 1.43	3.61 ± 4.12	0.81 ± 1.51	6.48 ± 6.97	D
SRNB	Santee River	North Bay	SC	33.16833	−79.24167	12	<MDL	0.19 ± 0.35	0.52 ± 1.11	1.18 ± 2.00	4.15 ± 2.33	<MDL	6.31 ± 4.83	D
HMBJ	Eureka	Humboldt Bay Jetty	CA	40.76417	−124.23750	14	0.32 ± 0.65	0.52 ± 1.14	0.32 ± 0.88	0.95 ± 1.15	3.98 ± 2.64	0.16 ± 0.29	6.28 ± 4.41	d
TBLB	Terrebonne Bay	Lake Barre	LA	29.25950	−90.59430	16	1.20 ± 2.81	0.37 ± 0.67	0.46 ± 0.93	1.49 ± 1.54	2.24 ± 1.47	0.47 ± 0.72	6.26 ± 5.60	D
EVFU	Everglades	Faka Union Bay	FL	25.90233	−81.51230	16	0.82 ± 1.87	0.87 ± 1.28	<MDL	1.30 ± 2.56	2.34 ± 1.51	0.75 ± 2.06	6.22 ± 8.30	D
YHFC	Yaquina Bay	Fogarty Creek	OR	44.83700	−124.05200	10	0.26 ± 0.30	0.24 ± 0.36	0.47 ± 1.11	0.81 ± 1.12	4.24 ± 2.78	0.17 ± 0.25	6.22 ± 3.86	D
EUSB	Eureka	Samoa Bridge	CA	40.82150	−124.17133	12	0.39 ± 0.34	0.53 ± 1.19	0.22 ± 0.38	0.89 ± 0.69	3.53 ± 1.53	0.26 ± 0.31	5.84 ± 3.06	d
TBHP	Tillamook Bay	Hobsonville Point	LA	45.54717	−123.90750	16	0.45 ± 0.99	0.31 ± 0.67	0.46 ± 1.13	1.08 ± 1.50	3.10 ± 1.86	0.39 ± 0.78	5.82 ± 5.00	nt
PDSC	Point Delgada	Shelter Cove	CA	40.02250	−124.07333	16	0.27 ± 0.46	0.41 ± 0.66	0.25 ± 0.56	0.64 ± 0.83	4.10 ± 1.83	<MDL	5.77 ± 2.74	D
SRWP	Suwannee River	West Pass	FL	29.32917	−83.17420	5	0.97 ± 1.46	0.12 ± 0.18	<MDL	1.06 ± 1.35	2.41 ± 2.38	1.03 ± 1.89	5.72 ± 7.10	D
WBLB	Winyah Bay	Lower Bay	SC	33.24333	−79.19717	13	0.32 ± 0.57	0.18 ± 0.26	0.22 ± 0.36	0.79 ± 0.98	4.07 ± 1.90	<MDL	5.63 ± 2.46	D
RBHC	Rookery Bay	Henderson Creek	FL	26.02700	−81.73880	15	0.61 ± 1.14	1.26 ± 1.72	<MDL	0.78 ± 1.69	2.65 ± 1.49	0.23 ± 0.34	5.63 ± 5.05	D
LOOR	Lake Ontario	Oswego River	NY	43.46833	−76.50973	1	0.87	0.15	0.28	1.34	2.31	0.56	5.51	?
LHTB	Lake Huron	Thunder Bay	MI	44.92217	−83.41350	7	0.95 ± 1.52	0.21 ± 0.47	0.29 ± 0.33	1.03 ± 0.90	2.07 ± 2.12	0.92 ± 1.14	5.48 ± 5.51	nt
LMSB	Lower Laguna Madre	South Bay	TX	26.04317	−97.17600	16	0.71 ± 1.48	0.22 ± 0.42	<MDL	0.40 ± 0.59	3.71 ± 3.28	0.28 ± 0.50	5.43 ± 4.53	D
TBLF	Terrebonne Bay	Lake Felicity	LA	29.26417	−90.39820	14	0.71 ± 1.47	0.65 ± 0.90	<MDL	1.19 ± 1.03	1.97 ± 1.13	0.63 ± 0.82	5.32 ± 3.55	D
PSCC	Puget Sound	Cavalero County Park	WA	48.17524	−122.47835	2	0.67 ± 0.94	<MDL	0.22 ± 0.02	0.45 ± 0.24	2.93 ± 0.43	0.95 ± 0.14	5.28 ± 0.20	?
BBTB	Barataria Bay	Turtle Bay	LA	29.51117	−90.08330	2	<MDL	0.67 ± 0.52	0.38 ± 0.38	1.74 ± 1.72	1.93 ± 0.47	<MDL	5.04 ± 2.02	?
PSHC	Puget Sound	Hood Canal	WA	47.83183	−122.68833	12	0.41 ± 0.65	<MDL	0.44 ± 0.69	0.67 ± 0.65	2.80 ± 1.82	0.54 ± 0.62	4.94 ± 3.58	d
PCFB	Pudding Creek	Fort Bragg	CA	39.46085	−123.80929	1	1.26	<MDL	<MDL	<MDL	3.02	<MDL	4.63	?
LSMP	Lake Superior	Minnesota Point	MN	46.71094	−92.02236	1	0.37	<MDL	0.34	1.64	1.26	0.62	4.34	?
MRCB	Matanzas River	Crescent Beach	FL	29.76400	−81.26183	15	0.25 ± 0.38	<MDL	0.17 ± 0.36	0.38 ± 0.66	2.32 ± 1.91	0.72 ± 1.47	3.97 ± 3.55	D
NBES	Nahku Bay	East Side	AK	59.45333	−135.33650	7	0.42 ± 0.62	<MDL	0.85 ± 0.79	0.24 ± 0.17	1.40 ± 0.61	0.70 ± 1.31	3.75 ± 2.74	nt
LOSL	Lake Ontario	St Lawrence River	NY	44.97987	−74.89162	1	1.56	<MDL	<MDL	0.63	1.00	0.22	3.41	?
PSHI	Puget Sound	Hat Island	WA	48.00950	−122.32592	1	0.51	<MDL	0.25	0.33	1.84	0.29	3.33	?
PSKP	Port Susan	Kayak Point	WA	48.13652	−122.36531	1	0.28	<MDL	0.26	0.39	1.74	0.46	3.22	?
SRDM	Sea Ranch	Fort Ross Cove	CA	38.73030	−123.48400	1	<MDL	<MDL	<MDL	0.29	2.76	<MDL	3.11	?
TBLL	Traverse Bay	Leelanau State Park	MI	45.20567	−85.53683	7	0.38 ± 0.38	<MDL	0.25 ± 0.31	0.51 ± 0.51	1.37 ± 1.42	0.46 ± 0.86	3.09 ± 2.86	d
KRFR	Klamath River	Flint Rock Head	CA	41.52717	−124.07967	1	1.27	0.23	<MDL	<MDL	1.29	<MDL	2.93	?
LEAR	Lake Erie	Ashtabula River	MI	41.91123	−80.78768	1	0.68	<MDL	<MDL	0.95	1.15	<MDL	2.87	?
ABBI	Ace Basin	Bass Island	SC	32.48936	−80.52833	2	<MDL	0.19 ± 0.26	<MDL	<MDL	2.35 ± 1.31	<MDL	2.67 ± 0.91	?
DRDP	Duxbury Reef	Duxbury Point	CA	37.89390	−122.70250	1	0.27	<MDL	<MDL	0.68	1.46	<MDL	2.62	?
KTMP	Ketchikan	Mountain Point	AK	55.29383	−131.54800	7	0.24 ± 0.33	<MDL	0.49 ± 0.74	0.30 ± 0.31	0.91 ± 0.41	0.24 ± 0.40	2.29 ± 1.51	d
PVMC	Port Valdez	Mineral Creek Flats	AK	61.13283	−146.46100	14	<MDL	0.27 ± 0.90	0.47 ± 1.19	<MDL	0.90 ± 0.97	0.21 ± 0.61	1.98 ± 2.54	nt
CIHS	Cook Inlet	Homer Spit	AK	59.61450	−151.44417	6	<MDL	0.52 ± 1.18	0.50 ± 1.15	<MDL	0.31 ± 0.24	0.40 ± 0.57	1.84 ± 2.39	nt
UISB	Unakwit Inlet	Siwash Bay	AK	60.96083	−147.64600	14	0.20 ± 0.71	0.35 ± 1.31	0.34 ± 0.90	<MDL	0.54 ± 0.56	<MDL	1.56 ± 2.59	nt
RBNR	Resurrection Bay	Nash Road	AK	60.10208	−149.36416	1	0.23	<MDL	<MDL	<MDL	0.49	0.48	1.45	?
BHKF	Florida Keys	Bahia Honda	FL	24.66117	−81.27300	10	<MDL	0.53 ± 0.71	<MDL	0.34 ± 0.61	0.31 ± 0.24	<MDL	1.33 ± 0.83	d
NGEK	Nushagek Bay	Nushagek Bay	AK	58.79610	−158.53248	1	0.43	0.27	<MDL	<MDL	0.46	<MDL	1.16	?
RBML	Resurrection Bay	Milliers Landing	AK	60.06488	−149.44000	1	<MDL	<MDL	<MDL	<MDL	0.49	0.30	0.79	?
RBMF	Resurrection Bay	Mud Flats	AK	60.11297	−149.37400	2	<MDL	<MDL	<MDL	<MDL	0.25 ± 0.04	0.18 ± 0.02	0.65 ± 0.32	?
CINK	Kachemak Bay	Nanwalek	AK	59.35800	−151.93000	1	<MDL	<MDL	<MDL	<MDL	0.55	<MDL	0.64	?
NINB	Ninilchik	North Beach	AK	60.05139	−151.66889	1	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	<MDL	?

[*] Trends are shown as significant decrease (D or d at the 95 or 90% level of confidence, respectively), no significant trend (nt), or not enough data point to analyze (?).
[**] Depending on the sample size, the typical Method Detection Limit (MDL) ranged from 0.05 to 0.30 ng/g dw for *o,p′*-DDT and *o,p′*-DDD.

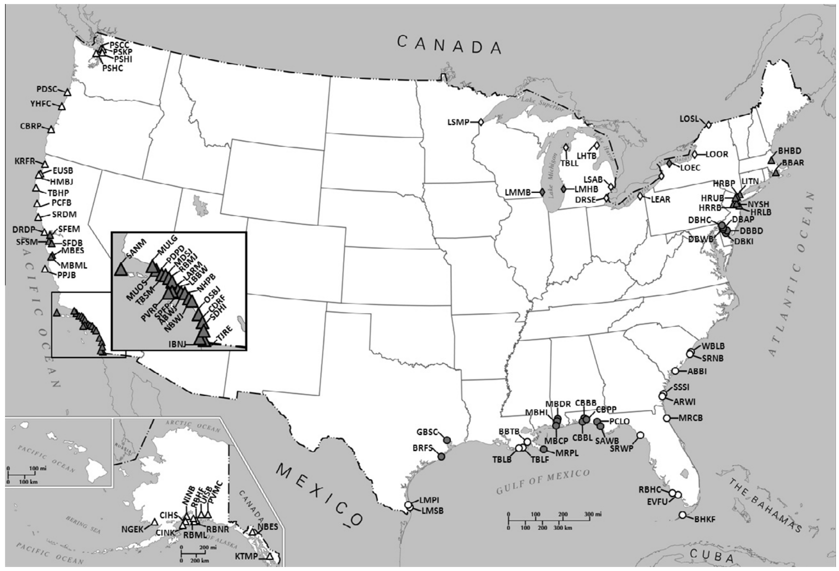

Fig. 5. Spatial distribution of ΣDDT concentrations above the national 85th percentile (closed markers) and concentrations below the national 15th percentile (open markers). See Fig. 2 for bivalves sampled at each location and Table 4 for site description and average concentrations.

In the Gulf of Mexico, NOAA's sites yielding bivalves with average ΣDDT concentrations above the national 85th percentile are concentrated on the northwestern (Panhandle) part of Florida and Alabama. In Mobile Bay, DDT, produced in a factory located on the northwestern edge of the Mobile-Tensaw Delta and used extensively by Alabama farmers, has washed off agricultural lands and ended up in the Bay (Hinck et al., 2009). The highest concentrations were detected in bivalves from the upper portion of the bay [Hollingers Island (MBHI) and Dog River (MBDR)] and decreased toward the Gulf of Mexico, Cedar Point Reef (MBCP), where most of local oyster harvesting occurs. Thus, DDT and breakdown products seem to contaminate the Mobile River before it reaches the Bay and their concentrations are diluted as the river water enters the Bay. All three locations in Mobile Bay revealed significantly decreasing trends in ΣDDT concentrations. Of the three sites in Choctawhatchee Bay [Postil Point (CBPP), Ben's Lake (CBBL), and Boggy Bayou (CBBB)], only Postil Point was sampled a sufficient number of times to discern a trend. The remaining locations on the Florida Panhandle [Little Oyster Bar near Panama City (PCLO) and Watson Bayou in St. Andrew Bay (SAWB)] also indicate that ΣDDT concentrations in the area are decreasing. Similar trends were detected at three isolated points in the Gulf of Mexico located on the mouth of the Mississippi (MRPL) and Brazos (BRFS) rivers, and upper Galveston Bay near the Houston Ship Channel (GBSC).

The lowest average ΣDDT concentrations in the continental U.S. were encountered principally in Alaska (all locations), on the northwestern and southeastern coasts of the USA, and on the western Gulf of Mexico coast with most of them showing decreasing trends.

4. Conclusions

While the use of DDT in the United States was banned or rigorously restricted forty years ago, this compound and breakdown products continue to be available to bivalves at virtually every location in the continental U.S. coastal areas sampled during the NOAA's NS&T "Mussel Watch" Program. The concentration in the coastal areas, as a whole, is decreasing with a half-life of about 10–14 years; faster in the south, slower in the north. Under these dissipation rates, average concentrations along the East, Gulf, and West coasts will decrease below 10% of today's concentrations by approximately 2050. This general decrease is also observed in many sampling areas; although, the distribution of sites with the highest concentrations is correlated to areas where DDT was produced or heavily used. In these areas (i.e., Southern California Bight or Delaware and Mobile Bays), surrounding agricultural soils, watersheds and marine sediments may continue to be a source of DDT and related compounds for many decades.

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