Developments in Environmental Science, Volume 7
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ISSN: 1474-8177/DOI:10.1016/S1474-8177(07)07015-5

Chapter 15

Persistent Organic Pollutants in Singapore's Marine Environment

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Abstract

Singapore is one of the busiest ports in the world and has many shipyards, petroleum refineries and pharmaceutical manufacturing plants are located on its coastline. Data on the prevalence of persistent organic pollutants (POPs) in Singapore's coastal ecosystems are therefore important in order to support research on the potential threats to the local marine environment and human health. This chapter presents data on the prevalence of POPs in the seawater, sediments, biota and mangrove habitats of Singapore. Data are presented for a range of POPs including polycyclic aromatic hydrocarbons (PAHs), organochlorine compounds (OCPs), polychlorinated biphenyls and polybrominated diphenyl ethers (PBDEs). Local studies confirm the ubiquity of POPs in the marine environment of Singapore. The prevailing ocean currents in the region govern the fate and transport of POPs in coastal waters, where the presence of localized high levels of POPs is likely to be a function of local shipping and industrial discharges. Land reclamation activities may also be a factor in the prevalence of POPs in seawater, where compounds are released from sediments and transported to the water column by seabed dredging. The land area under agricultural use in Singapore is negligible, and there is no direct application of OCPs in the country. However, pesticides may be easily transported through the atmosphere following volatilization from soil, and the presence of OCPs in Singapore's marine waters is likely to be a function of their use in neighbouring countries, with subsequent atmospheric transport and deposition. A biomagnification phenomenon was observed amongst the species collected and analysed from mangrove sites, but levels of POPs were not found to exceed relevant food safety standards. However, PCB concentrations in mangrove fish muscles were found to be higher than mean levels of PCBs found in seafood commonly

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consumed in Singapore. Overall, available data on the prevalence, fate and transfer of POPs in Singapore's marine environment highlights the ubiquity of these pollutants, and supports the need for a greater awareness on their fate, transport and bioaccumulation in local ecosystems.

15.1. Introduction

Persistent organic pollutants (POPs), including organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) are of significant concern due to their potential toxicity and prevalence in a wide range of environmental media, even at remote geographical locations (Iwata et al., 1994; Kallenborn et al., 1998). Ecotoxicological effects of POPs in the environment have caused much concern in recent years, and this has led to the control or complete ban on the use of these chemicals in many countries. PAHs are mainly derived from incomplete fossil fuel combustion and oil-related discharges to the environment (UNEP, 1997). When released to the aquatic environment, PAHs can adversely impact upon a wide variety of flora and fauna. Chlorinated organic compounds have a wide range of industrial and agricultural applications, and include the pesticides DDT (*p*,*p*′-dichlorodiphenyl trichloroethane) and Lindane (γ-hexachlorocyclohexane), as well as the PCBs. The latter have been used historically in an extensive range of industrial applications, including as dielectric fluids in electrical transformers. Such chemicals can readily enter the aquatic environment via atmospheric deposition, groundwater leaching, soil run-off and sewage discharge (Harrad, 2000), and subsequently impact the marine ecosystem.

POPs are known to adversely affect the endocrine system in both wild fauna and humans, have a propensity to bioaccumulate in the lipid fraction of biological tissues and are subject to biomagnification in both terrestrial and aquatic food webs (Li et al., 2006). POPs are also highly recalcitrant to chemical and biological degradation, and therefore persist in the environment for long periods. Humans are chronically exposed to environmental POPs via the ingestion and inhalation pathways (Duarte-Davidson and Jones, 1994), and many such compounds have been detected in a range of human tissues including serum, breast milk and adipose tissue (Dewailly et al., 1993; Newsome et al., 1995). In 2001, many countries signed the Stockholm Convention under the United Nations Environment Programme (UNEP) to implement measures to reduce and eliminate the release of POPs into the environment, including bans on production, import, export, and use of certain POPs (UNEP 2001).

Singapore is one of the busiest ports in the world with an annual visiting shipping tonnage in excess of 857 million gross tons per year, representing more than 140,000 ship movements (Nautilus, 1999). In addition, many ship building yards, petroleum refineries and pharmaceutical manufacturing plants are located along the coastline and are associated with both operational and fugitive discharges of POPs. Data on the prevalence of POPs in Singapore's coastal waters is therefore important in order to support research on potential threats to the local marine ecosystem and human health. This chapter presents data on the prevalence of POPS in the seawater, sediments, biota and mangrove habitats in Singapore's coastal marine environment.

15.2. POPs in seawater

Marine water samples within 1 km of the coastline of Singapore were analysed to determine prevalent concentrations of a range of POPs by Basheer et al. (2003a). Samples were collected from 0.5 m and mid-depth (approximately 14 m depth) at 22 coastal locations (Fig. 15.1). POPs analysed are classed as USEPA priority pollutants, and included: 16 PAHs; 8 PCBs; and 12 OCPs.

15.2.1. Polycyclic aromatic hydrocarbons

The individual and total concentrations of PAHs at surface (S) and mid-depth (M) for northeastern (NE) and southwestern (SW) sampling locations are shown in Tables 15.1a and 15.1b, respectively. All 16 PAHs were detected in all water samples from both depths at every sample location. Total PAH concentrations in seawater ranged from 93.0 to 1419.6 ng L^{-1} and from 88.4 to 1472.8 ng L^{-1} in the northeastern and southwestern region, respectively. The overall mean total PAH concentrations for seawater depth levels were as follows: surface, 235.1 ± 46.2 ng L^{-1}; and mid-depth, 343.1 ± 61.5 ng L^{-1}. The highest total PAH concentrations measured in the northeastern and the southwestern regions were at stations NE-02 and SW-02, respectively. These two stations are in the vicinity of shipyards and industrialized coastal areas (i.e., Jurong and Sembawang). The lowest concentrations of total PAHs for these regions were at NE-12 and SW-04. Both of these locations are remote from industrial areas and the water column is well mixed by strong oceanic currents. Among the 16 PAHs measured, the highest individual PAH concentrations measured were for six ring indeno[1,2,3-*cd*]pyrene i.e., 712.9 ng L^{-1} and 218.8 ng L^{-1} at northeastern and

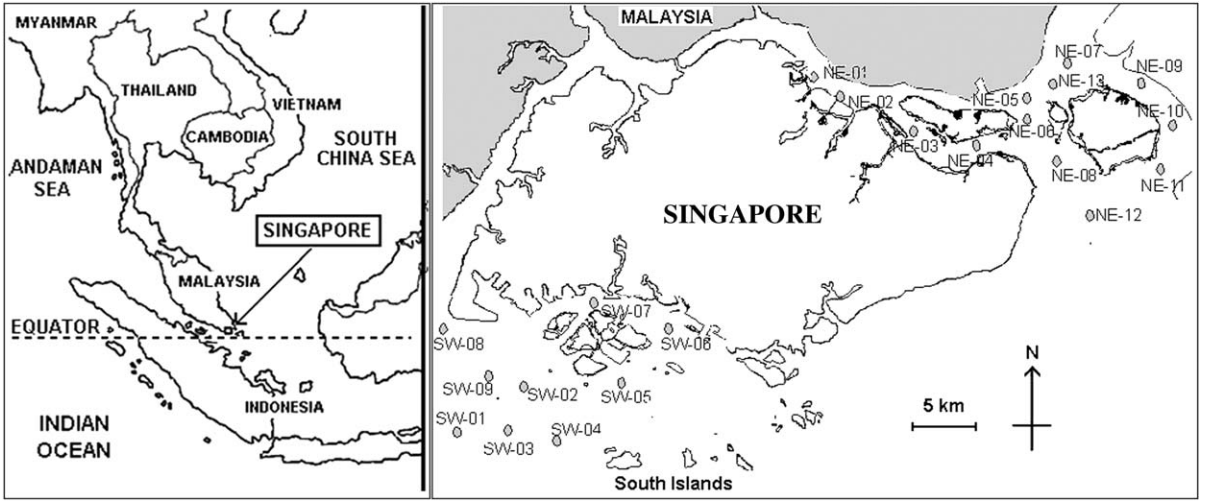

Figure 15.1. Sample locations in the northeastern and southwestern regions of Singapore's Coastal Waters.

Table 15.1a. Concentration of PAHs in seawater of the northeastern coastal region of Singapore (ng L^{-1})

	NE-01		NE-02		NE-03		NE-04		NE-05		NE-06	
	S	M	S	M	S	M	S	M	S	M	S	M
Naphthalene	0.68	2.41	2.62	4.32	2.20	1.92	2.49	3.87	2.07	2.19	2.85	0.22
Acenaphthylene	nd	0.66	0.64	0.52	0.69	0.18	0.73	0.64	0.17	0.61	0.51	0.30
Acenaphthene	0.12	1.11	0.93	1.17	0.58	0.43	0.96	1.14	0.64	0.23	1.01	0.03
Fluorene	0.85	0.39	0.49	1.14	1.12	1.19	1.14	0.61	0.68	0.85	1.63	0.68
Phenanthrene	2.73	4.06	5.87	4.49	10.82	1.00	6.50	3.63	4.50	1.96	2.87	1.72
Anthracene	16.68	30.03	48.64	37.41	73.66	8.73	45.56	22.33	23.42	6.48	24.32	11.81
Fluoranthene	1.18	4.53	2.04	2.44	3.37	1.11	2.30	0.97	5.18	0.84	0.86	1.00
Pyrene	5.96	19.41	3.27	6.20	16.96	5.61	14.85	4.91	5.26	4.21	4.33	5.03
Benz[*a*]anthracene	3.99	4.65	6.89	6.02	1.59	1.37	3.77	4.38	6.29	6.62	5.14	1.86
Chrysene	2.41	9.30	13.79	2.74	3.17	2.74	7.55	8.10	2.08	13.24	10.29	1.86
Benzo[*a*]fluoranthene	11.88	11.60	4.18	4.05	15.10	5.88	9.07	18.06	17.05	5.52	8.84	13.86
Benzo[*k*]fluoranthene	3.71	3.02	2.77	3.56	20.57	5.89	3.30	30.38	9.04	5.54	5.31	7.52
Benzo[*a*]pyrene	6.01	1.53	2.12	0.94	7.64	2.77	1.74	9.14	5.44	2.80	4.47	3.80
Indeno[1,2,3-*cd*]pyrene	54.51	86.48	22.89	189.68	607.92	97.75	136.26	526.53	66.13	214.03	343.02	53.42
Dibenz[*a*,*h*]anthacene	5.56	4.11	2.27	22.95	403.08	4.22	6.28	68.12	10.43	10.82	27.93	6.10
Benzo[*ghi*]perylene	5.41	2.63	2.27	22.93	251.10	9.70	13.52	68.06	6.56	7.21	34.03	4.94
Sum	121.69	185.93	121.69	310.52	1419.57	150.49	256.03	770.86	164.95	283.14	477.40	114.13

Table 15.1a. (*Continued*)

NE-07		NE-08		NE-09		NE-10		NE-11		NE-12		NE-13	
S	M	S	M	S	M	S	M	S	M	S	M	S	M
0.41	0.56	1.78	1.62	2.49	1.67	3.55	0.69	0.41	0.23	0.43	0.84	0.38	1.84
0.72	1.26	1.33	0.53	nd	0.57	0.44	0.17	0.40	0.51	0.35	0.12	0.15	1.01
0.23	0.22	0.18	0.56	0.57	0.66	1.30	0.32	0.34	0.16	0.45	0.14	0.26	0.58
1.36	1.16	2.69	0.33	0.28	1.44	1.87	0.34	1.28	0.62	0.92	0.29	1.03	0.58
3.46	2.72	11.55	1.65	1.37	2.87	7.66	3.23	4.95	1.68	1.48	3.86	3.01	3.23
14.05	15.46	77.96	18.99	8.09	21.30	49.02	26.76	23.93	15.40	31.70	34.52	23.10	17.52
1.56	2.81	5.01	5.25	2.92	0.51	1.07	1.97	0.70	3.09	0.67	2.92	2.09	1.51
5.03	10.17	25.26	4.56	14.73	8.19	5.38	9.94	3.51	15.55	3.39	14.73	6.78	10.06
3.77	5.96	1.59	3.17	2.52	3.72	2.52	0.88	2.68	4.87	5.63	8.48	4.65	5.74
7.55	7.77	3.17	16.53	4.82	7.44	5.14	7.66	1.31	9.74	4.27	16.96	9.30	3.17
17.58	3.85	6.74	2.99	17.96	5.29	1.19	6.36	18.52	7.40	9.45	9.42	11.02	15.45
9.45	13.79	6.76	7.24	15.01	5.31	29.28	1.73	18.57	7.42	3.10	5.21	11.05	6.76
7.50	5.28	3.41	0.90	7.58	2.68	5.28	2.36	9.37	3.74	1.56	4.38	5.58	3.77
91.21	133.36	194.40	328.13	107.92	437.50	418.97	180.23	712.94	119.91	24.71	23.26	308.87	41.79
5.45	13.24	19.30	32.58	9.85	43.44	78.84	9.20	70.79	4.73	2.45	2.31	30.67	4.15
4.51	4.00	19.28	4.72	9.84	43.40	78.76	9.19	70.72	5.44	2.45	2.31	30.64	4.15
173.83	221.63	380.43	429.75	205.94	586.00	690.28	261.03	940.41	200.49	93.02	129.77	448.58	121.30

Table 15.1b. Concentration of PAHs in seawater of the southwestern coastal region of Singapore (ng L^{-1})

	SW-01		SW-02		SW-03		SW-04		SW-05		SW-06		SW-07		SW-08		SW-09	
	S	M	S	M	S	M	S	M	S	M	S	M	S	M	S	M	S	M
Naphthalene	0.86	1.23	0.62	0.55	2.66	0.84	0.31	0.88	2.68	1.29	0.69	0.65	2.49	0.34	0.46	1.40	0.76	0.40
Acenaphthylene	0.80	0.85	0.17	0.77	1.20	nd	0.52	0.77	1.90	0.70	0.38	0.30	1.49	0.79	0.40	0.54	0.62	0.27
Acenaphthene	0.20	0.20	0.16	0.31	0.96	0.28	0.11	0.24	0.39	0.43	0.35	0.15	0.52	0.14	0.08	0.38	0.16	0.19
Fluorene	0.92	2.02	0.88	1.05	3.92	1.67	1.45	2.35	3.73	1.94	0.95	2.25	2.97	1.21	1.21	1.54	1.51	1.60
Phenanthrene	3.19	4.63	4.43	9.48	6.37	5.13	2.04	1.97	3.77	4.32	1.65	9.05	6.28	0.31	1.79	7.28	3.05	1.78
Anthracene	14.89	36.70	15.72	63.85	31.96	18.10	7.76	20.21	23.29	31.57	nd	59.42	37.28	11.36	16.30	59.93	24.38	17.77
Fluoranthene	2.09	2.99	3.74	1.37	4.62	2.79	3.39	3.04	4.76	1.81	1.62	1.56	4.02	0.81	2.97	5.32	1.44	0.97
Pyrene	10.52	3.86	11.11	9.47	23.27	13.80	12.63	15.32	23.97	9.12	8.19	16.14	20.23	4.09	4.91	26.78	7.25	4.91
Benz[*a*]anthracene	3.50	3.23	2.68	1.64	7.22	4.38	1.09	5.09	9.13	6.13	0.82	0.88	3.12	7.44	7.38	6.13	5.91	4.16
Chrysene	7.00	61.40	4.16	3.28	9.08	8.76	2.41	4.60	18.28	12.26	2.95	15.21	5.03	5.03	12.48	9.63	11.82	8.32
Benzo[*a*]fluoranthene	15.10	14.21	11.27	29.31	25.94	6.28	13.60	14.31	5.42	14.87	4.21	4.76	19.56	9.40	10.49	31.69	1.70	15.71
Benzo[*k*]fluoranthene	5.13	8.13	4.04	29.39	26.01	3.48	10.49	15.32	5.44	17.07	5.54	4.77	16.51	9.42	5.28	31.77	5.13	13.64
Benzo[*a*]pyrene	1.29	4.10	3.03	14.83	4.76	3.18	6.06	12.40	3.77	8.62	2.13	2.09	8.99	4.76	1.17	8.18	0.86	5.60
Indeno[1,2,3-*cd*]pyrene	86.85	196.22	93.75	73.75	218.75	76.67	22.17	200.94	38.15	198.04	140.26	103.92	175.51	90.48	90.12	85.76	146.80	210.76
Dibenz[*a*,*h*]anthacene	8.62	14.00	7.54	502.13	7.90	7.61	2.20	19.95	3.79	19.67	5.27	13.93	14.76	8.98	5.02	6.03	14.58	11.58
Benzo[*ghi*]perylene	8.62	19.47	7.53	731.56	7.89	4.29	2.20	19.93	3.78	7.32	11.03	11.68	17.41	3.78	8.94	16.55	14.56	9.84
Sum	169.58	373.24	170.84	1472.75	382.51	157.25	88.44	337.32	152.26	335.13	186.03	246.76	336.15	158.36	168.99	298.91	240.54	307.51

southwestern regions, respectively. The lowest concentrations of acenapthylene detected (three ring) were 1.3 ng L^{-1} and 1.9 ng L^{-1}, respectively. This distribution profile may reflect the different properties of low and high molecular weight PAHs, where low molecular weight compounds have higher vapour pressure and water solubility, and are therefore more readily volatilized and degraded by microbial activity. In contrast, higher molecular weight PAHs are more likely to be associated with the particulate phase within the water column and undergo sedimentation, thereby accounting for their higher concentration at mid-depth. Similar vertical distributions have been previously noted in a study of Baltic coastal waters by Broman et al. (1991). At mid-depth, PAHs were dominated by indeno[1,2,3-*cd*]pyrene, but other abundant compounds included dibenzo[*ah*]anthracene, benzo[*ghi*]perylene and anthracene.

Overall, higher molecular PAH compounds were more prevalent in Singapore coastal waters than lower molecular weight compounds. The prevailing ocean currents in the region most likely govern the fate of PAHs, and the presence of localized high levels of PAHs may be a function of petroleum discharges from shipping and the petrochemical industry where hydrocarbons rapidly become associated with both hydrophobic organic and suspended particulates (Capone and Bauer, 1992). In the coastal waters of Singapore, land reclamation activity may also be an additional factor in the increase of POPs in the water column. POPs are readily released from sediments and transported to the water column as a result of disassociation or and solubility of colloidal organic matter upon suspension.

The highest total PAH concentration detected in Singapore's coastal waters (i.e., 1472.8 ng L^{-1}) is greater than that reported for Xiamen Harbour, China (i.e., up to 945 ng L^{-1}; Zhou et al., 2000); the German-Baltic sea (i.e., 6.7 ng L^{-1}; Witt, 1995); the Coral Sea, Australia (i.e., 240 ng L^{-1}; Smith et al., 1987); Chesapeake Bay, USA (i.e., 14.05 ng L^{-1}; Ko and Baker, 1995); the Northwestern Black Sea, Ukraine (i.e., 0.7 ng L^{-1}; Maldonado et al., 1999); and Admiralty Bay, Antarctica (i.e., 80 ng L^{-1}; Bícego et al., 1996), but lower than concentrations reported for Rhode Island, USA (i.e., 115,000 ng L^{-1}) during an oil spill event (Reddy and Quinn, 1999). Overall, the data indicates extensive PAH contamination of Singapore's coastal waters.

15.2.2. Organochlorine pesticides (OCPs)

The individual and total concentrations of OCPs at surface and mid-depth for northeastern and southwestern sampling locations are shown in Tables 15.2a and 15.2b, respectively. In Singapore, extensive agricultural

Table 15.2a. Concentration of OCPs and PCBs in seawater of the northeastern coastal region of Singapore (ng L^{-1})

POPs	NE-01		NE-02		NE-03		NE-04		NE-05		NE-06	
	S	M	S	M	S	M	S	M	S	M	S	M
OCPs												
α-BHC	0.39	0.42	0.15	0.82	1.42	0.13	0.21	0.96	0.45	0.14	2.29	0.41
Lindane	0.02	0.06	0.02	0.02	0.21	0.03	0.03	0.14	0.07	0.02	0.34	0.06
β-BHC	11.41	4.92	4.14	3.12	2.31	2.39	5.01	8.22	8.74	6.37	3.24	2.59
Heptachlor	0.07	0.04	0.05	0.03	0.03	0.04	0.03	0.16	0.02	0.06	0.14	0.04
Aldrin	0.18	0.27	0.06	0.45	0.55	0.06	0.15	1.26	0.05	0.02	0.58	0.03
Dieldrin	3.84	1.12	2.27	1.92	3.48	3.48	0.34	1.71	2.03	2.74	0.96	1.68
Endrin	1.81	0.23	0.37	0.52	nd	0.48	0.42	0.34	1.71	1.87	1.13	0.71
Endosulfan II	0.07	0.14	nd	0.48	0.58	nd	nd	1.03	0.23	0.07	0.32	0.30
p,p′-DDD	0.06	0.06	0.09	0.32	0.52	0.03	0.08	0.76	0.05	0.08	0.22	0.02
p,p′-DDT	0.07	0.02	0.05	0.19	0.67	0.02	0.05	0.72	0.02	0.02	0.36	nd
Endrin aldehyde	0.13	nd	nd	nd	0.33	nd	0.21	nd	nd	nd	nd	nd
Methoxychlor	nd	nd	nd	nd	0.02	nd	nd	0.01	nd	nd	nd	nd
Sum	18.05	7.27	7.20	7.86	10.13	6.67	6.54	15.32	13.37	11.40	9.57	5.84
PCBs												
2-Chlorobiphenyl	0.01	0.03	0.01	0.02	nd	0.01	0.43	0.05	0.02	nd	0.43	nd
2,3-Dichlorobiphenyl	0.15	0.09	4.95	2.19	0.03	0.57	5.65	0.42	0.44	0.04	5.65	0.11
2,4,5-Trichlorobiphenyl	0.04	0.07	0.08	0.01	0.01	0.05	0.02	0.04	0.05	nd	0.13	0.04
2,2′,4,4′-Tetrachlorobiphenyl	0.12	0.03	0.10	0.08	0.09	nd	0.97	0.03	0.07	nd	0.97	0.06
2,2′,3′,4,6-Pentachlorbiphenyl	0.02	0.01	4.40	1.56	0.05	0.27	1.42	0.19	0.24	nd	4.17	0.10
2,2′,4,4′,5,6′-Hexachlorobiphenyl	0.02	0.01	1.33	0.19	0.08	0.06	0.54	0.04	0.04	nd	0.54	0.07
2,2′,3,3′,4,4′,6-Heptachlorobiphenyl	nd	0.08	0.05	0.11	nd	0.07	nd	0.03	0.04	nd	0.04	0.08
2,2′,3,3′,4,5′,6,6′-Octachlorobiphenyl	0.03	0.03	8.50	6.64	1.75	1.59	3.54	0.86	1.04	nd	3.54	0.12
Sum	0.38	0.35	19.42	10.79	2.01	2.62	12.55	1.65	1.94	0.04	15.46	0.57

Table 15.2a. (*Continued*)

NE-07		NE-08		NE-09		NE-10		NE-11		NE-12		NE-13	
S	M	S	M	S	M	S	M	S	M	S	M	S	M
OCPs													
nd	0.54	0.87	0.26	nd	0.77	0.97	0.22	0.29	0.14	0.55	0.25	0.50	0.84
nd	0.02	0.13	0.02	nd	0.12	0.15	0.04	0.04	0.07	0.03	0.04	0.07	0.13
3.72	3.99	5.93	6.70	1.93	6.36	6.42	6.11	15.16	10.24	6.97	1.68	17.87	5.72
0.03	0.04	0.04	0.06	0.03	0.03	0.05	0.10	0.03	0.06	0.05	0.02	0.02	0.09
0.03	0.11	0.58	0.06	0.06	0.65	1.48	0.10	0.38	0.05	0.03	0.02	0.03	1.08
1.92	2.63	2.45	1.21	3.41	3.40	6.91	2.06	2.28	2.05	0.76	1.61	1.68	1.42
0.24	0.33	1.21	0.34	nd	0.66	0.22	0.40	0.98	nd	0.61	0.26	1.33	0.58
nd	nd	0.53	nd	nd	1.29	1.77	nd	0.30	0.16	nd	nd	0.23	0.73
0.01	0.03	0.36	0.10	0.02	0.65	1.17	0.05	0.27	0.14	0.04	0.11	0.10	0.29
0.17	0.08	0.29	0.04	0.03	0.46	1.41	0.04	0.20	0.05	0.04	0.10	0.10	0.37
nd	nd	nd	nd	nd	nd	0.13	nd	0.15	nd	nd	nd	0.11	nd
nd	nd	nd	nd	nd	nd	0.01	nd	0.01	nd	nd	nd	0.02	nd
6.11	7.77	12.39	8.78	5.47	14.38	20.70	9.13	20.09	12.96	9.10	4.09	22.04	11.24
PCBs													
0.08	0.02	nd	0.01	nd	0.00	nd	0.03	0.04	0.02	0.02	0.06	0.06	0.04
0.01	0.06	0.07	0.10	0.04	0.13	0.03	0.08	1.77	0.06	0.65	0.09	12.16	4.17
0.02	0.41	0.09	0.07	0.03	0.01	0.01	0.04	0.16	0.04	1.00	0.08	0.59	0.04
0.30	nd	0.02	nd	0.07	0.02	0.01	nd	0.02	0.06	0.01	0.06	nd	nd
0.53	0.06	1.12	0.09	0.10	0.04	0.22	0.05	0.89	0.44	0.44	0.01	7.34	2.39
0.11	0.05	0.03	0.03	0.08	0.07	nd	0.04	0.14	0.03	0.04	nd	0.84	0.36
0.03	0.04	0.02	0.10	0.02	0.02	0.06	0.07	0.03	0.11	0.04	0.08	0.07	nd
2.66	2.19	0.19	0.18	2.12	0.09	0.30	0.13	5.11	0.21	1.23	0.07	40.71	12.10
3.75	2.82	1.54	0.57	2.46	0.38	0.65	0.43	8.15	0.97	3.44	0.45	61.76	19.11

Table 15.2b. Concentration of OCPs and PCBs in seawater of the southwestern coastal region of Singapore (ng L^{-1})

POPs	SW-01		SW-02		SW-03		SW-04		SW-05		SW-06		SW-07		SW-08		SW-09	
	S	M	S	M	S	M	S	M	S	M	S	M	S	M	S	M	S	M
OCPs																		
α-BHC	0.46	0.35	0.14	0.90	0.10	0.94	0.81	0.94	1.19	0.28	0.72	0.34	0.39	nd	0.12	1.10	0.77	0.63
Lindane	0.07	0.02	0.02	0.14	0.02	0.14	0.12	0.14	0.18	0.04	0.03	0.05	0.07	nd	0.02	0.17	0.12	0.01
β-BHC	2.32	6.49	1.85	6.29	11.45	3.38	8.14	3.38	10.12	3.36	3.14	1.11	4.95	2.89	2.09	15.08	6.36	6.69
Heptachlor	0.03	0.04	0.04	0.02	0.03	0.08	0.03	0.08	0.03	0.05	0.06	0.03	0.04	0.10	0.12	0.10	0.03	0.04
Aldrin	0.04	0.13	0.13	1.17	0.02	0.05	0.04	0.05	1.03	0.12	0.07	0.04	0.05	0.03	0.06	0.68	0.65	0.13
Dieldrin	1.72	0.82	0.22	5.21	0.75	1.14	0.33	1.14	2.78	1.48	0.58	2.44	1.33	1.23	0.91	2.52	3.40	0.88
Endrin	0.29	1.18	nd	0.32	0.31	0.24	nd	0.24	1.78	0.20	0.18	nd	0.50	0.36	0.30	0.21	0.66	0.81
Endosulfan II	nd	nd	0.52	1.24	nd	nd	nd	nd	0.95	nd	nd	nd	0.07	nd	nd	0.99	1.29	nd
p,p′-DDD	0.05	0.03	0.03	0.81	0.02	0.04	0.06	0.04	0.62	0.03	0.02	0.05	0.04	0.04	0.05	0.41	0.65	0.11
p,p′-DDT	0.06	0.05	nd	1.12	0.05	0.05	0.05	0.05	0.62	0.07	0.09	0.05	0.06	0.04	0.08	0.50	0.46	0.06
Endrin aldehyde	nd	nd	nd	0.16	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.16	nd	nd
Methoxychlor	0.01	0.01	nd	nd	nd	nd	nd	nd	nd	nd	nd	nd	0.00	nd	nd	0.01	nd	0.01
Sum	5.06	9.13	2.95	17.38	12.76	6.06	9.57	6.06	19.30	5.63	4.90	4.12	7.50	4.69	3.75	21.93	14.38	9.37
PCBs																		
2-Chlorobiphenyl	0.03	0.06	0.01	0.05	0.02	0.06	0.05	0.04	0.14	nd	0.03	0.03	0.08	0.04	0.03	0.02	0.04	0.02
2,3-Dichlorobiphenyl	0.18	0.14	0.46	0.03	0.09	0.05	0.02	0.06	0.06	0.06	0.06	0.04	5.98	0.07	0.33	0.04	0.40	4.01
2,4,5-Trichlorobiphenyl	0.01	1.10	0.07	0.01	0.01	0.03	0.02	0.07	0.02	0.03	0.01	0.84	0.02	1.12	0.02	0.02	0.04	0.21
2,2′,4,4′-Tetrachlorobiphenyl	nd	nd	2.78	0.09	nd	0.04	0.01	0.02	0.03	0.07	0.09	0.01	0.07	0.01	0.10	0.07	nd	nd
2,2′,3′,4,6-Pentachlorbiphenyl	nd	0.04	4.17	0.07	nd	0.10	0.02	0.08	0.02	0.04	0.08	0.06	4.62	0.06	0.22	0.02	0.24	0.28
2,2′,4,4′,5,6′-Hexachlorobiphenyl	nd	0.07	0.51	0.00	0.01	nd	nd	0.04	0.01	0.03	nd	0.05	0.83	0.02	0.02	0.04	0.05	0.42
2,2′,3,3′,4,4′,6-Heptachlorobiphenyl	nd	0.18	0.06	nd	0.11	nd	0.02	0.12	0.18	0.02	0.10	nd	0.11	0.02	0.07	0.04	0.02	0.05
2,2′,3,3′,4,5′,6,6′-Octachlorobiphenyl	nd	0.14	4.15	0.03	0.14	0.14	0.10	0.08	0.18	0.12	0.16	0.09	2.47	0.78	0.58	0.24	0.78	15.42
Sum	0.22	1.72	12.21	0.28	0.38	0.41	0.24	0.51	0.62	0.36	0.52	1.12	14.19	2.13	1.37	0.49	1.57	20.41

activities have been phased out for more than two decades. Although some minimal agricultural activities remain, they do not generally involve extensive use of the types of pesticides discussed in the present work. However, OCPs were detected in samples taken at both depths from all locations from both the northeastern and southwestern regions.

Total OCP concentrations ranged from 4.0 to 22.0 ng L^{-1} and 3.0 to 21.9 ng L^{-1} at the northeastern and southwestern regions, respectively. The highest total OCP concentrations occurred in the northeastern region at location NE-13 (i.e., 22.0 ng L^{-1}) and in the southwestern region at location SW-08 (i.e., 21.9 ng L^{-1}). Overall, higher concentrations were detected in the northeastern region, which is most likely due to the confined configuration of the coast, and the presence of the Pasir Gudong river estuary, which limits hydrodynamic dispersion of contaminants. This river runs across agricultural, commercial and industrial land in Malaysia and into the Straits of Johor, adjacent to Singapore. Lindane (BHC) and Dieldrin were the most abundant pesticides present and their levels ranged from 0.13 to 17.87 ng L^{-1} and 0.34 to 6.91 ng L^{-1} in northeastern and southwestern regions, respectively. In both regions, the highest concentrations recorded were: Lindane, 0.34 ng L^{-1}; Endrin, 1.97 ng L^{-1}; *p*,*p*′-DDT, 1.14 ng L^{-1}; and *p*,*p*′-DDD, 1.17 ng L^{-1}.

OCPs are, to a variable extent, insoluble in seawater (not more than 1 ppb), but are readily soluble in fat and adsorb strongly onto suspended particulates in the water column. The surface layer of the sea comprises a film of about 1 mm of thickness, which is known to contain surfactants, protein and lipids. Due to the lipophilic and persistent nature of OCP, accumulation in this surface layer is known to occur (Zhou and Rowland, 1997). OCP enrichment of the surface film may be of considerable importance to surface living organisms or to birds that skim food off the sea surface. Surface plankton and other organic particulates are readily associated with OCPs and undergo subsequent sedimentation. At the following locations higher amounts of OCPs were detected at mid-depth i.e., NE-01, NE-05, NE-09, and SW-01, SW-02, and SW-08. These locations are close to industries and shipping anchorages, where hydrodynamic dispersion is confined.

The land area under agriculture use in Singapore is negligible and there is no direct application of organochlorine pesticides in the country. However, pesticides may be easily transported through the ambient environment by different mechanisms including volatilization from soil and spray drift during application to crops (Dörfler and Scheunert, 1997). The presence of OCPs in Singapore's marine waters is probably a function of their use in neighbouring countries, including Malaysia and Indonesia.

Concentrations of OCPs measured in Singapore seawater are comparatively lower than those detected in water from the Selangor River in Malaysia i.e., Aldrin, up to 884.00 ng L^{-1}; Dieldrin, up to 850.00 ng L^{-1}; Endrin, up to 10,970.00 ng L^{-1}; α-Endosulfan, up to 8.90; β-Endosulfan, up to 12,270.00 ng L^{-1}; Heptachlor, up to 13,710.00 ng L^{-1}; Lindane, up to 4,0950.00 ng L^{-1}; *p,p′*-DDT, up to 44,770.00 ng L^{-1}; *p,p′′*-DDE up to 2310.00 ng L^{-1} (Mustafa et al., 2000); as well as the Surabaya river, Indonesia *p,p′*-DDT up to 49.63 ng L^{-1} (Dewi, 2000); Philippine coastal waters, α-BHC up to 21.00 ng L^{-1} and Aldrin at 7.00 ng L^{-1} of (Santiago, 2000).; the Dampha and Balat estuaries in Vietnam, DDT i.e., 30.00 ng L^{-1} (Viet et al., 2000); Bohai Sea, China, DDE, DDD and DDT up to 50.00 ng L^{-1} (Yeru and Hao, 2000). However, OCP levels in Singapore's coastal waters are higher than those found in the Coral Sea, Australia where total OCP concentrations have been measured at 1.21 ng L^{-1} (Tanabe et al., 1984), and 5.50 ng L^{-1} (Kurtz and Atlas, 1990).

15.2.3. Polychlorinated biphenyls (PCBs)

The individual and total concentrations of PCBs at surface and mid-depth for northeastern and southwestern sampling locations are shown in Tables 15.2a and 15.2b, respectively. PCBs represent a group of compounds that have been widely used in a range of industrial applications. All eight PCBs analysed in the study were detected at each region for the majority of sample stations.

Total PCB concentrations in seawater from both regions varied from 0.04 to 61.7 ng L^{-1} and 0.22 to 20.1 ng L^{-1} in northeastern and southwestern regions, respectively. The highest total concentrations of PCBs measured were at station (NE-13 i.e., 61.7 ng L^{-1}) and SW-09 (i.e., 20.1 ng L^{-1}). The prevalence of PCBs in these locations may be due to their proximity to contamination sources including the industrial sources in the southwest of Singapore, and the Bedok and Slater municipal wastewater treatment plants in the northeast. The highest measured concentration of an individual PCB congener i.e., 2,2′,3,3′,4,5′,6,6′-octachlorobiphenyl was 40.71 ng L^{-1} and 15.42 ng L^{-1} at the northeastern and southwestern regions, respectively.

Long-range atmospheric transport is likely to be a source of PCBs detected in remote waters and results in low-level concentrations in nearly all environmental matrices (Bidleman et al., 1989). However, higher levels can be associated with proximity to industry, as well as waste discharges from shipbuilding yards and municipal sewage plants located in coastal regions. PCBs are hydrophobic compounds with an octanol–water partition coefficient (K_{ow}) ranging from 4.5 to 8.2. The aqueous solubility is

less than $5\,mg\,L^{-1}$ for the more chlorinated congeners (i.e., >2 chloro group) (Patil, 1991). The distribution of PCBs in coastal waters contrasts with that of PAHs, where the surface was more contaminated than at mid-depth for the majority of sample locations. The concentration of PCBs in seawater samples shows that the northeastern coastal region of Singapore is more extensively polluted than the southwestern region. These variations are most likely due to historic and episodic inputs from industrial sources, hydrodynamic factors, as well as possible discharges form Malaysian and Indonesian coastal regions.

The maximum level of PCB contamination detected in Singapore coastal waters (i.e., $61.7\,ng\,L^{-1}$) is lower than that recorded from Xiamen, China and Victoria Harbour, Hong Kong (i.e., $151\,ng\,L^{-1}$; Hong et al., 1995); Jamaica-Kingston Harbour (i.e., $3500\,ng\,L^{-1}$; Mansing and Wilson, 1995); Doñana National Park, Spain (i.e., $237\,ng\,L^{-1}$; Fernàndez et al., 1992); and higher than levels measured in the Gulf of Mexico and Atlantic Ocean (North) (i.e., $<0.003\,ng\,L^{-1}$; Sauer et al., 1989), the Northern Pacific Ocean (i.e., $0.59\,ng\,L^{-1}$; Tanabe et al., 1984) and the Dutch Wadden Sea (i.e., $0.62\,ng\,L^{-1}$; Duinker and Hillebrand, 1983).

For seawater, it can be concluded that concentrations of PAHs measured in Singapore's coastal waters were generally higher than levels reported elsewhere, and this is most likely due to the presence of Singapore's extensive petroleum industry. Seawater concentrations of OCPs and PCBS are generally lower than the reported levels for other Asian countries, but higher than some levels reported elsewhere in the world. Overall, results from this survey clearly show the prevalence of POPs in Singapore's coastal waters, and the need for further research to fully evaluate their fate, transport and biological impact in the marine environment.

15.3. POPs in the sea-surface microlayer

The sea-surface microlayer (SML) of the ocean represents the boundary layer between the atmosphere and the ocean surface body, and has a typical thickness of 40–100 μm. The SML has a distinctive chemical composition compared to the bulk seawater underneath, including enrichment of naturally occurring surfactants, proteins and lipids. Hydrophobic organic pollutants, like POPs, tend to hyper accumulate in the SML in a range of 0.2–100 as reviewed by Wurl and Obbard (2004). The SML plays an important role in the transfer of material controlled by complex physicochemical processes and may have substantial effects on the global distribution of POPs as hypothesized by Wania et al. (1998). The role of the SML in the air–sea gas exchange of POPs was recently shown using data from field experiments (Wurl et al., 2006).

The first scientific data on POP concentrations and enrichment factors in the SML for Southeast Asia were reported from Singapore's marine environment (Wurl and Obbard, 2005a, 2006). SML samples were collected using a rotating glass drum sampler designed for trace organic analysis and described in details in the same reference. The concentration ranges for ΣHCH, ΣDDT and ΣPCB in the SML were 0.6–64.6 ng L^{-1} (mean 9.9 ng L^{-1}), 0.01–0.7 ng L^{-1} (mean 0.2 ng L^{-1}) and 0.07–12.4 ng L^{-1} (mean 1.3 ng L^{-1}), respectively. In subsurface seawater (1 m depth), the concentration ranges were 0.4–27.2 ng L^{-1} (mean 4.0 ng L^{-1}), 0.01–0.6 ng L^{-1} (mean 0.1 ng L^{-1}) and 0.05–1.8 ng L^{-1} (mean 0.5 ng L^{-1}) respectively. The enrichment factors (EF) in the SML relative to subsurface water for ΣHCH, ΣDDT and ΣPCBs were 0.8–6.9, 0.2–7.6 and 0.7–39.6 respectively, but generally ranged between 1.2 and 4. EF values were higher for temperate climatic zones and were in a range of 0.1–23.1 for an estuary in Argentina (Sericano and Pucci, 1984) and 0.2–93 in coastal offshore waters of Croatia (Picer and Picer, 1992). Comparable EF values of 1.7–3.5 were reported for SML samples collected in the sub-tropical zone of Alexandria (Abd-Allah, 1999). The temporal distribution of contaminants in the SML and subsurface water is related to the rainfall pattern of Singapore during the repeated sampling events between November 2003 and March 2004 (Figs. 15.2–15.4). In general the concentrations during the second survey (January–February) were lower by a factor of up to 50 compared to the first survey (November–December). The concentrations increased during the third survey in March by a factor of up to 10 compared to the previous survey. Rainfall patterns for Singapore in 2003 and 2004 show that most rain fell between October and December 2003 during the northeast monsoon (approximately 250 mm per month) and March 2004 (400 mm), whereas February 2004 had the lowest rainfall incidence (approximately 25 mm) (NEA, 2004). During this study SML and subsurface water prior to and after a heavy rainfall event could be collected for the first time. A load factor (LF) as the enrichment of contaminants before and after rainfall was defined. The LF was insignificant for the subsurface water, but between 2 and 5.9 greater for the SML. It can be concluded that wet deposition is a significant source of POPs to the SML in tropical regions, but not directly for the subsurface water. Removal mechanism of POPs accumulated in the SML may be diffusion to the subsurface water, volatilization into the atmosphere and microbial degradation. Microbial degradation of PCBs in aquatic environments were reported by Sugiura (1992) and elevated concentrations of microorganism and enrichment of inorganic nutrients in the SML (Zaitsev, 1997) may support the microbial degradation of POPs in the SML. Air–sea gas exchange rates between the compartment

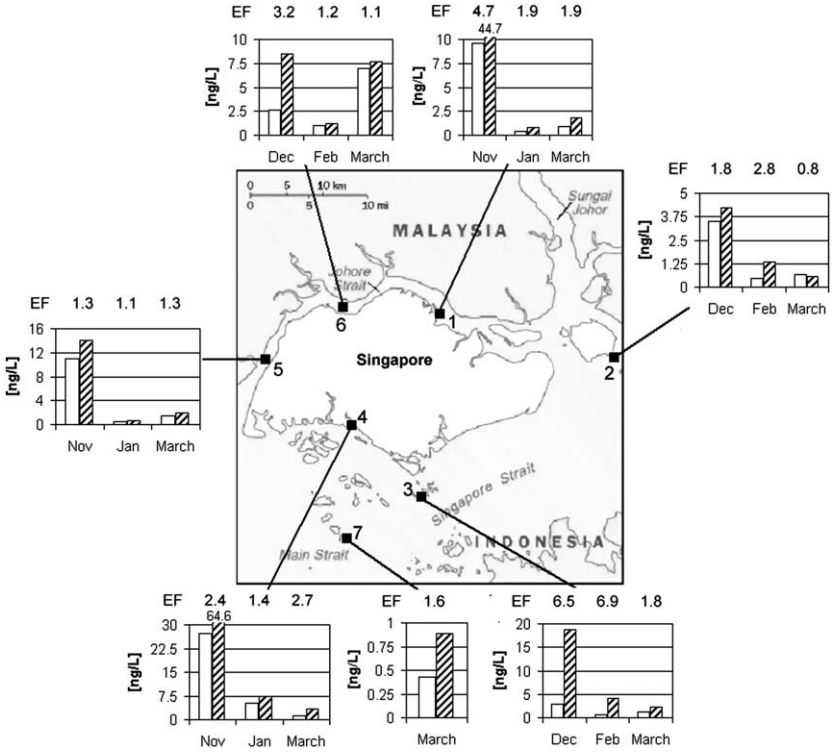

Figure 15.2. Concentrations and enrichment factors (EF) of ΣHCH in seawater (□) and SML (▨)

subsurface seawater and SML with the atmosphere were reported for selected PCBs, and HCHs using a classical two-layer model (Wurl et al., 2006). The model yields flux rates for ΣPCBs, ranging between 27 and 171 ng $(m^2\ day)^{-1}$, whereas the values of ΣHCH fluxes are −111–69 ng $(m^2\ day)^{-1}$ for the tropical region of Singapore. The negative flux data indicates adsorption of HCH isomers into the ocean. It was shown that physicochemical properties, like *n*-octanol/water partition coefficient and water solubility, may control the enrichment of POPs in the SML. Higher chlorinated PCBs tended to be enriched higher in the SML than lower chlorinated congeners (Wurl and Obbard, 2005a). Good correlations between the relationship between EF values and log K_{ow} support that the *n*-octanol/water partition coefficient is a key parameter for the enrichment of hydrophobic organic contaminants (Fig. 15.5).

In a further study the distribution of PCBs, HCHs and DDTs between the dissolved phase (DP) and suspended particulate matter (SPM) in the

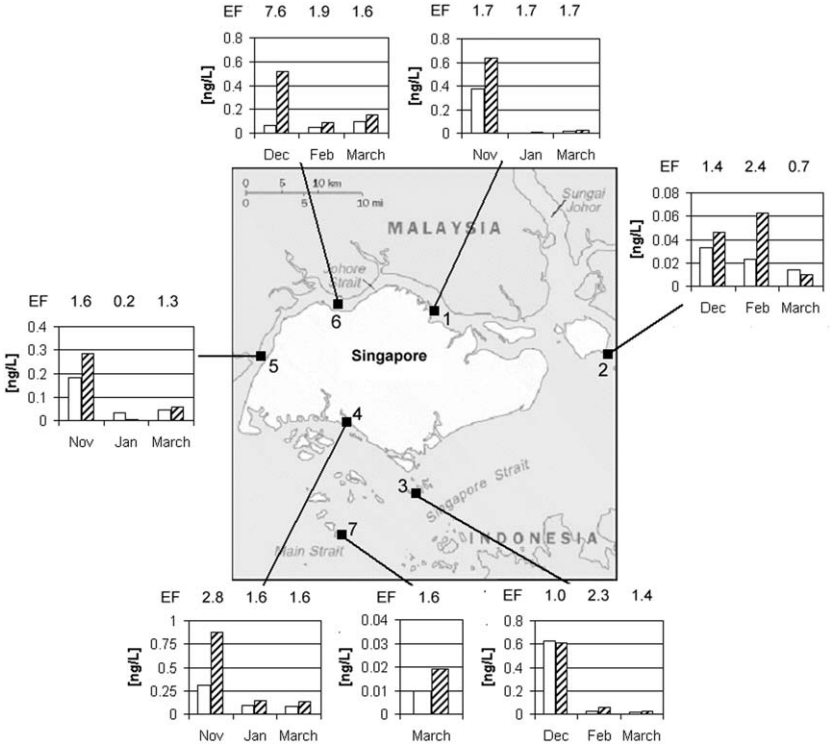

Figure 15.3. Concentrations and enrichment factors (EF) of ΣDDT in seawater (□) and SML (▨).

water column including the SML were investigated at two stations in Singapore's marine environment (Figs. 15.6, 15.7, and 15.8). It was reported that PCBs, HCHs and DDTs in the SML were in general dominant in the DP. The vertical distribution of ΣPCBs in the water column at both stations is similar (Fig. 15.6). Levels of ΣPCBs in the DP increase towards the surface. Enrichment of PCBs in the DP in the SML relative to subsurface water was found to be in the range of 1.3–7.2. Higher concentrations of ΣPCBs in the SPM were found in the bottom water with a trend of declining concentration to mid-depth. Near the surface and in the SML, the ΣPCB concentrations were enriched relative to water at mid-depth. The concentrations of ΣPCBs in the SPM were enriched in the SML by a factor (EF) of 1.2–7.2. This enrichment is most likely derived from small-size and/or flocculated particles floating on the water surface. Enrichment factors of ΣPCBs associated with the SPM in the bottom water at both stations were in the range of 2–3 compared to overlying

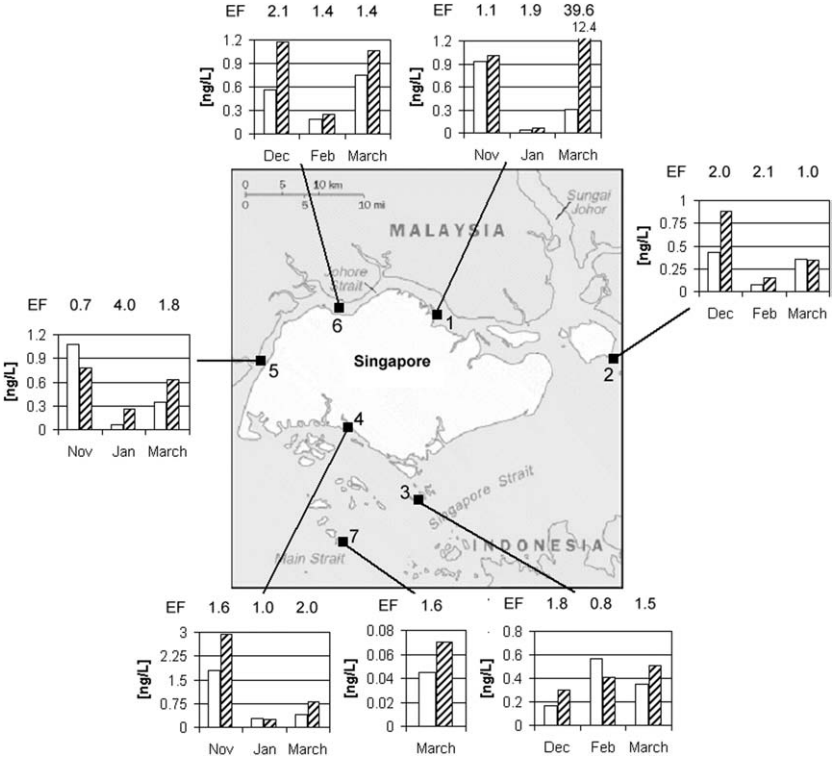

Figure 15.4. Concentrations and enrichment factors (EF) of ΣPCBs in seawater (□) and SML (▨).

water, but no enrichment of SPM was found in this layer at station 2. This phenomenon has also been reported recently for the Black Sea (Maldonado and Bayona, 2002). Different types and sizes of suspended particulates between water layers may lead to an enrichment of contaminant concentrations associated with the SPM, even though SPM itself is not enriched. The vertical distribution profile of OCs at both stations suggests that resuspension processes of contaminated sediments may be a key source of PCBs in the water column. However, wet and dry atmospheric deposition of OCs on the SML may act as secondary source, indicated by the enrichment in the SML.

The vertical profiles of ΣHCHs (Fig. 15.7) were characterized by a depletion in the DP of the bottom water compared to the overlying water at both stations. The trend of concentrations of dissolved HCHs decreased from the mid-depth to the subsurface layer. The vertical profiles for ΣHCHs associated with the SPM were similar at both stations with little

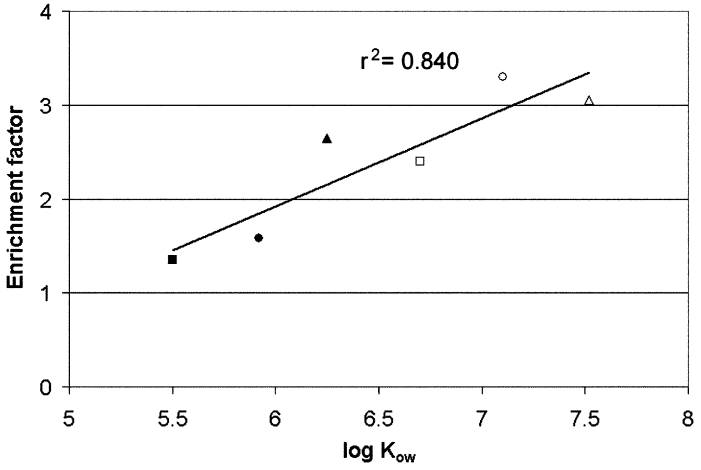

Figure 15.5. Correlation between average log K_{ow} value of tri- (■), tetra- (●), penta- (▲), hexa- (□), hepta- (○) and octa-chlorobiphenyls (Δ) and corresponding average enrichment factors (EF).

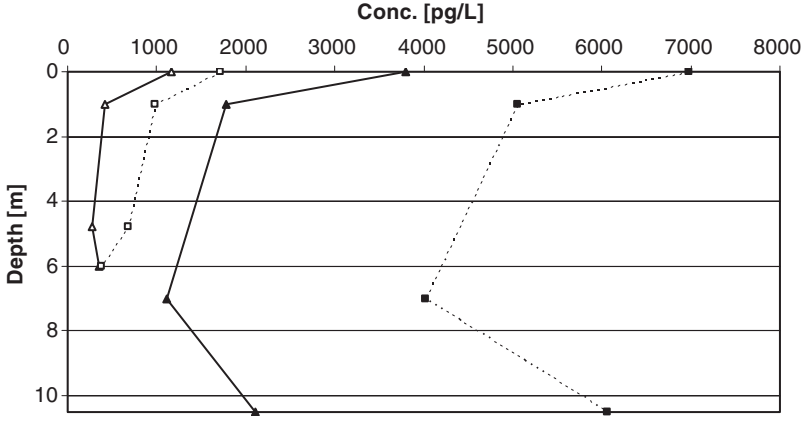

Figure 15.6. Vertical profiles of ΣPCBs in DP (□) and SPM (△) at station 1, and in DP (■) and SPM (▲) at station 2.

variation with depth. Exceptionally, an enrichment in the SML was found in a range of 1.1–7.2. In general the dissolved fraction occupies 60–99% of the total concentration of HCHs. Levels of ΣHCHs in the SPM were not enriched in the bottom water layer for both stations; although a high enrichment of SPM concentrations was apparent in this layer for station 1. This suggests that resuspension of sediments may not be a major source

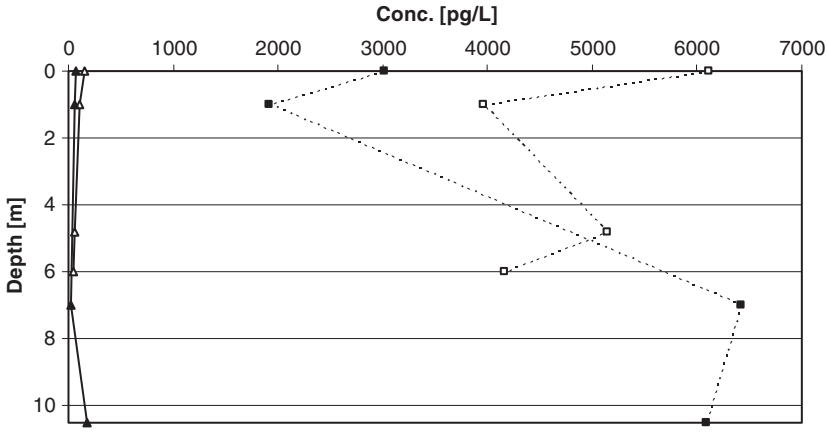

Figure 15.7. Vertical profiles of ΣHCHs in DP (□) and SPM (△) at station 1, and in DP (■) and SPM (▲) at station 2.

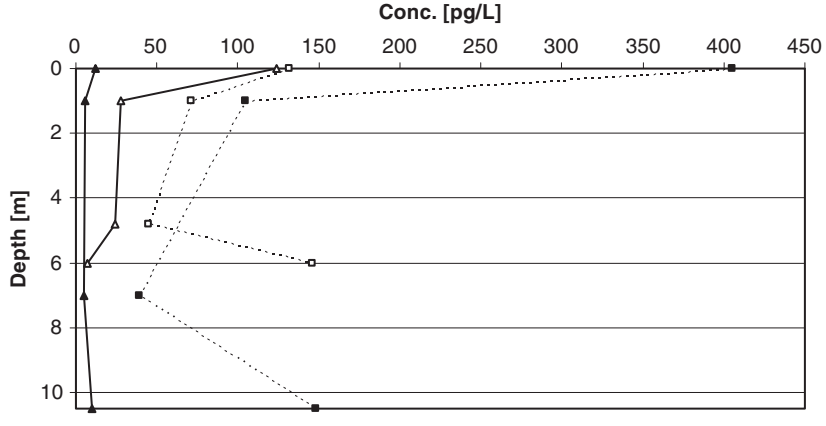

Figure 15.8. Vertical profiles of ΣDDTs in DP (□) and SPM (△) at station 1, and in DP (■) and SPM (▲) at station 2.

for HCHs into the water column. Enrichment in the surface layers of the water column and decreasing concentrations with the depths suggest that freshwater inputs, such as riverine discharge as well as wet and dry atmospheric deposition on the SML are the major inputs of HCHs into the marine water column.

Vertical distribution profiles for DDTs are similar at both stations (Fig. 15.8). An enrichment in the SML and bottom water of DDTs in the DP could be found in both water columns. The trend of concentrations

decreased from the surface to the mid-depth layer. From the sub-surface to the bottom water layer, the concentrations in the SPM varied little, but an enrichment in the SML (EF = 3.3–4.4) was found. Deposition of atmospheric particulates will result in the enrichment of the SML and represent a source of particulate DDT into the water column. The concentration of DDTs in the Singapore's marine sediments was low compared to levels of PCBs and HCHs (Wurl and Obbard, 2005b), meaning that resuspension processes are unlikely to act as a significant source of DDTs into the water column, where runoff and precipitation inputs are likely to account for the observed enrichment in the SML and subsurface layer. ΣDDTs are known for their lower water solubility and higher affinity to sediments and particles compared to HCH isomers. The fraction of ΣDDTs associated with the SPM were dominant (up to 90%) in the Pearl River Delta (Luo et al., 2004) but not for the Yangtze River, China (Jiang et al., 2000), Danube Estuary and open water in the Black Sea (Maldonado and Bayona, 2002), Mediterranean Sea and estuary areas of the Ebro and Rhone River (Dachs et al., 1996), and coastal area of east Java, Indonesia (Hillebrand et al., 1989). In this study, ΣDDTs in the dissolved fractions represented between 44 and 90% of the total concentration present. This distribution is not consistent with the high K_{ow} values for DDTs. Relatively low carbon content were found in the SPM in this study, and this may affect the adsorption behaviour of OCs on particulates. The small particles and colloids ($<1.0\,\mu m$) of the SPM passed through the filter can be considered as a dissolved fraction. The relative high water temperature in the tropical area of Singapore will increase the solubility of OCs and shift the distribution pattern towards the dissolved fraction. It may be these factors that account for the observed distribution.

The partitioning of HCHs and DDTs between the SPM and DP was qualitatively assessed using the distribution coefficient K'_d and this is illustrated in Fig. 15.9 for station 1 and 2 respectively. The K'_d values for the two stations vary with depth implying that either the water bodies are not well mixed or more likely that the distribution of SPM is hetereogenous. The profile at station 2 (Fig. 15.9) is similar among OCPs. An increase of the K'_d or log K'_d value from the SML to the subsurface water and a decline towards the bottom water is distinctive for the profile at station 2. This may indicate that the characteristics of the water body or the SPM in the SML and subsurface water are different. The SML may be enriched with small or voluminous floating particulates, whereas subsurface water will typically be dominated by denser, sinking particles. The profile at station 1 shows that more HCHs and DDTs in the SPM appear proportionally in the SML and subsurface layer than in the mid-depth layer relative to the DP. The different profiles in the lower

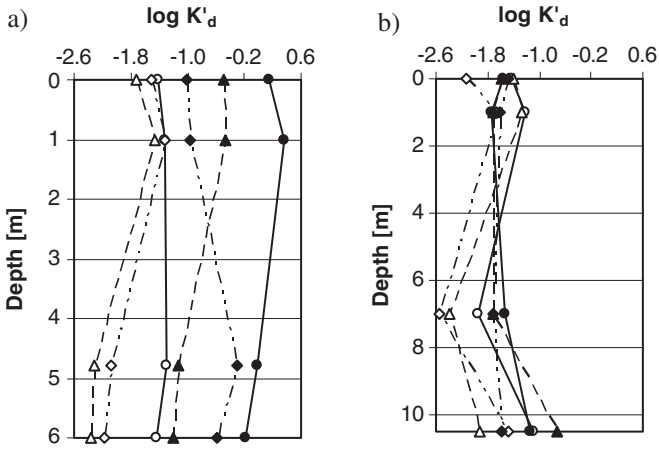

Figure 15.9. The distribution coefficient K'_d of α-HCH (○), γ-HCH (◇), β-HCH (△), *p*,*p*′-DDE (●), *p*,*p*′-DDD (◆) and *p*,*p*′-DDT (▲) with water column depth at station 1 (a) and 2 (b).

water bodies at stations 1 and 2 suggest that sediment resuspension processes at station 1 play a significant role in the release of OCPs into the water column, but only a minor role at station 2. However, the trends in the K'_d value from the SML to the subsurface layer suggest that the SML is another important compartment for the fate of OCPs, with more importance for the DP.

15.4. POPs in marine sediments

PAHs account for approximately 20% of total hydrocarbons present in crude oil, and are the most metabolically toxic of all the petroleum compounds (Neff, 1990). Oil spillage is a global problem where for example, in 1999 approximately 109,400 tons of oil was spilled worldwide into marine and terrestrial environments as the result of over 250 incidents. The Port of Singapore is one of the world's busiest ports, where the total cargo handled in January 2002 alone was in excess of 28.2 million tones (Nautilus, 2002). Singapore's coastal areas have also been extensively developed to support the petroleum industry, where the country is home to the world's third largest petroleum refining centre, with a processing capacity in excess of 1.3 M barrels of crude oil per day (SEDB, 1999). Singapore and the neighboring countries of Malaysia and Indonesia have experienced 10 major oil spill incidents between 1993 and 2002, including a major spillage of 28,500 tons of crude oil in the Singapore Straits in October 1997 (ITOPF, 2007). With continued industrial development and shipping

activity within the coastal region of Southeast Asia as a whole, there is clearly an increasing risk of adverse regional marine contamination.

A study was undertaken to determine the prevalence of PAHs in the marine sediments of Singapore's coastal environment by Basheer et al. (2003b). A total of 22 stations were sampled for sediment analysis, as for seawater analysis (see Fig. 15.1). These stations were located in the northeastern (stations NE-1–NE-13) and southwestern (stations SW-1–SW-9) regions of Singapore's coastal environment. Sampling stations were all within 1 km of the coast and adjacent to busy shipping lanes and coastal petroleum refineries. Surface sediments were collected using a Van Veen grab (1000 cm^2 sampling area). Total PAH concentrations in sediments are shown in Fig. 15.10.

All 16 PAHs were detected in all surface marine sediments taken from the 22 sample stations in both the northeastern and southwestern regions of Singapore's coastal environment–see Tables 15.3 and 15.4, respectively. The highest total PAH concentration measured in the northeastern region was 82.4 $\mu g\, g^{-1}$ (at NE-4) and 93.85 $\mu g\, g^{-1}$ in the southwestern region (at SW-9). The lowest total PAH concentration was detected at station NE-3 (i.e., 15.2 $\mu g\, g^{-1}$) and at station SW-4 (i.e., 12.6 $\mu g\, g^{-1}$) for each region respectively. Overall, the concentrations of PAHs in sediments sampled from the northeastern region are comparatively higher than those of the southwestern region (see Tables 15.3 and 15.4), and this is consistent with seawater quality data. In the northeastern coastal region of Singapore, the marine hydrodynamics are relatively confined, as the tidal exchange of Johor Straits is restricted. This is in contrast to the southwest region, where stronger oceanic currents and lower sedimentation rates facilitate particulate dispersion.

Among the 16 individual PAHs quantified, the highest concentrations measured were for four-ring chrysene and six-ring indeno[1,2,3-*cd*]pyrene which reached maximum concentrations of 3.7 $\mu g\, g^{-1}$ and 15. 2 $\mu g\, g^{-1}$ in the northeastern region, and 1.6 $\mu g\, g^{-1}$ and 7.5 $\mu g\, g^{-1}$ in the southwestern region (see Tables 15.3 and 15.4). Other abundant PAHs included benzo[*a*]fluoranthene, anthracene, dibenz[*ah*]anthracene and benzo[*ghi*] perylene. The lowest PAH concentration measured in sediments was for three-ring acenapthylene i.e., up to 0.3 $\mu g\, g^{-1}$ and up to 0.2 $\mu g\, g^{-1}$ in northeastern and southwestern regions, respectively.

The distribution of individual PAHs reflects the different physiochemical properties of low and high molecular weight PAHs, where lower molecular weight compounds have a relatively higher vapour pressure and water solubility, and are therefore more readily volatilized and degraded. Higher molecular PAHs are more readily adsorbed onto the particulate phase and undergo sedimentation. This is reflected in the

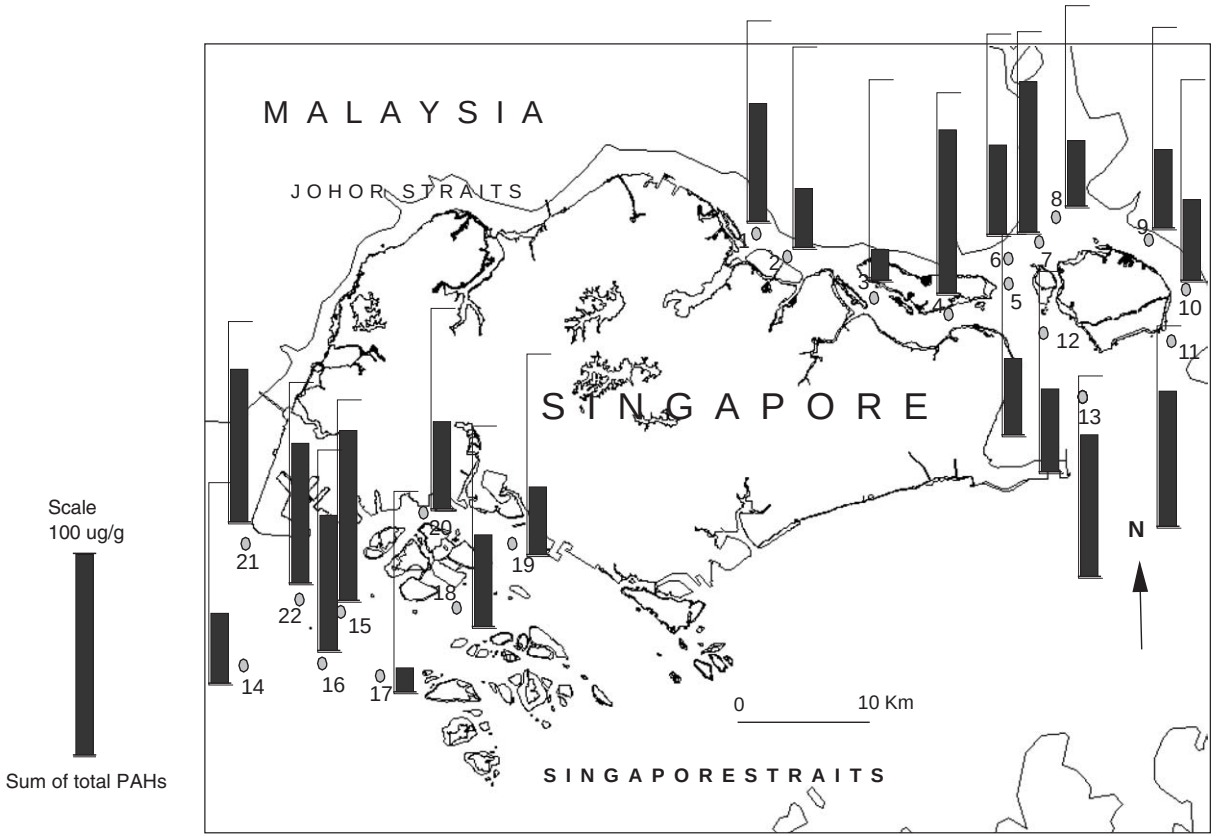

Figure 15.10. Total PAH concentrations in surface marine sediments of Singapore's coastal marine environment.

Table 15.3. Concentrations of PAHs in surface marine sediments of the northeastern coastal region of Singapore (μg g^{-1}, $n = 4$)

Stations	NE-01	NE-02	NE-03	NE-04	NE-05	NE-06	NE-07	NE-08	NE-09	NE-10	NE-11	NE-12	NE-13
Naphthalene	1.02	0.47	0.44	0.51	0.29	0.27	1.28	0.29	1.21	0.48	1.53	0.20	1.05
Acenaphthylene	0.17	0.23	0.05	0.08	0.08	0.09	0.23	0.08	0.24	0.06	0.19	0.06	0.22
Acenaphthalene	0.18	0.13	0.10	0.11	0.06	0.06	0.20	0.03	0.16	0.04	0.27	0.02	0.14
Fluorene	0.39	0.14	0.03	0.10	0.14	1.13	0.25	0.81	1.99	0.06	0.29	1.74	0.70
Phenanthrene	6.72	2.05	1.93	1.35	0.80	1.05	1.80	0.34	0.46	0.77	4.79	0.77	2.00
Anthracene	11.20	7.90	2.66	7.42	3.21	3.36	7.11	1.04	2.74	2.59	17.63	1.55	8.25
Fluoranthene	2.34	0.39	0.14	2.62	0.71	2.17	2.13	0.44	1.19	0.62	1.27	0.46	1.51
Pyrene	5.89	0.98	0.48	6.61	1.80	5.47	5.38	0.75	3.01	1.16	3.22	1.08	3.97
Benz[*a*]anthracene	7.42	0.60	0.03	18.95	5.15	7.18	14.39	0.58	8.06	3.53	7.36	2.89	11.81
Chrysene	7.42	1.27	0.11	25.27	7.07	9.58	19.21	0.77	10.76	4.71	9.82	11.24	15.74
Benzo[*a*]fluoranthene	1.94	3.53	1.46	2.75	3.80	4.47	6.08	3.79	1.62	1.70	1.73	3.05	3.11
Benzo[*k*]fluoranthene	1.94	2.84	2.46	2.48	3.81	2.24	6.09	3.65	1.31	1.72	1.85	3.29	2.82
Benzo[*a*]pyrene	0.98	1.44	1.24	1.25	1.92	1.13	0.99	1.84	0.51	0.86	0.87	0.77	1.57
Indeno[1,2,3-*cd*]pyrene	7.37	5.24	3.66	9.86	5.91	4.34	7.98	12.37	4.48	14.64	11.63	6.037	15.22
Dibenz[*a*,*h*]anthacene	1.70	1.31	0.40	1.54	1.76	1.97	1.09	3.37	1.06	3.95	2.73	1.40	1.81
Benzo[*ghi*]perylene	2.21	1.59	0.01	1.54	1.76	1.10	1.12	3.41	1.02	3.95	2.61	1.59	1.55
Total	58.88	30.12	15.19	82.42	38.25	45.61	75.33	33.58	39.83	40.82	67.78	30.09	71.45

Table 15.4. Concentrations of PAHs in surface marine sediments of the southwestern coastal region of Singapore (μg g^{-1}, $n = 4$)

Stations	SW-01	SW-02	SW-03	SW-04	SW-05	SW-06	SW-07	SW-08	SW-09
Naphthalene	0.45	0.26	0.95	0.14	0.74	0.21	0.30	0.90	0.46
Acenaphthylene	0.08	0.42	0.18	0.08	0.09	0.22	0.13	0.20	0.42
Acenaphthalene	0.09	0.06	0.12	0.05	0.08	0.02	0.03	0.16	0.09
Fluorene	1.35	0.21	1.08	0.09	0.86	0.08	0.28	1.43	2.87
Phenanthrene	1.72	1.01	1.36	0.57	0.75	0.46	1.81	3.84	2.46
Anthracene	6.41	4.15	4.76	2.09	2.98	1.05	6.04	5.26	9.45
Fluoranthene	0.58	4.50	9.33	0.16	2.10	0.42	2.52	1.44	6.53
Pyrene	2.71	22.66	5.43	0.80	10.18	2.14	12.68	6.84	17.71
Benz[*a*]anthracene	3.38	6.12	7.75	0.22	6.30	1.59	2.14	9.10	9.08
Chrysene	8.91	12.24	15.52	0.43	12.60	3.18	4.28	18.20	30.64
Benzo[*a*]fluoranthene	0.66	5.08	5.56	2.25	1.09	1.29	2.02	2.99	3.45
Benzo[*k*]fluoranthene	0.66	5.09	4.88	2.26	1.10	1.45	2.03	2.23	3.46
Benzo[*a*]pyrene	0.29	2.55	2.46	1.13	0.55	0.42	1.02	0.59	0.60
Indeno[1,2,3-*cd*]pyrene	3.92	7.29	7.12	1.63	4.55	5.65	5.05	7.52	3.78
Dibenz[*a*,*h*]anthacene	1.37	6.69	1.50	0.38	1.14	7.99	1.72	2.14	1.36
Benzo[*ghi*]perylene	2.79	6.59	0.37	0.38	1.03	7.98	1.72	13.88	1.48
Total	35.36	84.91	68.36	12.65	46.14	34.15	43.75	76.70	93.85

prevalence of higher molecular PAH found in sediment samples. The distribution of mean individual PAH concentrations is in the sequence of chyrsene > indeno[1,2,3-*cd*]pyrene > pyrene > benzo[*a*]anthracene > anthracene > fluoranthene > benzo[*ghi*]perylene > benz[*a*]fluoranthene > benzo[*k*]fluorantherne > dibenz[*a*]anthracene > phenanthrene > fluorine > benzo[*a*]pyrene > acenapthylene > acenaphthalene> naphthalene. Chrysene and fluoranthene are, therefore, prevalent in the coastal sediments of Singapore. Chrysene is the most abundant PAH and is believed to be a compound mainly derived from natural sources including pentacyclic triterpenes such as α- and β-amyrins which are, in turn, derived from epicuticular waxes of terrestrial plants via the process of microbial degradation (Wakeham et al., 1980). In contrast, fluoranthene is considered a good pollution indicator in environments impacted by urban and industrial activities (Chaudhry, 1994). The distribution pattern of fluoranthene in marine sediments from Singapore is similar and comparatively lower in concentration than previously detected in marine sediments from Australia (McCready et al., 2000).

As PAHs are widespread contaminants produced as a result of natural cycles (e.g., forest fires, plant decomposition and petrogenesis), as well as industrial activities, identification of anthropogenic PAHs contaminant sources is a challenge, particularly as atmospheric emissions are subject to long-range atmospheric transportation processes (Lockhart et al., 1992;

Peters et al., 1995). PAHs in marine sediments have been associated with various particulate sources where, for example, pyrene is typically found in particulates generated from hydrothermal sources, fluorene is associated with submarine volcanic extrusions and chrysene and benzo[*a*]pyrene are specific to the presence of oil (Chernova et al., 2001). Previous studies have vindicated the use of PAH isomer ratios for source identification (e.g., Kayal and Connell, 1989; Klamer and Fomsgaard, 1993). For example, as phenanthrene is a thermodynamically more stable three-ring aromatic isomer than anthracene, the phenanthrene:anthracene ratio has been used to differentiate PAH in the environment of petrogenic origin from those of pyrogenic origin (Klamer and Fomsgaard, 1993; Budzinski et al., 1997). Fluoranthene and pyrene are considered typical pyrogenic products generated from high temperature condensation of low molecular weight compounds (Soclo et al., 2000). During the combustion process, pyrene is more stable than fluoranthene, and hence pyrolytic products are usually characterized by a predominance of fluoranthene over pyrene at ratios greater than one (Baumard et al., 1999). Petrogenic PAH sources are generally characterized by a high ratio (>15), while those of pyrogenic origin have a lower ratio.

The kinetic/thermodynamic isomer ratios of phenanthrene:anthracene (three-ring) and pyrene:fluoranthene (four-ring) were derived from PAH marine sediment data for Singapore, and are shown in Fig. 15.11. An abundance of high molecular weight PAHs is typical of atmospheric

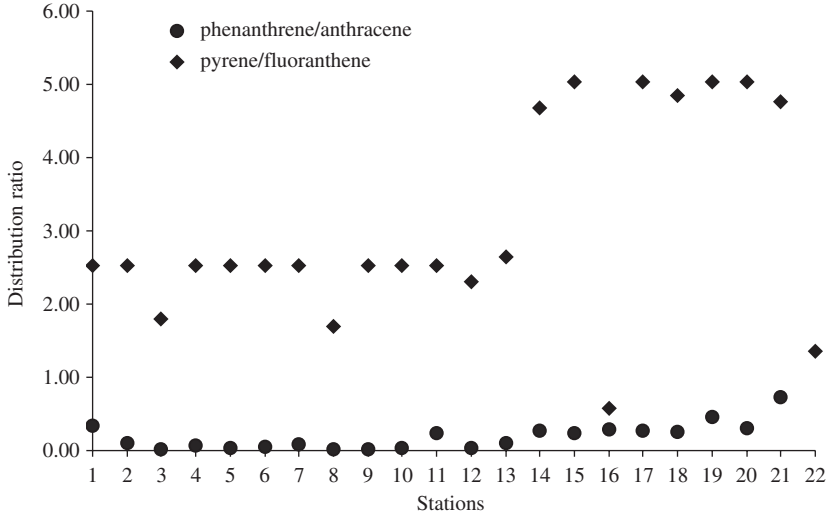

Figure 15.11. Distribution of PAH isomers ratios in surface marine sediments.

particles and urban aerosols (Ogata and Fujisawa, 1990), due to their low water solubility, low microbial degradation rate. The phenanthrene:anthracene ratio in sediments ranged from 0.02 to 0.73, and the pyrene:fluoranthene ratio ranged between 1.30 and 5.03. The relatively low isomer ratio suggests that PAHs of a pyrogenic origin are predominant in Singapore's coastal environment. The prevalence of four- and five-ring aromatic compounds in the sediments from both coastal regions in Singapore further suggests that PAH contamination in the coastal environment is a result of deposition of PAH-rich particulates produced by anthropogenic combustion processes (Muel and Saguem, 1985; Sicre et al., 1987). The survey also shows that the total concentrations of PAHs (based on dry weight) in Singapore's coastal marine sediments (i.e., 93.9 $\mu g\, g^{-1}$) are comparatively lower than that reported for sediments from Victoria Harbour, Hong Kong (i.e., up to 387 $\mu g\, g^{-1}$; Environmental Protection Department, 1997); Kitimat Harbour, Canada (i.e., up to 10, 000 $\mu g\, g^{-1}$; Simpson et al., 1996); Times Beach, New York, USA (i.e., up to 480 $\mu g\, g^{-1}$; Roper and Chery, 1994); Santander Bay, Spain (i.e., up to 344.6 $\mu g\, g^{-1}$; Viguri et al., 2002); Harbor of Refuge, Rhode Island Sound, USA (i.e., up to 730.0 $\mu g\, g^{-1}$; Ho et al., 1999); Sydney harbour, Australia (i.e., up to 380 $\mu g\, g^{-1}$; McCready et al., 2000); Kohtla-Järve, Estonia (i.e., up to 153.0 $\mu g\, g^{-1}$; Trapido, 1999), but higher than that reported for sediments in the Mediterranean Sea (i.e., up to 20.5 $\mu g\, g^{-1}$; Baumard et al., 1998); Pacific Coast, USA (i.e., up to 20 $\mu g\, g^{-1}$; Brown et al., 1998); Arkona Basin, Baltic Sea, Germany (i.e., up to 1.8 $\mu g\, g^{-1}$; Witt, 1995); and the Barents Sea, Svalbard, Norway (i.e., up to 2.0 $\mu g\, g^{-1}$; Stange and Klungsøyr, 1997); and Mai Po, Hong Kong (i.e., up to 1.0 $\mu g\, g^{-1}$; Zheng et al., 2000).

Overall, widespread PAH contamination in the coastal sediments of Singapore is apparent. PAH distribution profiles in both regions were dominated by pyrogenically derived PAH sources, signifying that inputs to the marine environment are dominated by the atmospheric deposition of contaminated particulates. With reference to studies conducted elsewhere, sediments in Singapore can be classified as moderately contaminated.

PCBs, polybrominated diphenyl ethers (PBDEs) and OCPs were reported in Singapore's coastal marine sediments by Wurl and Obbard (2005b) and concentration levels are summarized in Table 15.5 (PCBs and PBDEs) and Table 15.6 (OCPs). Sample stations are indicated in Fig. 15.12.Total PCB concentrations ranged widely from 1.4 to 329.6 $ng\, g^{-1}$. High concentrations were found in samples close to highly industrialized areas dominated by petrochemical plants. A notable decline in the concentration of ΣPCBs could be observed seawards from sample station SW3 over SW2 to SW1 (62.2–14.1 $ng\, g^{-1}$) (Fig. 15.12) and

Table 15.5. Sediment concentrations of major PCB congeners, ΣPCB and PBDE congeners in ng g^{-1} dw (mean ± sd, $n = 3$)

Station	SW1	SW2	SW3	SW4	SW5	NE1	NE2	NE3	NE4	NE5	NE6	NE7	NE8
PCB 28	<0.6	<0.6	<0.6	31.9±3.0	7.9±0.5	4.5±0.2	7.7±1.2	7.5±2.2	<0.6	<0.6	5.7±0.4	<0.6	<0.6
PCB 31	0.3±0.05	<0.03	8.0±0.7	8.9±0.5	<0.03	0.4±0.1	7.4±1.2	<0.03	7.2±0.8	<0.03	<0.03	1.0±0.1	0.2±0.01
PCB 33	<0.3	<0.3	14.3±2.1	11.4±1.2	<0.3	<0.3	12.7±0.5	1.7±0.2	<0.3	<0.3	<0.3	<0.3	<0.3
PCB 44	0.8±0.02	<0.2	<0.2	13.3±1.9	1.3±0.1	1.7±0.1	5.6±0.9	2.2±0.1	1.0±0.1	0.8±0.1	<0.2	3.4±0.2	<0.2
PCB 49	<0.3	<0.3	3.9±0.3	10.7±1.1	0.4±0.2	2.0±0.2	7.5±0.5	1.4±0.2	1.6±0.1	0.7±0.04	<0.3	<0.3	<0.3
PCB 53	0.63±0.1	<0.1	2.7±0.1	11.2±0.1	1.5±0.02	1.2±0.1	5.8±0.1	1.0±0.1	<0.12	<0.1	<0.1	2.7±0.3	<0.1
PCB 70	0.7±0.1	<0.2	3.2±0.6	13.3±1.6	1.2±0.4	1.5±0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2	<0.2
PCB 74	1.0±0.01	<0.3	<0.3	13.9±1.3	1.4±0.1	1.5±0.2	7.0±0.2	2.2±0.03	<0.3	<0.3	<0.3	3.0±0.5	<0.3
PCB 87	2.2±0.1	0.9±0.2	<0.3	13.3±0.4	<0.3	2.1±0.4	2.4±0.5	2.5±0.2	<0.3	1.2±0.2	<0.3	2.9±0.3	<0.3
PCB 118	1.5±0.1	<0.2	<0.2	13.6±1.6	<0.2	<0.2	4.0±0.9	<0.2	<0.2	<0.2	<0.2	3.4±0.7	1.2±0.1
PCB 128	<0.2	<0.2	1.3±0.3	13.1±0.1	1.2±0.2	<0.2	4.5±0.02	1.4±0.1	<0.2	<0.2	<0.2	1.8±0.1	<0.2
PCB 138	1.2±0.01	1.2±0.1	0.7±0.3	13.9±0.9	1.1±0.2	1.5±0.2	4.5±0.5	1.2±0.1	1.3±0.1	0.7±0.1	<0.4	1.8±0.1	<0.4
PCB 153	<0.2	13.3±3.3	19.4±2.1	0.6±0.2	0.9±0.2	21.9±1.6	13.6±2.7	1.7±0.04	2.2±0.2	<0.2	13.0±1.2	<0.2	<0.2
PCB 206 + 208	<0.4	10.0±0.7	<0.4	<0.4	41.0±2.4	16.7±0.9	21.3±0.9	36.1±2.0	15.0±1.8	35.8±3.7	15.5±1.0	27.7±1.8	<0.4
5PCB	14.1±0.7	29.1±3.6	62.2±2.7	329.6±7.7	72.5±1.8	60.6±7.8	138.5±5.8	79.8±2.8	34.6±2.4	44.7±4.4	38.3±0.5	55.4±0.8	1.4±0.1
BDE-47	3.4±0.9	4.0±0.6	11.1±0.9	4.5±0.5	4.7±0.9	3.7±0.7	13.8±1.1	5.1±0.7	4.0±0.6	3.8±0.3	5.5±0.5	4.3±0.3	12.7±1.2

Table 15.6. Sediment concentrations of OCPs in ng g^{-1} dw (mean $\pm$ sd, $n = 3$)

Station	SW1	SW2	SW3	SW4	SW5	NE1	NE2	NE3	NE4	NE5	NE6	NE7	NE8
α-HCH	4.4±0.8	4.9±0.5	7.2±0.5	0.9±0.2	2.0±0.2	2.6±0.1	8.4±0.7	5.8±1.6	6.4±0.8	4.6±0.6	9.3±1.6	2.9±0.6	2.4±0.4
β-HCH	3.2±1.6	5.3±1.4	4.6±0.6	0.7±0.2	2.6±0.7	4.3±0.1	11.1±0.9	5.2±0.2	6.0±0.4	4.1±0.7	4.2±0.4	4.3±0.4	2.7±0.4
γ-HCH	3.1±0.6	1.1±0.2	<0.7	0.8±0.2	3.5±1.1	2.7±0.5	13.4±0.9	8.8±2.8	5.7±0.3	7.7±0.5	<0.7	5.1±0.1	2.6±0.4
δ-HCH	3.1±0.3	1.4±0.4	7.2±0.8	0.9±0.2	3.3±1.1	5.3±0.9	13.3±1.3	2.9±1.4	5.2±0.6	4.0±0.6	8.0±0.9	3.8±0.2	2.3±0.7
ΣHCH	13.8±1.9	12.7±1.3	19.0±0.7	3.3±0.3	11.4±1.6	14.9±0.7	46.2±1.8	22.7±0.6	23.3±1.2	20.4±0.1	21.5±1.2	16.1±0.7	10.0±0.7
DDT	3.5±0.4	3.6±0.3	2.5±0.4	3.2±0.6	4.2±0.6	2.3±0.2	<0.5	2.1±0.7	<0.5	<0.5	0.6±0.2	1.9±0.2	2.2±0.3
DDD	2.8±0.2	1.5±0.2	3.5±0.4	4.0±0.6	3.7±1.1	1.8±0.4	1.3±0.2	2.4±0.4	1.5±0.2	1.5±0.2	2.1±0.2	3.2±0.3	3.8±0.6
DDE	2.5±0.4	0.6±0.3	2.7±0.8	4.7±0.9	2.6±0.5	1.1±0.1	0.9±0.3	1.6±0.3	0.7±0.1	1.2±0.1	1.6±0.3	4.1±0.5	3.7±0.3
ΣDDT	8.8±0.7	5.7±0.7	8.7±1.2	11.9±0.5	10.5±1.8	5.2±0.2	2.2±0.5	6.1±0.2	2.2±0.2	2.7±0.2	4.3±0.5	9.2±0.4	9.7±0.6
cis-Chlordane	0.8±0.2	1.0±0.2	<0.3	9.0±0.5	0.6±0.1	1.1±0.2	1.1±0.1	1.1±0.4	1.3±0.2	1.3±0.2	1.4±0.3	8.5±0.6	2.5±0.5
trans-Chlordane	1.5±0.1	2.9±0.4	1.4±0.1	9.7±0.5	1.2±0.4	1.7±0.2	1.5±0.2	1.3±0.1	1.4±0.2	1.9±0.1	1.3±0.3	10.0±0.6	4.0±0.4
Heptachlor	2.7±0.3	1.2±0.3	<0.9	0.9±0.2	2.0±0.2	4.2±0.7	<0.9	9.0±1.3	<0.9	2.3±0.6	<0.9	2.9±0.6	4.6±0.4
Heptachlor epoxide	6.8±0.6	2.9±0.2	<0.7	0.8±0.2	6.7±0.2	6.1±0.6	<0.7	5.9±0.9	<0.7	3.2±0.5	5.5±1.6	4.3±0.4	5.1±1.3
Aldrin	<0.3	<0.3	<0.3	<0.3	1.2±0.3	<0.3	<0.3	<0.3	0.6±0.1	1.4±0.4	1.1±0.4	1.0±0.4	0.5±0.2
Dieldrin	4.4±0.2	1.7±0.4	3.7±0.2	<1.2	1.9±0.4	3.4±0.5	3.8±0.2	4.0±0.4	2.2±0.2	3.8±0.1	2.3±0.7	3.9±0.2	1.6±0.3
Endrin	3.4±0.4	2.0±0.2	<1.3	<1.3	3.4±0.6	2.2±0.4	<1.3	<1.3	4.0±0.6	2.9±0.3	<1.3	<1.3	<1.3
Endrin aldehyde	2.9±0.5	3.0±0.4	<2.0	<2.0	2.5±0.2	3.4±0.6	2.1±0.3	<2.0	4.2±0.3	4.1±0.6	<2.0	<2.0	<2.0
Endosulfan I	<0.9	<0.9	<0.9	<0.9	2.2±0.2	1.0±0.2	<0.9	<0.9	<0.9	<0.9	<0.9	<0.9	<0.9
Endosulfan II	<1.0	<1.0	<1.0	<1.0	1.5±0.6	1.6±0.2	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Endosulfan sulfate	0.8±0.2	<0.7	<0.7	1.3±0.2	1.0±0.1	<0.7	<0.7	1.3±0.1	<0.7	<0.7	<0.7	<0.7	<0.7
Mirex	0.6±0.1	0.7±0.1	<0.1	0.6±0.1	<0.1	0.6±0.1	1.0±0.2	0.9±0.1	0.7±0.2	0.6±0.1	0.8±0.3	0.6±0.1	<0.1
Methoxychlor	1.2±0.2	<0.4	0.6±0.1	0.5±0.1	<0.4	0.6±0.1	0.5±0.3	1.2±0.3	<0.4	0.8±0.2	0.7±0.2	<0.4	<0.4

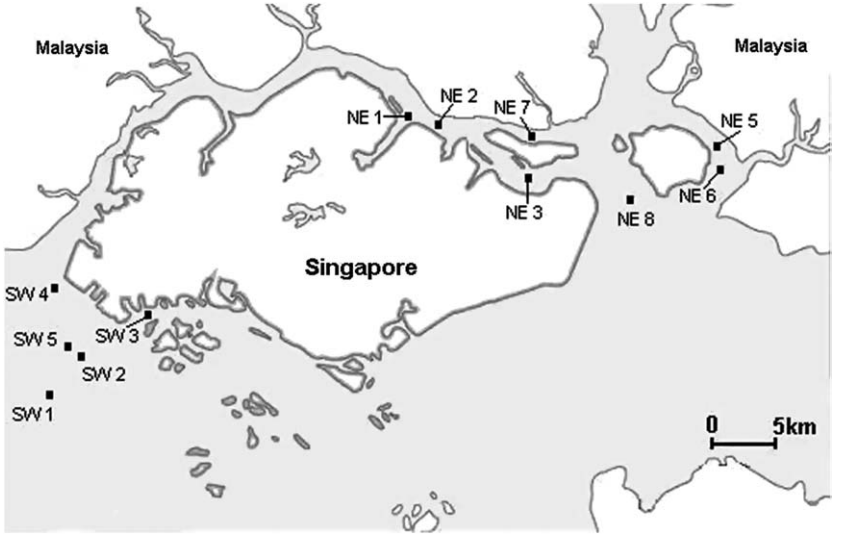

Figure 15.12. Sediment sample locations in the northwestern and southwestern regions of Singapore.

confirms a dispersion of PCBs moving seawards. A concentration of 138.5 ng g^{-1} was found in the Straits of Johor, northern part of Singapore (NE2). Shipyards and industry areas may be the sources for PCBs in this area. Lower concentrations of ΣPCBs between 1.4 and 55.4 ng g^{-1} were found in the northeastern part of Singapore, which is affected by agricultural and fish farming activities. The sediment sample corresponding to lowest concentration of 1.4 ng g^{-1} was mainly of mineral composition and showed the lowest TOC value. Significantly high concentration of higher chlorinated PCBs (62–69% chlorination grade) was found, which are less volatile and therefore more persistent.

Concentrations of ΣHCHs (sum of α-, β-, γ- and δ-HCH) ranged from 3.3 ng g^{-1} at station SW4 to 46.2 ng g^{-1} at station NE2. A slightly higher trend of concentrations of ΣHCH at sample stations in the NE region was observed, most likely due to the higher level of agricultural and fish farming activity to the north of Singapore. The levels are comparable to those found in Hong Kong (0.1–16.7 ng g^{-1}) reported by Richardson and Zheng (1999), but much higher than those reported for Daya Bay, China (0.3–4.2 ng g^{-1}) by Zhou et al., (2001) (Table 15.7). The mean percentage composition of α-,β-, γ-, δ-HCH to ΣHCHs for sediments analysed in our study are 27%, 25%, 23% and 25% respectively. The transformation of α- and γ-HCH to β-HCH, and the low degradation rate and lower vapor pressure of β-HCH (Wu et al., 1997; Willett et al., 1998) may be expected

Table 15.7. Comparison of OCPs, PCBs and PBDEs concentrations in sediments from coastal areas in Asia (ng g^{-1} dw)

Location	Year	ΣPCBs	ΣDDTs[g]	ΣHCH	Σchlordane[j]	ΣPBDEs[k]	Reference
Osaka Bay, Japan	1990	63–240[a]	2.5–11.9	4.5–6.2[h]	0.2–1		Iwata et al., 1994
Hong Kong	1997/98	n.d.–97.9[a]	0.3–14.8	0.1–16.7[i]	n.d.–11.3		Richardson and Zheng, 1999
Masan Bay, Korea	1997	1.2–41.4[b]	0.2–80.2	n.d.–1.3[i]	n.d.–1.7		Hong et al., 2003
North Coast of Vietnam	1997	1.1–66.4[c]	6.2–10.4	1.2–33.7[h]			Nhan et al., 1999
Minjiang River Estuary, China	1999	15.8–57.9[d]	1.6–13.1	3–16.2[i]			Zhang et al., 2003
Daya Bay, China	1999	0.9–11.2[c]	0.1–20.3	0.3–4.2[i]			Zhou et al., 2001
Osaka Bay, Japan	2000					0.2–651.9	Choi et al., 2003
Yangtze Estuary, China	2001	n.d.–19.0[e]	n.d.–0.6				Liu et al., 2003
Singapore	2003	1.4–329.6[f]	2.2–11.9	3.4–46.1[i]	1.4–18.7	3.4–13.8	This study

Note: n.d. = not detectable.
[a]PCB mixture basis (Aroclors).
[b]$2 \times \Sigma PCB_{18}$; twice the sum of PCB congeners 8,18, 28, 44, 52, 66, 101, 105, 118, 128, 138, 170, 180, 187, 195, 206 and 209.
[c]$5 \times \Sigma PCB_6$; sum of six PCB congeners 28, 52, 101, 138, 153 and 180 multiplied by a factor of 5 for 209 PCB mixture basis (Cullen et al., 1996).
[d]Sum of PCB congeners 1, 5, 28, 29, 47, 49, 77, 97, 101, 105, 118, 138, 153, 154, 169, 171, 180, 200, 204.
[e]Sum of PCB congeners 1, 11, 29, 47, 121, 136, 187.
[f]Sum of PCB congeners 18, 28, 31, 33, 44, 49, 53, 70, 74, 82, 87, 95, 99, 101, 105, 118, 128, 132, 138, 153, 156, 158, 169, 170, 171, 177, 180, 183, 187, 191, 194, 195, 199, 205, 206, 208, 209.
[g]Sum of *p,p′*-DDE, *p,p′*-DDD, and *p,p′*-DDT.
[h]Sum of α-HCH, β-HCH and γ-HCH.
[i]Sum of α-HCH, β-HCH, δ-HCH and γ-HCH.
[j]Sum of *cis*- and *trans*-chlordane.
[k]Sum of PBDE congeners 47, 99 and 100.

to lead to an accumulation of β-HCH in sediments and a much lower concentration of α- and γ-HCHs. However, this was not observed in this study and leads to the assumption that HCHs are still being introduced to the marine environment in the region of Singapore. Indonesia banned Lindane (γ-HCH) in 1991 and Thailand in 2002 (UNEP, 2002). However, Lindane may still be used in palm oil and coconut plantage in Malaysia (UNEP, 2002).

Concentrations of ΣDDT ranged from 2.2 to 11.9 ng g^{-1} in Singapore's coastal sediments compared to ranges in Hong Kong (Richardson and Zheng, 1999), Daya Bay (China, Zhou et al., 2001) and Osaka Bay (Japan, Iwata et al., 1994) of 0.3–14.8 ng g^{-1}, 0.1–20.3 and 2.5–11.9 ng g^{-1}, respectively (Table 15.7). The concentrations of DDT and its metabolites DDD and DDE are higher at sample locations around Jurong Island (SW series) by a factor of around 2 compared to sediments from the Straits of Johor (NE series), particularly concentrations of DDT. The highest concentrations for ΣDDT were found at sample locations SW4 and SW5 of 11.9 and 10.5 ng g^{-1}, respectively and the lowest at NE2 and at NE4 at 2.2 ng g^{-1}. However the ratio of DDT to the sum of DDE and DDD is much smaller than 1 (range of 0.2–0.8) at all locations, implying that there has been no recent introduction of DDT to the marine environment of Singapore. Bayen et al. (2004) made the same conclusion based on concentration levels of DDTs in the tissues of green mussels (*Perna viridis*). As DDT has been banned for a longer period than HCHs in many countries in Southeast Asia, DDT is less prevalent and present at a lower concentration. DDT was banned in Singapore in 1985, but later than this in other countries in the region and continues to be used in some countries such as the Philippines (UNEP, 2002).

Among the chlordane-related compounds (*cis*-Chlordane, *trans*-Chlordane, Heptachlor and Heptachlor epoxide), the concentration of ΣChlordane (*cis*- + *trans*-Chlordane) in sediments is high at sample locations SW4 and NE7 with 18.7 and 18.5 ng g^{-1} respectively, and higher by a factor of two compared with data from Hong Kong (Table 15.7). At the other sample locations, the concentration of ΣChlordane ranged between 1.4 and 6.5 ng g^{-1}, whereas the concentration of *cis*-Chlordane is either equal to or higher by a factor of two than the concentration of *trans*-Chlordane. Chlordane was banned in Singapore only in 1999, later than in some other Southeast Asian countries (UNEP, 2002). Heptachlor was detected at up to 9.0 ng g^{-1} and Heptachlor epoxide, a degradation product of Heptachlor, at a peak concentration of 6.8 ng g^{-1}. The concentrations of Heptachlor epoxide are higher by factors of 1.1–3.4 than Heptachlor, except for sediments from sample locations SW4 and NE3, where the concentration ratio of Heptachlor

epoxide to Heptachlor is less than 1. The data can be interpreted on the basis that there has been no recent introduction of Heptachlor into Singapore's marine environment. Heptachlor was banned in Singapore in 1985 and in Malaysia in 1990 (UNEP, 2002).

Aldrin was detected in 50% of all sediments collected, with a higher frequency in samples from the Straits of Johor (NE series) at a low concentration range (0.6–1.0 ng g^{-1}). Concentrations of Dieldrin were higher, ranging from below the limit of detection i.e., 1.2–4.4 ng g^{-1}. Endrin was detected in a range from below the limit of detection i.e., 1.3–4.0 ng g^{-1}. Endrin aldehyde, a metabolite of Endrin, was found in a range of below the limit of detection i.e., 2–4.2 ng g^{-1}. Endrin ketone, a second metabolite formed only by exposure to sunlight, could not be detected. Endrin was banned in Singapore in 1995, and later in other Southeast Asian countries (UNEP, 2002).

Endosulfan I, Endosulfan II and Endosulfan sulfate were only detected in a few sample locations at low concentrations of between 0.8 and 2.2 ng g^{-1}. Mirex and Methoxychlor were found frequently among sample locations at low concentrations ranging from 0.6 to 1 ng g^{-1} and 0.5 to 1.2 ng g^{-1}, respectively.

Among the PBDE congeners measured i.e., BDE-47, BDE-99 and BDE-100 (detection limit of 0.98 ng g^{-1}, 0.92 ng g^{-1} and 0.87 ng g^{-1} respectively), only BDE-47 could be detected in sediments, at a concentration range of 3.4–13.8 ng g^{-1} (Table 15.5). The concentration of PBDEs was lower and less variable among sample locations relative to PCBs, indicating a more disperse input of this compound into the marine environment around Singapore. The highest concentrations were found at sample locations SW3, NE2 and NE8. The main source of PBDEs at these sample locations are likely to be harbours and industrial activities, shipyards and intensive shipping traffic. Among the other sample locations, the concentrations of BDE-47 in sediments varied between 3.4 and 5.5 ng g^{-1}.

A comparison of the concentration of PCBs and OCPs in Singapore's coastal marine sediments with the sediment quality guideline specified by the USEPA (1997) and the Canadian Council of Ministers of the Environment (CCME, 2002) (Table 15.8), and comparison with levels of contaminants reported from other locations in Asia, Singapore's marine sediments can be classified as moderately contaminated with probable ecotoxicological impacts to marine organisms.

15.5. POPs in marine biota

An assessment of the fate and transport of POPs in the global environment requires baseline measurements of such compounds. Global

Table 15.8. Minimum, maximum and average concentrations of major organochlorine contaminants in ng g^{-1}, and corresponding sediment quality criteria

	C_{min}	C_{max}	$C_{average}$	ER-L[a]	% above ER-L[b]	ER-M[c]	% above ER-M[b]	TEL[d]	% above TEL[b]	PEL[e]	% above PEL[b]
∑PCBs	1.4	329.6	73.9	22.7	84.6	180	7.7	21.55	84.6	188.79	7.7
p,p′-DDT	<0.46	4.2	2.1	1	69.2	7	0	1.19	69.2	4.77	0
p,p′-DDD	1.3	4.0	2.5	2	61.5	20	0	1.22	100	7.81	0
p,p′-DDE	0.6	4.7	2.2	2.2	46.2	27	0	2.07	46.2	374.17	0
∑DDT	2.2	11.9	6.7	1.58	100	46.1	0	3.89	76.9	51.7	0
γ-HCH	<0.66	13.4	4.3					0.32	> 84.6	0.99	76.9
∑Chlordane[f]	<1.67	18.7	5.4	0.5	100	6	23.1	2.26	84.6	4.79	23.1

[a]Effects range-law value.
[b]percentage of samples above level.
[c]Effects range-median value.
[d]Threshold effects level.
[e]Probable effects level.
[f]Sum of cis- and trans-Chlordane.

distillation is the most probable theory to explain the widespread and ubiquitous nature of POPs in the environment. According to this theory, POPs are volatilized into the atmosphere at tropical and temperate latitudes and undergo long range transport prior to deposition in the polar regions of the globe (Vallack et al., 1998). Bioaccumulation of POPs in the polar foodchains represents a significant risk for tertiary consumers, where adverse effects on the reproductive health and well-being of a range or polar organisms has been recently reported (Bard, 1999). Despite the importance of the tropical region as a source of POPs in the global cycling of these compounds, there is a paucity of data on the prevalence of PCBs and organochlorine pesticides in the Asian environment. More recently, PBDEs, which are widely used in fire retarded materials have become of increasing concern due to their potential ecotoxicological impact.

Almost all studies on the behaviour of PBDEs in the environment are derived from Europe, North America, Japan and the Arctic Pole (de Wit, 2002). There is no reported data on the prevalence of PBDEs in the tropical environment of Southeast Asia. Although highly industrialized, Singapore still has a wide diversity of marine habitats including sandy beaches, mangroves, rocky shores and coral reefs. However, many of these habitats are threatened by land reclamation and intense port activity, as well as marine pollution (Thia-Eng et al., 2000). Although DDT residues have been detected in human blood serum in Singapore and partly attributed to seafood consumption (Luo et al., 1997), there is a lack of data on the prevalence of POPs in Singapore's marine environment. Polychlorinated biphenyl use has been banned for more than two decades in Singapore. To date, PBDEs have not been reported in the tropical marine environment of Southeast Asia, although a study has been completed on fish tissue in Japan. In 2001, Akutsu et al. (2001) found elevated levels of pentabrominated diphenyl ethers (penta-BDE), especially BDE-47, in marine fishes in the Inland Sea of Japan. [There are some recent studies in Hong Kong and mainland China.]

The Green Mussel, *P. viridis*, is a filter feeding organism and therefore readily accumulates POPs from its environment via the ingestion of waterborne organic particulates. Consequently, *P. viridis* has been previously used as a bioindicator species for various POPs including pesticides and polychlorinated biphenyls (Tanabe et al., 2000). The Blue Mussel, *Mytilus edulis*, has also been used for monitoring various POPs, and recently for PBDEs in Atlantic waters (Christensen et al., 2002). A study was undertaken on the levels of PCBs and PBDEs detected in *P. viridis* collected from Singapore's coastal marine environment by Bayen et al. (2003) and represents the first baseline study of such compounds in Singapore's marine

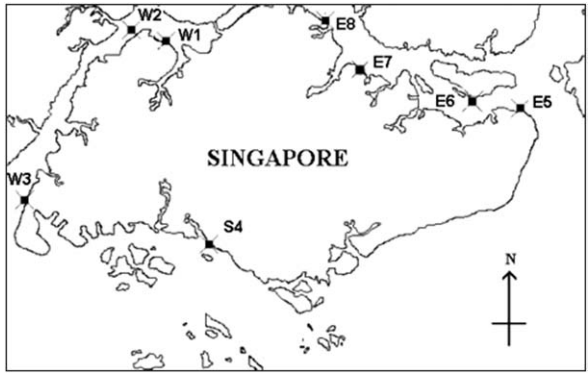

Figure 15.13. Sampling locations of *P. viridis* in Singapore's coastal environment.

biota, and is also the first report of the use of *P. viridis* for the monitoring of PBDEs in the tropical marine environment of Southeast Asia.

Specimens of *P. viridis* were collected from eight locations around Singapore's coastline between April and May 2002 (Fig. 15.13). The stations were chosen based on their proximity to industrial activities, and their position around the land-link causeway in the Straits of Johore. The causeway represents a physical barrier to marine hydrodynamics around Singapore's northern coast. Samples were collected from floating structures and shore defence walls. As the size of the individual is an important parameter affecting the level of pollutant bioaccumulation in *P. viridis* (Richardson et al., 2001), individuals were collected in the largest and most similar size range in all locations so as to target mature specimens for analysis.

15.5.1. PCB levels in P. viridis

Congener specific concentrations of PCBs in *P. viridis* samples on a dry-weight (dw) basis are given in Table 15.9. All PCB congeners in the analytical suite were detected in the mussel tissues. Total PCBs concentrations ranged from 6.1 to 82 ng g^{-1} dw. The highest concentrations were recorded in samples from E8 (i.e., 82 ng g^{-1} dw) and S4 (i.e., 44 ng g^{-1} dw) (Fig. 15.13), which are from locations adjacent to ship maintenance yards and busy shipping lanes located on the north (i.e., Sembawang) and east (i.e., Jurong) coasts of Singapore. Tissue levels of PCBs in stations W1, W2, W3, E5, E6 and E7 were lower (6.1–23 ng g^{-1} dw) with the lowest level recorded in *P. viridis* tissues from station W3. The causeway in the Straits of Johore separates hydrodynamic flow of marine water

Table 15.9. Concentrations for major PCB congeners in *P. viridis* tissue (ng g^{-1} dry basis)

	W1	W2	W3	S4	S4	E5	E6	E7	E7	E8
PCB 49	0.35	0.53	BLD	0.87	1.1	0.64	0.88	1.1	0.33	4.6
PCB 52	0.59	0.86	BLD	1.2	1.5	0.85	1.2	1.2	0.41	4.6
PCB 90/101	1.4	1.5	BLD	3.6	3.7	1.4	1.5	1.6	0.84	8.4
PCB 110	0.85	0.99	0.35	3.3	3.5	1.1	1.1	1.3	0.52	5.8
PCB 118	0.56	0.57	0.22	2.0	2.1	0.96	0.91	0.93	0.43	4.4
PCB 138	2.3	1.9	1.1	6.1	6.3	1.8	1.7	2.0	0.87	9.2
PCB 149	2.1	1.9	0.93	4.7	4.7	1.2	1.2	1.5	0.88	7.4
PCB 153	2.5	1.9	1.1	5.7	6.1	1.8	1.8	2.1	0.97	9.5
Total PCBs	17	23	6.1	44	47	15	15	18	7.1	82

Note: BLD – below limit of detection.

between the west and the east of the Straits of Johore, and this likely accounts for the observed differences in *P. viridis* tissue concentrations.

PCB data for *P. viridis* tissues are available for samples analysed elsewhere. On a wet weight basis (ww), PCB levels in Singapore (1.4–15 ng g^{-1} ww) are higher than values reported recently in neighbouring countries such as Malaysia (0.7–2.1ng g^{-1} ww), and Indonesia (0.2–2.7 ng g^{-1} ww) (Monirith et al., 2000). Hong Kong and Singapore have similarities in terms of size, human population and density and levels of industrial and shipping activities. However, the PCB burden in *P. viridis* reported from both clean and polluted marine zones of Hong Kong (Richardson et al., 2001) is higher than values reported here for Singapore, and range from 120 to 420 ng g^{-1} dw.

Pentachlorinated biphenyls and hexachlorinated biphenyls are the major PCB groups typically found in *P. viridis*. PCBs are sold commercially as technical mixtures, called Aroclors, each with a specific pattern of chlorination. Patterns have been determined for Aroclor mixtures 1221, 1232, 1242, 1248, 1254, 1260 and 1262 (Frame et al., 1996). Principal component analysis (PCA) was performed to compare the relative PCB congener profile of mussel tissues analysed in 2002 and the commercial Aroclor mixtures (Fig. 15.14). The closest match in the PCB data for *P. viridis* samples collected in Singapore from our study is the common Aroclor 1254. The slight discrepancy is due to the presence of PCB-149 in mussel tissue and a greater prominence of PCB-110 and -118 in Aroclor 1254. PCA analysis revealed that samples from the west Straits of Johore (W1, W2 and W3) contain more penta-CBs and less hexa-CBs than samples from the east Straits (E6, E7 and E8). The sample collected in the south of Singapore (S4) has an intermediary pattern of PCB contamination. A similar match has been observed in marine crabs and fishes

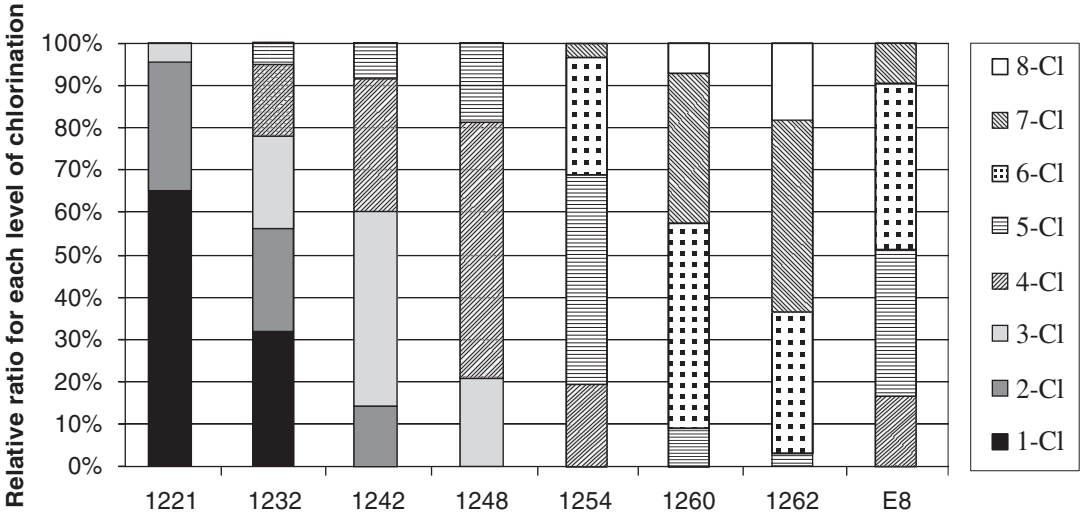

Figure 15.14. PCB profile for each level of chlorination in Aroclors compared to profile in *P. viridis* tissues from station E8. Nonachlorinated and decachlorinated PCBs were voluntary omitted as their amount is low compared to other levels of chlorination.

Table 15.10. Concentrations for major PBDE congeners in *P. viridis* tissue (ng.g^{-1} dry basis)

Congener	W1	W2	W3	S4	S4	E5	E6	E7	E7	E8
BDE 47	0.71	0.86	11	5.0	3.8	1.2	0.72	1.5	0.62	1.7
BDE 49	0.12	0.15	0.99	0.82	0.66	0.22	0.11	0.28	0.12	0.35
BDE 66	0.07	0.08	0.59	0.35	0.23	0.10	0.06	0.12	0.09	0.14
BDE 99	0.57	0.69	18	3.8	3.1	1.1	0.62	2.1	0.69	1.5
BDE 100	0.13	0.16	5.3	1.1	0.76	0.28	0.17	0.56	0.21	0.38
BDE 153	BLD	BLD	0.64	0.28	0.22	BLD	BLD	0.23	BLD	BLD
BDE 154	0.05	0.07	0.45	0.20	0.20	0.05	0.08	0.13	BLD	0.12
Total PBDEs	2.1	2.3	38	13	9.9	3.3	2.1	5.3	2.0	4.7
PCB/PBDE ratio	8.3	4.4	0.2	3.5	4.7	10	7.3	3.4	3.6	17

Note: BLD – below limit of detection.

in Hawaii (Miao et al., 2000). PCB 153, 138 and 149 are the major main congeners found in the tissues of *P. viridis* in Singapore, which are not classified as high-carcinogenic risk dioxin-like PCBs (Kumar et al., 2001).

15.5.2. PBDEs levels in P. viridis

Levels of PBDE congeners 17, 28, 32, 35, 37, 47, 49, 66, 71, 75, 77, 85, 99, 100, 120, 138, 153, 154, 166, 181 and 190 in *P. viridis* tissues are reported in Table 15.10. The sum total of individual congeners ranged from 2.0 to 38 ng g^{-1} dw and have a different geographical profile from PCBs. The highest concentration of PBDEs was found in tissue samples from station W3. This station is close to the industrial area of Tuas. station S4, which is adjacent to an industrialized area and shipping lane in the south of Singapore, also had an elevated PBDE concentration (13 ng g^{-1} dw). Tissue samples from the six other locations had lower concentrations and were similar in their PBDE patterns.

No comparative PBDE data are available for *P. viridis* outside Singapore, although PBDE levels have been previously reported in blue mussel (*M. edulis*) tissues from marine waters off Denmark and Greenland (Christensen and Platz, 2001; Christensen et al., 2002). Concentrations of the PBDEs 47, 99, 100 and 153 in *M. edulis* tissues ranged from 0.080 to 0.81 ng g^{-1} ww in Denmark and 0.11 ng g^{-1} ww in Greenland. On a wet weight basis *P. viridis* tissues from Singapore ranged from 0.29 to 8.6 ng g^{-1} ww. Therefore, in comparative terms, PBDE levels in mussel tissues from Singapore are up to an order of magnitude greater than the available data from elsewhere. PBDE levels found in this study are in the

range of concentrations recorded in a range of fish and marine mammals (de Wit, 2002).

BDE-99 and BDE-47 are the two main components of the commercial pentabrominated diphenyl ethers mixture such as Bromkal 70–5DE, with composition of 35% and 37% respectively (Sjödin et al., 1998) and DE-71, with compositions of 47% and 25% respectively (Dodder et al., 2002). These two PBDE congeners were dominant in the *P. viridis* samples, with a lower contribution of BDE-100. In most sample locations (except W3 and E7), BDE-47 was present at higher tissues concentrations than BDE-99. Gustafsson et al. (1999) reported that the uptake of BDE-47 and BDE-99 in *M. edulis* is virtually identical, but the depuration rate of BDE-99 is faster. As a consequence, an organism, exposed to penta-PBDE contamination, can be expected to have a higher tissue concentration of BDE-99 than BDE-47. When the organisms are no longer exposed, BDE-99 is likely to be excreted and BDE-47 therefore becomes the dominant congener. In samples collected in Tuas (station W3) and Sembawang (station E7) in Singapore, BDE-99 was more prevalent, which indicates a relatively recent contamination event. Fig. 15.15 shows the relative ratio of each BDE in *P. viridis* from station W3. BDE-99 and BDE-47 were present at composition percentages of 46% and 30%, respectively, which is close to the composition of the commercial mixture DE-71, if we neglect the contribution of other BDEs which were not monitored in this study. Similar conclusions can be deduced for tissue

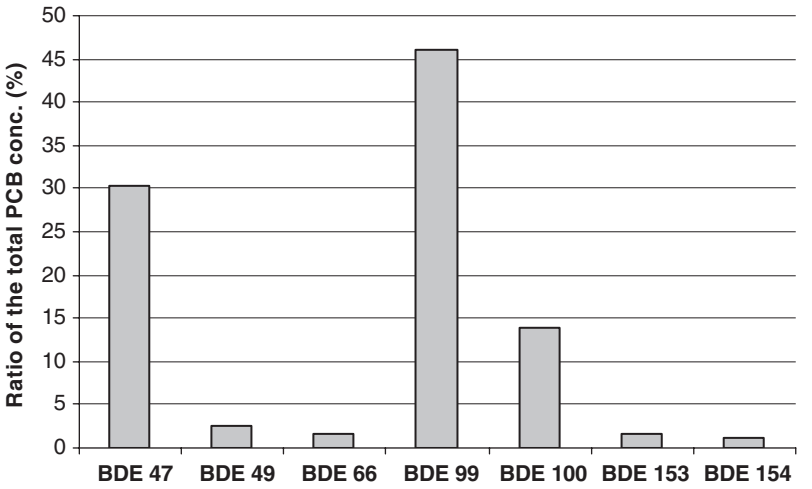

Figure 15.15. PBDEs congener profile in *P. viridis* tissues from station W3 as ratio of the total PBDE concentration.

samples from station E7 (i.e., 39% BDE-99 and 29% BDE-47). Amongst numerous studies summarized in a review by de Wit (2002), no reference reported this ratio for BDE-99 and BDE-47 in marine tissues. Important factor to consider: Debromination of BDE-209 occurs in marine organism forming lower brominated congeners, particular tetra- and penta-brominated congeners. BDE-209 is hardly reported in marine organism due to this effect. Commercial product DeBDE contains 97% of DeBDE and Asia demanded 42% of world market for DeBDE in 1999 (de Wit, 2002).

Ratios of total PCB:PBDE concentrations for *P. viridis* tissues have been calculated and are reported in Table 15.10. This ratio varies between 0.2 and 17. The mean ratio for samples from Singapore is close to 7, a value previously reported for Arctic biota samples (Christensen et al., 2002). However, stations W3 and E8, had respective ratios of 0.2 and 17, and therefore are likely affected by one or more specific local sources of contamination. The lack of correlation between these two pollutants in our study indicates that the sources of PCB and PBDE contamination are independent of each other. The difference may be due to the types of local industrial activities that are present, with emphasis on the electronics industry and shipping activities.

PCBs and PBDEs were detected in all *P. viridis* tissue samples collected from all sample locations in Singapore's coastal waters. Although the use of PCBs was banned several decades ago in Singapore, biological tissues still contain PCB congeners that have a similar profile to that of the commercial PCB Aroclor 1254. The busy shipping routes appear to be the most contaminated locations in Singapore's coastal waters, although levels are rather lower than recorded in Hong Kong (Richardson et al., 2001). *P. viridis* was also a useful bioindicator of polybrominated PBDEs in Singapore's marine environment. These POPs were also found in all tissue samples collected and measured levels were generally in the same concentration range as PCBs, although higher concentrations were measured in tissues collected from Tuas (station W3), adjacent to an industrial area. Tissue samples from all locations had elevated levels of pentabrominated diphenyl ethers which are characteristic of technical mixtures commonly used as flame retardants in a range of commercial and consumer goods. PBDEs can be readily emitted into the environment, where, for example, flame retardants may be discharged upon usage, and may also be leached from plastic and electronic wastes, construction material, textiles and sewage plants, or as direct emissions from industrial plants (de Wit, 2002). Levels of PBDEs in *P. viridis* tissue from Tuas (station W3) were an order of magnitude higher than the upper concentrations reported in *M. edulis* tissues from coastal waters adjacent to

Denmark (Christensen and Platz, 2001). Furthermore, individual congener analysis for BDE-47 and BDE-99 in tissue samples collected from stations W3 and E7 indicate that biotic exposure to PBDEs is current. Singapore, and more generally Southeast Asia, may represent a significant source of PBDEs on the global scale. As mussels are low in the food chain, higher concentrations of PCBs and PBDEs may be expected in organisms, such as marine mammals, at higher trophic levels.

15.6. POPs in mangrove habitats

Mangroves are an important, yet endangered tropical ecosystem in Southeast Asia. Mangroves once covered an estimated 13% of Singapore's total land area in 1820, but now represent less than 0.5% today (Ng and Sivasothi, 1999). They are unique ecosystems with a high level of biodiversity, and new species continue to be recorded in local mangrove habitats (Ng and Sivasothi, 1999). Mangroves also have an important socio-economic role in local aquaculture in Southeast Asia, and serve as nursing grounds for coastal fish stocks (Mumby et al., 2003). They also provide a wide range of food organisms that are commonly consumed by humans in Southeast Asia (e.g., mussels, rodong shell, crab, mullet and even monitor lizard) and aquaculture products (principally prawns and fish). The biomagnification of POPs in ecological food chains has been well documented for freshwater ecosystems (e.g., Stapleton et al., 2001), as well as marine food webs in polar (e.g., Dietz et al., 2000) and temperate regions (e.g., Law et al., 2003; Voorspoels et al., 2003). However, data on the prevalence of POPs in tropical mangrove ecosystems is restricted to just a few studies in Mexico (Páez-Osuna et al., 2002) and Hong Kong (Liang et al., 1999; Zheng et al., 2000), with sparse information on POP levels in biota.

For the purposes of study, organisms were collected within a one-week period in April 2004 at two mangrove sites in Singapore: Sungei Buloh and Sungei Khatib Bongsu (Fig. 15.16). These sites are located on each side of the land-link causeway between Singapore and Malaysia in the Straits of Johore. The causeway represents a physical barrier to marine hydrodynamics around Singapore's northern coast, where there is no exchange of seawater across the causeway. S. Buloh mangrove has been used in the past as a site for prawn aquaculture before being declared a protected nature reserve in 1989. S. Khatib Bongsu is not a protected site.

Criteria for species selection included abundant availability and trophic position within the mangrove food web. Fish were captured using a traditional cast net. Other organisms were collected directly from their

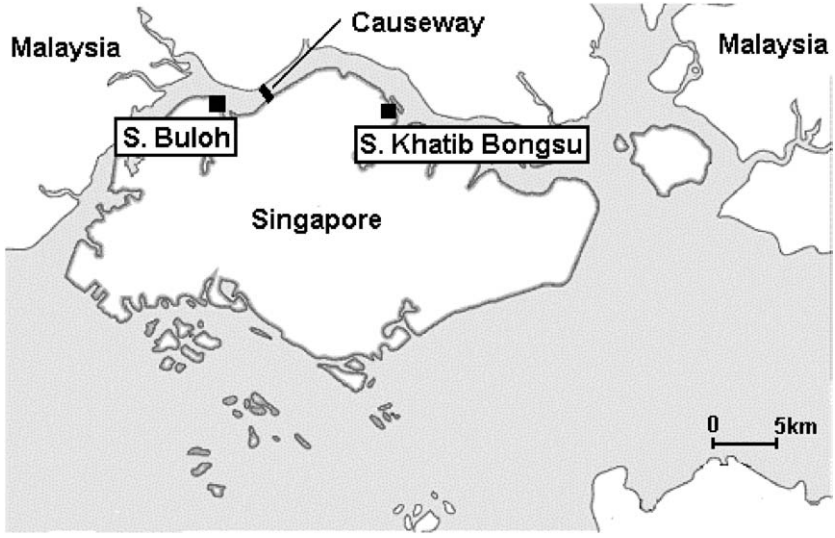

Figure 15.16. Location of Sungei Buloh and S. Khatib Bongsu mangroves in Singapore.

natural habitat. The number of individuals caught was different for each location and varied between 25 to 50 individuals for polychaetes, 6 to 40 individuals for molluscs, 5 to 170 individuals for crustaceans and 1 to 20 individuals for fish. All species sampled were available in large numbers, and a total of 15 species were common to both sites. Size and age are important parameters affecting the level of POPs accumulation (Stapleton et al., 2001), and organisms were selected for analysis in the most similar and available size range possible.

Levels of POPs in seawater and sediments in both mangrove sites are summarized in Table 15.11. All POPs, except PBDEs, were detected in subsurface seawater, SML and sediments. PCBs were the dominant POPs in subsurface seawater and SML with levels ranging from 0.1 to 6.7 ng L^{-1} and 2.4 to 26 ng L^{-1}, respectively. The PCB congener profile in subsurface seawater and SML samples was dominated by PCB 151, which represented more than 80% of the total PCB load at both mangrove sites. HCHs were the dominant OCP in subsurface seawater and SML samples with levels ranging from 0.1 to 2.3 ng L^{-1} and 0.9 to 9.0 ng L^{-1}, respectively. Levels DDTs were generally two orders of magnitude higher than HCHs. *p*,*p*′-DDT and β-HCH dominated the DDT and HCH profiles of seawater at both sites.

The PCB congener profile in mangrove sediments was dominated by trichlorinated biphenyls in S. Buloh (66% of the total PCB load), whereas

Table 15.11. Levels of POPs in sediments, subsurface seawater and sea surface microlayer collected from mangroves in Singapore

	POPs level in sediments (ng.g^{-1} dw)		POPs level in subsurface seawater (pg.L^{-1})		POPs level in sea surface microlayer (pg.L^{-1})		Enrichment factor microlayer/ subsurface	
	SB	SKB	SB	SKB	SB	SKB	SB	SKB
PCBs	0.59–1.14 (0.88)	0.80–1.86 (1.33)		140–1500 (910)			0.3–3.9 (2.1)	2.3–10.9 (6.7)
			6700–7100 (6900)		2400–26000 (14000)	1500–7300 (4100)		
PBDEs	n.d.	n.d.	<80	<80	<80	<80	n.a.	n.a.
DDTs	<0.1–0.93 (0.29)	0.56–0.85 (0.70)		3–72 (32)			1.2–33 (17)	1.4–8.3 (4.0)
			18–23 (21)		21–170 (95)	27–99 (61)		
HCHs	1.8–6.0 (3.9)	1.2–1.6 (1.4)		110–1100 (770)			1.7–22 (11)	0.8–8.8 (3.6)
			2000–2300 (2100)		4000–9000 (6500)	880–1400 (1100)		
Endosulfan	<0.2	<0.2		25–40 (30)			2.8	2.4–9.7 (4.9)
			280–630 (450)		330–1800 (1000)	59–240 (140)		
Chlordanes	0.01–0.04 (0.02)	0.02–0.06 (0.04)		<1–3 (2)		<1	1.3	n.a.
			4–8 (6)		<1–5			

Note: n.d.– not detected; n.a.– not applicable.

hexachlorinated (54%) and pentachlorinated biphenyls (27%) were dominant in sediments in S. Khatib Bongsu. HCHs were the dominant organochlorine pesticide in mangrove sediments with levels ranging from 1.2 to 6.0 ng g^{-1} dw.

The concentrations of PCBs, PBDEs, Chlordanes, DDTs, HCHs, and Endosulfans in all mangrove biota samples are reported on a wet weight basis (ww) in Table 15.12. Concentrations in mangrove organisms ranged from BLD to 45 ng g^{-1} ww for Chlordanes, BLD to150 ng g^{-1} ww for DDTs, 0.6–190 ng g^{-1} ww for PCBs, BLD to 9.9 ng g^{-1} ww for PBDEs, BLD to 2.8 ng g^{-1} ww for HCHs and BLD to 25 ng g^{-1} ww for Endosulfans. The lowest concentrations of POPs were found generally in the algal species (*Chaetomorpha gracilis* and *Catellana* sp.), the tube worms (*Dioptra neopolitana*), the nerite snail (*Nerita lineata*), prawn species (*Aphoristia microrhynchus* and *Penaus* sp.) and the tree-climbing crabs (*Episesarma* sp.). The highest concentrations of POPs were found in the soft tissues of the thunder crab (*Myomenippe hardwicki*) and the fishes. Levels of POPs in fish liver and fish eggs were generally one to two orders of magnitude higher than in the muscle tissue. On a lipid weight (lw) basis, POPs in fish livers ranged from 41 to 270 ng g^{-1} lw for Chlordanes, 27 to 860 ng g^{-1} lw for DDTs, 65 to 1600 ng g^{-1} lw for PCBs, 1.8 to 87 ng g^{-1} lw for PBDEs, 1.8 to 17 ng g^{-1} lw for HCHs and 2 to 350 ng g^{-1} lw for Endosulfans.

Concentrations of Chlordanes, DDTs, PCBs and PBDEs, on a dry weight basis (dw), are summarized for the 15 species common to both sites in Fig. 15.17. Higher concentrations of PCBs and PBDEs were generally found in biota from S. Khatib Bongsu and Chlordanes in biota from S. Buloh. However, the differences between the two sites are generally less than three standard deviations and are not significant.

PBDE profiles for mangrove biota samples collected in S. Buloh and S. Khatib Bongsu are presented in Figs. 15.18a b respectively. BDE-47 was the most abundant PBDE congener in the samples accounting for $71 \pm 18\%$ of the total PBDE load. The general contribution to total load is BDE-47 > BDE-99 > BDE-100 > BDE-154 > BDE-153, where BDE-47, -99 and -100 represent $96 \pm 11\%$ of the total load. The percentage of BDE-99 as the total PBDE load in polychaetes, rodongs, oysters, marine prawns and tree-climbing crabs at S. Khatib Bongsu was significantly higher than all other species at this mangrove site, and all species in S. Buloh (Mann-Whitney, $p < 0.05$). Organisms higher in the food chain, especially in S. Khatib Bongsu, appear to have lower levels of BDE-99. BDE-99 was absent in lokan clams (*Polymesoda expansa*) at both sites. With the exception of mullet, BDE-47 was present in lower proportions in fish livers than in muscles.

Table 15.12. POPs concentrations (ng g^{-1} ww) in the mangrove biota samples collected in April 2004

Common name	Site[a]	CHLs	DDTs	PCBs	PBDEs	HCHs	Endosulfans
Green algae	W	0.37±0.12	0.18±0.08	1.1±0.9	BLD	BLD	0.51±0.73
	E	BLD	2.3±1.9	0.71±0.94	BLD	BLD	BLD
Red algae	W	0.13±0.01	0.09±0.04	0.63±0.89	BLD	0.12±0.01	BLD
Nereid worm	W	0.80±0.03	1.1±0.1	1.2±0.2	0.09±0.00	0.14±0.00	0.22±0.03
	E	1.2±0.1	0.75±0.06	3.5±0.1	0.11±0.01	BLD	0.19±0.00
Tube worm	W	1.2±0.0	0.61±0.06	1.3±0.3	0.04±0.02	BLD	0.15±0.00
	E	0.14±0.01	0.34±0.02	0.91±0.26	0.05±0.01	0.05±0.07	0.05±0.06
Nerite snail	W	0.36±0.02	0.75±0.18	1.0±0.6	0.06±0.04	BLD	BLD
	E	0.55±0.20	0.23±0.05	0.53±0.26	BLD	BLD	BLD
Drill shell	W	11±2	5.1±0.8	10.1±2.2	0.74±0.03	0.21±0.00	2.5±0.3
	E	0.60±0.11	0.73±0.00	2.2±0.0	0.16±0.16	BLD	0.08±0.12
Rodong shell	W	0.28±0.00	1.6±0.6	4.8±0.1	0.01±0.02	BLD	BLD
	E	1.2±0.0	4.7±0.1	10.7±2.8	0.17±0.01	0.09±0.12	0.22±0.30
Green mussel	W	1.5±0.1	1.1±0.0	2.7±1.1	0.05±0.01	0.09±0.13	2.4±0.3
Lokan (clam)	W	3.8±0.6	3.1±0.7	4.5±1.9	0.07±0.00	0.22±0.07	4.3±2.7
	E	1.5±0.1	1.0±0.1	3.6±0.8	0.12±0.02	0.06±0.08	0.12±0.16
Leaf oyster	W	5.6±0.9	1.8±0.2	3.6±0.8	0.12±0.15	0.33±0.16	2.2±0.4
Mangrove oyster	E	4.5±0.3	2.5±0.6	8.5±0.5	0.16±0.01	0.21±0.30	1.0±0.1
Barnacles	W	3.1±0.1	0.70±0.05	4.4±2.0	0.09±0.03	BLD	0.67±0.05
	E	3.2±0.2	0.92±0.07	4.8±0.3	0.22±0.04	0.10±0.15	0.35±0.49
Snapping prawn	W	0.10±0.02	BLD	3.5±0.7	BLD	BLD	BLD
	E	0.06±0.00	BLD	2.8±1.0	BLD	0.09±0.12	BLD
Marine prawn	E	0.29±0.10	0.57±0.20	1.9±0.7	0.31±0.07	BLD	BLD
Tree climbing crab	W	0.18±0.00	0.25±0.02	0.59±0.59	0.09±0.00	0.05±0.06	BLD
Muscle	E	2.7±0.0	1.0±0.1	3.8±1.0	0.11±0.01	0.9±0.13	0.19±0.06
Tree climbing crab	W	0.33	1.3	5.3	0.27	0.50	BLD
Egg	E	2.8	7.3	31	1.1	0.68	0.60

Table 15.12. (*Continued*)

Common name	Site[a]	CHLs	DDTs	PCBs	PBDEs	HCHs	Endosulfans
Thunder crab	W	4.1±0.1	1.8±0.1	4.1±0.7	0.22±0.01	0.24±0.02	0.83±0.11
	E	15±4	5.8±2.9	23±11	2.0±1.2	0.14±0.06	2.1±0.6
Half-beak	W	1.9±0.2	2.3±0.9	4.0±0.9	0.30±0.04	BLD	1.3±0.5
	E	1.5±0.5	2.0±0.6	6.7±2.0	0.35±0.20	0.05±0.06	1.6±1.7
Mudskipper (muscle)	W	BLD	0.17±0.03	0.22±0.25	BLD	BLD	BLD
Eggs		1.5	18	34	0.70	0.2	0.12
Goby (muscle)	E	1.2±0.3	2.4±2.1	24±27	0.35±0.01	BLD	BLD
Liver	E	6.8	70	93	1.8	2.8	0.35
Eggs	E	14	17	70	4.5	0.59	4.1
Glass perchlet	W	1.6±0.3	1.5±0.1	2.6±1.3	0.07±0.07	BLD	0.64±0.40
Cardinalfish (muscle)	E	0.58±0.10	1.6±1.4	2.3±1.3	0.18±0.01	BLD	BLD
Liver	E	45	40	190	9.9	1.9	11
Mullet (muscle)	W	18±2	6.2±0.4	6.7±0.2	0.59±0.07	0.26±0.37	12±8
Liver	W	7.0	3.0	7.1	0.2	0.19	11
Muscle	E	4.7±0.2	4.0±0.3	9.2±1.1	0.42±0.24	0.33±0.04	1.0±0.1
Archer fish (muscle)	W	2.5±1.5	4.8±3.0	5.1±3.1	0.46±0.35	0.13±0.13	3.8±2.7
Liver	W	8.8	15	20	1.2	0.85	25
Muscle	E	1.1	6.8	3.4	0.32	BLD	0.31
Chromide (muscle)	W	0.46±0.16	1.7±1.2	2.3±2.3	0.23±0.12	BLD	BLD
Liver	W	14	47	90	4.8	0.73	4.2
Muscle	E	1.5±0.5	6.7±4.3	7.6±3.8	0.33±0.22	0.18±0.27	0.4±0.3
Liver	E	37	150	150	4.0	2.1	8.0

Note: BLD – below limit of detection.
[a]W: Sungei Buloh, E: Sungei Khatib Bongsu.

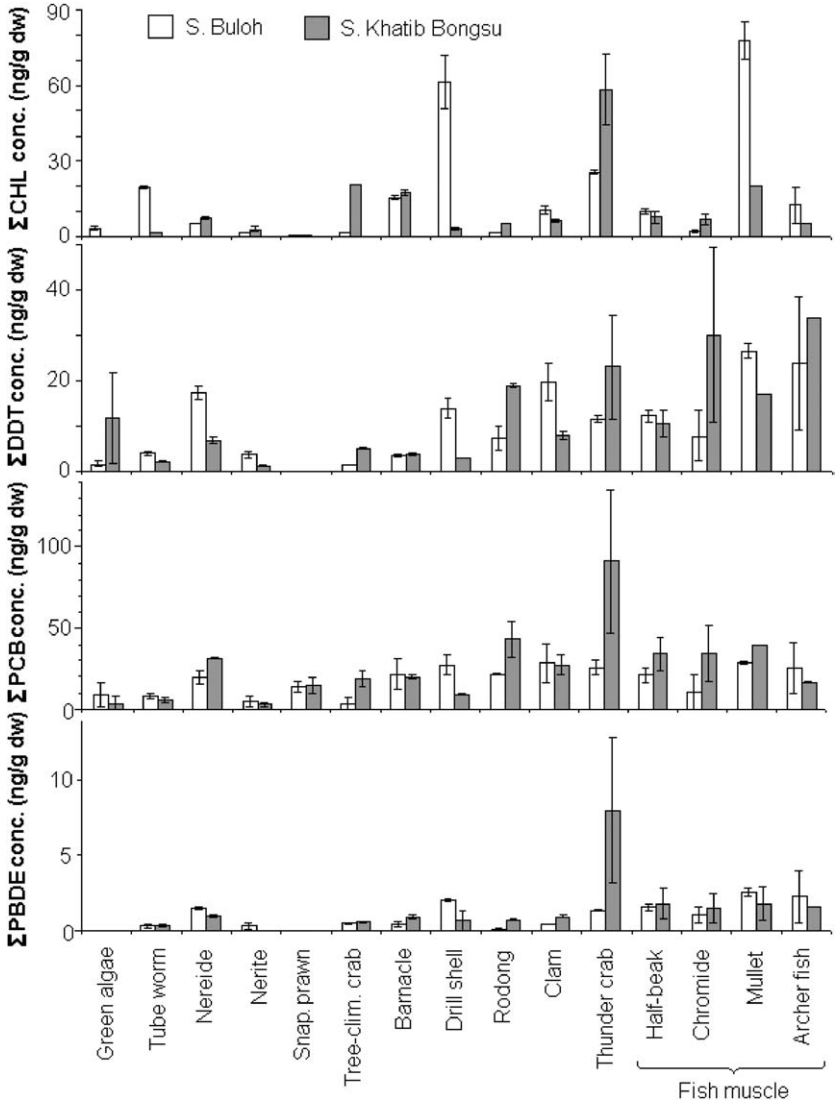

Figure 15.17. Concentrations of POPs in mangrove biota in Singapore (ng g^{-1} dw).

Hexachlorinated and pentachlorinated biphenyls dominated the PCB congener profile with an average of $39 \pm 12\%$ and $28 \pm 10\%$ respectively. PCB profiles were similar at both mangrove sites (See Figs. 15.19a b), where no clear difference in the PCB profile was discernable between the

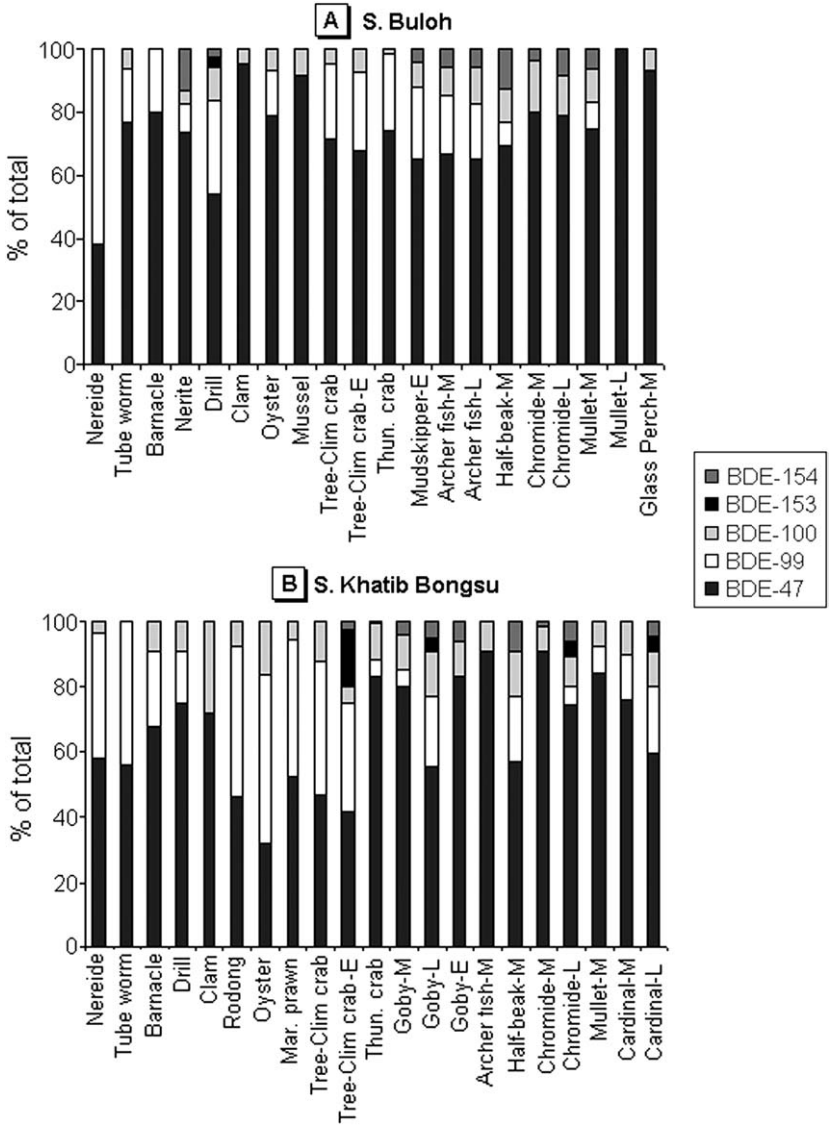

Figure 15.18. PBDE profiles in mangrove biota samples collected in Singapore. M: muscle; L: liver; E: eggs.

various mangrove species. The closest match in the PCB data for mangrove biota samples collected in this study is Aroclor 1254. α- and γ-Chlordane generally dominated the Chlordane congener profile of mangrove biota with an average of $38 \pm 16\%$ and $39 \pm 16\%$, respectively

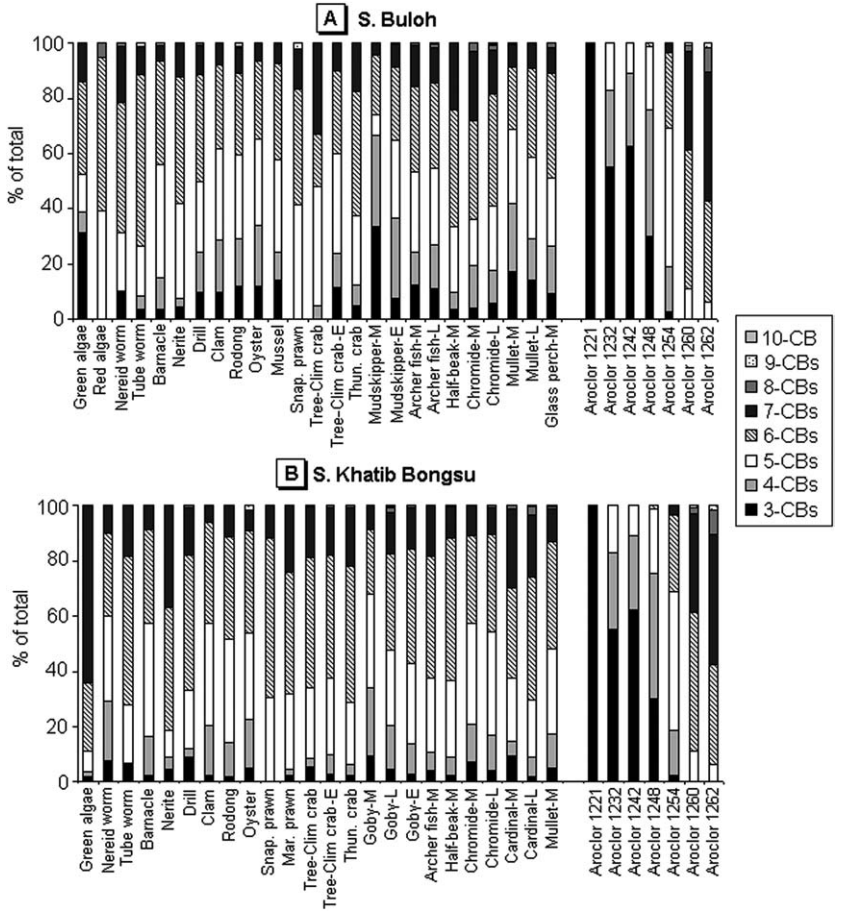

Figure 15.19. PCB profiles in mangrove biota samples collected in Singapore and in commercial Aroclor products. M: muscle; L: liver; E: eggs.

(See Figs. 15.20a b). However, a significantly lower percentage of α- and γ-Chlordane was found in the two crab species (Mann-Whitney, $p < 0.05$), and Heptachlor epoxide dominated the profile in crab species. Gobies, mudskipper and shrimp tissues also contained a significantly higher percentage of Heptachlor epoxide (Mann-Whitney, $p < 0.05$). p,p'-DDE was the dominant DDT congener in all samples, except in the green algae in S. Khatib Bongsu, representing an average of $84 \pm 15\%$ of the total DDT load. β-HCH was the dominant HCH congener, representing an average of $72 \pm 37\%$ of the total HCH load Fig. 15.21.

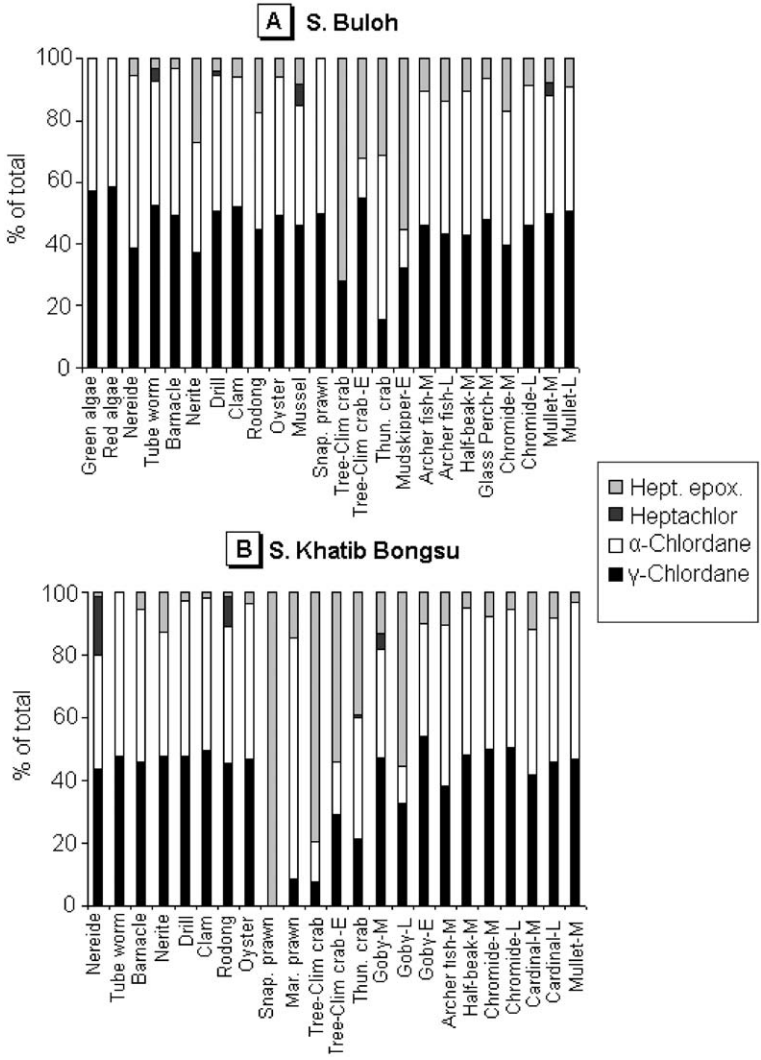

Figure 15.20. Chlordane profiles in mangrove biota samples collected in Singapore. M: muscle; L: liver; E: eggs.

Overall, the levels of POPs in mangrove sediments can be regarded as low (generally below or in the ng g^{-1} dw range), i.e., lower than what has been recorded in the marine sediments in Singapore (Wurl and Obbard, 2005b). Other studies have also reported that mangrove sediments have PCB levels lower or comparable to those in marine sediments, for

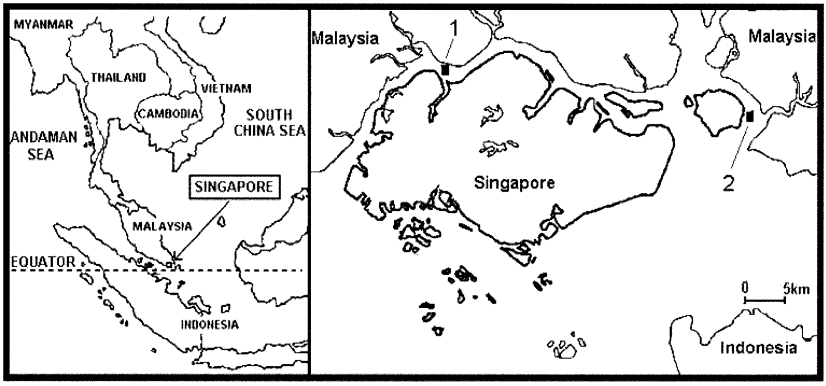

Figure 15.21. Sample stations.

example in Hong Kong (Tam and Yao, 2002; Zheng et al., 2000). Except for similarities between biota and sediments at S. Khatib Bongsu, the PCB congener profile was generally different for seawater, sediments and biota samples. *p*,*p*′-DDT was dominant in seawater and sediments, while the metabolite, *p*,*p*′-DDE, was generally dominant in biota samples. This difference may reflect an ability of the biomass to metabolize the parent DDT pesticide, still present in Singapore's marine environment. HCH concentrations in seawater samples were relatively high compared to other POPs, but were relatively low in biota. Biomagnification is not expected for contaminants with a K_{ow} lower than 5 and will only be significant for a $K_{ow} > 6.3$ (van der Oost et al., 2003). HCHs have a K_{ow} of 3.8 therefore have less propensity for bioaccumulation than PCBs and DDTs.

The dominant group of POPs in Singapore's mangrove biota was the PCBs, followed by the organochlorine pesticides DDT and Chlordane. PBDEs and HCHs were present at concentrations generally one or two orders of magnitude less than PCBs. This observation is consistent with the findings presented in a previous study using *P. viridis* as a bioindicator of marine contamination in Singapore (Bayen et al., 2003). Concentrations of POPs in food webs is related to the feeding habits and trophic level of organisms (Dietz et al., 2000; Kidd et al., 2001). The highest POPs concentrations were found in higher trophic status predator species (e.g., thunder crabs, drill shell, archer fish, chromide) and detritivores (e.g., rodong shell), while organisms at lower trophic levels (e.g., tube worm, nerite, prawns) had, in general, the lowest levels of POPs. In particular, the predator thunder crab (*M. hardwicki*) had higher tissue levels

of POPs than found in leaf-eater crabs (*Episesarma* spp.), although organisms were collected from the same habitat. A similar observation is valid when comparing the predator polychaete, *Neanthes glandicincta*, and the filter-feeder tube worm (*Diopatra neapolitana*). Differences in POP concentrations between the invertebrates and fish were generally one to two orders of magnitude, which is consistent with other studies (Dietz et al., 2000; Boon et al., 2002). Major biomagnification stage is usually associated between the trophic level of fish and mammals.

Comparison of PBDE levels with other studies is more straightforward by limiting the comparison to the single congener always present in the highest concentration, i.e., BDE-47 (Boon et al., 2002; Voorspoels et al., 2003). The levels of BDE-47 detected in this study (BLD-5.9 ng g^{-1} ww for all mangrove samples; 1.8–69 ng g^{-1} lw in fish livers) cover the middle range of data reported from Europe, Japan and Canada (de Wit, 2002; Law et al., 2003; Voorspoels et al., 2003). Levels of BDE-47 in mangrove fish muscles were higher or in the range of levels recorded for tuna muscle tissue in the China Sea (Ueno et al., 2004). The prevalence of BDE-47 in the PBDE profile, and the relative percentages of other PBDE congeners, are consistent with previous studies (Boon et al., 2002; Voorspoels et al., 2003). BDE-47 was present in lower proportions in the liver of mangrove fish compared to muscle tissue, which is consistent with a previous report on marine fish in Belgium (Voorspoels et al., 2003), but no clear explanation is available. Higher proportions of BDE-99 were present in the PBDE profile of organisms at lower trophic levels in S. Khatib Bongsu (i.e., polychaetes, oyster, rodong shell, marine prawn, tree-climbing crab). In a previous study using *P. viridis* as a bioindicator (Bayen et al., 2003), BDE-99 levels in green mussel tissues were reported as greater than those for BDE-47 in samples collected in Punggol, i.e., near S. Khatib Bongsu, but not mussels collected in S. Buloh. The source of PBDE contamination observed in Punggol is also likely to impact the mangrove at S. Khatib Bongsu. As a result, organisms at low trophic levels in S. Khatib Bongsu (polychaetes, oysters, rodong shell, marine prawn and tree-climbing crab), with a high uptake and/or poor metabolism capability of PBDEs, will also present higher levels of BDE-99. Organisms at higher trophic levels (e.g., thunder crab, fish) may have lower loads of BDE-99 as a result of different uptake/metabolic processes. Gustafsson et al. (1999) showed that the uptake of BDE-47 and BDE-99 were virtually identical for the mussel *M. edulis*. However, Stapleton et al. (2004) demonstrated, in a controlled experiment, that a rapid assimilation of BDE-47 occurred in juvenile carps, whereas no assimilation of BDE-99 was observed. Such differences are likely to explain contrasting PBDE patterns observed in the mangrove organisms of S. Khatib Bongsu.

PCBs concentrations found in shrimps and fish in the mangroves of Singapore are similar to those reported in Hong Kong (Liang et al., 1999). PCB concentrations in mangrove fish correspond to the upper range of what has been recorded in various species of fish elsewhere in the Asia-Pacific region (UNEP, 2002). The PCB profile was generally dominated by penta and hexachlorinated biphenyls, matching the profile of the commercial product Aroclor 1254, one of the main components of Askarel, the common PCB-containing product previously used in transformers and capacitors (Erickson, 1992).

Concentrations of Chlordanes (sum of α- and γ-) are higher than reported values for other marine food webs in the Baltic Sea (Falandysz et al., 2001). Chlordane concentrations in mangrove fish correspond to the upper range of what has been recorded in various species of fish elsewhere in the Asia-Pacific region (UNEP, 2002). The Chlordane profile was dominated by α- and γ-Chlordane in all samples except for crabs and gobies. Falandysz et al. (2001) also noted differences in Chlordane levels between crabs and other marine organisms which were attributed to the potency of crabs to metabolize Chlordane into Oxychlordane.

DDT concentrations in mangrove fish correspond to the middle range of what has been recorded in various species of fish elsewhere in the Asia-Pacific region (UNEP, 2002). *p*,*p*′-DDE was the dominant DDT congener in mangrove biota, suggesting that no input of DDT has occurred to these ecosystems recently.

Many biological and environmental parameters affect the uptake of contaminants in marine biota. Therefore, a simple interpretation of pollutant data for a large number of organisms is not readily feasible when comparing the two mangrove sites studied. For example, organisms at S. Khatib Bongsu were generally smaller in size than those at S. Buloh. As POP concentrations are known to increase with organism size in some species, such as fish (Liang et al., 1999; Makarevicz et al., 2003), the comparison of the two sites may include a size-related bias. However, mangrove organisms collected in S. Khatib Bongsu generally have higher levels of PCBs and PBDE. This observation is consistent with the findings of previous studies (Bayen et al., 2003) where *P. viridis* was used as a bioindicator, where higher concentrations of PCBs and PBDEs were recorded in the organisms collected in the east Strait of Johore relative to those in the west.

Levels of POPs in marine mammals and birds liver are generally one to three orders of magnitudes higher than in fish liver which, in turn, are four orders of magnitude lower than human adipose tissues (Liang et al., 1999; Dietz et al., 2000). Mammals, such as smooth-coated otters (*Lutrogale perspicillata*), and over 100 species of birds have been recorded in the

mangroves in Singapore (Ng and Sivasothi, 1999). If we use this species richness as a guide for mangrove food webs in Singapore, POPs levels in the μg g^{-1} ww range can be predicted in bird and mammal livers. The analysis of POPs in the eggs of fish-eating birds in Hong Kong, such as heron and egrets, revealed toxicological risks to breeding success (Connell et al., 2003). As levels in the present studies are comparable to those in Hong Kong, it is recommended that the risk for organisms at higher trophic levels in the mangroves of Singapore be evaluated.

As noted in Table 15.12 VI-1, at least 10 species of organisms analysed in the present study are commonly consumed by humans in Southeast Asia. Concentrations of POPs did not exceed the food safety standards of Singapore (Government of Singapore, 1990). However, PCB concentrations in mangrove fish muscles are also higher than the mean levels of PCBs in seafood commonly consumed in Singapore, which was found to increase the cancer incidence risk over a human lifetime (Bayen et al., 2004). Therefore, consumption of organisms from mangroves of Singapore should be considered carefully.

The study confirms the ubiquity of POPs, including PBDEs, in the marine environment of Singapore. A biomagnification phenomenon was observed amongst the species collected and analysed from both mangrove sites studied. Overall, available data on the prevalence, fate and transfer of POPS highlights the ubiquity of these pollutants in Singapore's coastal marine environment, and supports the need for a greater awareness of POP bioaccumulation and toxicity, particularly for organisms cultured locally and destined for human consumption.

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