

Marine Pollution Bulletin, Vol. 51, No. 8-12, 2005, pp. 960-965

Characterization of Transboundary POP Contamination in Aquatic Ecosystems of Pearl River Delta

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Abstract

During the past two decades, the rapid development of the Pearl River delta leads to substantial accumulation of various toxic organic compounds. This study aims to give a preliminary characterization of the existing state of contamination in this region and to provide insight into the possible fate of persistent organic pollutants (POPs) in this estuary. The available data on POPs in water, river, estuarine sediments, soil, and marine organisms within the Pearl River Delta are compiled. It is shown that it may lead to transboundary POP pollution problems at both Hong Kong and Macau Special Administration Regions located at the downstream end of the region. It is noted that the levels of DDTs and HCHs in various environmental media are at alerting levels and that fresh DDT might still be applied illegally within the region. A systematic research is required to determine both the temporal and spatial variations of all POPs in various carrying media of the Pearl River delta as a whole.

Keywords: Aquatic ecosystem; Pearl River Delta; POP contamination; South China; Toxicology characterization; Transboundary

1. Introduction

The Pearl River is the largest river system in Southern China. Currently, the Pearl River delta is a significant and quickly developing economic zone in China, covering eight large cities in China: Dongguan; Foshan; Guangzhou; Huizhou; Jiangmen; Shenzhen; and, Zhongshan. Figure 1 shows the bathymetry of the Pearl River delta. During the past two decades, the rapid economic boom and growing agricultural, industrial and municipal development in the region leads to substantial accumulation of toxic organic compounds and a significant environmental impact has been imposed on the ambient conditions. Untreated sewage is discharged in an expeditious rate and large quantities of various pollutants have been released, largely via five outlets, namely, Hu men, Jiao men, Hongqi men, Heng men, and Shenzhen River. The water quality within the delta has been deteriorating and the frequency of the occurrence of algae blooms has been increasing (Chau and Sin, 1992; Sin and Chau, 1992; Jin et al., 1998).

Amongst others, persistent organic pollutants (POPs) are, by their very nature, extremely hazardous because of their toxicity for human beings, their long-term persistence in the environment, their impact on non-target organisms, and their bioaccumulation capability in the tissues of animals and humans via the food chain. Although adverse environmental impacts are usually associated with high contaminant concentrations formed in close proximity to input sources, POPs may be

transported for long distances by air, rivers, and ocean currents (Tanabe, 1991). These pollutants may be transported from these outlets towards the entrance of the estuary, through the interaction of tidal effects as well as runoff discharge.

The impact due to transboundary pollution from the inner Pearl River delta may be very significant since Hong Kong Special Administrative Region (HKSAR) and Macau are located on the eastern and western downstream ends of the outlet, respectively. It will potentially add an additional dimension and complication to the environmental protection tasks in both HKSAR and Macau. However, up to date, only limited information is available in the estuary with regard to the prevalent conditions of the POP that were reported to have detrimental effects elsewhere. Hence, necessity arises to determine the impacts of these pollutants from the Pearl River delta on the water quality in the ambient seawaters of HKSAR in both quantitative and qualitative terms. In particular, after the restoration of the sovereigns and administration of Hong Kong the People Republic of China in 1997, increasing interactions as well as number of projects are undertaken which in turn lead to more attention to the region (Chau, 2001 & 2002). Previously, several two dimensional or three-dimensional models have been developed and employed to simulate the environmental hydraulics in Pearl River delta (Chau and Jiang, 2001 & 2002).

In this paper, the available data on POPs in water, river, estuarine sediments, soil, and marine organisms (fish, shrimp and green-lipped mussel) within the Pearl River delta are gleaned and compiled. An objective of this study is to give a preliminary characterization of the existing state of contamination in this region and to provide insight into the possible fate of POPs under a typical sub-tropical environment. It will serve as an assessment of the current status, distribution, severity, fate, and environmental consequence of detected contaminants, thus furnishing possible insight as well as future directions for global and strategic environmental management of the region.

2. Persistent organic pollutants (POPs) in Pearl River Delta

Table 1 shows a summary of concentrations of various POPs in different carrying media of Pearl River delta from existing literature (Cheng et al., 2000; Fang et al., 2001; Fu et al., 2001; Hong et al., 1999; Kang et al., 2000; Mai et al., 2001; Wu et al., 1999; Yuan et al., 2001; Zheng et al., 2001; Zhou et al., 2001). Owing to the apparent significance of sediment phase in the fate and transport of contaminants, previous research efforts have mainly been focused on surficial sediments in assessing the pollution of water bodies. Moreover, it can be observed from Table 1 that most previous research is limited to a few kinds of POP compounds and that information on many combinations of POP type and carrying medium are still lacking.

2.1 Organochlorine pesticides

Most commonly detected chlorinated pesticides, with the exception of Dichloro diphenyl trichloroethane (DDT) and related compounds, dichloro diphenyl dichloroethane (DDD) and dichloro diphenyl ethane (DDE), and hexachlorocyclohexanes (HCH), are not significant in the sediment samples. The concentrations of DDT and HCH range from 3 to 163 ng/g and from 1 to 101 ng/g, respectively. The ratios of DDT/(DDD+DDE) and DDT/DDE are about 6 and 26,

respectively whilst HCH exhibits a relatively high composition of β -HCH (67% β -HCH, 22% γ -HCH, 8% δ -HCH, and 3% α -HCH) (Wu et al., 1999). Concentrations of HCH in surface sediments are comparable with other river/estuary systems in other coastal areas of the world, such as in Japan, Europe and North America, except some polluted areas. However, concentrations of DDTs and its metabolites appear to be relatively higher (Fowler, 1990; Iwata et al., 1994).

The relatively high concentrations of DDTs and its metabolites recorded indicate that the estuary has received significant amount of DDTs from surface runoff and/or river discharge. Since DDT undergoes natural and dehydrochlorinated degradation to DDD and DDE under anaerobic and aerobic conditions, respectively, the relatively high ratio of DDT/(DDD+DDE) may be brought about by slow degradation of DDT or more recent fresh illegal input of DDT compounds within this region. It is also possibly due to the DDT application for the control of malaria in southern China. This deduction is further reinforced by the unusual high ratio of DDT/DDE since aerobic degradation may be deterred by high concentration of DDT source in the vicinity (Pereira et al., 1996).

The levels of DDTs and HCHs in differently cultivated and non-cultivated soils from the Pearl River Delta are found to range from 15-125 ng/g and 2-30 ng/g, respectively (Zhang et al., 2001). In particular, their concentrations are higher in the cultivated soils than the non-cultivated soils. This strongly suggests their former or even currently illegal use in agriculture or for fish cultivation purposes.

DDTs concentrations in aerosols range from 65-109 pg/m^3 (Cheng et al., 2000). No data is available for HCHs concentrations in aerosols. An air migration process may take place and the air particulate may be originated from agricultural soils containing high levels of DDTs. Levels of DDTs and HCHs concentrations in surface water are determined to be 27-976 ng/l and 36-1,229 ng/l, respectively (Zhou et al., 2001). The ranges of DDTs and HCHs concentrations in marine organisms are 9-191 ng/g and 0-1 ng/g, respectively (Fang et al., 2001). All these values in different carrying media suggest a rather critical condition for organochlorine pesticides in the Pearl River delta.

2.2 Polycyclic aromatic hydrocarbons (PAHs)

PAHs were prevalent contaminants as a result of the heavy anthropogenic and industrial activities in the Pearl River delta. The recorded concentrations of total PAHs in sediment samples range from 156 to 10,811 ng/g (Fu et al., 2001; Mai et al., 2001). The concentration of total PAHs in Pearl estuaries are found to be low compared with those reported for other industrialized countries, indicating that the PAH pollution in the sediment samples are moderate compared to developed countries (Yuan et al., 2001). The alkylated/non-alkylated ratio is about 2 and the high molecular weight (HMW)/low molecular weight (LMW) ratio is 1 or so. Here, HMW and LMW denote four to six-ring PAHs and two to three-ring PAHs, respectively. It is noted that refined petroleum products comprises primarily LMW PAHs and alkylated PAHs. The relatively high alkylated/non-alkylated ratio and relatively low HMW/LMW indicate that the major PAH pollution was contributed by petroleum input (Yuan et al., 2001). This phenomenon might be explained by the heavy harbor commercial as well as industrial shipping activities, including ship discharges and accidental oil spills in the estuary.

PAHs concentrations in aerosols range from 0-200 ng/m^3 (Fu et al., 1997). It may be associated with industrial and heavily trafficked activities. The range of PAHs

concentrations in marine organisms is 28-74 ng/g (Richardson et al., 2003). No data are available for PAHs concentrations in soil or surface water.

2.3 Polychlorinated biphenyls (PCBs)

In general, the observed concentrations of PCB in the sediment samples are low with the range from 10 to 486 ng/g (Hong et al., 1999; Kang et al., 2000). The PCB congeners identified are principally moderately-chlorinated biphenyls, i.e., tetra- to hepta-chlorinated, rather than lower chlorinated congeners. The average concentration is much lower than those reported for other coastal areas in the world (Fowler, 1990; Iwata et al., 1994). Thus, PCBs might not be major contaminants and might not be a major environmental concern in most of the Pearl River delta, except at certain highly industrialized and urbanized areas such as Huangpujiang. This may be attributed to the high assimilative and self-purification capabilities of the estuary against anthropogenic activities and pollution impact via large runoff discharge during wet season (10,524 m³/s), enormous sediment loads and supplementation by biogeochemical processes including volatilization, biodegradation, etc.

No data is available for PCBs concentrations in soil. PCBs concentrations are found to range from 297-537 pg/m³ in air (Li et al., 2001). This level is comparable to or is lower than that in many other countries (Iwata et al., 1994). Levels of PCBs concentrations in surface water are determined to range from 91-1353 ng/l (Zhou et al., 2001). The range of PCBs concentrations in marine organisms is 83-615 ng/g (Fang et al., 2001). The relatively high values in surface water and marine organisms may be due to the combined effects of soil runoffs, wastewater discharge, sewage outfalls and shipping activities.

2.4 Butyltins

Organotin and other butyltin compounds, in particular Tributyltin (TBT), are commonly used as active ingredient of the antifouling agent in paints of ships. Sources of TBT in coastal water as well as sediments can be primarily attributed to the leaching from ship paint. The concentrations of TBT found in the sediment samples range from 2 to 380 ng/g (Yuan et al., 2001). Moreover, concentrations of dibutyltin (DBT) and monobutyltin (MBT), as the naturally degraded forms of TBT, have values of 2 ng/g and 13 ng/g, respectively. This may be due to various shipping activities in this major navigation channel with capacity to accommodate large domestic and foreign vessels.

No data are available for organotin concentrations in soil or air. The TBT concentration is found to range from 21-39 ng/l in surface water and 5-19 ng/g in marine organisms, respectively (Yuan et al., 2001). These relatively low values may suggest that TBT contamination is not particularly serious in this estuary.

2.5 Polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF)

Existing information on the contamination status, extent, level, and fate of polychlorinated dibenzo-p-dioxins (PCDD) and polychlorinated dibenzofurans (PCDF) in different media of the Pearl River delta are very lacking. The concentrations of PCDD/Fs in the surface sediment of the area are determined to be between 472 to 2502 pg/g of dry weight (Zheng et al., 2001). It may be attributed to the extensive use

of sodium pentachlorophenate (Na-PCP) previously in the upper reaches of the Pearl River (Zheng et al., 2001). Although Na-PCP is no longer used in recent years, the persistent PCDD/Fs have been accumulated and remained in sediments.

No data are available for PCDD/Fs concentrations in soil, air, surface water or marine organism. It can be observed that extremely limited studies are available in the scientific literature on levels of PCDD/Fs in environment of the Pearl River Delta. More effort is to be spent in this specific domain.

3. Analysis and Discussions

In China, DDT was extensively used during previous years and its usage has been banned since 1983. It is natural to anticipate the records should exhibit a gradual reduction of DDT concentrations since the presence of DDTs in the sediment should predominantly be residue from previous usage. It is the author's opinion that, in order to align with the international standards and be comparable with the POPs pollution in other coastal areas of the world, the non-compliance should become a priority issue and more stringent controls should be imposed on further disposal of DDTs and HCHs.

Similarly, PCBs were banned in 1983, yet a large proportion of PCBs still remain in use at present in older transformers and capacitors. Thus, it is strongly believed that, in order to avoid the deterioration of the existing situation and to comply with the global standards, rigorous surveillance and monitoring on the disposal of these organic chemicals is entailed and future research should be conducted to report infringement instances for prevention of further contamination to the environment.

The concentration of PAHs in the Pearl River delta is in general high compared with other industrialized countries. It can be considered a seriously contaminated location. Whilst the bioavailability and bioconcentration of PAHs by organisms can be significant, biomagnification through the food chain is relatively low due possibly to rapid metabolism. Thus PAH data in marine organisms located in the top of food chain is still low. Nevertheless, the author strongly trusts that it might be cautious to further investigate the impact of PAHs to different types of marine organisms in the Pearl River delta.

The relatively low TBT concentration and high MBT concentration in the sediment in Pearl River delta reveal that the butyltins have largely gone through chemical and microbial degradation. Moreover, the short half-life of TBT may be attributed to the highly contaminated water which facilitates the degradation process (Hattori et al., 1988). It is deduced that continuous but more systematic monitoring of concentrations of various forms of organotin should be conducted in the Pearl River delta, particularly in areas with frequent shipping activities such as Victoria Harbor and Huangpu Harbor, so as to determine whether or not alarming level is already reached and whether or not new legislation should be enacted.

Although information on PCDDs and PCDFs in most media of the Pearl River delta are lacking, there is increasing evidence that these organic micro-polluting compounds are extremely hazardous to aquatic ecosystems, especially in view of their capability of biomagnification through the food chain (Loonen et al., 1996). The author strongly believes that further research should be entailed to monitor contamination levels of PCDD/Fs in other media, other than surface sediment, including atmosphere, industrial discharge, outfall from sewage treatment plant, storm drain, sea water, marine organism, etc.

Figure 2 shows the spatial distribution of various POPs in surface sediments of the

Pearl River delta. The spatial patterns of DDT, PAH and PCB concentrations with the distance from the Humen outlet are very similar. In overall, the concentrations of POPs are higher near Humen outlet and declined gradually with distance from the outlet, which is possibly due to existence of localized anthropogenic inputs at the upstream end and the dilution effect of the estuary. From this Figure, the impact due to transboundary pollution from the inner estuary is apparent in both HKSAR and Macau SAR located at the downstream end of the Pearl River Delta region.

Based on an extensive literature review, Long and Morgan (1990) has correlated observed biological effects with different concentrations of sediment-sorbed contaminants. Data from all these studied articles were reviewed and ranked from low to high to represent different degrees of adverse biological effects. The 10th and 50th percentiles are termed effects range low (ER-L) and effects range median (ER-M), respectively. Whilst ER-L values indicate possible toxic biological effects for benthic organism, ER-M values denote high possibility to pose detrimental biological effects on the benthic organisms. Figure 3 shows the concentrations of various contaminants in sediments of the Pearl River delta in comparison with their corresponding ER-L and ER-M values. It can be observed that the total PAH and PCB concentrations in the sediments are below the ER-L values, whilst the concentration of DDT is above the corresponding ER-L value. This situation will, in particular, be crucial during the dry season when the runoff discharge may not be large enough to dilute and flush out the organochlorine compounds.

The main finding of this study is that the existing data are very limited and do not cover all the combinations of POP type and carrying medium. In particular, studies analyzing levels of POPs in soil and in PCDDs and PCDFs are lacking.

Thus, prior to further deterioration of the existing condition in the Pearl River delta, a more extensive and systematic research is required in order to determine both the temporal and spatial variations of all POPs in various carrying media for ultimate formulation of effective strategic environmental management measures. This deduction prompts us a promising research to explore in the future.

4. Summary

In this paper, existing data on POPs in water, river, estuarine sediments, soil, and marine organisms within the Pearl River Delta are gleaned and compiled. A preliminary characterization of the existing state of contamination in this region and insight into the possible fate of POPs are studied. It is demonstrated that POPs may be transported for long distances by air, rivers, and ocean currents, thus leading to transboundary pollution problems at both Hong Kong and Macau Special Administration Regions located at the downstream end of the Pearl River Delta region. It is observed that fresh DDT might still be applied illegally within the region and that the prevalent levels of DDTs and HCHs in various environmental media are likely to pose detrimental biological effects on the benthic organisms. This should become a priority issue and more stringent controls should be imposed on further disposal of DDTs and HCHs. Moreover, further research will be entailed to monitor contamination levels of PAH, PCB, TBT and PCDD/Fs in other carrying media. It is concluded that a more extensive as well as systematic research is required in order to determine both the temporal and spatial variations of all POPs in various carrying media of the Pearl River delta as a whole, for ultimate formulation of effective strategic and integrated environmental management measures.

Acknowledgment

This research was supported by the Internal Competitive Research Grant of Hong Kong Polytechnic University (A-PE26). The author would also like to express his sincere thanks to the anonymous referees for their invaluable comments and constructive suggestions on the paper.

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Table 1. Concentrations of various POPs in different media of Pearl River delta

Conc.	DDT	HCH	PAH	PCB	TBT	PCDD/F
Soil (ng/g)	15-125	2-30				
Air (pg/m ³)	65-109		0-200,000	297-537		
Water (ng/l)	27-976	36-1,229		91-1,353	21-39	
Marine organisms (ng/g)	9-191	0-1	28-74	83-615	5-19	
Sediments (ng/g)	3-163	1-101	156-10,811	10-486	2-380	0.472-2.502

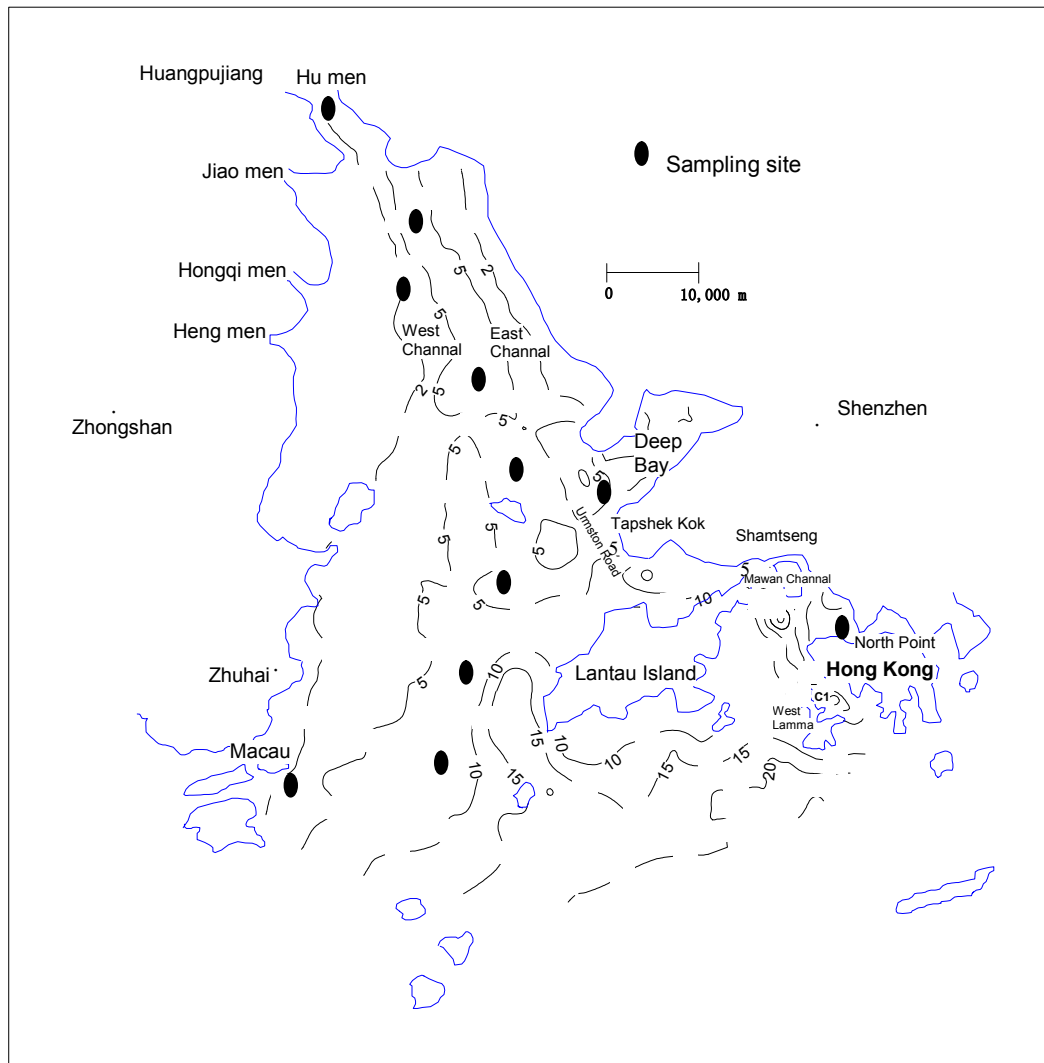


Figure 1. The bathygraphy of the Pearl River delta

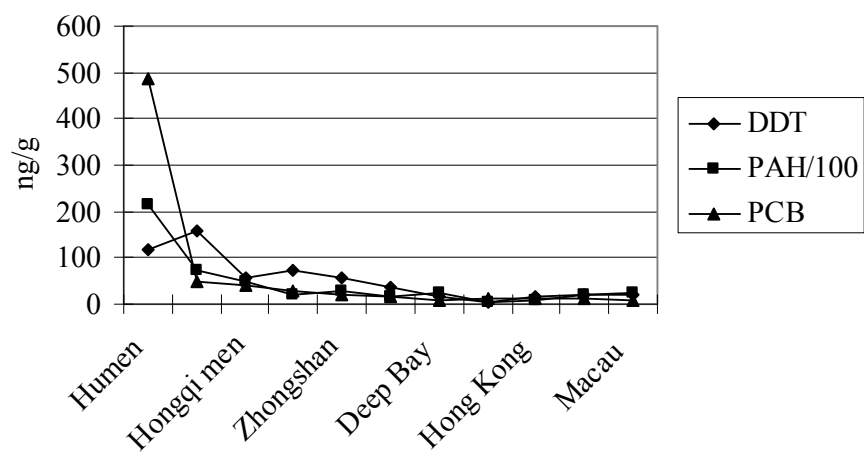


Figure 2. Spatial distribution of various POPs in surface sediments of the Pearl River delta (Fu et al., 2001; Hong et al., 1999; Kang et al., 2000; Mai et al., 2001; Wu et al., 1999)

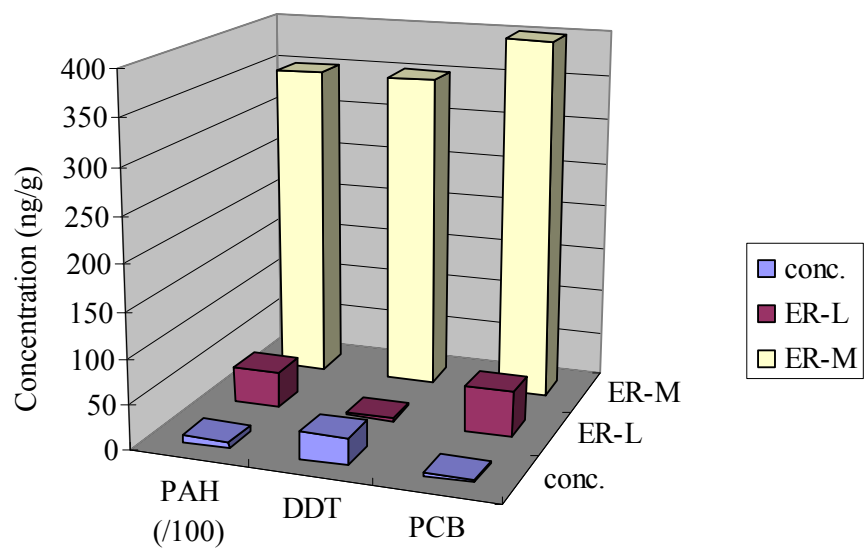


Figure 3. Concentrations of various contaminants in sediments of the Pearl River delta in comparison with their corresponding ER-L and ER-M values