

Atmospheric Deposition and Fluxes of Organochlorine Pesticides and Coplanar Polychlorinated Biphenyls in Aquatic Environments of Hong Kong, China

HIU LAM WONG,[†] JOHN P. GIESY,^{†,‡} AND PAUL KWAN SING LAM^{*†}

Department of Biology and Chemistry, City University of Hong Kong, 83 Tat Chee Avenue, Kowloon, Hong Kong, and Department of Zoology, National Food Safety and Toxicology Center, and Center for Integrative Toxicology, Michigan State University, East Lansing, Michigan 48824-1311

Concentrations of organochlorine (OC) pesticides and coplanar (dioxin-like) polychlorinated biphenyls (PCBs) in bulk deposition were measured at the Mai Po Marshes Nature Reserve (MPMNR) and A Chau, which are both important habitats for waterbirds in Hong Kong. OC pesticides exempted from the Stockholm Convention were present in greater concentrations than those that have been restricted for use in the region. Among the OC pesticides, HCB, Σ DDTs, and Σ HCHs were detected. Concentrations of HCB were greater at MPMNR than at A Chau, and this finding suggests short-range transport of this compound, which is different from the other OC pesticides. Several environmental factors including seasonal variations in temperature, particulate matter, and rainfall may influence the flux of OC pesticides. Since sources of HCB often coexist with sources of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (dioxins and furans), the presence of HCB may be a useful surrogate for monitoring airborne dioxins and for understanding their deposition potential. The contribution of atmospheric deposition to the OC pesticide input to the two study sites was small. Concentrations of most OC pesticides in surface waters were greater than would be predicted based on the inputs from atmospheric deposition and sedimentation. The mass balance calculation suggests a net flux of OC pesticides from bottom sediments to the overlying water column.

Introduction

By 2002, the manufacture and use of nine common persistent organic pollutants (POPs) had been totally banned in China following the Stockholm Convention. Residues of these POPs are, however, still present in the environment. The atmosphere remains a potential source for persistent, semivolatile organic compounds (SVOC) (1). Net fluxes of chemicals in the environment are determined by the relative rates of deposition (wet and dry), sedimentation, volatilization, and washout in rainfall (2). Atmospheric deposition of POPs has

been well-characterized in the North American Great Lakes (3), but less information is available for Asian environments, particularly Hong Kong. Since rain washout plays an important role in removing POPs present in the atmosphere, the unique climate of Hong Kong with alternating wet and dry seasons results in significant seasonal variations in deposition rates. Recently, OCs (e.g., hexachlorocyclohexanes (HCHs), dichlorodiphenyltrichloroethanes (Σ DDTs), hexachlorobenzene (HCB), polychlorinated biphenyls (PCBs), and dioxins) were detected in the local air of Hong Kong (4, 5), and PCBs, DDTs, and HCHs were detected in the air over the whole Pearl River Estuary (6).

Despite the small size of Hong Kong and the pressing demand for land resources and development, Hong Kong has a wetland of international importance, the Mai Po and Inner Deep Bay Ramsar site. The Mai Po Marshes Nature Reserve (MPMNR), occupying an important area for biodiversity conservation within the Ramsar site, was originally designated as a Site of Special Scientific Interest in 1976 and has been managed by the World Wide Fund for Nature Hong Kong since 1984. Despite its conservation value, the area is under severe pollution stress. The water quality of Deep Bay is deteriorating due to increasing pollution loadings from the Shenzhen River, Yuen Long-Kam Tin catchments, and the Pearl River (7, 8). A Chau is another important habitat for waterbirds situated in the northeastern part of Hong Kong and is relatively remote from the Pearl River Delta (Figure 1). POPs such as Σ DDTs, Σ HCHs, cyclodienes, PCBs, and dioxins and furans have been measured in MPMNR (9–11). There has, however, been no attempt to investigate the relative importance and interrelationship of various pollutants from different environmental compartments such as air, water, and sediment. Specifically, there have been very few atmospheric deposition measurements.

To develop an effective management plan for areas of high conservation value, it was necessary to make clear the magnitudes of fluxes of OC pesticides and other residues into and out of the aquatic environments over and above the OC pesticide concentrations in biota, seawater, and sediment. For this reason, a comprehensive study of the current status and sources of OC pesticides and dioxin-like PCBs was undertaken at MPMNR and A Chau. Specifically, fluxes from atmospheric deposition and re-emission from local sources were investigated to determine seasonal trends and influences of atmospheric phenomena on the cycling of important POPs.

Materials and Methods

Collection of Deposition Samples. Samples of bulk deposition were collected in stainless steel samplers with an inner diameter of 30 cm and a height of 30 cm. The samplers were set up such that the top edges were 120 cm above ground. The sampler at Mai Po was located outside the World Wildlife Fund for Nature (WWFHK) Education Centre, and the sampler at A Chau was placed at the Sha Tau Kok Fish Farm. Before sampling, 9 L of double-distilled, deionized water was added to each sampler to cover the bottom to act as a reservoir for the deposited particles to reduce the loss of particulates by wind in the open container over a prolonged sampling period. Copper sulfate was added to prevent algal growth (12). Water samples were collected monthly. Water samples were filtered through precleaned glass fiber filters (TOYO, GC-50) immediately upon return to the laboratory. Filters were weighed before and after the filtration, and differences in weight were recorded. Filters were then lyophilized for subsequent analysis. Meteorological information for Mai Po and A Chau were obtained from the closest automatic weather

* Corresponding author telephone: +852 2788 7681; fax: +852 2788 7406; e-mail: bhpksl@cityu.edu.hk

[†] City University of Hong Kong.

[‡] Michigan State University.

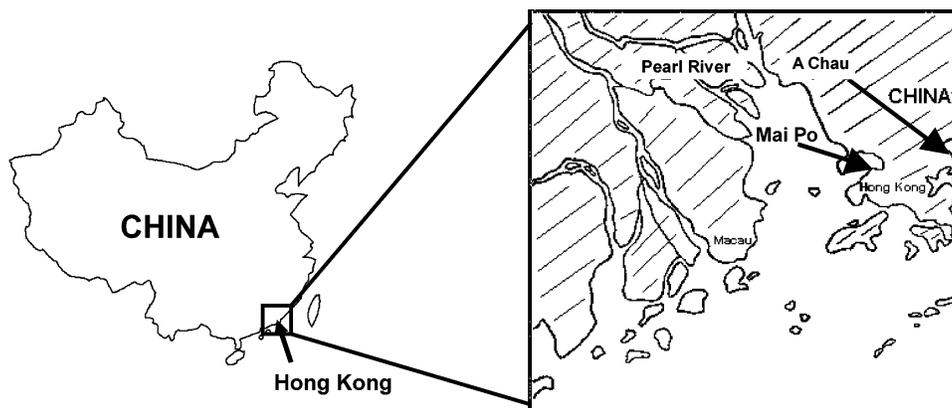


FIGURE 1. Map of the sampling sites: Mai Po and A Chau in Hong Kong.

TABLE 1. Predicted Sedimentation Rates at Selected Indicator Points in the Mai Po Marshes Nature Reserve^a

	sedimentation rate (mm/yr)	wt of sedimentation/yr (kg) in whole area (W_{sed})	suspended particles (mg/L)	av wt of suspended particles (kg) in whole area (W_{ss})	ratio for sedimentation/av wt of suspended particles (f')
Mai Po	16.2	1.2×10^8	56.95	6.5×10^6	17.92

^a Data from ref 16.

stations (<5 km) of the Hong Kong Observatory at Lau Fau Shan and Ta Kwu Ling, respectively.

Analysis of the Samples. OC pesticides were analyzed in the particulate and liquid fractions to determine the magnitude of total deposition. OC pesticides collected on filters were Soxhlet extracted with 400 mL of dichloromethane with 25% hexane for 16 h. The filtrates were extracted 20 times with 400 mL of dichloromethane. Volumes of the extracts were then reduced to 1 mL in a rotatory evaporator at 40 °C. Extracts were further fractionated by Florisil (Sigma, F9127) columns (13). Concentrations of OC pesticides and PCBs in the eluants were quantified using a Hewlett-Packard 5890 series II gas chromatograph (Palo Alto, CA) equipped with an electron capture detector (GC-ECD). A fused silica capillary column (60 m × 0.25 mm in diameter) coated with DB-5MS (J & W Scientific, Folsom, CA) at 0.25 μm film thickness was used. The column oven was programmed from an initial temperature of 100 °C with 3 min holding time to 210 °C at a rate of 25 °C/min and was then ramped at a rate of 1 °C/min to 270 °C, with a final hold time of 60 min. The injector and detector temperatures were maintained at 250 and 280 °C, respectively. Dioxin-like PCBs were analyzed by GC-ECD after acid cleanup. Again, a DB-5MS fused silica capillary column was used. The column oven, injector, and detector temperatures were the same as above. The total mass of *o,p'*- and *p,p'*-isomers of DDD, DDE, and DDT; HCB; α-, β-, and γ-HCHs; cyclodienes (aldrin, dieldrin, eldrin, endosulfans I and II, heptachlor, kepone, and mirex); and PCBs (IUPAC; 28, 37, 52, 71, 81, 105, 114, 118, 126, 156, 157, 167, and 189) were determined by summing of the mass of each OCs in the filtrate and the particulate fractions. Toxic equivalencies (TEQ_{WHO}) for various organisms developed by World Health Organization were reported in this study (14). Since the OCs in the particulates or dusts interacted with the water continuously over a prolonged period, the total combined mass of the OC residues in particulates and water was used. All the meteorological information for reference was obtained by on-site measurement or data from local monitoring authority (15).

Recovery tests were performed in solvent-washed, double-distilled, deionized water spiked with OC standards to a final concentration of 50 ng/L. Recoveries of OC pesticides in the particulates were 94.6 ± 3.57% (min: 80.3 ± 2.6% for mirex;

max: 105.4 ± 2.6% for γ-chlordane). Recoveries of dioxin-like PCBs were 96.5 ± 2.8%. Recoveries for filtrates were 65.3 ± 12.9% and 71.4 ± 6.3% for OC pesticides and dioxin-like PCBs, respectively. The method detection limits (MDL) for the particulates and the filtrates were 1 and 10 ng/L, respectively. Concentrations measured by instrumental analyses were not corrected for recoveries.

The total masses of residues in bulk deposition samples were used to determine fluxes, which were applied in mass balance calculations to estimate the exchange of OC pesticides and PCBs between the water and the atmosphere. Concentrations of OC pesticides and PCBs in rainfall, surface water, and bulk deposition were divided by the cross-section area of the sampler (m) and duration of sampling (d) (flux (mg/m²/d) = amount of OCs (mg)/cross section of the container (m²)/time (d)). The particulate fluxes were estimated from the standardized freeze-dried weight of the filtered precipitates.

Mass Balance Analysis. The 380-ha MPMNR with a relatively stable, known sedimentation rate was the focus of the mass balance study. This investigation uses a compartmentation model (16) to describe major material exchange in the study area based on measured concentrations of OCs in deposition, seawater, and sediment samples. The following parameters were obtained from a previous study (16): average depth of the marsh (*d*, m), area of the marsh (*A*, m²), average sedimentation rate (R_{sed} , mm/year), average concentration of suspended particles (SS, mg/L), and dry weight of sediment per m³ (W_{dw} , kg/m³). The average weight of suspended particles (g) at the site was calculated from an average concentration of suspended particles (mg/L) as listed in Table 1. Sedimentation (mass of sediment deposited on the top surface sediment layer) was estimated as the product of the sedimentation rate (mm/yr), the area of the mudflat, and the dry weight of the sediment. Concentrations of OC pesticides in water ($[X]_{water}$ (wet or dry season), ng/L) and in sediments ($[X]_{sediment}$ (wet or dry season), ng/g dw) in the wet and dry seasons were calculated (Table 2). Depositions were measured in the wet and dry seasons (flux($X_{(wet or dry season)}$), ng m⁻² wet or dry seasons⁻¹), and the weighted seasonal depositions were calculated based on daily depositions in the dry and wet season separately (eqs 1–10).

TABLE 2. OC Pesticides in Water (ng/L ± SD) and Sediment (ng/g dw ± SD) Samples from Mai Po (MP) and A Chau (AC) in the Dry (D) and Wet (W) Seasons^a

	total DDTs		total HCHs		HCB		total cyclodienes		dioxin equivalency (I-TEQ _{bird})	
	seawater	sediment	seawater	sediment	seawater	sediment	seawater	sediment	seawater	sediment
MP (D)	1.5 × 10 ⁻¹ ± 6.4 × 10 ⁻³	1.0 × 10 ¹ ± 8.3 × 10 ⁻¹	8.6 × 10 ⁻² ± 3.3 × 10 ⁻²	5.4 ± 4.0 × 10 ⁻¹	1.4 × 10 ⁻¹ ± 5.7 × 10 ⁻²	7.5 ± 3.8 × 10 ⁻¹	1.2 × 10 ⁻² ± 1.2 × 10 ⁻³	1.3 × 10 ¹ ± 1.2	1.8 × 10 ⁻¹ ± 2.3 × 10 ⁻²	1.8 × 10 ⁻² ± 2.3 × 10 ⁻²
AC (D)	2.0 × 10 ⁻¹ ± 2.0 × 10 ⁻²	1.4 ± 5.6 × 10 ⁻²	nd	4.1 × 10 ⁻¹ ± 5.4 × 10 ⁻²	1.2 × 10 ⁻¹ ± 1.5 × 10 ⁻²	1.5 ± 1.6 × 10 ⁻¹	1.4 × 10 ⁻¹ ± 5.8 × 10 ⁻²	8.0 × 10 ⁻² ± 5.4 × 10 ⁻²	1.6 × 10 ⁻¹ ± 1.0 × 10 ⁻³	1.6 × 10 ⁻² ± 1.0 × 10 ⁻³
MP (W)	1.3 × 10 ⁻¹ ± 4.3 × 10 ⁻²	2.9 × 10 ¹ ± 9.3 × 10 ⁻¹	8.9 × 10 ⁻² ± 6.7 × 10 ⁻²	9.0 ± 4.4 × 10 ⁻¹	4.7 × 10 ⁻² ± 3.0 × 10 ⁻²	1.5 ± 2.4 × 10 ⁻¹	3.9 × 10 ⁻² ± 2.0 × 10 ⁻²	1.9 × 10 ¹ ± 4.1 × 10 ⁻¹	2.0 × 10 ⁻³ ± 1.0 × 10 ⁻³	2.6 × 10 ⁻² ± 1.7 × 10 ⁻³
AC (W)	9.7 × 10 ⁻² ± 1.5 × 10 ⁻²	2.0 ± 2.6 × 10 ⁻¹	7.8 × 10 ⁻² ± 6.4 × 10 ⁻³	0.0 ± 0.0	6.5 × 10 ⁻² ± 4.7 × 10 ⁻²	3.3 ± 5.4 × 10 ⁻²	3.8 × 10 ⁻² ± 1.4 × 10 ⁻²	4.0 × 10 ⁻¹ ± 3.8 × 10 ⁻¹	nd	nd

^a The Mai Po sediments were collected from the mudflat of the outermost part of Mai Po Marshes Nature Reserve. PCB dioxin equivalencies were calculated based on the multiplication of concentration of dioxin-like PCB with the TEF for bird (pg TEQ_{WHO-bird}/g dw) (14). nd indicates samples with OC concentrations less than the method detection limits.

Thus, the following parameters were estimated based on data from ref 16:

volume of the marsh (V , m³):

$$V = d \times A \text{ (m}^3\text{)} \quad (1)$$

annual total suspended particulates marsh ($W_{t_{ss}}$, g):

$$W_{t_{ss}} = SS \times V/1000/0.1^3 \text{ (g)} \quad (2)$$

annual total weight of surface sediment from sedimentation (W_{ssed} , g):

$$W_{ssed} = A \times R_{sed} \times W_{dw} \text{ (g)} \quad (3)$$

Thus, the ratio (R) of sediment from sedimentation to the total suspended particle in the marshes:

$$R = W_{ssed}/W_{t_{ss}} \quad (4)$$

R can act as a conversion factor for the amount of residues in water to the amount of residues deposited by sedimentation. The sedimentation rate was assumed to be the same in both seasons, which was the same as the sedimentation rate of OC pesticides. Therefore, the half-year sedimentation rates were $R_{1/2}$.

Therefore, the average OC pesticide concentrations in the water column of the marshes (W_{sea-x} , g) in the wet or dry season were estimated from eq 5:

$$W_{sea-x(\text{wet or dry season})} = [X]_{\text{water(wet or dry season)}} \times V/0.001 \times 10^{-9} \text{ (g)} \quad (5)$$

Sedimentation of each OC pesticide from seawater (W_{w-s} , g) to the marshes in the wet or dry season was estimated from eq 6:

$$W_{w-s(\text{wet or dry season})} = R \times W_{sea-x(\text{wet or dry season})} \text{ (g)} \quad (6)$$

In this study, total concentrations of OC pesticides in surface sediments (W_{sed} , g) were calculated from the measured concentrations of OCs using eq 7:

$$W_{sed} = [X]_{\text{sedimentation(wet or dry season)}} \times A \times R_{1/2} \times 10^{-9} \text{ (g)} \quad (7)$$

Weighted seasonal deposition mass ($W_{dep(\text{wet or dry season})}$, g) was estimated by eq 8 with measured flux($x_{(\text{wet or dry season})}$) from the deposition sampler in this study:

$$W_{dep(\text{wet or dry season})} = \text{flux}(x_{(\text{wet or dry season})}) \times A \times 10^{-9} \text{ (g)} \quad (8)$$

Therefore, the ratios used to compare seasonal masses of deposition were calculated by eqs 9 and 10.

Air-sediment was calculated as the ratio of OC deposited from atmosphere to the total surface sediment mass ($R_{air-sediment}$):

$$R_{air-sediment} = W_{dep(\text{wet or dry season})}/W_{w-s(\text{wet or dry season})} \quad (9)$$

Water-sediment was calculated as the ratio of OC deposited from seawater to the total surface sediment mass ($R_{water-sediment}$):

$$R_{water-sediment} = W_{sea-x(\text{wet or dry season})}/W_{w-s(\text{wet or dry season})} \quad (10)$$

Results and Discussions

Weather Conditions. General weather conditions were similar between the two study sites, but some local events were different (Table 3). Temperatures were similar at the

TABLE 3. Meteorological Information during Periods of Bulk Deposition Sampling^a

sampling period	period (d)	pptn (mm)	av temp (°C)	prevailing wind direction	wt of pptn in deposition samples (g, dw)	monthly particulate flux (g m ⁻² month ⁻¹)
Mai Po						
8/1/02–8/30/02	2.9 × 10 ¹	3.0 × 10 ²	2.8 × 10 ¹ ± 1.0	8.0 × 10 ¹	0.400–0.4301	1.5 ± 7.5 × 10 ⁻²
8/30/02–9/29/02	3.0 × 10 ¹	3.0 × 10 ²	2.9 × 10 ¹ ± 2.2	8.0 × 10 ¹	0.3296–0.4401	1.4 ± 2.8 × 10 ⁻¹
9/29/02–10/26/02	2.7 × 10 ¹	8.6 × 10 ¹	2.5 × 10 ¹ ± 2.4	8.0 × 10 ¹	0.5020–0.5446	3.4 ± 2.8
10/26/02–11/26/02	3.0 × 10 ¹	2.5 × 10 ¹	2.1 × 10 ¹ ± 2.4	7.0 × 10 ¹	0.2122–0.2321	7.8 × 10 ⁻¹ ± 5.0 × 10 ⁻²
11/26/02–12/28/02	3.2 × 10 ¹	7.4 × 10 ¹	1.8 × 10 ¹ ± 4.5	8.0 × 10 ¹	0.3434–0.3468	1.2 ± 9.0 × 10 ⁻⁴
12/28/02–1/26/03	2.8 × 10 ¹	1.2 × 10 ¹	1.5 × 10 ¹ ± 2.7	8.0 × 10 ¹	0.3623–0.3776	1.3 ± 3.8 × 10 ⁻²
1/26/03–2/26/03	3.1 × 10 ¹	5.0	1.8 × 10 ¹ ± 2.8	8.0 × 10 ¹	0.2052–0.2182	7.5 × 10 ⁻¹ ± 3.3 × 10 ⁻²
2/26/03–3/27/03	2.9 × 10 ¹	3.8 × 10 ¹	1.9 × 10 ¹ ± 3.4	8.0 × 10 ¹	0.1991–0.2028	7.1 × 10 ⁻¹ ± 9.0 × 10 ⁻³
3/27/03–4/24/03	2.8 × 10 ¹	1.5 × 10 ¹	2.3 × 10 ¹ ± 2.5	8.0 × 10 ¹	0.1575–0.1855	6.1 × 10 ⁻¹ ± 7.0 × 10 ⁻²
4/24/03–5/27/03	3.3 × 10 ¹	2.5 × 10 ²	2.6 × 10 ¹ ± 3.6	1.4 × 10 ²	0.2392–0.2693	9.0 × 10 ⁻¹ ± 7.5 × 10 ⁻²
5/27/03–6/25/03	2.9 × 10 ¹	3.3 × 10 ²	2.7 × 10 ¹ ± 1.4	1.4 × 10 ²	0.1953–0.2012	7.0 × 10 ⁻¹ ± 1.5 × 10 ⁻²
6/25/03–7/23/03	2.8 × 10 ¹	8.9 × 10 ¹	2.9 × 10 ¹ ± 0.6	1.4 × 10 ²	0.3865–0.5226	1.6 ± 3.4 × 10 ⁻¹
A Chau						
8/2/02–8/31/02	2.9 × 10 ¹	4.2 × 10 ²	2.8 × 10 ¹ ± 1.0	1.0 × 10 ²	0.1005–0.1038	3.6 × 10 ⁻¹ ± 8.0 × 10 ⁻²
8/31/02–9/30/02	3.0 × 10 ¹	5.2 × 10 ²	2.6 × 10 ¹ ± 1.2	1.0 × 10 ²	0.0840–0.1003	2.9 × 10 ⁻¹ ± 9.1 × 10 ⁻²
9/30/02–10/28/02	2.7 × 10 ¹	4.3 × 10 ¹	2.4 × 10 ¹ ± 2.3	1.1 × 10 ²	0.0311–0.0802	2.0 × 10 ⁻¹ ± 1.2 × 10 ⁻¹
10/28/02–11/28/02	3.0 × 10 ¹	3.2 × 10 ¹	2.0 × 10 ¹ ± 2.5	1.0 × 10 ¹	0.0895–0.1023	3.4 × 10 ⁻¹ ± 3.2 × 10 ⁻²
11/28/02–12/28/02	3.2 × 10 ¹	5.4 × 10 ¹	1.7 × 10 ¹ ± 4.8	1.1 × 10 ²	0.1003–0.1009	3.6 × 10 ⁻¹ ± 2.0 × 10 ⁻³
12/28/02–1/28/03	2.8 × 10 ¹	1.9 × 10 ¹	1.4 × 10 ¹ ± 2.7	1.1 × 10 ²	0.1001–0.0997	3.5 × 10 ⁻¹ ± 1.0 × 10 ⁻²
1/28/03–2/28/03	3.1 × 10 ¹	5.5	1.7 × 10 ¹ ± 2.9	1.1 × 10 ²	0.0854–0.0911	3.1 × 10 ⁻¹ ± 1.4 × 10 ⁻²
2/28/03–3/28/03	2.9 × 10 ¹	3.5 × 10 ¹	1.8 × 10 ¹ ± 3.4	1.1 × 10 ²	0.0891–0.0903	3.2 × 10 ⁻¹ ± 3.0 × 10 ⁻³
3/28/03–4/28/03	2.8 × 10 ¹	2.1 × 10 ¹	2.3 × 10 ¹ ± 2.7	1.1 × 10 ²	0.0792–0.0942	3.1 × 10 ⁻¹ ± 3.8 × 10 ⁻¹
4/28/03–5/29/03	3.3 × 10 ¹	4.2 × 10 ²	2.6 × 10 ¹ ± 1.4	1.1 × 10 ²	0.048–0.053	1.8 × 10 ⁻¹ ± 1.3 × 10 ⁻¹
5/29/03–6/27/03	2.9 × 10 ¹	4.1 × 10 ²	2.7 × 10 ¹ ± 1.2	1.1 × 10 ²	0.0921–0.0110	1.8 × 10 ⁻¹ ± 2.0 × 10 ⁻¹
6/27/03–7/25/03	2.8 × 10 ¹	1.2 × 10 ²	2.9 × 10 ¹ ± 0.7	1.1 × 10 ²	0.0231–0.0245	8.4 × 10 ⁻² ± 4.0 × 10 ⁻³

^a All samples were collected between 0930 and 1030 h at both sites.

two sites: highest in June and lowest in January. Significant seasonal differences in rainfall and mass of the particulate matter were observed at both sites. More rainfall was recorded at A Chau (417–522 mm) than at Mai Po (252–298 mm) in the wet season, but the amount of rainfall was similar at both locations during the dry season. Deposited mass was greater at Mai Po than A Chau, which may be due to differences in rainfall and/or the source of airborne particulates. The prevailing wind direction at Mai Po was different between the wet and dry seasons. At Mai Po, the wind was predominantly ESE (140°) during the wet season, while it was NNE (80°) during the dry season. At A Chau, the wind was primarily from the same direction ESE (100°) in all seasons (15).

Spatial and Seasonal Differences in OC Fluxes. Concentrations of HCB, DDTs and HCHs were generally greater in deposition samples from Mai Po than A Chau, and concentrations observed were independent of the weather conditions in both sites (Table 4). The magnitude and pattern of the deposition flux of each OC pesticide varied throughout the year at Mai Po. No significant seasonal differences for OC deposition, except for the greater HCB and HCH depositions in the dry season at Mai Po and in the wet season at A Chau, respectively. Except for HCB, deposition fluxes of most OC pesticides were constant throughout the year (nd–2.5 ng m⁻² d⁻¹). Lesser concentrations of OC pesticides in deposition at A Chau were found, with most of the deposition occurring between February and June, except for *o,p'*-DDE, *o,p'*-DDT, and HCB. Correlations between loadings of OCs and environmental factors such as rainfall were poor (multiple regression: *p* > 0.05 for all factors), suggesting that none of these parameters were the primary controlling factors for the deposition rates. This was not unexpected because atmospheric deposition flux of OCs is a complicated process (17).

Relatively small OC depositions were found in the study sites when compared with similar investigations elsewhere. Fluxes of OC pesticides at both sites were less than those

reported from the North American Great Lakes, except for HCB and chlordanes (Table 5). At Mai Po, deposition fluxes of chlordanes were occasionally greater than those of the Great Lakes. Since HCB, DDTs, and HCHs were the dominant OC pesticides in this study, these compounds will be discussed separately from cyclodienes and dioxin-like PCBs.

HCB. HCB was the dominant OC in atmospheric deposition at both sites throughout the year. This is consistent with the results of a recent study on OC concentrations in ambient air in Hong Kong (0.046–0.232 ng m⁻³ d⁻¹) (5). HCB accounted for 60–80% of the atmospheric deposition fluxes at Mai Po. During both the wet and dry seasons, deposition fluxes of HCB were 4-fold greater (Mann–Whitney test, *p* < 0.05) at Mai Po than at A Chau (Table 6). Since there is relatively little industrial activity near A Chau, the concentrations observed are likely to be indicative of the baseline concentration of HCB in the local environment due to long-range atmospheric transport. HCHs and DDTs were also found in the deposition samples but at concentrations that were less than those of HCB. No significant differences in HCHs and DDTs were observed between the two sites during both seasons.

The relatively great proportion of HCB as compared to other OC pesticides or dioxin-like PCBs is likely due to the physical properties of HCB and the environmental input in both areas. HCB concentrations in atmospheric deposition were greater during the dry season than the wet season (Mann–Whitney test, *p* < 0.05) at Mai Po, with seasonal fluxes of 9.4 × 10¹ and 5.3 × 10¹ ng m⁻² season⁻¹, respectively. These differences in daily fluxes were likely due to differences in rainfall and the amount of suspended particulate matters in air and/or dry atmospheric deposition. The pattern of the daily flux from atmospheric deposition was inversely proportional to the daily flux of dried precipitate in the sampler during the period between August 2002 and July 2003 (Figure 2). HCB has a relatively long half-life of 6 yr in water, soil, and sediment and 2 yr in air, which is longer than the half-lives of most other OC pesticides (except DDEs), ranging

TABLE 4. Daily Deposition Fluxes ($\text{ng m}^{-2} \text{d}^{-1}$) of OC Pesticides at Mai Po and A Chau from August 2002 to July 2003^a

	Section A: Mai Po									
	α -chlordane	γ -chlordane	<i>o,p'</i> -DDD	<i>p,p'</i> -DDD	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	endosulfan I	α -HCH	γ -HCH	HCB
Aug	nd	nd	nd	nd	nd	$1.5 \pm 2.1 \times 10^{-1}$	$6.0 \times 10^{-1} \pm 8.0 \times 10^{-2}$	nd	nd	$3.0 \pm 6.2 \times 10^{-1}$
Sept	7.6 ± 2.4	nd	$1.8 \pm 5.7 \times 10^{-1}$	nd	$1.7 \pm 4.2 \times 10^{-1}$	2.6 ± 1.1	nd	1.1 ± 1.6	$6.8 \times 10^{-1} \pm 5.3 \times 10^{-1}$	$2.0 \times 10^1 \pm 9.5$
Oct	nd	nd	nd	nd	nd	$2.6 \pm 2.9 \times 10^{-1}$	$5.4 \times 10^{-1} \pm 3.8 \times 10^{-1}$	nd	nd	$1.4 \times 10^1 \pm 5.7$
Nov	$3.0 \times 10^{-1} \pm 4.2 \times 10^{-1}$	nd	nd	nd	$9.2 \times 10^{-1} \pm 2.1 \times 10^{-2}$	$1.8 \pm 2.1 \times 10^{-1}$	$6.1 \times 10^{-1} \pm 1.0 \times 10^{-2}$	$2.7 \times 10^{-1} \pm 1.3 \times 10^{-1}$	$3.8 \times 10^{-1} \pm 5.3 \times 10^{-1}$	$2.2 \times 10^1 \pm 3.6$
Dec	$3.2 \times 10^{-1} \pm 4.5 \times 10^{-1}$	nd	nd	nd	$7.7 \times 10^{-1} \pm 2.7 \times 10^{-1}$	$1.7 \pm 5.7 \times 10^{-1}$	$6.6 \times 10^{-1} \pm 2.0 \times 10^{-2}$	nd	$3.2 \times 10^{-1} \pm 9.0 \times 10^{-2}$	$2.6 \times 10^1 \pm 2.4$
Jan	nd	nd	1.8 ± 2.6	nd	$5.9 \times 10^{-1} \pm 4.4 \times 10^{-1}$	2.0 ± 2.3	$1.7 \pm 2.0 \times 10^{-2}$	nd	nd	9.7 ± 6.1
Feb	nd	nd	$3.4 \times 10^{-1} \pm 4.4 \times 10^{-2}$	$3.6 \times 10^{-1} \pm 6.6 \times 10^{-2}$	$3.6 \times 10^{-1} \pm 6.6 \times 10^{-2}$	$9.3 \times 10^{-1} \pm 4.4 \times 10^{-1}$	$1.1 \pm 5.9 \times 10^{-1}$	$9.8 \times 10^{-1} \pm 2.2 \times 10^{-2}$	nd	$1.1 \times 10^1 \pm 1.3 \times 10^1$
Mar	nd	nd	nd	nd	$5.8 \times 10^{-1} \pm 8.2 \times 10^{-1}$	1.2 ± 1.6	nd	$6.5 \times 10^{-1} \pm 5.1 \times 10^{-1}$	$3.0 \times 10^{-1} \pm 2.0 \times 10^{-2}$	5.2 ± 1.2
Apr	nd	$9.5 \times 10^{-1} \pm 2.4 \times 10^{-1}$	nd	nd	$5.7 \times 10^{-1} \pm 2.0 \times 10^{-2}$	$9.1 \times 10^{-1} \pm 3.0 \times 10^{-1}$	$4.0 \times 10^{-1} \pm 4.9 \times 10^{-1}$	$1.7 \pm 2.0 \times 10^{-1}$	nd	3.2 ± 1.5
May	nd	nd	nd	nd	nd	$2.3 \pm 4.7 \times 10^{-1}$	nd	nd	nd	$1.1 \times 10^1 \pm 4.7 \times 10^{-1}$
June	nd	$6.4 \times 10^{-1} \pm 4.1 \times 10^{-2}$	nd	nd	$1.5 \pm 6.2 \times 10^{-1}$	$2.5 \pm 2.1 \times 10^{-1}$	nd	$1.0 \pm 6.2 \times 10^{-1}$	nd	$3.8 \pm 5.1 \times 10^{-1}$
July	nd	nd	$5.3 \times 10^{-1} \pm 7.9 \times 10^{-2}$	$3.1 \times 10^{-1} \pm 4.0 \times 10^{-2}$	$5.7 \times 10^{-1} \pm 2.0 \times 10^{-2}$	$1.4 \pm 4.0 \times 10^{-1}$	nd	$2.5 \pm 4.0 \times 10^{-1}$	nd	$1.2 \times 10^1 \pm 9.9 \times 10^{-1}$

	Section B: A Chau							
	<i>o,p'</i> -DDE	<i>p,p'</i> -DDE	<i>o,p'</i> -DDT	endosulfan I	endosulfan II	α -HCH	γ -HCH	HCB
Aug	nd	$1.1 \times 10^{-2} \pm 2.1 \times 10^{-3}$	nd	nd	nd	$4.5 \times 10^{-2} \pm 2.1 \times 10^{-2}$	nd	$1.8 \times 10^{-1} \pm 1.0 \times 10^{-3}$
Sept	nd	$9.3 \times 10^{-2} \pm 3.2 \times 10^{-2}$	nd	nd	nd	$9.5 \times 10^{-2} \pm 4.9 \times 10^{-2}$	nd	$1.2 \times 10^{-1} \pm 7.1 \times 10^{-2}$
Oct	nd	$1.2 \times 10^{-1} \pm 3.2 \times 10^{-2}$	nd	$3.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	nd	nd	nd	$8.3 \times 10^{-2} \pm 2.5 \times 10^{-2}$
Nov	nd	$7.2 \times 10^{-2} \pm 1.4 \times 10^{-3}$						
Dec	nd	nd	nd	$1.1 \times 10^{-2} \pm 1.4 \times 10^{-3}$	nd	nd	nd	$7.6 \times 10^{-2} \pm 9.1 \times 10^{-2}$
Jan	nd	$2.1 \times 10^{-2} \pm 1.6 \times 10^{-2}$	nd	$2.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	$3.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	$2.3 \times 10^{-2} \pm 1.8 \times 10^{-2}$	nd	$1.1 \times 10^{-1} \pm 7.1 \times 10^{-2}$
Feb	nd	$3.0 \times 10^{-2} \pm 7.1 \times 10^{-4}$	nd	$1.4 \times 10^{-2} \pm 6.4 \times 10^{-3}$	nd	nd	nd	$5.5 \times 10^{-2} \pm 2.1 \times 10^{-2}$
Mar	$5.8 \times 10^{-2} \pm 1.8 \times 10^{-2}$	$5.3 \times 10^{-2} \pm 2.5 \times 10^{-2}$	$1.1 \times 10^{-1} \pm 1.1 \times 10^{-1}$	$3.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	nd	$5.0 \times 10^{-2} \pm 4.2 \times 10^{-2}$	$8.0 \times 10^{-2} \pm 2.8 \times 10^{-2}$	$1.3 \times 10^{-1} \pm 1.1 \times 10^{-2}$
Apr	$2.2 \times 10^{-2} \pm 1.4 \times 10^{-3}$	$1.9 \times 10^{-1} \pm 4.9 \times 10^{-2}$	nd	$5.3 \times 10^{-2} \pm 1.8 \times 10^{-2}$	nd	$8.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	$2.0 \times 10^{-2} \pm 1.4 \times 10^{-2}$	$1.1 \times 10^{-1} \pm 3.5 \times 10^{-3}$
May	$4.0 \times 10^{-2} \pm 2.8 \times 10^{-2}$	$1.0 \times 10^{-1} \pm 2 \times 10^{-2}$	nd	$4.3 \times 10^{-2} \pm 1.8 \times 10^{-2}$	nd	nd	$5.5 \times 10^{-2} \pm 4.9 \times 10^{-2}$	$1.1 \times 10^{-1} \pm 7.1 \times 10^{-3}$
June	nd	$3.0 \times 10^{-2} \pm 2.8 \times 10^{-2}$	nd	nd	nd	$1.5 \times 10^{-2} \pm 2.1 \times 10^{-2}$	nd	$2.1 \times 10^{-2} \pm 7.1 \times 10^{-4}$
July	nd	$5.0 \times 10^{-2} \pm 4.2 \times 10^{-2}$	nd	nd	nd	$7.5 \times 10^{-2} \pm 3.5 \times 10^{-2}$	nd	$7.0 \times 10^{-2} \pm 3.5 \times 10^{-2}$

^a Only detectable OCs were included in this table; the others were below the detection limit throughout the sampling period. nd indicates samples with OC concentrations below detection limit.

TABLE 5. Daily Atmospheric Deposition Fluxes (ng m⁻² d⁻¹) in Mai Po and A Chau Compared with the Great Lakes (3)^a

Great Lake	daily flux (bulk deposition ng m ⁻² d ⁻¹)				
	HCHs	HCB	DDTs	endosulfan I and II	chlordanes
Superior	0.698	0.02	0.312	0.28	0.238
Michigan	1.542	0.039	0.55	0.91	0.813
Huron	7.6	0.043	0.68	1.5	0.237
Erie	0.289	0.024	0.309	0.065	0.664
Ontario	7.9	0.08	1.03	4	0.25
Mai Po	nd-0.060	0.105-0.675	0.05-0.202	nd-0.060	nd-0.253
A Chau	nd-0.130	0.06-0.180	nd-0.215	nd-0.105	nd

^a nd indicates samples with OC concentrations below detection limit.

TABLE 6. Season-Weighted Deposition Flux ± SD (ng m⁻² season⁻¹) of OC Pesticides at Mai Po and A Chau^a

	DDTs				HCHs		HCB	endosulfan I and II
	total	DDTs (%)	DDD	DDEs	total	α/γ		
Mai Po								
dry	1.6 × 10 ¹ ± 2.5	nd	15.8(%)	84.2(%)	2.9 ± 1.3	1.9	9.4 × 10 ¹ ± 1.1 × 10 ¹	4.6 ± 1.0
wet	1.8 × 10 ¹ ± 1.6	nd	14.9(%)	85.1(%)	7.0 ± 3.3	9.4	5.3 × 10 ¹ ± 1.4 × 10 ¹	9.3 × 10 ⁻¹ ± 4.0 × 10 ⁻¹
A Chau								
dry	1.1 × 10 ¹ ± 4.1	27.9	15.3(%)	56.8(%)	4.4 ± 2.5	0.90	1.6 × 10 ¹ ± 4.5	4.6 ± 3.1
wet	1.6 × 10 ¹ ± 1.9	nd	12.3(%)	87.7(%)	1.1 × 10 ¹ ± 2.9	3.76	1.8 × 10 ¹ ± 3.4	2.1 ± 3.6

^a nd indicates samples with OC concentrations below detection limit.

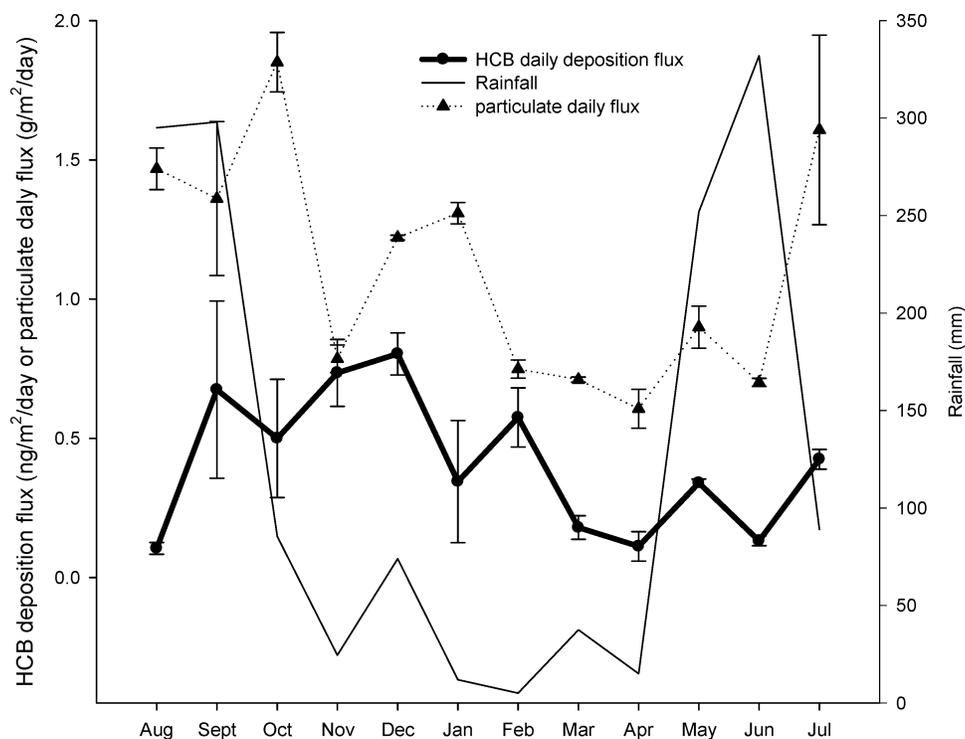


FIGURE 2. Relationship between daily deposition flux of HCB, rainfall, and particulate daily flux in the deposition samples at Mai Po.

from a few days to a few months in air (18). Since HCB is comparatively resistant to photooxidation, it is relatively stable in the sampler, even under direct sunlight. Furthermore, HCB has a high affinity toward organic matter and, therefore, can be adsorbed onto suspended particulate matters, soil, etc. Thus, the fluxes between soil and air are relatively great, especially under dry and windy conditions, such as those that occur during the dry season in Hong Kong (5). Rainfall reduces the amount of airborne suspended particulate matters and transport distance and, therefore, effectively reduces deposition fluxes. Alternatively these

factors might also explain the small flux of HCB from atmospheric deposition at A Chau, where the flux of particulates from atmospheric deposition was small. China has listed HCB as an exemption from the Stockholm Convention for use as an intermediate in the production of chlorinated organic solvents such as tetrachloroethylene (PCE), trichloroethylene (TCE), and carbon tetrachloride (19). HCB can also be produced as a byproduct during waste incineration, chlorination, and metallurgical processes, which are known to generate airborne HCB and other dioxin/furan-like compounds with ashes. Thus, there is still a continuous

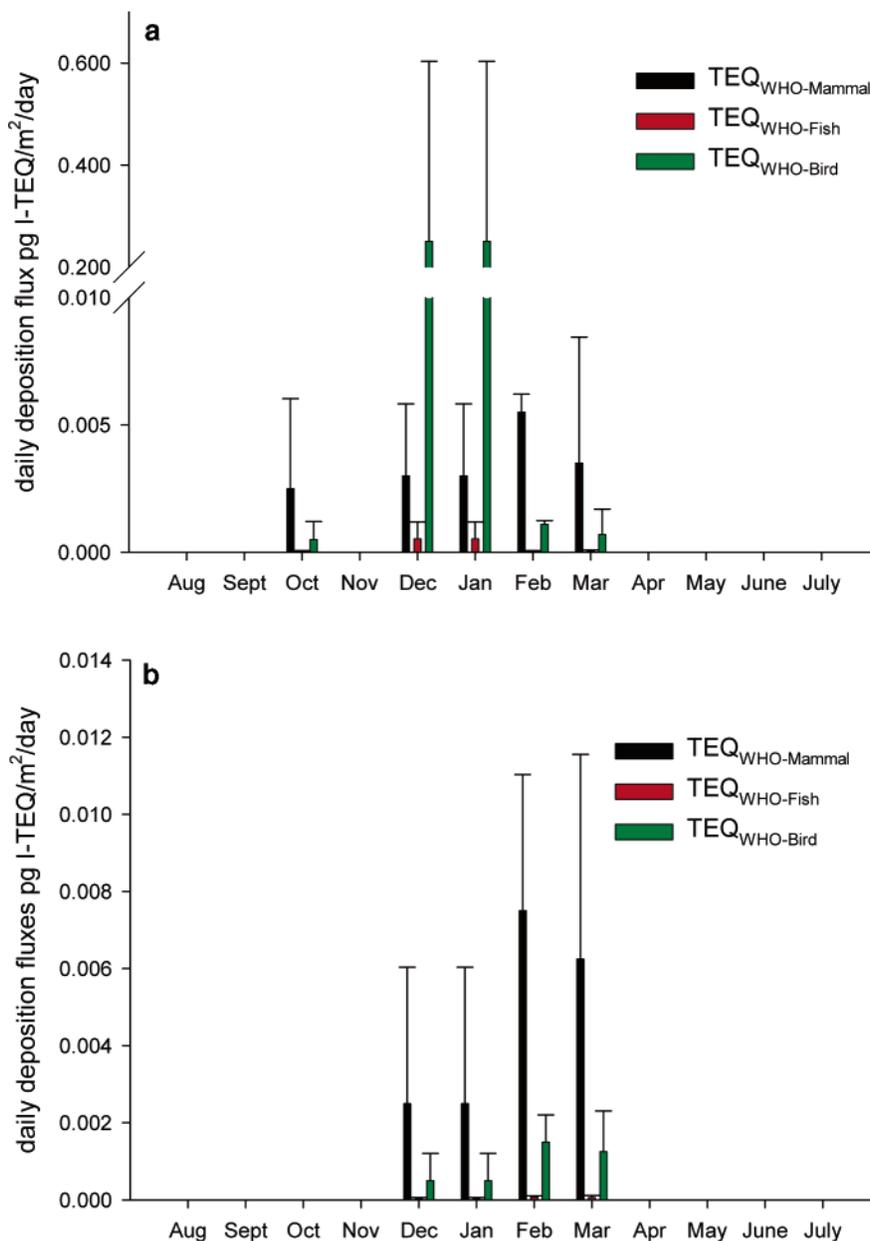


FIGURE 3. Daily deposition fluxes of PCBs, expressed as toxic equivalency (TEF_{WHO}) based on mammals, birds, and fish. (a) Mai Po. (b) A Chau.

input of HCB into the environment in China, especially in industrial areas along the Pearl River Estuary. A previous study has identified the correlation of HCB and dioxin concentration in air (20), suggesting that many sources of HCB are also the source for dioxin compounds. It might be instructive to monitor airborne dioxin concentrations in areas with great HCB deposition rates.

DDTs. In general, less than one-fifth of the OCs in atmospheric deposition were Σ DDTs, with DDEs as the predominant metabolites, from both sites in both seasons, which were smaller than the data from the Great Lakes (Table 5). The ratio of Σ DDTs to their metabolites was different between the two study sites (Table 6). At A Chau approximately 30% of Σ DDTs was *p,p'*-DDT, but DDTs were not detected in atmospheric deposition at Mai Po. Relatively great concentrations of *p,p'*-DDT (3.4–825 ng/L in seawater and >0.01–19.5 ng/g in sediment) have been reported to occur in some areas of Daya Bay, which is close to A Chau (21). The nearby farming areas might be a potential source of fresh DDTs to this remote area. Fresh DDTs were not

found in Mai Po deposition samples, and this might be related not to the absence of farmlands but to industrial areas nearby. Most of these values were less than those observed in other urban areas such as Galveston Bay, TX ($5.3 \text{ ng m}^{-2} \text{ d}^{-1}$) (22) and $0.15\text{--}0.46 \text{ ng m}^{-2} \text{ d}^{-1}$ in Niigata, Japan, except in February, July, and November (23).

HCHs. Relatively small depositions of HCHs, mainly technical mixtures, were found to be dominant in the wet season in both areas. This phenomenon might be related to the application pattern of these pesticides in the area. Higher ratios of α/γ -HCH isomers were found at both sites during the wet season, in comparison with those reported in the literature (23). This result is different from that observed in Northern Europe, where a lower ratio of α/γ -HCH isomers were reported in the summer (24). Despite a ban on agricultural applications in China in 1983, the seasonal fluctuations of HCHs and the higher α/γ -HCH ratios suggest that small-scale applications of technical mixtures may still be occurring or that there is substantial remobilization of HCHs from a contaminated location. This supposition is

TABLE 7. Estimation of Ratio of OCs from Atmospheric Deposition and Sedimentation on a Seasonal Basis

			HCB	total DDTs	total HCHs
dry season	atmospheric deposition	deposition rate (ng m ⁻² yr)	93.990	15.920	2.890
		deposition amt (g/yr) (<i>W_{dep}</i>)	0.357	0.060	0.011
wet season	atmospheric deposition	deposition rate (ng m ⁻² yr ⁻¹)	53.410	18.020	7.020
		deposition amt (g/yr)	0.203	0.068	0.027
dry season	estimation by [OC] in water	seawater concn (ng/L) (<i>W_{s-x}</i>)	0.165	0.147	0.086
		OC amt deposited by sea (g/yr)	16.804	15.030	8.828
		ratio of atmos deposition/sedimentation from water	0.021	0.004	0.001
	estimation by [OC] in sediment	sediment concn (ng/g)	7.460	10.005	5.375
		OC in deposited surface layer (g) (<i>W_{sed}</i>)	43.397	58.206	31.267
		ratio of atmos deposition/amount in surface layer	0.008	0.001	0.000
wet season	estimation by [OC] in water	seawater concn (ng/L)	0.247	0.132	0.089
		OC amt deposited by sea (g/yr)	13.963	13.488	9.060
		ratio of atmos deposition/sedimentation (surface sediment)	0.015	0.005	0.003
	estimation by [OC] in sediment	sediment concn (ng/g)	1.468	29.229	8.967
		OC in deposited surface layer (g)	8.539	170.040	52.164
		ratio of atmos deposition/amt in surface layer	0.024	0.000	0.001
		ratio of sedimentation from water/amt in surface layer	1.635	0.079	0.174

supported by the composition of HCHs in both sediment and seawater. Conversely, volatilization of HCHs tends to be positively related to temperature, which might explain the domination of HCH deposition in the wet season. At lower temperatures, HCHs tend to bind with the particulate matters in soil or sediment, which would decrease the atmospheric concentrations of HCHs. Moreover, photochemical transformation of γ - to α -HCH might also contribute to this process (24). The α/γ -HCH ratios were, in general, similar to atmospheric ratios of HCHs (>1) in the Northern Hemisphere. Atmospheric deposition fluxes of HCHs measured in this study were less than those observed in North America (>10 ng m⁻² d⁻¹ for α -HCH and 0.1–1.0 ng m⁻² d⁻¹ for γ -HCH), in Galveston Bay (4.7 ng m⁻² d⁻¹) and in open sea (8.3–6.7 × 10¹ ng m⁻² d⁻¹) (19, 22, 24). This result might partly be due to photolysis and volatilization of HCHs during the prolonged sampling period under direct sunlight. Interestingly, β -HCH was not detected in atmospheric deposition in this study, although this HCH isomer was found in water and sediment samples from both areas.

Cyclodienes. Data in this study suggest that local and regional emission sources of cyclodienes, such as endosulfans, are likely to be major contributing factors for the deposition of these pollutants in both areas. Greater atmospheric fluxes were observed during January (1.7 ng m⁻² month⁻¹) and February (1.1 ng m⁻² month⁻¹) at Mai Po. In contrast, at A Chau endosulfans were mainly detected between December to May (nd–1.5 ng m⁻² month⁻¹) with maximum concentrations of 1.5 and 1.4 ng m⁻² month⁻¹ in April and May, respectively. Greater concentrations of endosulfans were detected in Daya Bay, which is close to A Chau (21). This suggests that Daya Bay may have a local source of these OCs. Seasonal fluctuations in concentrations might be related to the seasonal variations in utilization pattern of these pesticides in the nearby farming area. Since A Chau is close to Daya Bay and Huizhou, it might have a large potential for receiving these atmospherically deposited OCs, especially during the wet season. Since the manufacture and use of endosulfans have not been restricted in China, endosulfans are consistently found in seawater and sediment samples (21, 25). This observation is also consistent with the trace amounts of endosulfans detected in the Hong Kong atmospheric environment (5). The concentrations of cyclodienes found in atmospheric depositions at both Mai Po and A Chau were, however, smaller than those reported to occur in the North American Great Lakes (3).

PCBs. Small amounts of dioxin-like PCBs were found at both sites, especially in the dry season. In this study, only 15 dioxin-like PCB congeners with non-ortho, mono-ortho, and

di-ortho structures were measured. PCB congeners (IUPAC) 28, 37, 77, 114 (0.01–0.02 ng m⁻² d⁻¹) were detected in atmospheric deposition at Mai Po. Concentrations of TEQ_{WHO-Bird} were greater than concentrations of TEQ_{WHO-Fish} or TEQ_{WHO-Mammal}. PCB 28, 37, and 114 (~0.01 ng m⁻² d⁻¹) were found in the A Chau samples. Fluxes of Σ PCBs in atmospheric deposition ranged from <5 × 10⁻³ to 2.0 × 10⁻² pg TCDD EQs m⁻² d⁻¹ (Figure 3). Most of the dioxin-like PCBs were detected in the dry season. This phenomenon might be related to the amount of suspended particulate matters in the atmosphere. Since PCBs have relatively large *K_{ow}* values they have a high affinity for particles. Thus, dry and strong winds in the winter might facilitate the long-range transport from remote areas in the Pearl River Delta. Measured concentrations of PCBs were however small as compared to fluxes of other OC pesticides. Concentrations of PCBs in air have been reported to be in the range from around 0.1 pg TEQ_{WHO}/m³ in Hong Kong (5) to several hundred picograms of PCB/m³ in the Pearl River Delta (6), reflecting historical utilization or disposal of PCBs in the whole area.

Mass Balance Analysis. In this study, a new designed mass balance analysis was conducted for the 380-ha MPMNR. The dry weight of the marine sediment was determined to be 189 kg/m³. The average depth of water in the selected mudflat area was determined to be 2 m. The small ratio of atmospheric deposition and sedimentation indicates that material exchange by deposition via air was less than sedimentation (Table 7). In general, the masses of OC pesticides deposited to sediments were less than those released from the surface sediments, which indicate that a large proportion of OC pesticides could have been released from the deeper sediments. For HCHs and DDTs, the ratios of sedimentation to the mass of these compounds in the sediment were greater during the dry season than during the wet season, and this might be due to the discharge of organic-carbon-rich sewage from the Shenzhen River in the dry season. Since little tidal flushing occurred during the dry season, OC residues from the Shenzhen River tended to accumulate in the sediment. The sedimentation rate (>14 mg/L) was greater during the dry season (16). The results of the mass balance calculations suggest that HCHs were transported to the MPMNR primarily by seawater rather than via the atmospheric route. This might also be partly due to the relatively high temperature of the local environment causing evaporation of HCHs in the deposition samplers. Concentrations of HCB in water and sediment were significantly greater during the wet season when the atmospheric deposition of HCB was least. The estimated amount of

seawater-related HCB deposition was greater than the mass in the surface sediment. During the wet season, HCB was released from sediment to the overlying water (Table 7).

Acknowledgments

This study was supported by a Central Allocation Grant (8730020) awarded by the Research Grants Council, Hong Kong, and the Area of Excellence Scheme under the University Grants Committee of the Hong Kong Special Administration Region, China (Project AoE/P-04/2004).

Literature Cited

- (1) Bedos, C.; Cellier, P.; Calvet, R.; Barriuso, E. Occurrence of pesticides in atmosphere in France. *Agronomie* **2002**, *22*, 35–49.
- (2) Shatalov, V.; Malanichev, A.; Nadezhda, Berg T.; Manø, S. *Assessment of POP Transport and Accumulation in the Environment*; EMEP Report 4/2001; MSC-EAST: Moscow, Russia, 2001.
- (3) Integrated Atmospheric Deposition Network (IADN), Environment Canada. http://www.msc-smc.ec.gc.ca/iadn/Resources/loadings9798/final_9798_loadings_report_e.html (accessed May 2004).
- (4) Sin, D. W. M.; Choi, J. Y. Y.; Louie, P. K. K. A study of polychlorinated dibenzo-*p*-dioxins and dibenzofurans in the atmosphere of Hong Kong. *Chemosphere* **2002**, *47*, 647–653.
- (5) Louie, P. K.; Sin, D. W. M. A preliminary investigation of persistent organic pollutants in ambient air of Hong Kong. *Chemosphere* **2003**, *52*, 1937–1403.
- (6) Fu, J.; Sheng, G.; Chen, Y.; Wang, X.; Min, Y.; Peng, P.; Lee, S.; Chang, L.; Wang Z. Preliminary study of organic pollutants in air of Guangzhou, Hong Kong and Macao. In *Molecular Markers in Environmental Geochemistry*; Eganhouse, R. P., Ed.; ACS Symposium Series 671; American Chemical Society: Washington, DC, 1997; pp 164–176.
- (7) WWF (HK). www.wwf.org.hk (accessed May 2004).
- (8) Shutes, R. B. E. Artificial wetlands and water quality improvement. *Environ. Int.* **2001**, *26*, 441–447.
- (9) Liang, Y.; Wong, M. H.; Shutes, R. B. E.; Revitt, D. M. Ecological risk assessment of polychlorinated biphenyl contamination in the Mai Po Marshes Nature Reserve, Hong Kong. *Water Res.* **1999**, *33*, 1337–1346.
- (10) Müller, J. F.; Gaus, C.; Prange, J. A.; Pöpke, O.; Poon, K. F.; Lam, M. H. W.; Lam P. K. S. Polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans in sediments from Hong Kong. *Mar. Pollut. Bull.* **2002**, *45*, 372–378.
- (11) Richardson, B. J.; Zheng, G. J. Chlorinated hydrocarbon contaminants in Hong Kong surficial sediment. *Chemosphere* **1999**, *39*, 913–923.
- (12) Ogur, I.; Masunaga, S.; Nakanishi, J. Atmospheric deposition of polychlorinated dibenzo-*p*-dioxins, polychlorinated dibenzofurans and dioxin-like polychlorinated biphenyls in the Kanto region, Japan. *Chemosphere* **2001**, *44*, 1473–1487.
- (13) Khim, J. S.; Villeneuve, D. L.; Kannan, K.; Lee, K. T.; Snyder, S. A.; Koh, C. H.; Giesy, J. P. Alkylphenols, polycyclic aromatic hydrocarbon, and organochlorines in sediment from Lake Shihwa, Korea: Instrumental and bioanalytical characterization. *Environ. Toxicol. Chem.* **1999**, *18*, 2424–2432.
- (14) Van den Berg, M.; Birnbaum, L.; Bosveld, A. T. C.; Brunström, B.; Cook, P.; Feeley, M.; Giesy, J. P.; Hanberg, A.; Hasegawa, R.; Kennedy, S. W.; Kubiak, T.; Larsen, C. J.; van Leeuwen, F. X. R.; Liem, A. K. D.; Nolt, C.; Peterson, R. E.; Poellinger, L.; Safe, S.; Schrenk, D.; Tillitt, D.; Tysklind, M.; Younes, M.; Wærn, F.; Zacharewski T. Toxic equivalency factors (TEFs) for PCBs, PCDDs, PCDFs for human and wildlife. *Environ. Health Perspect.* **1998**, *106*, 775–792.
- (15) Hong Kong Observatory. *Extract of Meteorological Observations for Hong Kong*; 2003.
- (16) Highways Department. Agreement No. CE 39/2001 Shenzhen Western Corridor—Investigation and Planning, Environmental Impact Assessment Report. 2002.
- (17) Lee, S. C.; Chiu, M. Y.; Ho, K. F.; Zou, S. C.; Wang, X. M. Volatile organic compounds (VOCs) in urban atmosphere of Hong Kong. *Chemosphere* **2002**, *48*, 375–382.
- (18) Mackay, D.; Shiu, W. Y.; Ma, K. C. *Illustrated Handbook of Physical–Chemical Properties and Environmental Fate for Organic Chemicals. Vol. 5, Pesticide Chemicals*; Lewis Publishers: Chelsea, MI, 1992.
- (19) UNEP. Central and North East Asia Regional Report. In United Nations Environment Programme (Chemicals). 2002.
- (20) Öberg, T.; Bergström, J. G. T. Hexachlorobenzene as an indicator of production from combustion. *Chemosphere* **1985**, *14*, 1081–1086.
- (21) Zhou, J. L.; Maskaoui, K.; Qiu, Y. W.; Hong, H. S.; Wang Z. D. Polychlorinated biphenyl congeners and organochlorine insecticides in the water column and sediment of Daya Bay, China. *Environ. Pollut.* **2001**, *113*, 373–384.
- (22) Park, J. S.; Wade, T. L.; Sweet, S. T. Atmospheric deposition of PAHs, PCBs and organochlorine pesticides to Corpus Christi Bay, Texas. *Atmos. Environ.* **2002**, *36*, 1707–1720.
- (23) Takase, Y.; Murayama, H.; Mitobe, H.; Aoki, T.; Yagoh, H.; Shibuya, N.; Shimizu, K.; Kitayama, Y. Persistent organic pollutants in rain at Niigata, Japan. *Atmos. Environ.* **2003**, *37*, 4077–4085.
- (24) Willett, K.; Ulrich, E.; Hites, R. Differential toxicity and environmental fates of hexachlorocyclohexane isomers. *Environ. Sci. Technol.* **1998**, *32*, 2197–2207.
- (25) Zhang, Z.; Dai, M.; Hong, H.; Zhou, J. L.; Yu, G. J. Dissolved insecticides and polychlorinated biphenyls in the Pearl River Estuary and South China Sea. *J. Environ. Monit.* **2002**, *4*, 922–928.

Received for review June 29, 2004. Revised manuscript received August 13, 2004. Accepted October 8, 2004.

ES049021+