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Contamination of soil, leaves and vegetables by polychlorinated biphenyls in Xiamen region, China

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Abstract: The paper aimed to identify the primary of polychlorinated biphenyls(PCBs) in the Jiulong River Estuary, investigate the spatial distribution of PCBs contamination in the environment, localize the atmospheric source and evaluate ongoing PCBs emissions by analyzing soil samples collected along the Jiulong River region. In addition, the accumulation of PCBs in the human food chain was quantified by analyzing leaf of orange trees and vegetable samples collected along a gradient of soil/atmospheric contamination moving away from the source. Consequently, the impact on the human health and the ecosystem was quantified, different management options were proposedto reduce this impact and to carry out research on organic contaminants along the Jiulong River and Xiamen region.

Keywords: Jiulong River; PCBs; soil; vegetable; contamination

Introduction

The governments, donor agencies and commercial firms around the world were meeting in Geneva, 10 June 2004 for two days to promote international efforts to rid polychlorinated biphenyls (PCBs) in the world, one of 12 highly toxic chemicals targeted for elimination by the Stockholm Convention on Persistent Organic Pollutants (POPs). Polychlorinated biphenyls (PCBs), a class of synthetic organic chemicals that are amongst the most widespread of all environmental pollutants, were found worldwide in air, water, soil, food and the fatty tissues of humans and animals. Due to their low flammability, PCBs have been used extensively in electrical equipment such as transformers and large capacitors used in power lines and major facilities. They have also been used as additives in paint, carbonless copy paper, and plastics. Many hundreds of thousands of tonnes of PCBs have been commercially manufactured since 1929. Annual world production peaked in the late 1960s at close to 60000 ton showed by UNEP Report of 2004.

Although production is now banned under the Stockholm Convention, PCBs continue to pose a risk to human health and the environment because of the wide array of PCBs-containing electrical equipment is still in service. Tons of wastes containing PCBs or contaminated by PCBs are also being held at temporary storage sites, particularly in developing countries.

The recent studies in Xiamen coastal sediments have shown that elevated concentrations of PCBs have been recorded in some locations. The sewage outfalls, industrial discharges and sewage waste dumping are a major source of PCBs(Maskaoui, 2000; 2002; Zhou, 2000). In Daya Bay the major source of pollutants is related to urban/industrialized discharges and atmospheric fallout (Zhou, 2001). The detailed data of the level of these organic pollutants from different proposed sources of contamination still need to be investigated. Also we are unaware of many reports concerning assessment of their levels in the soil, plants, leaves and vegetables since large numbers of people have been exposed to PCBs through food contamination. Consumption of PCBs-contaminated rice oil in Japan in 1968 and in Taiwan in 1979 caused pigmentation of nails and mucous membranes and swelling of the eyelids, along with fatigue, nausea, and vomiting.

The purpose of this work is to make a comparison of vertical profiles of PCBs around Xiamen area with different source background. With the lack of data about the other sites, an investigation program of these micropollutants in sediments from aquaculture area, wastewater discharges, agriculture locations, industrial effluents will be conducted in some sites around Xiamen Western Bay. To estimate the sources and transport pathways of the POPs, sediment cores, soil and vegetable samples will be taken from contrasting marine and land locations affecting Xiamen water quality. So, the sampling in the agricultural locations were chosen in Xinglin closed to Xiamen harbor and Longhai along Jiulong River as the most activated field in this region.

1 Materials and methods

1.1 Geographic features, climates and rainfall distribution

The total land area of the Xiamen Municipality's administrative jurisdiction, including islands, is 1516 km^2; it is located in the southern coast of the Fujian Province(Fig. 1). The bay has a complex structure with several islands and different seas, i.e. the West Harbour, Maluan Bay, Tong'an Bay, Jiulong River Estuary, the Southern Sea and the Eastern Sea. The average annual temperature is 20.9℃; the warmest month is July/August with 28.4℃ and the coldest period is January/February with an average 12.6℃. Rainfall has an annual average of 1143 mm. The cultivated soil in the Municipality of the Xiamen area was 427 km^2. The major crops are rice(29.8 km^2), peanuts(12.6 km^2), vegetable (22.2 km^2) and fruits(32 km^2).

1.2 Chemical standards

Reference PCBs (12 compounds, each at 10 μg/ml) were obtained from Supelco. Working standard was prepared by diluting the stock solution in cyclohexane. They were further diluted with cyclohexane to prepare calibration solutions for capillary GC analyses in the range 0.01—2

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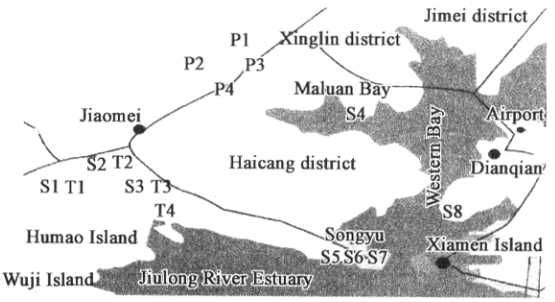

Fig.1 Map of Xiamen region showing the sampling sites

ng/μl. All solvents used for sample processing and analyses (dichloromethane, ethyl acetate, acetone, hexane, cyclohexane, and methanol) were analytical grade and further distilled twice to remove impurities. Deionized water was taken from a Milli-Q system(Millipore, Watford).

1.3 Sampling and sample treatment

Samples of top soil in the cultivated areas along the Juilong River and subsurface sediment from different areas of Western Xiamen Sea (Maluan and Yuandang Lake) were collected during surveys on June 26, 2002. The grid reference of each sampling site is given in Table 1. Samples were taken using pre-cleaned glass bottles. After the surface sediment samples were collected, the top 1 cm surface layer was carefully removed with a stainless steel spoon and stored in pre-washed glass bottles. After returning to the laboratory, the sediments were then stored at −20℃ till extraction.

Table 1 The grid reference of each sampling site

Sample No.	Time	Source
S1	14: 00 pm	Soil from orange trees field
S2	14: 20 pm	Soil from orange trees field
S3	14: 35 pm	Soil from orange trees field
S4	9: 30 am	Aquacultural area(Maluan)
S5	12: 30 pm	Electrical Industrial Factory(Songyu)
S6/S7	13: 00 pm	Electrical Industrial Factory(Songyu)
S8	8: 30 am	Urban Sewage(Wastewater Treatment Station of Yuandang Lake)
T1	14: 10 pm	Leaves from orange trees
T2	14: 15 pm	Leaves from orange trees
T3	14: 20 pm	Leaves from orange trees
T4	14:30 pm	Leaves from orange trees
P1	15: 00 pm	Vegetable(Xinglin district)
P2/P3	15: 15 pm	Vegetables(Xinglin district)
P4	15: 30 pm	Vegetables(Xinglin district)

1.4 Sample extraction

Soil, sediment, leaves and vegetable samples were extracted in an ultrasonication bath, where SPM or sediment samples(approximately 10 g dw) were extracted twice in 100 ml of hexane: ethyl acetate, 2∶1 (v∶v) for 30 min(Zhou, 2000; Maskaoui, 2002). The extracts were blown down to 0.5 ml, and purified in a silica gel column(4 mm i.d. × 90 mm). The column was then eluted first with 3.5 ml of hexane and the solution discarded. Further elution was by dichloromethane (3.5 ml) to obtain PCBs (Hong, 1995; Zhou, 2000). All the extracts were concentrated by gentle N_2 blow-down to about 100 μl. Good recovery was obtained using this procedure as shown by Zhou *et al*.

1.5 Analyses

A Hewlett-Packard GC 5890 with an electron capture detector(ECD), an autosampler, and Chemstation software were used for determining the level of PCBs in soil and sediment samples. The capillary column used for the analyses was BPX-5(HP-5 equivalent, 45 m × 0.22 mm i.d. × 0.25 μm film thickness). The oven temperature of analyses was programmed from 60℃(initial time, 1 min) to 140℃ at a rate of 20℃/min. 140℃ to 236℃ at a rate of 3℃/min. 236℃ to 290℃ at a rate of 4℃/min, and held at 290℃ for 1 min.

Before analysis, relevant standards were run to check column performance, peak height and resolution, and the limits of detection. With each set of samples analysed, a solvent blank, a standard mixture and a procedural blank were run in sequence to check contamination, peak of identification and quantification. Compounds were identified mainly by their retention times. Selected samples were analysed by full scan GC/MS for confirmation. All results for the samples were reported on a day-weight basis.

1.6 Quality assurance

All analytical data were subject to strict quality control. Spiked soil, sediment vegetable and leaves samples were determined with good precision, and recoveries ranged from 60% ± 8% to 94% ± 10% for the samples(Zhou, 2000; 2003; Maskaoui, 2000; 2002). In addition, the errors involved in sampling were assessed by carrying out triplicate sampling at the same site and the analysis of sample extracts. Results showed good reproducibility of the sampling process.

2 Results and discussion

2.1 Major sources of PCBs

The total PCBs in sediment samples ranged from 2.141—6.065 ng/g dw, with a mean concentration of 4.007 ng/g dw(Table 2, Fig.2). The highest concentration, which was found in the location S8, has showed that the most polluted samples were related to the sewage. High concentration was also found at station S4, which is close to the fish culture area (Maluan Bay). The current environmental contamination by PCBs in the sediments is therefore due to the intense shipping activity near station S4, discharge of untreated sewage from Xinglin and Jimei districts, atmospheric deposition and dumping of toxic and industrial wastes from Xiamen's surrounding land (as Haicang, Jimei and Xinglin).

Table 2 Concentration of PCB compounds from some sampling sites in Xiamen region(ng/g dw)

Compound	Aquaculture S4	Industry S5	Industry S6/S7	Sewage S8
CB 18	0.073	0.001	0.001	0.005
CB 31 + 28	0.003	0.001	0.001	0.003
CB 52	0.376	0.111	0.076	0.209
CB 44	0.001	0.001	0.001	0.003
CB 101	0.040	0.002	0.001	0.005
CB 149	0.072	0.039	0.027	0.189
CB 118	0.005	0.003	0.003	0.012
CB 153	0.971	0.298	0.275	0.847
CB 138	0.069	0.048	0.051	0.197
CB 180	0.262	0.171	0.125	0.956
CB 194	3.820	1.453	1.58	3.639
Total PCBs	5.692	2.128	2.141	6.065

The total PCBs levels in these samples were higher than those observed in Xiamen Harbour and Jiulong River Estuary (< 0.01—0.32 ng/g dw), (Zhou, 2000; Maskaoui, 2000), but lower than those in the sediments of Daya Bay (0.85—27.37 ng/g dw) (Maskaoui, 2000; Zhou, 2001), those of the New Territories of Hong Kong(43—461 ng/g dw) (Zhou, 1999) and those in Pearl River Delta and Macao Harbour(11.9—158 ng/g dw) (Fu, 1999). The total PCBs levels in the samples collected from the Electrical factory in Songyu were lower than S4 and S8 but they are still more significant since the samples were collected from only three different locations, two of them sited around the factory, only one is inside but not really from the installation building. Regarding these results, the major sources of PCBs inputs to the Xiamen environment are still focused on these two stations S5, S6, S7 and S8.

In terms of individual PCBs congener distributions, as shown in Table 2, most congeners were present in all samples. The predominance of highly chlorinated biphenyls (CBs) congeners CB 153, 180 and 194 in samples was clearly observed, especially CB 194 (with 8 chlorine substitution). On average the three compounds collectively accounted for 90% of total PCBs concentration. Other congeners(mainly less chlorinated) generally made up a small individual contribution to total PCBs loading, except for CB 52 which contributed 6% at station S4. The observed finding of congener distribution towards more highly chlorinated compounds is consistent with previously published data indicating the preferential retention of these less volatile and more lipophilic compounds in the marine environment (de Voogt, 1990). Differences in congener composition in the aquatic systems may also be attributed to a decline in the proportion of less chlorinated PCBs that are more susceptible to losses through volatilization, sedimentation, and possibly microbial degradation (Brown, 1987; Quensen, 1988; MacDonald, 1992). Moderately and highly chlorinated PCBs may therefore remain persistent in the aquatic environment because they are less volatile and more soluble in lipids, adsorbed more readily to sediment, and are more resistant to microbial degradation (Shiu, 1986; Connell, 1988; Tyler, 1996).

2.2 Distribution and transport of PCBs

Organic contaminants, such as polycyclic aromatic hydrocarbons(PAHs) and PCBs are transported long distance in the atmosphere and enter surface water via different deposition processes (Bidleman, 1988; McVeety, 1988; Baker, 1990; Iwata, 1993). Atmospheric fluxes often dominate the input of POPs to lakes and oceans both far and near from source areas (Warmenhoven, 1989; Eisenreich, 1981; Barrie, 1992).

The compounds mentioned above are included in term of persistent organic pollutants(POPs) and they are considered to be demi-volatile, which means that they are transported in the atmosphere both in the gas and particle phases(Pankow, 1987; Bidleman, 1988). The distribution between the phases depends on factors such as the vapour pressure of the compound, the ambient temperature, particle concentration in the air and the nature of the particles(Bidleman, 1988). The distribution of the compound between the phases is of great importance for the deposition process.

Wet and dry deposition which includes scavenging of both gas and particle phases as well as vapour exchange across the air-water interface are major transport processes for atmospheric input of POPs to a water body (Eisenreich, 1981; Duinker, 1989). Unlike wet and dry deposition via particles, vapour exchange across the air-water interface takes place in two directions(Achman, 1993; McConell, 1993). Deposition fluxes are highly variable and are controlled by the properties of the compound, the particle concentration in the air, and the particle size and meteorological factors such as temperature and wind speed(Tsal, 1991).

In this work, the atmospheric input transport and deposition of PCBs to Xiamen agricultural areas have been estimated. These compounds represent compounds with somewhat different chemical and physical properties such as volatility and polarity. The different classes of compounds originate from different sources, for instance PAHs are mainly emitted in combustion processes, PCBs spread via industrial chemicals.

In order to estimate the deposition of PCBs to the area, measurements in soil, leaves, vegetable samples have been carried out at Jiulong River catchment.

The soil leaves and vegetable samples were collected from different locations in Xiamen regions. These sites are considered as far from the direct input sources of PCBs. The sampling sites are shown in the Fig.1. The samples analyses for the determination of the target compounds made us have a well understanding of the level of the probable contamination by atmospheric transport from the most polluted areas. Table 2 shows the concentration of these pollutants in different samples. As shown in Table 2, it is very clear that the high concentration was found in the oranges leaves samples. This may be also related to the absorption of PCBs by the soil and then transport to the leaves. Since there was no study so far on the PCBs levels with air sample, this suggestion has also been taken in to consideration. The range concentration of total PCBs is 0.003—0.239 ng/g dw in vegetable samples, 8.835—12.73 ng/g in soil samples and 564.9—1510 ng/g dw in leaves samples (Table 3). With these concentrations detected especially in food, it is important to note that the pollution degree is slightly higher and need to be more monitored since it affects human health by consuming these contaminated vegetables.

There are many reports revealing the enrichment of low molecular weight PCBs congeners, i.e. tri- to penta-chlorinated biphenyls in sediment, water and biological samples from high latitudes (Muir, 1990; 1992; Iwataet, 1993).

This enrichment might be explained by the higher vapour pressures of lower-chlorinated PCBs resulting in a more efficient atmospheric transport from sources in lower latitude countries and large air-seawater fluxes in the colder regions (Iwata, 1993; 1994a; 1994b). The fact that the compositions of PCBs homologues in the leaves, vegetable and soil samples suggests local or nearby sources of PCBs into this area. It is important to note that the concentrations of PCBs detected in leaves of orange trees were very higher than any other sample. Compared the level of PCBs in soil

samples with that in sewage, it can be concluded that the both levels were almost similar the same. So, it was easy to find out that the soil is seriously contaminated by these contaminants in this agricultural and this may cause hazardous effects on human health via food-web(Fig.2).

Table 3 Concentration of PCBs from soil, leaves of orange, vegetable samples in Xiamen region

	P1	P2/P3	P4	S1	S2	S3	T1	T2	T3	T4
CB 18	0.003	0.001	0.004	Nd	Nd	0.001	0.040	0.045	0.337	0.159
CB 31 + 28	0.194	Nd	0.232	0.003	0.001	0.001	0.092	0.094	0.684	0.322
CB 52	Nd *	Nd	Nd	0.380	0.089	0.550	37.13	29.61	62.55	29.50
CB 44	Nd	Nd	Nd	0.004	0.001	0.045	0.955	0.810	0.415	0.196
CB 101	Nd	0.002	0.003	0.005	0.002	0.002	0.168	0.169	3.411	1.609
CB 149	Nd	Nd	Nd	0.330	0.037	0.155	76.97	67.36	50.91	24.01
CB 118	Nd	Nd	Nd	0.110	0.003	0.006	7.978	0.977	3.170	1.495
CB 153	Nd	Nd	Nd	0	0.309	0.470	41.54	39.09	83.37	39.32
CB 138	Nd	Nd	Nd	0.558	0.115	0.097	107.7	90.34	123.2	58.10
CB 180	Nd	Nd	Nd	1.576	0.424	0.208	9.782	9.411	129.3	60.97
CB 194	Nd	Nd	Nd	9.711	11.75	7.300	439.8	309.0	1053	496.4
Total PCBs	0.197	0.003	0.239	12.68	12.73	8.835	722.2	546.9	1510	712.1

Note: * Nd: not determined(< 0.001)

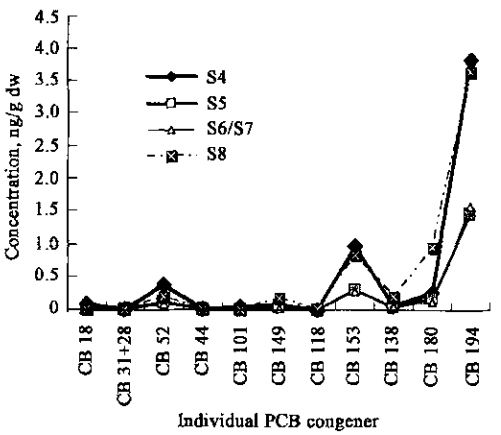

Fig.2 Distribution of individual PCBs congeners in different samples

Starting from the global scale, we examined emission histories and sources for selected contaminants focusing especially on the PCBs. Physical and chemical properties, transport processes in the environment (e.g. winds, currents), partitioning, and models are then used to identify, understand and illustrate the connection of the contaminant sources in industrial and agricultural regions in the south and eventual arrival of contaminants in remote regions. Within the Xiamen region, these contaminations showed how contaminants impacted on marine and terrestrial pathways and how they subsequently either removed to sinks or remained where they could enter the biosphere. A comparison PCBs sources within our sampling sites with PCBs imported through long-range transport, a comparison of natural vs. anthropogenic sources of PCBs in the Xiamen environment should be done to get a global understanding. The research and syntheses provide compelling evidence for close connectivity between the global emission of contaminants from industrial and agricultural activities and the local contamination.

For compounds that partition strongly onto particles, and for which the soil reservoir is most important, we have seen a quick in their arrival in Xiamen and some fractionation toward more volatile compounds(e.g. lower-chlorinated PCBs).

3 Conclusions

The total PCBs in sediment samples ranged from 2.141—6.065 ng/g dw. The concentration of total PCBs is 0.003—0.239 ng/g dw in vegetable samples, 8.835—12.73 ng/g in soil samples and 564.9—1510 ng/g dw in leaf samples. This contamination has been scientifically recognized as a manifestation of long-range transport and absorption of pollutants to the Xiamen areas. Problems caused by contaminants depend on their distinctive characteristics and how these interact with biogeochemical pathways. Since PCBs have been produced and released to the environment in large quantities, there is an urgent need to develop a robust monitoring program for their continued surveillance, to maintain an adequate level of environment protection and consequently to adopt an efficient risk assessment strategy to determine the likely impacts these compounds may have on the local ecosystems.

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