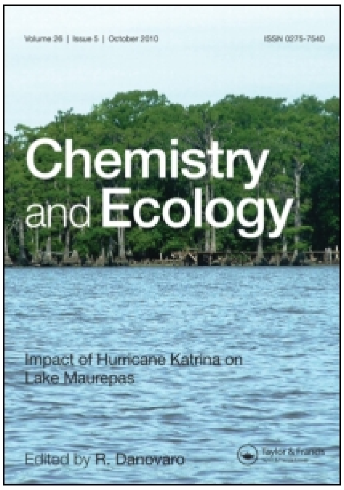

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The Distribution and Impacts of Dioxins and Polychlorinated Biphenyls in the Taiwan Er-Jen River

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THE DISTRIBUTION AND IMPACTS OF DIOXINS AND POLYCHLORINATED BIPHENYLS IN THE TAIWAN ER-JEN RIVER

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The distribution and impacts of polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/DFs) and polychlorinated biphenyls (PCBs) in the Taiwan Er-Jen River were investigated by monitoring fish distribution and measuring PCDD/DFs and PCBs levels in sediment and fish samples. Most of the fish collected are pollution-resistant species such as *Oreochromis* spp, *Liza macrolepsis*, *Channa* spp and *Megalops cyprinoides*. The most polluted river sections are in the upstream where large amount of waste effluents from households, livestock rearing and nearby dumping sites were discharged into the river, and the lower reaches of the river where metal recovery activities including open burning of waste electrical wire/scrap occurred. Sediment samples collected near the burning site show 2,3,7,8-TCDD toxicity equivalents (TEQ) ranging from 0.014 to 14.2 ng g^{-1} by PCDD/DFs and from 0.015 to 1.03 ng g^{-1} by coplanar PCBs (Co-PCBs). The fish samples show TEQ ranging from 0.029 to 0.615 ng g^{-1} by PCDD/DFs and from 0.012 to 0.12 ng g^{-1} by Co-PCBs. Possible PCDD/DFs sources are discharged PCBs, open burning of waste wire/scrap, and pentachlorophenol. The consumption of these fish will cause an average intake of 54 pg kg^{-1} d^{-1} TEQ. The prevention of direct discharge of livestock and the dumping of waste effuents into River Er-Jen as well as the control of metal recovery activities appears to be the first step toward the restoration of River Er-Jen.

KEY WORDS: Dioxins, polychlorinated biphenyls, fish, sediment, Taiwan river.

1 INTRODUCTION

The River Er-Jen in west southern Taiwan (Fig. 1) is among one of the heavily polluted rivers in Taiwan. A number of metal recovery activities, including acid washing and open burning of waste electrical wire/scrap, waste motors and electrical capacitors/transformers was considered previously the main polluting source. Waste water from households, livestock rearing and nearby dumping sites worsened the pollution further. Waste effluents containing large amount of pollutants ultimately found their way into River Er-Jen since the late 1960s. The extent to which these pollutants affect the ecosystem depends largely upon the quantity and nature of the particular compounds involved. With increasing fish breeding activities downstream, pollution in River Er-Jen has been recently the subject of intense public concern. Halogenated

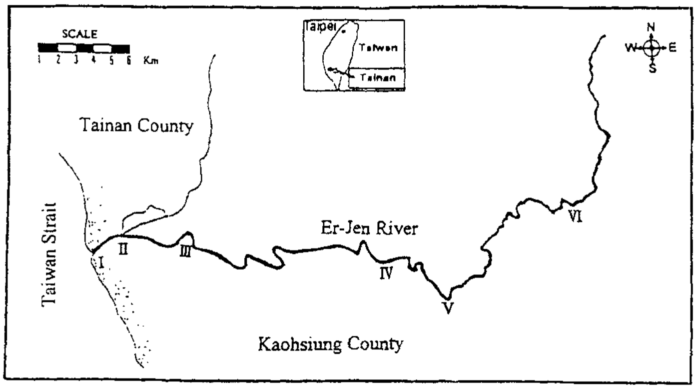

Figure 1 Sampling stations in River Er-Jen.

hydrocarbons, i.e., persistent and widely distributed polychlorinated dibenzo-*p*-dioxins and dibenzofurans (PCDD/DFs), and polychlorinated biphenyls (PCBs) (Travis and Hester, 1991) have received much attention. The effects of these compounds on aquatic ecosystems have been studied (Cooper, 1989; Cook *et al.*, 1991).

Previous works were done on PCDD/DFs in soils (Huang *et al.*, 1992; Lee *et al.*, 1994), sediments (Lee *et al.*, 1994; Lu *et al.*, 1994; Ling *et al.*, 1995) and in fish (Soong *et al.*, 1994; Ling *et al.*, 1994; Ling *et al.*, 1995) collected from River Er-Jen to determine the degree of pollution. Recent work has extended to other chlorinated compounds such as PCBs and organochlorine pesticides in fish (Ling and Huang, 1995) to gain a more comprehensive understanding about organochlorine pollution in the river Er-Jen. Other polluting sources were also investigated in a recent fish distribution survey (Wang *et al.*, 1994). This research attempts to understand the distribution and impacts of PCDD/DFs and PCBs in this river by monitoring fish distribution and measuring PCDD/DFs and PCBs levels in sediment and fish samples. The aim is to assess the ecological status of river and suggest means of restoration. The possible health impact upon man was assessed based on 2,3,7,8-TCDD toxicity equivalents (TEQ), defined as the summation of the toxicity times concentration for the 17 toxic PCDD/DFs (Safe, 1990) and three toxic coplanar PCBs (Co-PCBs).

EXPERIMENTAL METHODS

For the fish distribution study, fish were collected seasonally from 6 sampling stations along the River Er-Jen on January, April, and June 1993. For PCDD/DFs and PCBs study, a total of 11 sediment samples (S1 to S11, i.e., S1, S2, ..., S11) and 7 fish samples (F1 to F7) were collected. The sampling stations are shown in Figure 1. Most sediment samples were collected from the surface (depth 0–5 cm), except samples S4 and S7 (depth 40–50 cm) as well as S5 and S8 (depth 90–100 cm). Sample S1 was taken 0.5 km downstream of the burning site and was at the entrance of river/sea area (station I). Sample S2 was taken from the lower reach of the river near the burning site (station II). Samples S3, S4 and S5 were taken from the southern side of the river; whereas samples

S6, S7 and S8 were from the northern side of the river, all at station II. Samples S9, S10 and S11 were collected at station V where little burning activities were observed. Fish samples F3 (*Oreochromis* spp), F4 (*Liza macrolepsis*) and F6 (*Oreochromis* spp) were composites of 5 to 7 smaller fish. Other samples were of single fish. Samples F1 and F2 (*Megalops cyprinoides*), F3 and F4 were collected at station I. Samples F5 (*Megalops cyprinoides*) and F6 were collected near the burning site (station II). Sample F7 (*Clarias fuscus*) was collected at station V. The samples were analyzed using GC/MS for total PCBs, and isotope-dilution GC/MS for PCDD/DFs and Co-PCBs as described in the previous study (Ling *et al.*, 1995).

RESULTS AND DISCUSSION

Distribution of Fish Number and Species

The species and number of fish collected from the 6 sampling stations are listed in Table I. A total of 9 fish species were observed in downstream stations (I and II) and stations in the upper reaches of the river (III, IV, and V). By contrast, only 3 fish species were observed in upstream station VI. In the downstream, *Liza marcolepsis*, *Megalops cyprinoides* and *Oreochromis* spp, appear most frequently. The former two are peripheral freshwater fish. They live in estuarine or coastal water and enter river during the period of their lifecycle. The last one is a secondary freshwater fish. It can survive in either fresh water or sea water. In the upper reaches of the river, *Carassius auratus auratus*, *Clarias fuscus*, *Oreochromis* spp and *Channa* spp appear most frequently. They are all primary freshwater fish and have their lifecycle entirely in fresh water, except

Table I Distribution of fish species and number in the River Er-Jen.

	Fish number at Station					
Fish species	*I*	*II*	*III*	*IV*	*V*	*VI*
*Cyprinus carpio**	–	–	–	1	–	–
*Ctenopharyngodon idellus**	–	–	1	–	–	–
*Hemiculter kneri**	–	–	–	1	2	1
*Clarias fuscus**	–	–	4	2	8	–
*Carassius auratus auratus**	–	–	1	20	5	–
Channa spp*	–	–	–	9	7	14
*Tricogaster trichopterus**	–	–	1	–	–	–
*Anguilla japonica***	–	1	–	1	1	–
Oreochromis spp**	16	31	5	8	3	–
*Lates calcarifer****	–	1	–	–	–	–
*Scatophagus argus****	2	–	–	–	–	–
*Chanos chanos****	–	2	–	–	–	–
*Leiognathus splendens****	2	1	–	–	–	–
*Ambassis urotaenia****	2	1	–	–	–	–
*Megalops cyprinoides****	9	15	4	–	–	–
*Liza marcolepsis****	38	11	–	–	–	–

*Primary freshwater fish: fish have their lifecycle entirely in fresh water.
**Secondary freshwater fish: fish survive in either fresh water or sea water.
***Peripheral freshwater fish: fish live in either estuarine or coastal water and enter river during period of their lifecycle.

Oreochromis spp which is secondary freshwater fish. In the upstream, primary freshwater fish, *Channa* spp, appear most frequently. This distribution of fish species is consistent with the extent of tidal intrusion. The downstream is within the tidal head during high tide period. Consequently, the fish collected in these stations are mostly peripheral and secondary freshwater fish. Most of the fish collected in this study are not native and are known to adopt themselves well to polluted environment. The relatively smaller number and species of fish collected at station VI, indicating that the water quality around this river section is severe. The colour of the water appears dark with a dissolved oxygen value < 2 ppm. This might be related to the direct discharge of large amount of waste effluents from households, livestock rearing and dumping sites into this section of river.

Concentration of PCDD/DFs and PCBs in Sediment

Table II summarizes the concentrations of PCDD/DFs, total PCBs, Co-PCBs (i.e., PCB # 77, 126, and 169) and the TEQ in sediment samples. The TEQ values from 0.002 ng g^{-1} in sample S10 to 14.2 ng g^{-1} in sample S2, taken from the middle of the river near the burning site. The 14.2 ng g^{-1} TEQ value is much higher than those reported in

Table II Concentrations (ng g^{-1}, dried matter) of PCDD/DFs, total PCBs, Co-PCBs and the TEQ in sediment samples.

Analyte	*Sample No.*										
	S1	*S2*	*S3*	*S4*	*S5*	*S6*	*S7*	*S8*	*S9*	*S10*	*S11*
2,3,7,8-TCDD	–	0.115	0.001	0.005	0.003	0.002	0.003	0.003	0.002	–	0.003
1,2,3,7,8-PeCDD	–	0.807	0.006	0.010	0.011	0.005	0.23	0.006	0.003	0.001	0.007
1,2,3,4,7,8-HxCDD	0.001	1.434	0.008	0.009	0.007	0.015	0.022	0.013	0.006	0.003	0.009
1,2,3,6,7,8-HxCDD	–	2.604	0.007	0.010	0.020	0.028	0.36	0.029	0.003	0.001	0.006
1,2,3,7,8,9-HxCDD	–	4.313	0.007	0.021	0.042	0.056	0.068	0.048	0.006	–	0.006
1,2,3,4,6,7,8-HpCDD	0.013	19.63	0.031	0.113	0.218	0.365	0.334	0.254	0.014	0.013	0.040
OCDD	0.023	15.81	0.053	0.166	2.155	0.549	0.473	0.396	0.028	0.022	0.086
2,3,7,8-TCDF	0.006	11.27	0.015	0.045	0.057	0.034	0.121	0.058	0.002	–	0.019
1,2,3,7,8-PeCDF	–	4.029	0.008	0.016	0.044	0.027	0.047	0.040	0.004	–	0.009
2,3,4,7,8-PeCDF	0.004	13.39	0.005	0.029	0.066	0.055	0.95	0.065	0.004	0.001	0.014
1,2,3,4,7,8-HxCDF	0.006	22.81	0.019	0.088	0.203	0.260	0.285	0.229	0.003	0.002	0.023
1,2,3,6,7,8-HxCDF	0.001	7.535	0.006	0.026	0.065	0.086	0.093	0.071	0.003	0.001	0.007
1,2,3,7,8,9-HxCDF	–	0.420	0.002	0.002	0.024	0.021	0.026	0.023	0.002	–	0.002
2,3,4,6,8,9-HxCDF	0.003	11.06	0.008	0.037	0.076	0.105	0.130	0.100	0.002	–	0.011
1,2,3,4,6,7,8-HpCDF	0.017	39.52	0.036	0.138	0.371	0.735	0.570	0.489	0.020	0.014	0.051
1,2,3,4,7,8,9-HpCDF	0.001	2.682	–	0.010	0.032	0.158	0.048	0.041	0.003	–	0.006
OCDF	0.016	10.57	0.024	0.085	0.156	0.695	0.207	0.145	0.023	0.026	0.022
TEQ	0.004	14.2	0.014	0.052	0.102	0.108	0.152	0.106	0.009	0.002	0.024
PCB # 77	*	24.23	1.09	5.76	2.94	0.90	2.76	3.74	–	–	0.53
PCB # 126	*	7.56	0.06	0.44	0.26	0.05	0.19	0.28	–	–	0.03
PCB # 169	*	6.00	0.005	0.03	0.01	0.01	0.02	0.02	–	–	0.004
Total PCBs	1555	13615	1492	*	4639	871	4431	4768	8	4	1555
TEQ	*	1.03	0.017	0.103	0.056	0.015	0.048	0.066	–	–	0.009

*Not determined.

other rivers (Tong *et al.*, 1990; Kjeller *et al.*, 1990, Götz *et al.*, 1990, 1993). The low TEQ values in samples S9, S10 and S11 is understandable since little burning activities occurred in the upstream river area. The unexpected low TEQ value (0.004 ng g^{-1}) in downstream sample S1 might be due to inadequate sampling since S1 is a surface sediment and reflects only the recent pollution. The flowing water might continuously drift the surface sediment away and yielded low TEQ value. This speculation is supported by the low TEQ value (0.014 ng g^{-1}) in another surface sediment sample S3. In comparison with S4 and S5 (or S7 and S8), the relatively low TEQ value in sample S3 (or S6) also reflects recent pollution is less serious than it was before. The lack of sediment deposition histories prevents us from estimating the period during which the surface sediment was deposited. Nevertheless, the ban on importing waste metal in general by the R.O.C. government at the beginning of 1993 accords with this finding. In comparison with samples S3 and S4, the relatively high TEQ value in samples S6, S7 and S8 reflects that most burning activities occurred on the northern side of River Er-Jen. The extremely high TEQ value in sample S2 might be ascribed to the strong leaching from sediments due to high flow that is characteristic of rivers in the west coast of Taiwan.

The PCDD/DFs patterns in sediment are shown in Figure 2. Samples with TEQ < 0.010 ng g^{-1}, i.e., S1, S9, and S10, have similar profiles. The total concentrations of PCDFs are about 2 to 3 times higher than those of PCCDs with the same degree of chlorination. Similar patterns were found in emissions from municipal solid waste incinerators. Therefore, the calculated PCDD/DFs are considered as background (Rappe 1994). Samples S2 to S8 show similar patterns and reflect the fact that they were taken near the burning site. The ratios of tetra-, penta-, and hexa-CDFs concentration to similarly chlorinated PCDDs resemble those in Aroclor 1242 and 1254. In Taiwan, most electrical capacitors contain Aroclor 1242 and electrical transformers contain Aroclor 1242 and 1254. Hence, it is to assign that PCBs as the additional polluting source. The high level of OCDD in these samples might be due to the wide use of pentachlorophenol in Taiwan before its ban in 1989.

Comparing the calculated toxic effects between PCDD/DFs and Co-PCBs, the former contributes more toxicity in samples taken near the burning site, i.e., samples S2, S5, S6, S7, and S8 (Fig. 3). This fact indicates that open burning is the main organochlorine polluting source in the Er-Jen. PCDD/DFs are responsible for more than 90% of the calculated toxicity in the most contaminated sample, S2. Figure 3 shows that the TEQ value of Co-PCBs to PCDD/DFs increases from recently deposited sediment, S3 (or S6), to previously deposited sediments S4 (or S7 and S8). This finding indicates that the amount of Aroclor mixture from disposed electrical transformers/capacitors discharged into the Er-Jen has been reduced recently. This fact reflects the ban on using PCBs and strict regulation for storing waste electrical transformers/capacitors by the R.O.C. government in 1988.

Concentration of PCDD/DFs and PCBs in Fish

Table III summarizes the concentrations of PCDD/DFs, total PCBs, Co-PCBs (i.e., PCB # 77, 126, and 169) and the TEQ values in fish samples. The TEQ values range from 0.029 ng g^{-1} in sample F2 to 0.615 ng g^{-1} in sample F7. The TEQ values found

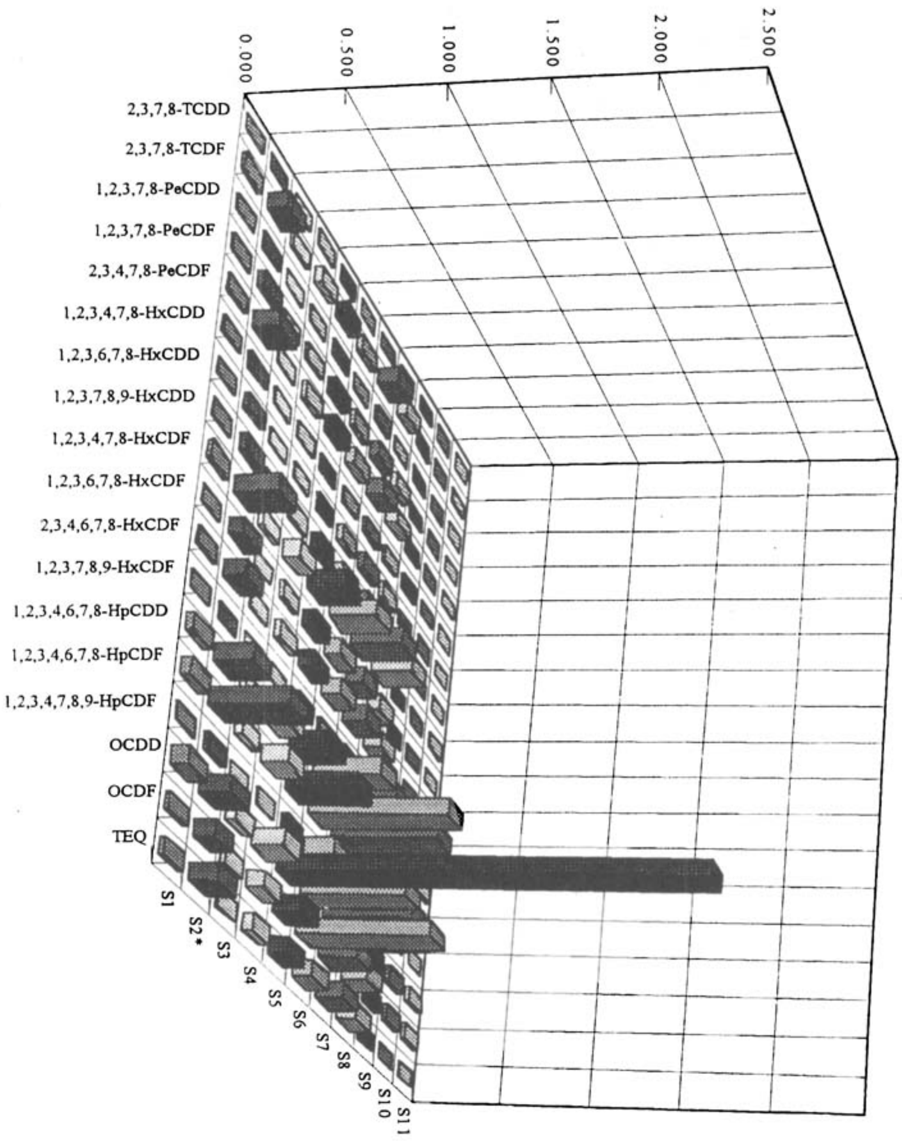

Figure 2 PCDD/DFs patterns in sediment samples. (*S2 concentration is much higher than the other samples and is scaled down by 100).

are much higher than those found in fish (0.008 to 0.039 ng g^{-1}) from river Rhine and Neckar in south-west Germany (Frommberger, 1991). The average TEQ value in fish from this study is 0.188 ng g^{-1} which is much higher than the 0.012 ng g^{-1} from a recent U.S.A. national survey study (Kuehl *et al.*, 1994). Although the difference in fish species and size might cause the difference in TEQ values, the higher TEQ values found in this

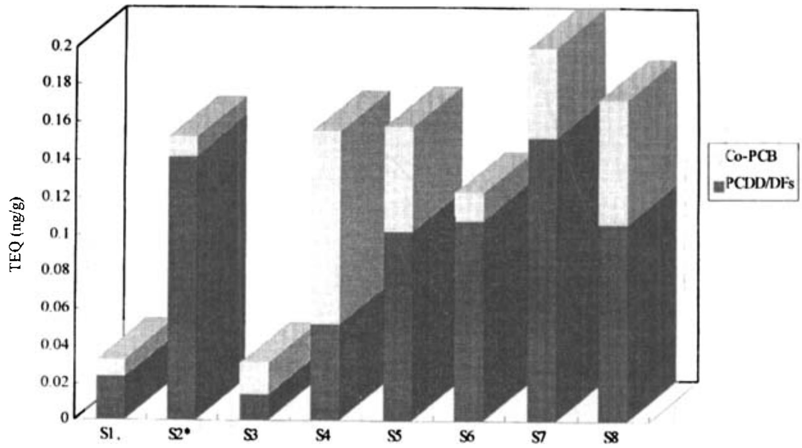

Figure 3 TEQ by Co-PCBs and PCDD/DFs in sediment samples. (*S2 TCDD-EQ ratio is much higher than the other samples and is scaled down by 100).

Table III Concentrations (ng g^{-1}, dried matter) of PCDD/DFs, total PCBs, Co-PCBs and the TEQ in fish samples.*

Analyte	*Sample No.*						
	F1	*F2*	*F3*	*F4*	*F5*	*F6*	*F7*
2,3,7,8-TCDD	0.016	0.007	0.018	0.009	0.041	0.087	0.008
1,2,3,7,8-PeCDD	0.007	0.011	0.022	0.017	0.112	0.071	0.040
1,2,3,4,7,8-HxCDD	0.017	0.028	0.041	0.036	0.139	0.145	0.132
1,2,3,6,7,8-HxCDD	0.010	0.007	0.017	0.018	0.116	0.013	0.281
1,2,3,7,8,9-HxCDD	0.011	0.020	0.023	0.053	0.111	0.007	0.314
1,2,3,4,6,7,8-HpCDD	0.064	0.059	0.593	1.083	0.452	0.268	16.457
OCDD	0.324	0.723	84.78	10.93	1.298	3.418	156.8
2,3,7,8-TCDF	0.010	0.008	0.018	0.006	0.009	0.078	0.007
1,2,3,7,8-PeCDF	0.007	0.006	0.024	0.022	0.093	0.061	0.011
2,3,4,7,8-PeCDF	0.003	0.007	0.023	0.082	0.033	0.003	0.009
1,2,3,4,7,8-HxCDF	0.014	0.015	0.012	0.019	0.071	0.127	0.070
1,2,3,6,7,8-HxCDF	0.004	0.006	0.004	0.011	0.033	0.005	0.045
1,2,3,7,8,9-HxCDF	0.003	0.012	0.006	0.011	0.066	0.006	0.015
2,3,4,6,8,9-HxCDF	0.009	0.011	0.118	0.017	0.023	0.012	0.111
1,2,3,4,6,7,8-HpCDF	0.068	0.059	0.397	0.732	0.447	0.246	10.278
1,2,3,4,7,8,9-HpCDF	0.025	0.019	0.027	0.032	0.089	0.010	0.027
OCDF	0.142	0.166	8.221	3.734	0.998	0.937	60.19
TEQ	0.031	0.029	0.168	0.110	0.188	0.177	0.615
PCB# 77	0.96	1.09	0.62	2.67	0.58	0.77	0.09
PCB# 126	0.79	0.73	0.17	0.86	0.53	0.26	0.10
PCB# 169	0.07	0.29	0.02	0.08	0.06	0.07	0.02
Total PCBs	1924	2598	315	2352	902	502	123
TEQ	0.092	0.098	0.024	0.12	0.062	0.037	0.012

*F1, F2 and F5 (*Megalops cyprinoides*), F3 (*Orechromis* spp), F4 (*Liza macrolepsis*), F6 (*Oreochromis* spp, F7 (*Clarias fuscus*).

study nevertheless indicate that River Er-Jen is heavily polluted by PCDD/DFs. Unlike the sediment samples, no clear relationship between TEQ values and sampling locations was found. this might be ascribed to the migratory characteristics of the fish grown in River Er-Jen.

The PCDD/DFs patterns in fish samples are shown in Figure 4. Samples F1, F2 and F5, all *Megalops cyprinoides* collected from the same area, show similar PCDD/DFs

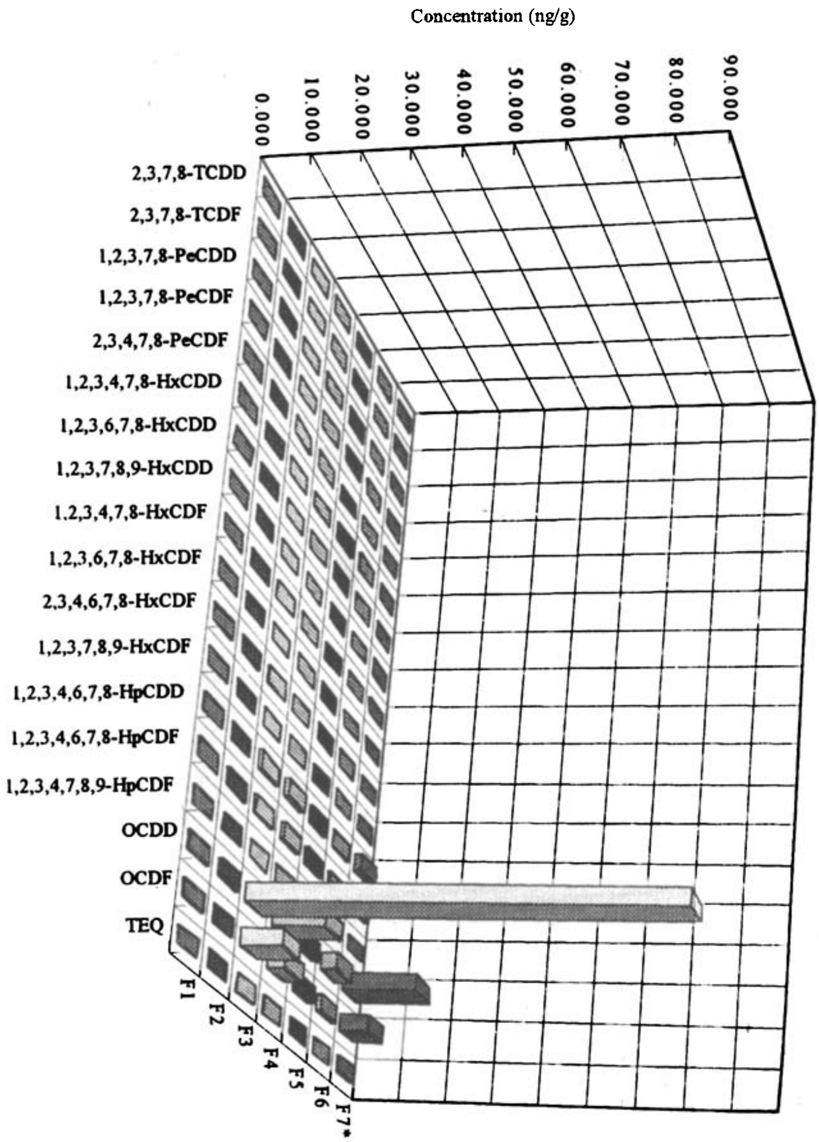

Figure 4 PCDD/DFs patterns in fish samples. (*F7 concentration is much higher than the other samples and is scaled down by 100).

patterns. Samples F5 and F6 show relatively higher levels of tetra- and penta-CDDs and CDFs, indicating that PCBs might be the source of PCDD/DFs. This finding accords with the fact that these two samples were collected near the burning site. Other samples show different PCDD/DFs patterns. Upon close inspection, more contaminated fish such as F3, F5 and F7 show higher level of hepta- and octa-CDDs and CDFs. These differences might be due to the species-specific metabolism and/or intake of the different congeners in fish. Further investigations are needed to clarify this speculation. The concentration of PCDDs are generally higher than those of PCDFs. Similar observations were reported before (Frommberger, 1991). The unusually high level of OCDD found in sample F7 collected upstream far away from the burning site, indicating that PCP might be the source of PCDD/DFs. Based on an average daily fish consumption of 15 g for an adult with 70 kg body weight, an average intake is estimated to be 40 pg $kg^{-1}d^{-1}$ (daily consuming 40 pg of fish per kg body weight of the consumer). The consumption of these fish will exceed the tolerable daily intake (TDI) of 10 pg $kg^{-1}d^{-1}$ recommended by the WHO (Kello and Yrjanheikki, 1992).

Unlike sediment samples, the concentrations of total PCBs and Co-PCBs do not parallel to those of PCDD/DFs in these fish samples. Samples F1, F2 and F4 show relatively high concentrations of total PCBs ranging from 1924 to 2598 ng g^{-1} and TEQ values from about 0.092 to 0.12 ng g^{-1} PCBs are responsible for 50 to 75% of the calculated toxicity in these samples (Fig. 5). Taking the TEQ from PCBs into consideration, the consumption of these fish will cause an average intake of 54 pg kg^{-1} d^{-1}, which is 5 times of the TDI recommended by WHO. It is important to note that normal background exposure viz. common food consumption, which is not currently available in Taiwan, and exposure to the consumption of other contaminated species such as the 4,4′-DDE present in these fish samples (Ling and Huang, 1995) will further elevate the daily intake value of TEQ. Restrictions on the consumption of fish caught from the river Er-Jen is therefore warranted.

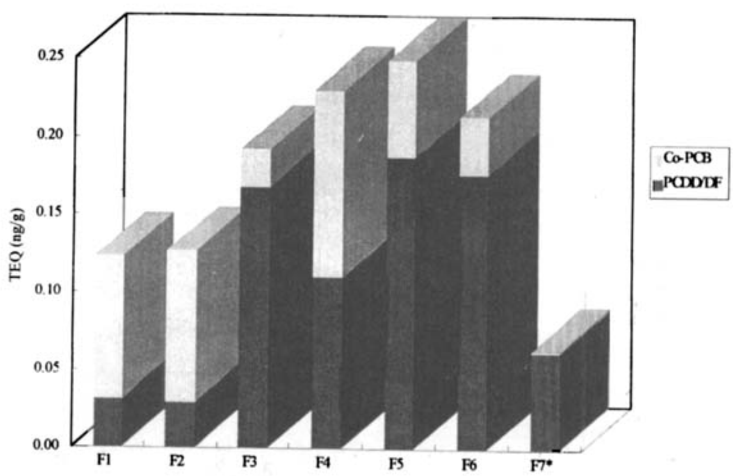

Figure 5 TEQ by Co-PCBs and PCDD/DFs in fish samples. (*F2 TCDD-EQ ratio is much higher than the other samples and is scaled down by 100).

CONCLUSIONS

Ecological status of River Er-Jen, i.e., due to pollution from both livestock rearing and dump waste effluents, was investigated. Most of the fish collected are non-native and pollution-resistant species such as *Oreochromis* spp, *Liza macrolepsis*, *Channa* spp and *Megalops cyprinoides*. Sediment samples collected near the burning site show TEQ values from 0.014 to 14.2 ng g^{-1} by PCDD/DFs and from 0.015 to 1.03 ng g^{-1} by Co-PCBs. The fish samples show TEQ values from 0.029 to 0.615 ng g^{-1} by PCDD/DFs and from 0.012 to 0.12 ng g^{-1} by Co-PCBs. Possible PCDD/DFs sources are discharged PCBs, open burning activities and pentachlorophenol. The consumption of these fish will cause an average intake of 54 pg kg^{-1} d^{-1} TEQ. Restrictions on the consumption of fish caught from the River Er-Jen is therefore warranted. The prevention of direct discharge of livestock rearing and dumping waste effluents into River Er-Jen as well as the control of metal reclamation activities appear to be the first step toward the restoration of River Er-Jen.

Acknowledgements

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