



# Identifying the potential sources of di-(2-ethylhexyl) phthalate contamination in the sediment of the Houjing River in southern Taiwan

Chitsan Lin<sup>a,\*</sup>, Cheng-Jin Lee<sup>a,1</sup>, Wen-Ming Mao<sup>a,1</sup>, Farhad Nadim<sup>b,2</sup>

<sup>a</sup> National Kaohsiung Marine University, Department of Marine Environmental Engineering, 142, Haijhuang Road, Nanzih District, Kaohsiung 81157, Taiwan

<sup>b</sup> University of Connecticut, Department of Civil and Environmental Engineering, Unit 2037, Storrs, CT, 06269-2037, USA

## ARTICLE INFO

### Article history:

Received 9 February 2008

Received in revised form 14 March 2008

Accepted 18 March 2008

Available online 22 March 2008

### Keywords:

River pollution prevention

Sediment contamination

Endocrine disruptor

Environmental forensics

Contamination sources characterization

## ABSTRACT

Sediment samples were analyzed for di-(2-ethylhexyl) phthalate (DEHP), an organic endocrine disruptor, in Houjing River in southern Taiwan. The average DEHP concentration at 10 sampling locations, spanning from upper, middle, and lower segments of the stream, was calculated at  $3.81 \pm 6.36 \text{ mg kg}^{-1}$  dry wt. Highest concentration was recorded at the Jhongsing Bridge ( $20.22 \text{ mg kg}^{-1}$  dry wt.) near the Dashe Industrial Park, followed by the Renwu Bridge ( $8.93 \text{ mg kg}^{-1}$  dry wt.) near the Renwu Industrial Park. The surface sediment concentration of DEHP was found to be higher in the dry season (October and December), and lower in the wet (flood) season (August), indicating that sources of DEHP remained active and continued to recharge the Houjing River. Vertical sediment core analysis revealed that highest concentration occurred at the depth of 40–60 cm, indicating that historical discharges of DEHP may have been higher than recent years. Domestic comparison of DEHP concentrations in sediment from highest to lowest could be categorized as northern, southern, central, and eastern Taiwan, respectively, and seemed to be positively correlated with population density and/or industrial activity. Compared to other countries, DEHP concentration of the Houjing River was relatively higher than rivers studied in Japan, Germany, Italy, and Malaysia, and was relatively lower than the Aire and Trent Rivers in the United Kingdom.

© 2008 Elsevier B.V. All rights reserved.

## 1. Introduction

Di-(2-ethylhexyl) phthalate (DEHP) is a very popular synthetic product, used widely as a plasticizer in plastic industry. Annual production of phthalate esters (PAEs) in European Union countries is about 1 million tons, and global production is about 4.3 million tons annually [1–2]. Approximately 50% of PAEs are DEHP. Owing to its environmental impacts and health concerns, DEHP was identified as an organic endocrine disruptor [1–4] and is regulated in many countries. Numerous investigations have revealed that DEHP had been widely spread in our living environment.

Problems associated with DEHP have been identified in river and estuary systems of the world in the past decade, and have led to worldwide attention to this problem. In a monitoring network carried out to survey 45 domestic major rivers by the Environmental Protection Agency of Japan [5], DEHP concentrations in the sediment samples were identified in the

range of  $0.009\text{--}3.5 \text{ mg kg}^{-1}$  dry wt. In Malaysia, Tan [6] reported  $0.49\text{--}15.0 \text{ mg kg}^{-1}$  dry wt. DEHP in Klang River estuary sediment. In Germany, Fromme et al. [7] reported DEHP sediment concentration of  $0.21\text{--}8.44 \text{ mg kg}^{-1}$  dry wt. range for 35 rivers. Vitali et al. [8] reported  $0.058\text{--}0.487 \text{ mg kg}^{-1}$  dry wt. DEHP concentration for various rivers in central Italy. Ray et al. [9] reported  $0.04\text{--}16.0 \text{ mg kg}^{-1}$  dry wt. DEHP in Nueces estuary, in Texas in southern United States.

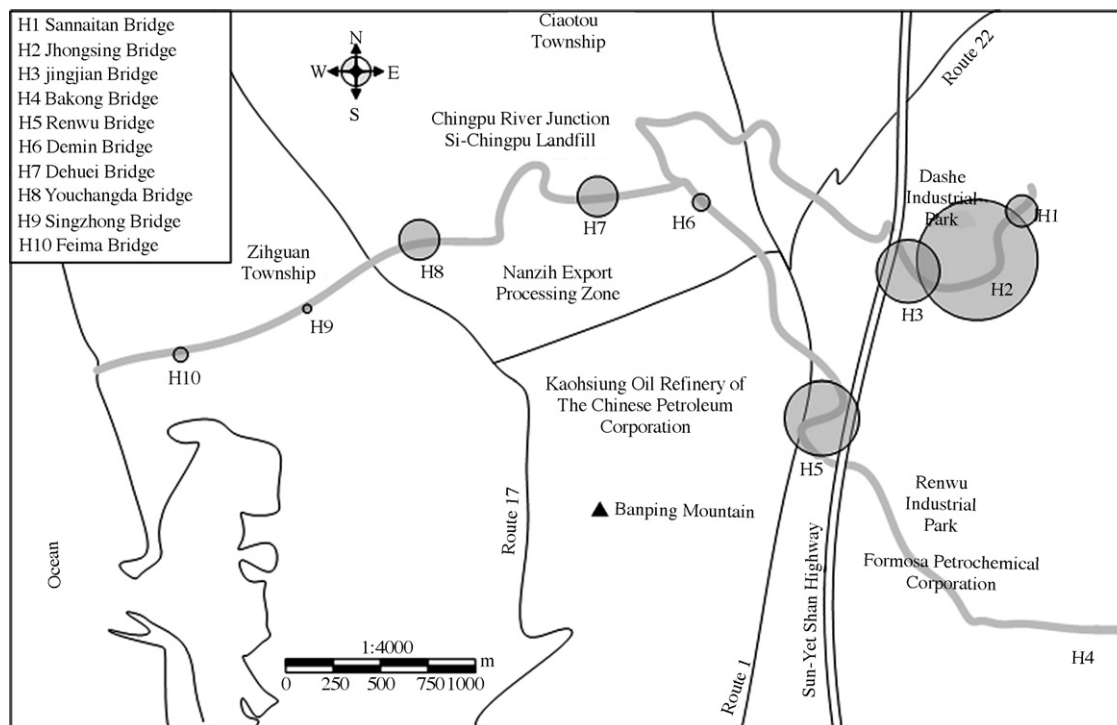
Huang et al. [10] analyzed river sediments in northern Taiwan, and found that DEHP ( $\text{ND}\text{--}8.97 \text{ mg kg}^{-1}$  dry wt.) was mainly distributed in the downstream sections of rivers where population is relatively higher. In our previous study [11], 128 semi-volatile organic compounds (SVOCs) in the Houjing River in southern Taiwan were surveyed, and the results indicated that of the total SVOCs, PAEs accounted for 57%, followed by phenols (38%) > chlorinated hydrocarbons (3%) > others (1%)  $\geq$  poly aromatic hydrocarbons (1%). The highest level of PAEs found in the Houjing River sediments, was identified as DEHP at 84%, followed by dibutylphthalate (DBP) at 15%, and DEP (diethylphthalate) at 1%. A river water quality survey project, carried out by the Taiwan National Institute of Environmental Analysis (Taiwan NIEA) [12], also documented the fact of DEHP contamination in rivers in Taiwan. Contamination of riparian systems in Taiwan necessitates the establishment of a DEHP discharge elimination policy

\* Corresponding author. Tel.: +886 7 3651472; fax: +886 7 3651472.

E-mail addresses: [ctlin@mail.nkmu.edu.tw](mailto:ctlin@mail.nkmu.edu.tw) (C. Lin), [fan02001@engr.uconn.edu](mailto:fan02001@engr.uconn.edu) (F. Nadim).

<sup>1</sup> Tel.: +886 7 3651472; fax: +886 7 3651472.

<sup>2</sup> Tel.: +1 860 4874625.



**Fig. 1.** Geographical distribution of sampling locations and spatial characterization of DEHP contamination in the sediment of Houjing River. (Note: H1 = 1.37, H2 = 20.22, H3 = 3.68, H4 = 0.07, H5 = 8.93, H6 = 0.26, H7 = 1.47, H8 = 1.82, H9 = 0.09, H10 = 0.18; unit =  $\text{mg kg}^{-1}$  dry wt.)

that should include identification of the sources of discharges and establishment of discharge control regulations. In this research study, the authors applied gas chromatograph/mass spectrometer (GC/MS) analytical technique to survey SVOC contaminations in the Houjing River sediments, and utilized spatially and temporally forensic characterization techniques to unveil the potential discharge sources, and to establish DEHP sediment contamination database for southern Taiwan.

## 2. Methodology

### 2.1. Sampling design

Houjing River originates from two upstream branches (Fig. 1). The Dashe and Renwu branches that flow through the Dashe and Renwu Industrial Parks, respectively. The two streams merge at the Si-Chingpu Landfill, and extend all the way down to the Taiwan Strait. There are three sampling locations in each upstream branch and four sampling locations after the two streams merged (Fig. 1). Sampling locations were selected to cover the upstream and downstream sections of all potential sources of DEHP and to characterize concentration changes. For example, sampling stations H4 and H5 were chosen to assess the background and post-discharge levels of DEHP from the Renwu Industrial Park, respectively. Similarly, H1, H2 and H3 were designed to characterize the contribution of the Dashe Industrial Park to the river system.

### 2.2. Sediment sampling method

The Taiwan National Institute of Environmental Analysis (NIEA) Method – S104.30C [13] was adopted to collect surface (0–15 cm) sediment samples. For each sampling location, three sediment samples were collected randomly in the stream and composite into one sample, sealed with Teflon-lined cap in a 500 mL open-mouth brown glass bottle. Sediment cores were collected with

Teflon-lined sampling tubes with direct push method. Samples were well preserved and stored in a 4 °C environment before analysis.

### 2.3. Sample treatment and analysis

Taiwan NIEA M167.00C [14] and M731.00C [15] methods were adopted to extract SVOCs from sediment and followed by a GC/MS system for compound separation, identification and quantification. Ten grams of room/cool dried sediment was used for extraction, followed by adding 20 g of anhydrous sodium sulfate to remove water, and 10  $\mu\text{L}$  of surrogate standard solution was added before performing the dichloromethane (DCM) ultrasonic extraction procedure. After three ultra sonic extraction processes, all DCM phases were collected and concentrated by a Turbo Vap II concentrator before being injected into the GC/MS system (Agilent 6890N GC equipped with an Agilent 5973N MS) for analysis.

Injection port temperature was set at 250 °C; carrier gas was helium at 1  $\text{mL min}^{-1}$  rate; and splitless mode was adopted for GC operation. Capillary column used for analysis was HP-5 MS, 30 m  $\times$  0.25 mm  $\times$  0.25  $\mu\text{m}$ . The system also included an Agilent 6873 automatic injector that injects 1  $\mu\text{L}$  per injection. The oven temperature was programmed to start at 45 °C for 1 min and was raised to 310 °C at a rate of 10 °C  $\text{min}^{-1}$  and maintained for 7 min. Temperature was then raised to 320 °C at a rate of 10 °C  $\text{min}^{-1}$  and maintained for 2.5 min. All chemicals used in this study are reagent grade and all standards were NIST traceable quality. Activated carbon was used to remove organic constituents from the de-ionized water.

### 2.4. Quality assurance and quality control practices

Both Taiwan NIEA M731.00C [15] and USEPA Method 8270 [16] QA/QC protocols were followed to ensure the quality of the data. Daily quality control practices mainly included the following:

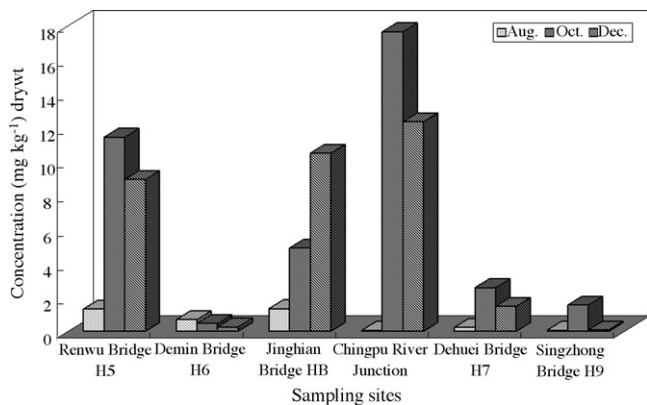


Fig. 2. Comparison of sediment DEHP concentration in dry and flood season.

1. Daily performance of Decafluorotriphenylphosphine (DFTPP) standard check to calibrate the mass spectrum detector (MSD) before the start of analysis.
2. Performing system proficiency calibration check (SPCC) once every 12 h of analysis. Relative response factors must be greater than 0.05.
3. Performing calibration curve verification check to ensure %deviation of each compound is less than 20%.
4. For every 10 samples or every batch of samples, performing blank tests, sample duplicate or matrix spike duplicate, and spike recovery test.
5. Checking the surrogate recovery rates.
6. Internal standards were added before injection into GC/MS for quantification.

Note that DEHP is very common in laboratory plastic wares (e.g., bottles, pipettes, plastic gloves, and filter papers) and it can be difficult to obtain clean blanks. Laboratory dishes and flasks used during this study were free of plastic to minimize potential DEHP contamination. In this research, blanks showed nondetectable levels of DEHP indicating that there was no contamination during sampling,

storage, or sample extraction procedures. The sample duplicates (RPD < 20%), blank spike recoveries ( $88 \pm 17\%$ ), and matrix spike and matrix spike duplicate recoveries ( $77 \pm 29\%$ ) were all within reasonable range.

### 3. Results and discussion

#### 3.1. Comparison of sediment DEHP concentration during wet (flood) and dry seasons

The DEHP concentration monitored at seven sampling locations (bridges) during the month of August, October, and December in the year of 2005 are presented in Fig. 2. In the summer of 2005 (July, August, and September), seven typhoons landed in Taiwan. Total precipitations of the three months were recorded at 934 mm for a 30-y average. In most occasions, typhoons were associated with plenty of precipitation that led to cleaning of the surface sediment. If DEHP discharging sources remained active, sediment concentrations would be expected to accumulate when dry season comes. On the other hand, the sediment concentrations would not be expected to rise, if the discharging sources were inactive. The results indicate DEHP concentration in October and December (dry season) were much higher than that of August (flood season), especially at the sampling locations downstream of potential sources, for example, Renwu Bridge, Jingjian Bridge, and Chingpu River junction. The above observation indicates that current DEHP sources remain active with continuous discharging of DEHP into the Houjing River. To eliminate the sources of DEHP discharge into the Houjing River, pollution prevention plans must be developed and enforced.

#### 3.2. Spatial characterization of DEHP contamination of the Houjing River

DEHP spatial characterization samplings were performed in December 2006, for the 10 sampling locations (H1–H10). Using the area of the shaded circle to represent DEHP concentration in relative scales, Fig. 1 implies that the highest concentration occurred at H2 (Jhongsing Bridge,  $20.22 \text{ mg kg}^{-1} \text{ dry wt.}$ ) in

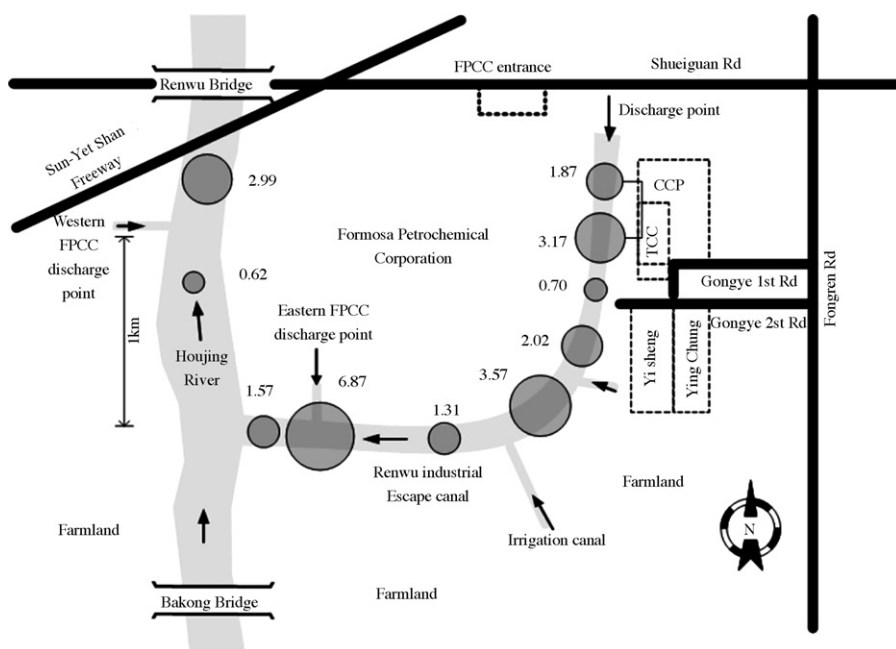


Fig. 3. Spatial characterization of DEHP sediment contamination at the Renwu Industrial Park.

the Dashe Industrial Park area where as H1 (Sannaitan Bridge,  $1.37 \text{ mg kg}^{-1}$  dry wt.) the background site was relatively low in concentration. The second highest concentration occurred at H5 (Renwu Bridge,  $8.93 \text{ mg kg}^{-1}$  dry wt.) located at the downstream end of the Renwu Industrial Park where as H4 (Bakong Bridge) the background site was nondetectable. Therefore, it is conclusive that the potential DEHP discharging sources are located at the Dashe Industrial Park, in between H1 (Sannaitan Bridge) and H2 (Jhongsing Bridge) that are only 600m apart and with four industrial entities present. Studying the Renwu branch, the potential DEHP discharging sources were found to be located at the Renwu Industrial Park, in between Bakong (H4) and the Renwu Bridges (H5) that are about 1.5 km apart and encompass the entire Renwu Industrial Park that requires further detail investigation for the potential DEHP sources. Also shown on Fig. 1, it seems that DEHP concentration in the Houjing River is non-monotonic with the maximum concentration very close to the discharging source. The observed transport behavior may lead to their environmental concerns localized, thus affect on the future cleanup consideration.

### 3.3. Spatial characterization of DEHP contamination at the Renwu Industrial Park

To further pinpoint potential DEHP discharging sources in the Renwu Industrial Park, the authors carried out another stage of spatial characterization sampling, and the analytical results are depicted in Fig. 3. The DEHP concentrations are shown with shaded circles in relative scales. It is clear that the concentration at the background location (Bakong Bridge, H4) was nearly negligible ( $0.07 \text{ mg kg}^{-1}$  dry wt.). But at the downstream point, near the Renwu Bridge (H5), DEHP concentration increased to  $2.99 \text{ mg kg}^{-1}$  dry wt. indicating that the potential sources were located in between, including all industries discharging their effluents into the Renwu Industrial Escape Canal that merges into the Houjing River. Spatial characterization of the Renwu Industrial Escape Canal indicates rather complicated distributions with fluctuating concentrations. However, it can also be emphasized that the sediment collected at the effluent discharge point of the eastern Formosa Petrochemical Corporation (FPCC) was recorded as the highest value at  $6.87 \text{ mg kg}^{-1}$  dry wt. Also in the Houjing River, the sample before western FPCC discharge point was recorded at  $0.62 \text{ mg kg}^{-1}$  dry wt. while as the point after was increased to  $2.99 \text{ mg kg}^{-1}$  dry wt.

### 3.4. Vertical DEHP distribution of sediment cores collected at the Dehuei Bridge (H7)

Vertical sediment cores were collected to investigate the historical release of DEHP into the Houjing River. An important factor to consider while collecting core sample was to make sure that the core was not disturbed during the time of preservation. Moreover, the deeper the core, the more the data would be provided. After a field survey trip, a dead corner close to the Dehuei Bridge (H7) where one can obtain deeper core sediment was selected. At this sampling location, the sediment did not indicate any sign of previous disturbance.

The DEHP concentration in depth (below ground surface, bgs) is presented in Fig. 4. As indicated in Fig. 4, the higher concentration occurred at the depth of 40–60 cm bgs, and the lower concentration occurred at the depth in between 0 and 30 cm bgs. Higher concentrations in deeper depths may be related to more DEHP discharge in the past most likely due to less stringent regulatory control. Lower concentrations in the upper layer suggested that the current discharges might have been reduced. Presence of higher DEHP concentrations in deeper cores corresponds well with the fact that

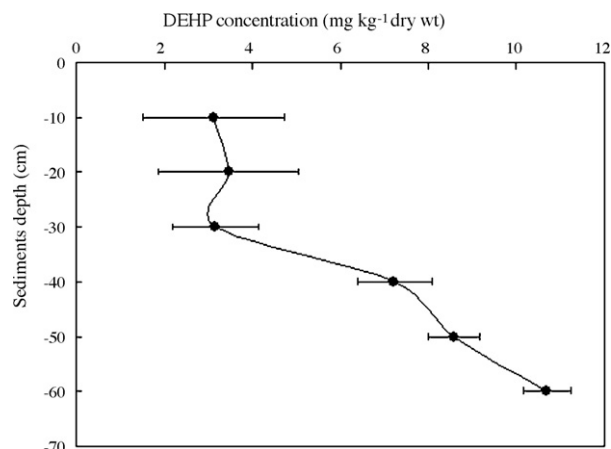


Fig. 4. Vertical DEHP distribution of sediment cores collected at the Dehuei Bridge.

DEHP could accumulate and is resistant to degradation. The above finding also explains why DEHP was regulated as one of the major organic endocrine disruptors. For future studies, collection of more than one sediment core at more than one sampling location and application of age dating techniques are suggested. Multiple sampling locations will provide data that could depict the sediment age versus depth, and delineates the historical distribution of DEHP in river sediment.

Furthermore, it is worth mentioning that about 300 and 1000 m downstream of the sediment core sampling location (H7, Dehuei Bridge), there are two water gates that divert the Houjing River water for irrigation of 750 ha of agriculture where about 80% of the areas are paddy fields, and about 20% of the land is allocated for growing vegetables and limited animal husbandry activities. Findings of this study prompt the need for a complete future survey for safe usage of the river water intended for agriculture and irrigation purposes where livestock and people, specifically farmers are frequently exposed to the river water.

### 3.5. Comparison of DEHP contamination in domestic river sediments

The DEHP contamination data of this study were compared with other domestic sediment contamination data in Taiwan. Huang et al. [10] reported DEHP sediment contamination data for some major rivers in Taiwan. Wang et al. [12] also reported DEHP

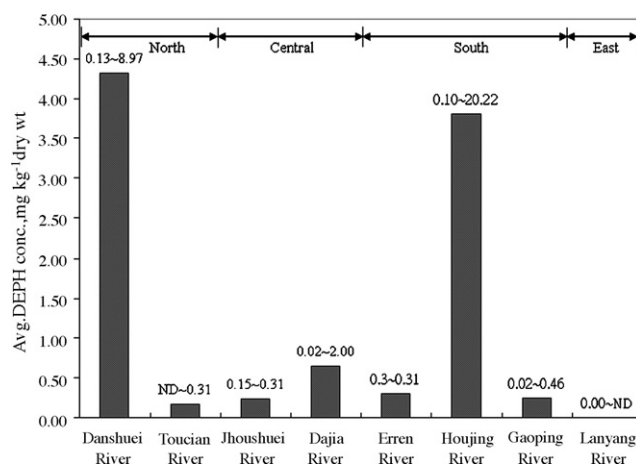


Fig. 5. Comparison of domestic DEHP river sediment contamination versus the Houjing River of this study.



**Table 1**  
Comparison of the Houjing River sediment DEHP contamination data with other geographical areas

River and estuaries	DEHP of sediment (mg kg <sup>-1</sup> dry wt.)	Reference
Aire River (UK)	7.89–115	[17,21]
Trent River (UK)	0.84–31.0	[17,21]
Klang River (Malaysia)	0.49–15.0	[6,21]
Various rivers in central (Italy)	0.058–0.487	[8,21]
Nueces Estuary (Texas)	0.04–16.0	[9,21]
Mersey Estuary (UK)	1.20	[18,21]
Rhine (Westphalia, Germany)	0.21–8.44	[7]
Furu River (Toyama, Japan)	1–2	[19]
Zhonggang, Keya, Erren, Gaoping, Donggang, Danshui Rivers (Taiwan)	0.5–23.9	[20]
Houjing River (Taiwan)	0.10–20.22	This study

data for some major rivers in an organic contaminants survey project. The averaged Houjing River sediment DEHP concentration measured in this study was compared with other rivers' sediment data reported by Huang et al. [10]. In Fig. 5, from left to right data for Danshuei River and Toucian River (representing northern Taiwan); Jhuoshuei River and Dajia River (representing central Taiwan); Erren River, Houjing River and Gaoping River (representing southern Taiwan); and Lanyang River (representing eastern Taiwan) are presented. It can be seen that, in general, DEHP contamination potential can be ordered as the following sequence: northern  $\geq$  southern  $>$  central  $>$  eastern Taiwan. The above sequence corresponds well with the population density distribution in Taiwan. This is not surprising because more population demands more plastic industrial activities and introduces more DEHP containing commercial products into the environment. However, for the Houjing River scenario, local plastic manufacturing plants located in the industrial parks may account for the high concentrations.

### 3.6. Comparison of sediment DEHP contamination with other geographical areas

Table 1 compares Houjing River sediment DEHP data with similar studies carried out in other countries. Among the studies, only the Aire River (7.89–115 mg kg<sup>-1</sup> dry wt.) and the Trent River (0.84–31.0 mg kg<sup>-1</sup> dry wt.) in the United Kingdom [17] had reported higher concentration than that of the Houjing River (0.10–20.22 mg kg<sup>-1</sup> dry wt.) in southern Taiwan. Other rivers, such as various rivers in central Italy [8], Mersey estuary in the United Kingdom [18], the Rhine River in Westphalia, Germany [7], the Furu River system in Toyama City, Japan [19] were relatively lower than that of the Houjing River in this study. Moreover, Nueces estuary [9] in Texas, United States, the Klang River [6] in Malaysia, and the six river sediments monitored by Yuan et al. [20] in Taiwan indicated DEHP contamination similar to the Houjing River reported in this study.

## 4. Conclusions

Spatial characterization study of the Houjing River sediment DEHP contamination indicated that Dashe and Renwu Industrial Parks were the potential sources of DEHP contamination input into the upstream sections of the Houjing River. Elevation of DEHP concentration from the wet (flood) to the dry season indicated that current sources were still active, and remained as continuous sources of DEHP to the Houjing River sediment. To avoid DEHP contamination and to improve river sediment quality, an effective pollution prevention strategy should be developed and enforced. Before the application of any effective remediation measures taken, continuous monitoring is suggested to ensure that wide spread

organic endocrine disruptors is not damaging the river ecosystem. Meanwhile, a DEHP effluent discharge elimination system should be employed to reduce the current discharges of DEHP into the rivers. More understanding in the fate and transport behavior of DEHP is warranted to develop a cost effective pollution prevention strategy.

## Acknowledgment

This research was funded by the National Science Council (NSC) of Taiwan, under the grant NSC-94-2211-E-022-001. The authors thank its financial support.

## References

- [1] J.K.H. Cheung, R.K.W. Lam, M.Y. Shi, J.-D. Gu, Environmental fate of endocrine-disrupting dimethyl phthalate esters (DMPE) under sulfate-reducing condition, *Sci. Total Environ.* 381 (2007) 126–133.
- [2] Council of the European Union (CEU), Implementation of the community strategy for endocrine disruptors – a range of substances suspected of interfering with the hormone systems of humans and wildlife (COM-1999-706), Document no. 14341/04, Brussels, Belgium, 2004.
- [3] U.S. Environmental Protection Agency, Removal of endocrine disruptor chemicals using drinking water treatment processes, EPA/625/R-00/015, Washington, DC, USA, 2001.
- [4] O.S. Fatoki, F. Vernon, Phthalate esters in rivers of the greater Manchester area, U.K., *Sci. Total Environ.* 95 (1990) 227–232.
- [5] EPAJ (Environmental Protection Agency of Japan), Chemicals in the environment. Report on Environmental Survey and Wildlife Monitoring of Chemicals in F.Y. 1993.
- [6] G.H. Tan, Residue levels of phthalate esters in water and sediment samples from the Klang River basin, *Bull. Environ. Contam. Toxicol.* 54 (1995) 171–176.
- [7] H. Fromme, T. Kuchler, T. Otto, K. Pilz, J. Muller, A. Wenzel, Occurrence of phthalates and bisphenol A and F in the environment, *Water Res.* 36 (2002) 1429–1438.
- [8] M. Vitali, M. Guidotti, G. Macilenti, C. Cremisini, Phthalate esters in freshwaters as markers of contamination sources—a site study in Italy, *Environ. Int.* 23 (1997) 337–347.
- [9] L.E. Ray, H.E. Murray, C.S. Giam, Analysis of water and sediment from the Nueces Estuary/Corpus Christi Bay (Texas) for selected organic pollutants, *Chemosphere* 12 (1983) 1039–1045.
- [10] P.C. Huang, C.Y. Hsieh, Y.T. Tsai, K.C. Peng, C.C. Lee, The distribution of di-(2-ethyl-hexyl) phthalate (DEHP) in major rivers in Taiwan, in: The Annual Conference of Environmental Analytical Chemistry, National Institute of Environmental Accreditation, Zhongli City, Taiwan, R.O.C., 2005.
- [11] C. Lin, C.J. Lee, W.J. Hsiao, W.M. Mao, Characterization of semi-volatile organic compound contamination in the sediment of Houjing River, in: Association of the Chinese Institute of Environmental Engineering, the Third Soil and Groundwater Remediation Conference, National Central University, Zhongli City, Taiwan, R.O.C., 2005.
- [12] S.K. Wang, Y.H. Kuo, S.C. Chuang, Investigation of organic contaminations in the major rivers of Taiwan, in: The 2005 Annual Conference of Environmental Analytical Chemistry, National Institute of Environmental Accreditation, Zhongli City, Taiwan, R.O.C., 2005.
- [13] NIEA Taiwan, Sediment sampling guidelines, NIEA S104.30C, National Institute of Environmental Accreditation (NIEA), Zhongli City, Taiwan, R.O.C., 2003.
- [14] NIEA Taiwan, Ultrasonic extraction procedures for the preparation of SVOC samples, NIEA M167.00C, National Institute of Environmental Accreditation (NIEA), Zhongli City, Taiwan, R.O.C., 2002.
- [15] NIEA Taiwan, Analysis method for semivolatile organic compounds by gas chromatography/mass spectrometry (GC/MS): capillary column technique, NIEA

- M731.00C, National Institute of Environmental Accreditation (NIEA), Zhongli City, Taiwan, R.O.C., 2002.
- [16] USEPA, Method 8270D: semivolatile organic compounds by gas chromatography/mass spectrometry (GC/MS), SW-846, Test Methods for Evaluating Solid Waste, Physical/Chemical Methods, official compendium of analytical and sampling methods issued by Office of Solid Wastes, U.S. Environmental Protection Agency.
- [17] J.L.A. Long, W.A. House, A. Parker, J.E. Rae, Micro-organic compounds associated with sediments in the Humber Rivers, *Sci. Total Environ.* 210 (1998) 229–253.
- [18] M.R. Preston, L.A. Al-Omran, Phthalate ester speciation in estuarine water, suspended particulates and sediments, *Environ. Pollut.* 62 (1989) 183–193.
- [19] E. Yuwatini, N. Hata, S. Taguchi, Behavior of di(2-ethylhexyl) phthalate discharged from domestic waste water into aquatic environment, *J. Environ. Monitor.* 8 (2006) 191–196.
- [20] S.Y. Yuan, C. Liu, C.S. Liao, B.V. Chang, Occurrence and microbial degradation of phthalate esters in Taiwan river sediments, *Chemosphere* 49 (2002) 1295–1299.
- [21] A. Turner, M.C. Rawling, The behaviour of di-(2-ethylhexyl) phthalate in estuaries, *Mar. Chem.* 68 (2000) 203–217.