Contamination levels and potential sources of organic pollution in an Asian river

Chitsan Lin, Kim Anh Nguyen, Chi Thanh Vu, Delia Senoro and Maria Ching Villanueva

ABSTRACT

The Houjing River has long been an environmental victim of economic development. Industries that have settled along the bank of this river may have largely contributed to severe organic wastes pollution. This study collected water and sediment samples at various points along the river and measured concentrations of 61 volatile organic compounds (VOCs) and 128 semi-volatile organic compounds (SVOCs) for a period of 16 months (Feb 2014–June 2015). Our analyses show that elevated levels of VOCs were observed near two industrial areas, Dashe and Renwu industrial parks. High SVOC concentrations were found in the vicinities of the Nanzih Export Processing Zone (NEPZ) and CingPu station, possibly due to considerable effluent discharges of adjacent industrial and residential areas. Comparing this study's findings with the standard values of different governmental agencies and studies similar to this one, the ecosystem of the Houjing River was seriously contaminated. This study could be used by the government as a basis for future and urgent pollution prevention actions aimed at protecting this ecosystem and reducing the negative impacts of these contaminants on the health and well-being of the local residents and the environment.

Key words | BTEX, chlorinated solvents, PAEs, pollution prevention

Chitsan Lin
Kim Anh Nguyen
Chi Thanh Vu (corresponding author)
Center of Environmental Analysis Service,
National Kaohsiung Marine University,
Kaohsiung 81157,
Taiwan
E-mail: thaanhchivu@gmail.com

Delia Senoro
Sustainable Development Research Office,
Mapúa University,
Manila 1002,
Philippines

Maria Ching Villanueva
IFREMER Centre de Bretagne,
Unité de Sciences et Technologies Halieutiques,
Pointe du Diable B.P. 70,
Plouzané 29280,
France

INTRODUCTION

Rivers are highly dynamic systems that change as a function of their catchments and water flows. These ecosystems are strongly influenced by the inflow of their effluents and climate change. River-basin monitoring programs require a continuous tracking of several environmental parameters in order to evaluate the state of the local environment. Organic contamination in densely populated areas is often investigated, as this has been proven to be highly dangerous to human health (Lee et al. 2002; Ost et al. 2003; de Gennaro et al. 2013; Senthil Kumar et al. 2017; Chen et al. 2016; Kristin et al. 2016). Therefore, any effort to assess and manage organic contaminant emissions, especially in populated industrial areas, should be extolled.

Anthropogenic activities that commonly take place near or along water bodies around the world have contributed to the rapid degradation of these natural resources (Lekkas et al. 2004; Moliner-Martinez et al. 2013; Hassanzadeh et al. 2014; Liu et al. 2016). River basins have been exposed to diffuse pollution when persistent, toxic and deleterious compounds are released by urban, industrial and agricultural activities (Huang et al. 2014; Villanueva & Ibarra 2016). The use of chlorinated consumer products has been reported as a potential source of organic contamination (Odabasi et al. 2014). Water, soil and sediments have often been used to measure organic contaminants and analyze water body quality (Moliner-Martinez et al. 2013; Hassanzadeh et al. 2014) and to assess contamination in areas' aquatic systems (Moliner-Martinez et al. 2013; Hassanzadeh et al. 2014; Liu et al. 2015).

Taiwan has made remarkable social and political gains since the 1950s due to its huge economic and material progress. The country has gone through a long period of industrial development and is now recognized as home to a world-class semiconductor industry (Lin et al. 2009). The perils of such enormous growth, however, have consequently contributed to the severe degradation of its

environment (Williams & Chang 2008), especially its scarce water resources (Hung *et al.* 2007). Water bodies around Taiwan have been used as waste dumps, and thus are heavily contaminated with organic compounds (Hung *et al.* 2007; Wang *et al.* 2015b).

Before the industrialization, the Houjing River was formerly a reliable source of clean water for human use and agricultural activities such as fish and crop farming (Sun & Tsai 2009). The establishment of petrochemical, plastic, and semiconductor industries has contributed to the region's rapid economic expansion (Sun & Tsai 2009; Patterson 2014). Currently, water from the river is unsuitable for household, recreational or agricultural use. Local residents are now exerting strong pressure on the city government in order to restore the ecosystem's water quality.

Organic contamination of the ecosystem may be attributed to the establishment of numerous manufacturing plants in four industrial areas along the river: Dashe and Renwu industrial parks, the Kaohsiung Oil Refinery, and Nanzih export processing zone (NEPZ). Industries located in the two parks and the oil refinery specialize in petrochemical productions. These factories were suspected of flushing high concentrations of toxic compounds such as benzene, toluene, ethylbenzene, and xylene (or BTEX) into the river (Hsieh *et al.* 2006). Runoff from the Formosa Plastics Corporation, located at the Renwu industrial park, could be responsible for the formation of chlorinated toxic by-product compounds such as vinyl chloride, methylene chloride, 1,1-dichloroethane, chloroform, 1,2-dichloroethane, etc. (Huang *et al.* 2014). Metal processing, chemical manufacture, and plastics resin production industries are common in the NEPZ, Dashe and Renwu industrial parks. These industrial effluents, coupled with commercial and domestic wastes in the Nanzih District, could be the source of phthalate esters (PAEs) pollution (Staples *et al.* 1997).

The goal of this study was to measure the contamination levels of volatile organic compounds (VOCs) and semi-volatile organic compounds (SVOCs) in water and sediment samples collected from various sites in the Houjing River to evaluate current ecosystem quality and suggest potential sources of pollution or toxic hotspots. Results of this study could be used to locate ecosystem pollution hotspots necessary to help mitigate the severe negative impact of river contamination on human health and provide the basis for local government to implement more stringent regulations for wastewater and sludge treatment, disposal and recycling.

MATERIALS AND METHODS

Study site

The Houjing River is located in north-west Kaohsiung City, an industrial harbor city in southwestern Taiwan (22°69′ – 22°73′, 120°25′ – 120°33′). The river stems from two streams that flow from Dashe from the northeast (Ciaotou District) and Renwu (Renwu District) from the southeast (Figure 1). These river branches meet at Si-Chingpu Landfill. Except for a few branches fluxing around the area of Nanzih District, the river flows directly toward the Taiwan Strait. It has a length of about 21 km. The climate in the basin is characterized by long summers and winters, while springs and autumns are relatively short. The river used to be a major water source for irrigation and fish farming. However, the progressive installation and expansion of industrial areas along the river, coupled with lenient legislations against illegal wastewater and sludge discharges, have contributed to serious contamination of this ecosystem (Lin *et al.* 2010). Recent studies showed that the system toxicity has been contaminating biological resources directly consumed by humans (Sun & Tsai 2009; Patterson 2014).

Sampling

In order to cover all possible contamination point sources of the river, nine specific sampling sites were chosen:
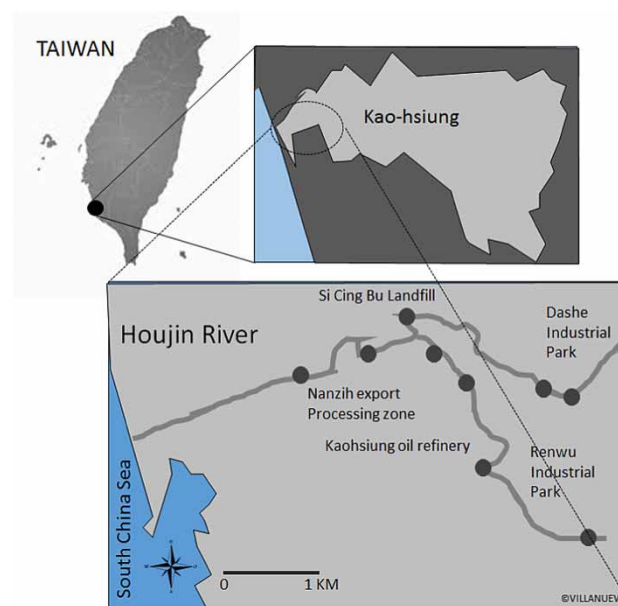

Figure 1 | Location and map of the Houjing River in Taiwan showing the nine stations considered in this study.

Sannaitan, Jingjian, Bakong, Renwu, Hueifeng, Deming, CingPu, Dehuei and Youchangda. The stations considered were located in four potential contamination source zones. Sannaitan and Jingjian stations were chosen to investigate the potential impact of effluents from the Dashe industrial park. The Bakong, Renwu and Hueifeng stations were considered to examine potential contamination of industries at the Renwu industrial park and the Kaohsiung Oil Refinery. Similarly, Deming, CingPu, Dehuei and Youchangda stations were considered to examine potential contamination of industrial activities at NEPZ. Water and sediment samples were collected based on the United States Environmental Protection Agency (USEPA) methods (Lin *et al.* 2007; Lin *et al.* 2009; USEPA 2013, 2014). Water samples were used for VOC analyses. Surface water was collected using a bucket in the middle of the river (about 5 m from the river banks) and kept in 40 mL amber screw VOC vials, which were cleaned with deionized water before the sample collection. Sediment samples were collected for SVOC analyses. Surface sediments (about 10–15 cm from the surface) were collected using an Ekman dredge and stored in pre-cleaned 500 mL amber screw bottles. At each site, two water and sediment replicates were collected. All samples were kept in an ice cooler box during transport. At the laboratory, they were stored and maintained for 3 days at $-4\,^\circ C$ until the analysis. Sixty-one VOCs and 128 SVOCs were measured and analyzed for a period of 16 months (Feb 2014 – June 2015). Sample collections were usually attempted to be carried out during sunny days to avoid the potential dilution effect caused by rainfall, runoff and rapid water currents. However, climate instabilities might have affected samples and introduced some biases in our analyses.

Instrumental analysis

Volatile organic compounds

The USEPA Method 8260B was used in this study to quantify VOCs that have boiling points below $200\,^\circ C$. The most common sample-preparation technique for 8260B is provided by USEPA 5030, which recommends a Purge-and-Trap technique. Both the OI Analytical model 4552 autosampler and the Eclipse 4660 Purge-and-Trap sample concentrator were employed to preprocess samples for analysis by gas chromatography/mass spectrometry (GC/MS). An HP 6890 GC coupled with the 5973N MS system was used for VOC analysis. A total of 61 compounds was analyzed, including BTEX and chlorinated VOCs. Before

beginning the 8260B analysis, the GC/MS system should be conditioned and prepared for use. The Purge-and-Trap optimization conditions employed were a 5 mL purge vessel (VOCARB 4560 Trap), with a $40\,mL\cdot min^{-1}$ helium purge flow at purge-ready temperature of $20\,^\circ C$ for 13 minutes. The purged samples were desorbed at $180\,^\circ C$ for four minutes then baked for 15 minutes at $200\,^\circ C$. For the GC/MS system, a capillary separation column (HP-624, 60 m $\times$ 0.25 mm $\times$ 1.5 μm film thickness) was installed. The oven was pre-heated at $40\,^\circ C$ for one minute before increasing the temperature to $180\,^\circ C$ at a rate of $8\,^\circ C\cdot min^{-1}$ and finally to $220\,^\circ C$ at a rate of $10\,^\circ C\cdot min^{-1}$ for 10 minutes.

Deionized (DI) water was used for the preparation of standards and test samples. All samples and calibration standards were then spiked with an internal standard/surrogates mixture. The mixture used is composed of non-deuterated fluorobenzene as the internal standard and non-deuterated 4-bromofluorobenzene with deuterated 1,2-dichlorobenzene as surrogates. Non-deuterated fluorobenzene gave the closest retention time to the target analyte, and was used in the response factor (RF) calculation for each compound. As a minimum, the 8260B method requires a five-point calibration curve. In this study, calibration levels at 2, 5, 20, 40 and 100 μg·mL^{-1} were considered. The correlation coefficients (*r*-value) of curves estimated were >0.995. Water blanks from laboratory and field samples were analyzed to establish a baseline. Seven samples with the same concentration were prepared to estimate the method detection limits (MDLs). Each individual analyte's MDL was obtained by multiplying three times the standard deviation (SD) by the appropriate one-sided 99% t-statistic. The MDL values for VOCs ranged from 0.3 to 0.9 μg·L^{-1}. Water, laboratory and field samples showed non-detected (ND) values. For every batch of 10 samples, standard mid-point and spiked-sample checks were performed. Standard mid-point and spiked sample percent recoveries were from 90 to 110% and from 75 to 94%, respectively. Accuracies of the measures were $100 \pm 10\%$.

Semi-volatile organic compounds

A low concentration method was performed by mixing a 10-gram dried sediment sample with 20 g anhydrous sodium sulfate to form a free-flowing powder. USEPA Method 3550C was the ultrasonic process used to extract nonvolatile and SVOCs with solvent three times at three minute intervals. A 10-μL surrogate standard solution was added before performing the dichloromethane (DCM) ultrasonic extraction. The DCM extract was collected and

concentrated by a Turbo Vap II concentrator prior to injection into the GC/MS system for analysis. The Method 8270D was the GC/MS used to determine SVOC concentrations. A total of 120 compounds were analyzed. Sediment sample preparation and analysis employed were based on Lin *et al.* 2009.

Standard SVOC mixtures and deuterated internal standards (1,4-dichlorobenzene, naphthalene, acenaphthene, phenanthrene, chrysene, perylene) were purchased from SUPELCO (Bellefonte, Pennsylvania, USA). Six calibration levels (1.25, 5, 10, 20, 40 and 100 $\mu g \cdot mL^{-1}$) were considered for the Method 3550C. Relative RFs of all compounds were above 0.05, while the percentage of deviation of each compound was <20%. In order to ensure good data quality, a decafluorotriphenylphosphine (DFTPP) standard check was performed to calibrate the mass selective detector (MSD) before analysis. A system proficiency calibration check (SPCC) was also performed once every 12 hours of analysis. Blank and spike recovery tests were carried out afterwards. Similar to VOC measurements, MDLs were estimated from blanks and seven samples of the same concentration. Again, an individual MDL analyte was calculated from the product of the SD by the appropriate one-sided 99% t-statistic. The MDL values for SVOCs ranged from 0.1 to 0.5 $mg \cdot kg^{-1}$ dry weight. The blanks showed ND values. Standard mid-point and spiked-sample checks were performed per 10-sample batch. Standard mid-point and spiked-sample recoveries were from 90 to 108% and

from 79 to 85%, respectively. Accuracies of the measures were 100 ± 10%.

All the chemicals were of reagent grade and all standards were NIST traceable quality. DI water was used in all analyses.

RESULTS AND DISCUSSION

Spatial distribution and potential sources of VOC contamination

Figure 2 shows the spatial distribution of the total VOC concentrations in different sampling sites of the Houjing River. VOCs were detected in all stations except Bakong and Sannaitan. High SDs around mean values could be due to variation in findings over time. Total VOC concentrations in samples collected at Jingjian and Renwu stations were unusually high (102.7 ± 184.5 and 131.2 ± 105.1 $\mu g \cdot L^{-1}$, respectively). Observed concentrations at CingPu and Hueifeng stations (16.3 ± 18.3 and 42.4 ± 27.9 $\mu g \cdot L^{-1}$, respectively) were considerably lower. VOC concentrations continued to decline at the Dehuei (17.9 ± 15.2 $\mu g \cdot L^{-1}$) and Youchangda (9.8 ± 7.3 $\mu g \cdot L^{-1}$) stations. These two latter stations were located near the river mouth, where freshwater was constantly mixed with seawater. Based on these results, the potential main sources that contributed to severe total VOC contamination of the ecosystem were
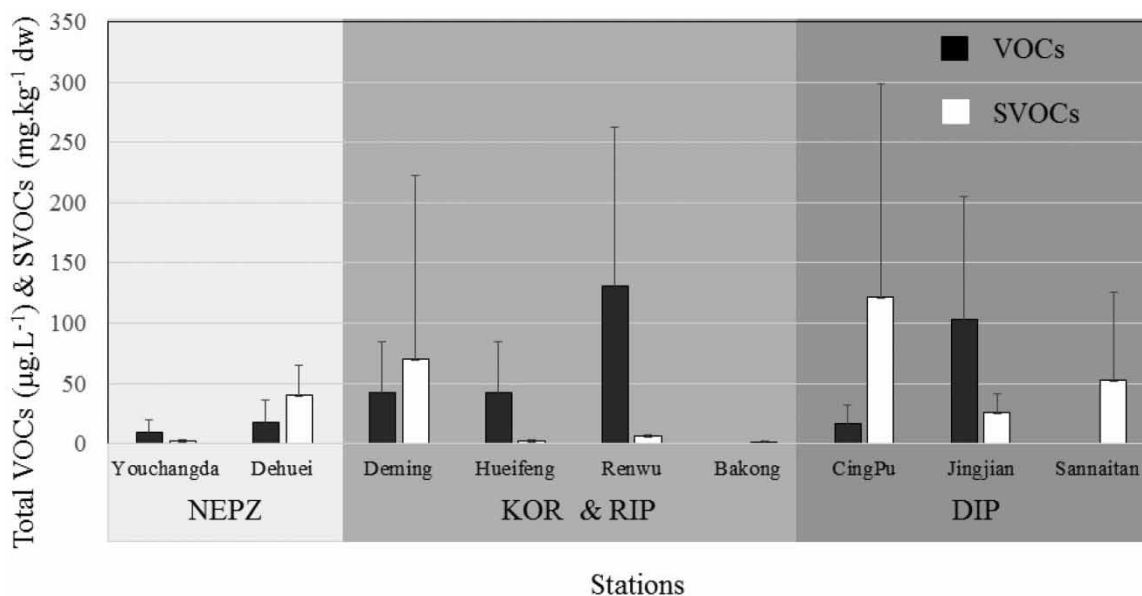
Figure 2 | Total VOC ($\mu g \cdot L^{-1}$) and SVOC ($mg \cdot kg^{-1}$ dry weight) concentrations in water and sediment samples collected from the Houjing River, respectively. NEPZ: Nanzih Export Processing Zone; KOR: Kaoshing Oil Refinery; RIP: Renwu industrial park; DIP: Dashe industrial park.

most likely located around the vicinity of Jingjian and Renwu stations. These two stations were located near the Dashe and Renwu industrial parks, which included numerous petrochemical and plastic manufacturers. Total VOCs decreased progressively towards the downstream areas of the river, possibly due to the natural dilution effect caused by seawater. Special attention was given to BTEX, a well-known carcinogen, and chlorinated VOC pollutants, which are harmful to both human and environmental health, and results are further discussed below.

BTEX contamination

Samples collected from the Jingjian station showed the highest BTEX value of $102.7 \pm 184.5\,\mu g \cdot L^{-1}$, which may be due to petrochemical industry effluents in the Dashe industrial park. A comparison with established standards by the USEPA (USEPA 2015) and Japan's Ministry of Environment (JMOE) (MOE 1993) was provided for a more comprehensive view of the current BTEX river contamination (Table 1). Benzene, toluene and ethylbenzene were detected in water samples from the five BTEX compounds analyzed. The ethylbenzene maximum value was estimated at $270.6\,\mu g \cdot L^{-1}$, which is four times higher than the accepted USEPA limit ($68\,\mu g \cdot L^{-1}$). In this study, the more stringent value of USEPA for toluene ($57\,\mu g \cdot L^{-1}$) was selected for the comparison. Highest toluene concentration at the Jingjian station ($17.0\,\mu g \cdot L^{-1}$) estimated was found to be within the acceptable USEPA limit. The mean concentration of benzene ($44.5\,\mu g \cdot L^{-1}$) exceeded the Japan MOE's limit ($10\,\mu g \cdot L^{-1}$) and was more than twentyfold higher than the upper limit set by USEPA's range ($0.58–2.1\,\mu g \cdot L^{-1}$). Ethylbenzene concentrations ranged from 0 to $270.6\,\mu g \cdot L^{-1}$, with some far

Table 1 | Min-max BTEX concentrations ($\mu g \cdot L^{-1}$) in water samples at the Jingjian station ($n = 8$)

Compound name	MDLs	Min	Max	Mean	USEPA	JMOE
Benzene	0.4	0.9	158.0	44.5	0.58–2.1	10
Toluene	0.3	0	17.0	3.7	57	600
Ethylbenzene	0.6	0	270.6	54.5	68	NA
p,m-Xylene	0.5	ND	ND	ND	NA	400
o-Xylene	0.6	ND	ND	ND	NA	400

USEPA (United States Environmental Protection Agency): National Recommended Water Quality Criteria – Human Health Criteria Table.
JMOE (Japan's Ministry of Environment): Environmental Quality Standards for Water Pollution.
MDLs: method detection limits; NA: not available; ND: not detected.

above the USEPA limit ($68\,\mu g \cdot L^{-1}$) while others were lower (mean value $54.5\,\mu g \cdot L^{-1}$). Meanwhile, no xylene compounds were observed.

A highly elevated benzene concentration ($158.0\,\mu g \cdot L^{-1}$) at Jingjian station was observed from this study. Similar to ethylbenzene at the same station, this exceeded the standard limits established by both USEPA ($2.1\,\mu g \cdot L^{-1}$) and JMOE ($10\,\mu g \cdot L^{-1}$). This is also one of the worst and highly extensive hazardous types of air pollution, especially in Taipei (Williams & Chang 2008). Benzene is considered a very good solvent and has been used widely in the petrochemical industry since the early 1990s (Chung et al. 2010; Uragami et al. 2016). This industry is recognized as an important source of emissions of chemical substances with adverse health results to the residents (Chen et al. 2016). Many studies proved benzene to be highly poisonous (Casale et al. 2016; Hosseinzadeh & Moosavi-Movahedi 2016) and carcinogenic (Maltoni et al. 1983; McMichael 1988; Smith et al. 1989; Golding & Watson 1999). Despite the evidence of its toxicity to human and environmental health (Weaver et al. 1983; Lee et al. 2007; Chung et al. 2010), this industrial chemical still has been utilized extremely by petrochemical plants in many countries (Yin et al. 1987; Liu et al. 2009; Chung et al. 2010).

Chlorinated-VOC contamination

The Renwu station ($131.2 \pm 105.1\,\mu g \cdot L^{-1}$) was possibly the main source of chlorinated-VOC contamination due to a lower observed estimation in Hueifeng ($42.4 \pm 27.9\,\mu g \cdot L^{-1}$) and the absence of background contamination at Bakong (Table 2). Concentration estimated at the Deming ($42.3 \pm 25.2\,\mu g \cdot L^{-1}$) station was slightly lower than the value obtained in Huiefeng. A progressive decline in concentrations were noted at the Dehuei ($17.9 \pm 15.2\,\mu g \cdot L^{-1}$), CingPu ($16.3 \pm 18.3\,\mu g \cdot L^{-1}$) and Youchangda ($9.8 \pm 7.3\,\mu g \cdot L^{-1}$) stations, which might be due to the freshwater dilution effect. No chlorinated-VOCs were found in water samples from Bakong, Sannaitan and Jingjian stations.

Chlorinated-VOCs were examined only in five stations located in the Renwu branch and the river mouth (Table 3). Our results showed that unusually high contamination levels were only observed in one or two of these stations. Among the stations considered, Renwu had the highest concentrations of three of the five chlorinated-VOCs examined. These were vinyl chloride ($29.0 \pm 26.6\,\mu g \cdot L^{-1}$), chloroform ($108.3 \pm 102.2\,\mu g \cdot L^{-1}$) and 1,1-dichloroethane ($6.2 \pm 5.2\,\mu g \cdot L^{-1}$). The pollution rate decreased for stations located near the mouth of the river. Again, may be

Table 2 │ Chlorinated-VOC concentrations ($\mu g \cdot L^{-1}$) in water samples ($n = 8$ per station) collected from the Houjing River

Sampling location	Dashe branch			Renwu branch				To the ocean	
	Sannaitan	Jingjian	CingPu	Bakong	Renwu	Hueifeng	Deming	Dehuei	Youchangda
Range	ND	ND	0–44.6	ND	1.2–331.6	1.3–76.7	5.8–69.4	0–39.2	0.7–21.0
Mean ± S.D	ND	ND	16.3 ± 18.3	ND	131.2 ± 105.1	42.4 ± 27.9	42.3 ± 25.2	17.9 ± 15.2	9.8 ± 7.3
Median	ND	ND	8.0	ND	100.7	39.5	45.3	14.7	10.2

ND: not detected.

Table 3 │ Mean concentrations ($\mu g \cdot L^{-1}$) of chlorinated-VOCs observed at some stations of the Houjing River

Compound name	MDLs	Renwu branch			Near the ocean	
		Renwu	Hueifeng	Deming	Dehuei	Youchangda
Vinyl chloride	0.3	29.0 ± 26.6	9.7 ± 3.9	5.5 ± 3.2	2.2 ± 1.0	1.7 ± 0.9
Chloroform	0.4	108.3 ± 102.2	34.0 ± 10.2	33.5 ± 12.2	20.2 ± 15.8	8.6 ± 6.2
1,1-Dichloroethane	0.4	6.2 ± 5.2	ND	ND	ND	ND
Methylene chloride	0.3	12.4 ± 7.1	4.5 ± 4.4	14 ± 8.4	3.0 ± 2.1	2.1 ± 1.9
1,2-Dichloroethane	0.5	11.8 ± 9.8	6.3 ± 1.9	15.7 ± 10.0	ND	ND

MDLs: method detection limits.

due to the fresh-saltwater mixing effect. For the 1,1-dichloroethane, this contaminant was only observed in the Renwu station and was not detected in the other sites. Methylene chloride ($14.0 \pm 8.4 \mu g \cdot L^{-1}$) and 1,2-dichloroethane ($15.7 \pm 10.0 \mu g \cdot L^{-1}$) were highest at the Deming station located near NEPZ, and this might be caused by the computer chip packaging plants prevalent in this zone. On the other hand, 1,2-dichloroethane was absent at the Dehuei and Youchangda stations.

Huge amounts of chlorinated-VOC effluents were released by plastic manufacturers, who used this substance as plastic building blocks, as solvents and as scavenging or degreasing agents (Huang *et al.* 2014). Other manufacturing activities might also produce chlorinated-VOCs in solid and liquid forms (Huang *et al.* 2014). Based on the results of this study, it might be reasonable to infer that industrial factories at Renwu industrial park and NEPZ might contribute largely to chlorinated-VOC river contamination. Due to their adverse health effects in humans, (Yu & Chou 2000; Nagano *et al.* 2006; Gwinn *et al.* 2011), the use of some chlorinated-VOCs has been highly regulated or even banned following the Montreal Protocol because they belong to a list of compounds that highly contribute to ozone layer depletion, global warming and photochemical smog formation (UNEP 1987; Michalik-Zym *et al.* 2015).

Spatial distribution and potential sources of SVOC contamination

Figure 2 presents the spatial distribution of total SVOCs in the different sampling areas. For this analysis, sediment samples were collected to measure total SVOCs with high SDs around mean values resulting from differences over time. SVOCs were detected in all nine stations considered in the study. Our results showed low levels of SVOCs at the Bakong (1.5 ± 0.4 mg.$^{-1}$), Renwu (6.5 ± 1.1 mg·kg^{-1}), Hueifeng (2.3 ± 1.0 mg·kg^{-1}) and Youchangda (2.1 ± 0.8 mg·kg^{-1}) stations. These suggested that Renwu industrial park and the Kaohsiung Oil Refinery did not contribute heavily to the total SVOC contamination. Some sites showed elevated SVOC concentrations. This was the case for the two stations at the Dashe river branch: Jingjian (25.6 ± 16.0 mg·kg^{-1}) and Sannaitan (52.5 ± 72.8 mg·kg^{-1}). A considerably high concentration was observed at the Deming (70.0 ± 152.1 mg·kg^{-1}) station along the Renwu river branch, probably due to the occurrence of many semiconductor industries in the area. SVOC occurrence at the Dehuei (40.3 ± 24.3 mg·kg^{-1}) might be due to cumulated impacts of industrial, commercial and residential activities. Among all the stations considered, the CingPu station showed a considerably high SVOC pollution (122.0 ± 176.2 mg·kg^{-1}).

PAEs accounted for more than 90% of the total SVOCs in this study. These synthetic compounds are used as additives to improve production quality of plastics (Van Wezel *et al.* 2000; Gao *et al.* 2014), pharmaceutical, detergents, pesticides, toys and cosmetics (Staples *et al.* 1997). PAEs are carcinogens (Murahashi *et al.* 2003; Diamanti-Kandarakis *et al.* 2009) and are suspected to be endocrine disrupters (Van Wezel *et al.* 2000). The potential carcinogenic and mutagenic effects of PAEs have been well studied (Hauser & Calafat 2005; Meng *et al.* 2014). PAE exposure could impair human metabolism and reproductive system functioning (Bell 1982; Jurewicz & Hanke 2011) and they have been associated with miscarriages and sterility complications (i.e., ovulation problems, teratogenicity, and low semen quality) (Wang *et al.* 2015a). Table 4 shows the contamination levels of the 5 PAEs analyzed. Among these, di (2-ethylhexyl)phthalate (DEHP) was present at all sampling sites. It was the principal PAE pollutant, and levels observed overshadowed the other phthalates considered in this study. Used globally as a plasticizer (plastic softener) in many polyvinyl chloride (PVC) products (Hauser *et al.* 2005; Van Vliet *et al.* 2011), DEHP does not bind chemically to PVCs and leaches out of the product to which it is added (Van Vliet *et al.* 2011). The highest mean DEHP concentrations were observed at the CingPu (120.3 mg·kg^{-1}) and Deming (86.3 mg·kg^{-1}) stations. Lower concentrations were observed at Sannaitan (25.3 mg·kg^{-1}), Dehuei (22.1 mg·kg^{-1}) and Jingjian (21.4 mg·kg^{-1}) sites. Food processing and packaging factories near the Deming and Dehuei stations were the most likely culprits in river DEHP pollution.

In 2006, DEHP was listed as an important potentially dangerous chemical recognized as one of the priority existing chemicals to be treated by the National Industrial Chemicals Notifications and Assessment Scheme (NICNAS) under the 1989 Industrial Chemicals Act. Like many other PAE compounds, DEHP was reported to be an endocrine disruptor and is known to cause many human cardiovascular and reproductive complications (Tickner *et al.* 2001). The USEPA has issued a limit of 6 ppb for DEHP in drinking water (USEPA 2008). The Occupational Safety and Health Administration (OSHA) restricted occupational exposure to DEHP to less than 5 mg·m^{-3} of air (OSHA 2004). The 'plasticizer' DEHP has been banned under European Union (EU) law since 2015 after being listed among the toxic substances in Annex XIV under the EU's REACH (Registration, Evaluation, Authorization and Restriction of Chemicals) regulations (EU 2011). As can be seen in Table 5, DEHP concentrations in the Houjing River were high, but lower that those reported for the Songhua River (China) by Gao *et al.* (2014) and those reported for the Kaveri River (India) by Selvaraj *et al.* (2015). However, the concentrations in this study were well above the acceptable environmental risk limit in sediment of 1 mg·kg^{-1} fresh weight as suggested by Van Wezel *et al.* (2000). These findings suggested that DEHP contamination was very serious in the Houjing River and should also be monitored regularly and regulated by national authorities.

Comparative study of VOCs and SVOCs with other ecosystems

Table 5 shows a summary of primary VOCs and SVOCs observations in different countries. Maximum levels of total VOC pollutants analyzed in this study exceeded tremendously observations made in other freshwater ecosystems. Except for xylene, benzene (158.0 µg·L^{-1}), toluene (17.0 µg·L^{-1}), and ethylbenzene (270.6 µg·L^{-1}) pollutions in the Houjing River were highly elevated

Table 4 │ PAE concentrations (mg·kg^{-1} dry weight) in sediment samples (*n* = 6 per station) of the Houjing River

| Compound name | MDLs | Dashe branch | | | Renwu branch | | | | Near the ocean | |
		Sannaitan	Jingjian	CingPu	Bakong	Renwu	Hueifeng	Deming	Dehuei	Youchangda
Diethyl phthalate (DEP)	0.1	0.1 ± 0.2	0.3 ± 0.2	0.1 ± 0.1	0.2 ± 0.2	ND	ND	ND	0.4 ± 0.3	0.5 ± 0.5
Diphenylamine (DPA)	0.1	ND	ND	ND	ND	0.2 ± 0.1	ND	0.1 ± 0.1	ND	ND
Di-n-butyl phthalate (DnBP)	0.1	0.1 ± 0.2	3.6 ± 2.9	1.1 ± 1.0	0.4 ± 0.3	2.4 ± 2.0	1.4 ± 1.1	0.4 ± 0.3	15.5 ± 10.0	1.2 ± 1.2
Di (2-ethylhexyl) phthalate (DEHP)	0.2	25.3 ± 18.6	21.4 ± 19.7	120.3 ± 85.7	0.7 ± 0.5	3.7 ± 2.9	0.8 ± 0.7	86.3 ± 65.1	22.1 ± 11.8	0.5 ± 0.4
Di-n-octyl phthalate (DnOP)	0.1	ND	0.2 ± 0.3	0.5 ± 0.4	0.2 ± 0.1	0.2 ± 0.2	0.1 ± 0.1	0.3 ± 0.2	2.2 ± 1.4	ND

MDLs: method detection limits; ND: not detected.

Table 5 | Comparison of estimated BTEX (μg·L^{-1}), chlorinated-VOCs (μg·L^{-1}) and DEHP (mg·kg^{-1} dry weight) concentrations in the Houjing River with other ecosystems

Geographical areas	Types of water bodies	BTEX					Chlorinated VOCs	DEHP	Reference
		Benzene	Toluene	Ethylbenzene	m + p – Xylene	o – Xylene			
Bangladesh	River	0.35–0.38	0.20–0.27	0.12–0.17	0.21–0.14[a]		–	–	Mottaleb *et al.* (2003)
Swiss Lake	Lake	0–0.47	0–1.02	0–0.39	0–2.05[a]		–	–	Schmidt *et al.* (2004)
India	River	0–1.50	0–0.82	0–0.07	0–0.16	0–0.15			Lokhande *et al.* (2009)
Portugal	Rivers	0.01–0.70	0.01–4.77	0.09–4.75	0.01–27.61	0.02–2.16	–	–	Moliner-Martinez *et al.* (2013)
Canada	River	–	–	–	–	–	0–30.00	–	Kaiser & Comba (1983)
Greek	Rivers	–	–	–	–	–	0–40.00	–	Kostopoulou *et al.* (2000)
Japan	Rivers	–	–	–	–	–	0–55.60	–	Yamamoto *et al.* (2001)
Germany	Rivers	–	–	–	–	–	–	0.21–8.44	Fromme *et al.* (2002)
The Netherlands	Rivers	–	–	–	–	–	–	0–19.26	Peijnenburg & Struijs (2006)
China	River	–	–	–	–	–	–	227.08–566.54	Gao *et al.* (2014)
Iran	River	–	–	–	–	–	–	2.00–1,438.00	Selvaraj *et al.* (2015)
Taiwan	River	0.9–158.0	0–17.0	0–270.6	ND	ND	0–331.6	0.5–120.3	This study

Concentrations are expressed as min–max values. ND: not detected.
[a]Sum for all xylene isomers.

compared to other rivers and lakes. The chlorinated-VOC concentrations in the Houjing River were much higher than those reported for the Welland River (Kaiser & Comba 1983), northern rivers and lakes in Greece (Kostopoulou *et al.* 2000) and Osaka (Yamamoto *et al.* 2001). In terms of DEHP, the pollution level was higher compared to results observed in several rivers and lakes in Germany (Fromme *et al.* 2002) and the Netherlands (Peijnenburg & Struijs 2006). This phthalate compound, however, was lower compared to the observations of Gao *et al.* (2014) and Selvaraj *et al.* (2015) in China and Iran, respectively.

CONCLUSIONS

This study has provided pertinent data on the levels of both volatile and semi-volatile organic contaminants in the Houjing River. The Renwu station had the highest level of total VOC toxicity, while the highest level of total SVOCs was observed at the CingPu station. Petrochemical industries, particularly, might be involved in VOC contamination in the river, particularly BTEX and chlorinated VOCs. These pollutants were detected in water samples with concentrations considerably elevated compared to other freshwater ecosystems. Stations located near the Dashe and Renwu industrial parks showed the highest VOC pollution, originating from numerous petrochemical and plastics industries. For SVOCs, especially DEHP, the highest contamination levels were observed at the Deming and the Cingpu stations, mainly originating from sewage and industrial wastewater, sewage sludge application, discarded plastic effusion and probably atmospheric deposition. The concentrations of BTEX, chlorinated VOCs and DEHP suggested that the river is a highly contaminated ecosystem in urgent need of environmental management plans and waste discharge regulations at both local and national levels. Local and national authorities should take action to regulate and limit pollution if they want to restore the

river's diminished water quality. Regular water, sediment and wastewater monitoring and assessments should be implemented.

REFERENCES

Bell, F. P. 1982 Effects of phthalate esters on lipid metabolism in various tissues, cells and organelles in mammals. *Environmental Health Perspectives* **45**, 41–50.

Casale, T., Sacco, C., Ricci, S., Loreti, B., Pacchiarotti, A., Cupelli, V., Arcangeli, G., Mucci, N., Antuono, V., De Marco, F., Tomei, G., Tomei, F. & Rosati, M. V. 2016 Workers exposed to low levels of benzene present in urban air: assessment of peripheral blood count variations. *Chemosphere* **152**, 392–398.

Chen, W. H., Chen, Z. B., Yuan, C. S., Hung, C. H. & Ning, S. K. 2016 Investigating the differences between receptor and dispersion modeling for concentration prediction and health risk assessment of volatile organic compounds from petrochemical industrial complexes. *Journal of Environmental Management* **166**, 440–449.

Chung, E.-K., Shin, J.-A., Lee, B.-K., Kwon, J., Lee, N., Chung, K.-J., Lee, J.-H., Lee, I.-S., Kang, S.-K. & Jang, J.-K. 2010 Characteristics of occupational exposure to benzene during turnaround in the petrochemical industries. *Safety and Health at Work* **1** (1), 51–60.

De Gennaro, G., Farella, G., Marzocca, A., Mazzone, A. & Tutino, M. 2013 Indoor and outdoor monitoring of volatile organic compounds in school buildings: indicators based on health risk assessment to single out critical issues. *International Journal of Environmental Research and Public Health* **10** (12), 6273–6291.

Diamanti-Kandarakis, E., Bourguignon, J.-P., Giudice, L. C., Hauser, R., Prins, G. S., Soto, A. M., Zoeller, R. T. & Gore, A. C. 2009 Endocrine-Disrupting chemicals: an endocrine society scientific statement. *Endocrine Reviews* **30** (4), 293–342.

EU 2011 *Chemicals/REACH: Six Dangerous Substances to be Phased out by the EU*. European Union, Brussels, Belgium. http://europa.eu/rapid/press-release_IP-11-196_en.htm?locale=en (accessed 16 May 2017).

Fromme, H., Kuchler, T., Otto, T., Pilz, K., Muller, J. & Wenzel, A. 2002 Occurrence of phthalates and bisphenol A and F in the environment. *Water Research* **36** (6), 1429–1438.

Gao, D. W., Li, Z., Wen, Z. D. & Ren, N. Q. 2014 Occurrence and fate of phthalate esters in full-scale domestic wastewater treatment plants and their impact on receiving waters along the Songhua River in China. *Chemosphere* **95**, 24–32.

Golding, B. T. & Watson, W. P. 1999 Possible mechanisms of carcinogenesis after exposure to benzene. *IARC Sci Publ* **150**, 75–88.

Gwinn, M. R., Johns, D. O., Bateson, T. F. & Guyton, K. Z. 2011 A review of the genotoxicity of 1,2-dichloroethane (EDC). *Mutation Research/Reviews in Mutation Research* **727** (1–2), 42–53.

Hassanzadeh, N., Sari, A. E., Khodabandeh, S. & Bahramifar, N. 2014 Occurrence and distribution of two phthalate esters in the sediments of the Anzali wetlands on the coast of the Caspian Sea (Iran). *Marine Pollution Bulletin* **89** (1–2), 128–135.

Hauser, R. & Calafat, A. M. 2005 Phthalates and human health. *Occup. Environ. Med.* **62** (11), 806–818.

Hauser, R., Williams, P., Altshul, L. & Calafat, A. M. 2005 Evidence of interaction between polychlorinated biphenyls and phthalates in relation to human sperm motility. *Environmental Health Perspectives* **113** (4), 425–430.

Hosseinzadeh, R. & Moosavi-Movahedi, A. A. 2016 Human hemoglobin structural and functional alterations and heme degradation upon interaction with benzene: a spectroscopic study. *Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy* **157**, 41–49.

Hsieh, L. T., Yang, H. H. & Chen, H. W. 2006 Ambient BTEX and MTBE in the neighborhoods of different industrial parks in southern Taiwan. *Journal of Hazardous Materials* **128** (2–3), 106–115.

Huang, B. B., Lei, C., Wei, C. H. & Zeng, G. M. 2014 Chlorinated volatile organic compounds (Cl-VOCs) in environment – sources, potential human health impacts, and current remediation technologies. *Environment International* **71**, 118–138.

Hung, C. C., Gong, G. C., Chen, H. Y., Hsieh, H. L., Santschi, P. H., Wade, T. L. & Sericano, J. L. 2007 Relationships between pesticides and organic carbon fractions in sediments of the Danshui River estuary and adjacent coastal areas of Taiwan. *Environmental Pollution* **148** (2), 546–554.

Jurewicz, J. & Hanke, W. 2011 Exposure to phthalates: reproductive outcome and children health. A review of epidemiological studies. *Int. J. Occup. Med. Environ. Health* **24** (2), 115–141.

Kaiser, K. L. E. & Comba, M. E. 1983 Volatile contaminants in the Welland River watershed. *Journal of Great Lakes Research* **9** (2), 274–280.

Kostopoulou, M. N., Golfinopoulos, S. K., Nikolaou, A. D., Xilourgidis, N. K. & Lekkas, T. D. 2000 Volatile organic compounds in the surface waters of Northern Greece. *Chemosphere* **40** (5), 527–532.

Kristin, S., Roland, B., Christian, J., Wolfram, B., Thorsten, W., Jens, N., Gunda, L., Steffen, F. & Irene, N. 2016 Comparison of volatile organic compounds from lung cancer patients and healthy controls – challenges and limitations of an observational study. *Journal of Breath Research* **10** (4), 046007.

Lee, C. R., Yoo, C. I., Lee, J. H., Kim, S.-R. & Kim, Y. 2002 Hematological changes of children exposed to volatile organic compounds containing low levels of benzene. *Science of the Total Environment* **299** (1–3), 237–245.

Lee, E. H., Do Eum, K., Cho, S. I., Cheong, H. K. & Paek, D. M. 2007 Acquired dyschromatopsia among petrochemical industry workers exposed to benzene. *Neurotoxicology* **28** (2), 356–363.

Lekkas, T., Kolokythas, G., Nikolaou, A., Kostopoulou, M., Kotrikla, A., Gatidou, G., Thomaidis, N. S., Golfinopoulos, S., Makri, C., Babos, D., Vagi, M., Stasinakis, A., Petsas, A. & Lekkas, D. F. 2004 Evaluation of the pollution of the surface waters of Greece from the priority compounds of list II, 76/464/EEC Directive, and other toxic compounds. *Environment International* **30** (8), 995–1007.

Lin, C., Mao, W. M. & Nadim, F. 2007 Forensic investigation of BTEX contamination in the Houjing River in Southern Taiwan. *Journal of Environmental Engineering and Management* **17** (6), 395–402.

Lin, C., Lee, C. J., Mao, W. M. & Nadim, F. 2009 Identifying the potential sources of di-(2-ethylhexyl) phthalate contamination in the sediment of the Houjing River in southern Taiwan. *Journal of Hazardous Materials* **161** (1), 270–275.

Lin, C. E., Kao, C. M., Jou, C. J., Lai, Y. C., Wu, C. Y. & Liang, S. H. 2010 Preliminary identification of watershed management strategies for the Houjing river in Taiwan. *Water Science and Technology* **62** (7), 1667–1675.

Liu, H., Liang, Y. X., Bowes, S., Xu, H. Z., Zhou, Y. M., Armstrong, T. W., Wong, O., Schnatter, A. R., Fang, J. B., Wang, L. M., Nie, L. P., Fu, H. & Irons, R. 2009 Benzene exposure in industries using or manufacturing paint in China-A literature review, 1956–2005. *Journal of Occupational and Environmental Hygiene* **6** (11), 659–670.

Liu, B., Chen, L., Huang, L., Wang, Y. & Li, Y. 2015 Distribution of volatile organic compounds (VOCs) in surface water, soil, and groundwater within a chemical industry park in Eastern China. *Water Science and Technology* **71** (2), 259–267.

Liu, B., Li, Y., Ma, J., Huang, L. & Chen, L. 2016 Detection of semi-volatile organic compounds (SVOCs) in surface water, soil, and groundwater in a chemical industrial park in Eastern China. *Water Science and Technology* **73** (5), 1175–1189.

Lokhande, P. B., Patil, V. V. & Mujawar, H. A. 2009 Multivariate statistical study of seasonal variation of BTEX in the surface water of Savitri River. *Environmental Monitoring and Assessment* **157** (1–4), 51–61.

Maltoni, C., Conti, B. & Cotti, G. 1983 Benzene: a multipotential carcinogen. Results of long-term bioassays performed at the Bologna Institute of Oncology. *Am. J. Ind. Med.* **4** (5), 589–630.

Mcmichael, A. J. 1988 Carcinogenicity of benzene, toluene and xylene: epidemiological and experimental evidence. *IARC Sci. Publ.* **85**, 3–18.

Meng, X.-Z., Wang, Y., Xiang, N., Chen, L., Liu, Z., Wu, B., Dai, X., Zhang, Y.-H., Xie, Z. & Ebinghaus, R. 2014 Flow of sewage sludge-borne phthalate esters (PAEs) from human release to human intake: implication for risk assessment of sludge applied to soil. *Science of the Total Environment* **476–477**, 242–249.

Michalik-Zym, A., Dula, R., Duraczyńska, D., Kryściak-Czerwenka, J., Machej, T., Socha, R. P., Włodarczyk, W., Gaweł, A., Matusik, J., Bahranowski, K., Wisła-Walsh, E., Lityńska-Dobrzyńska, L. & Serwicka, E. M. 2015 Active, selective and robust Pd and/or Cr catalysts supported on Ti-, Zr- or [Ti,Zr]-pillared montmorillonites for destruction of chlorinated volatile organic compounds. *Applied Catalysis B: Environmental* **174–175**, 293–307.

MOE 1993 *Environmental Quality Standards for Water Pollution*. Ministry of Environment, Japan. https://www.env.go.jp/en/water/wq/wp.pdf (accessed 26 March 2017).

Moliner-Martinez, Y., Herraez-Hernandez, R., Verdu-Andres, J., Campins-Falco, P., Garrido-Palanca, C., Molins-Legua, C. & Seco, A. 2013 Study of the influence of temperature and precipitations on the levels of BTEX in natural waters. *Journal of Hazardous Materials* **263**, 131–138.

Mottaleb, M. A., Abedin, M. Z. & Islam, M. S. 2003 Determination of benzene, toluene, ethylbenzene and xylene in River water by solid-phase extraction and gas chromatography. *Analytical Sciences* **19** (10), 1365–1369.

Murahashi, T., Sasaki, S. & Nakajima, T. 2003 Determination of endocrine disruptors in automobile exhaust particulate matter. *Journal of Health Science* **49** (1), 72–75.

Nagano, K., Umeda, Y., Senoh, H., Gotoh, K., Arito, H., Yamamoto, S. & Matsushima, T. 2006 Carcinogenicity and chronic toxicity in rats and mice exposed by inhalation to 1,2-dichloroethane for two years. *J. Occup. Health* **48** (6), 424–436.

Odabasi, M., Elbir, T., Dumanoglu, Y. & Sofuoglu, S. C. 2014 Halogenated volatile organic compounds in chlorine-bleach-containing household products and implications for their use. *Atmospheric Environment* **92**, 376–383.

Osha 2004 *Di-(2-Ethylhexyl)phthalate. Chemical Sampling Information*. Occupational Safety and Health Administration, USA. https://www.osha.gov/dts/chemicalsampling/data/CH_235155.html (accessed 16 March 2017).

Ost, D., Shah, R. D., Fein, D. & Fein, A. M. 2003 To screen or not to screen: a volatile issue in lung cancer. *Chest* **123** (6), 1788–1792.

Patterson, A. 2014 Is ASE No Longer a 'Serial Polluter'? EE Times. http://www.eetimes.com/document.asp?doc_id1324554 (accessed 26 March 2016).

Peijnenburg, W. J. G. M. & Struijs, J. 2006 Occurrence of phthalate esters in the environment of the Netherlands. *Ecotoxicology and Environmental Safety* **63** (2), 204–215.

Schmidt, T. C., Haderlein, S. B., Pfister, R. & Forster, R. 2004 Occurrence and fate modeling of MTBE and BTEX compounds in a Swiss Lake used as drinking water supply. *Water Research* **38** (6), 1520–1529.

Selvaraj, K. K., Sundaramoorthy, G., Ravichandran, P. K., Girijan, G. K., Sampath, S. & Ramaswamy, B. R. 2015 Phthalate esters in water and sediments of the Kaveri River, India: environmental levels and ecotoxicological evaluations. *Environmental Geochemistry and Health* **37** (1), 83–96.

Senthil Kumar, M., Sivasankar, V. & Gopalakrishna, G. V. T. 2017 Quantification of benzene in groundwater sources and risk analysis in a popular South Indian Pilgrimage City – a GIS based approach. *Arabian Journal of Chemistry* 10, S2523–S2533.

Smith, M. T., Yager, J. W., Steinmetz, K. L. & Eastmond, D. A. 1989 Peroxidase-dependent metabolism of benzene's phenolic metabolites and its potential role in benzene toxicity and carcinogenicity. *Environ. Health Perspect.* 82, 23–29.

Staples, C. A., Peterson, D. R., Parkerton, T. F. & Adams, W. J. 1997 The environmental fate of phthalate esters: a literature review. *Chemosphere* 35 (4), 667–749.

Sun, P. L. & Tsai, S. S. 2009 Intersex tilapia (Oreochromis spp.) from a contaminated river in Taiwan: a case study. *Toxins (Basel)* 1 (1), 14–24.

Tickner, J. A., Schettler, T., Guidotti, T., Mccally, M. & Rossi, M. 2001 Health risks posed by use of Di-2-ethylhexyl phthalate (DEHP) in PVC medical devices: a critical review. *American Journal of Industrial Medicine* 39 (1), 100–111.

UNEP 1987 *The Montreal Protocol on Substances that Deplete the Ozone Layer*. Ozone Secretariat, United Nations Environment Programme. http://ozone.unep.org/new_site/en/index.php (accessed 26 March 2017).

Uragami, T., Fukuyama, E. & Miyata, T. 2016 Selective removal of dilute benzene from water by poly(methyl methacrylate)-graft-poly(dimethylsiloxane) membranes containing hydrophobic ionic liquid by pervaporation. *Journal of Membrane Science* 510, 131–140.

USEPA 2008 *Integrated Risk Information System*. United States Environmental Protection Agency, Washington, DC, USA. https://www.epa.gov/iris (accessed 26 March 2017).

USEPA 2013 *Surface Water Sampling*. United States Environmental Protection Agency, Washington, DC, USA. https://www.epa.gov/sites/production/files/2015-06/documents/Surfacewater-Sampling.pdf (accessed 26 March 2017).

USEPA 2014 *Sediment Sampling*. United States Environmental Protection Agency, Washington, DC, USA. https://www.epa.gov/sites/production/files/2015-06/documents/Sediment-Sampling.pdf (accessed 26 March 2017).

USEPA 2015 *National Recommended Water Quality Criteria*. Human Health Criteria Table, United States Environmental Protection Agency, Washington, DC, USA. https://www.epa.gov/wqc/national-recommended-water-quality-criteria-human-health-criteria-table (accessed 26 March 2017).

Van Vliet, E. D. S., Reitano, E. M., Chhabra, J. S., Bergen, G. P. & Whyatt, R. M. 2011 A review of alternatives to di (2-ethylhexyl) phthalate-containing medical devices in the neonatal intensive care unit. *Journal of Perinatology* 31 (8), 551–560.

Van Wezel, A. P., Van Vlaardingen, P., Posthumus, R., Crommentuijn, G. H. & Sijm, D. T. 2000 Environmental risk limits for two phthalates, with special emphasis on endocrine disruptive properties. *Ecotoxicol. Environ. Saf.* 46 (3), 305–321.

Villanueva, M. C. & Ibarra, A. A. 2016 Assessing the ecological stress in a Garonne River stretch, southwest France. *Ecological Indicators* 67, 466–473.

Wang, J., Chen, G., Christie, P., Zhang, M., Luo, Y. & Teng, Y. 2015a Occurrence and risk assessment of phthalate esters (PAEs) in vegetables and soils of suburban plastic film greenhouses. *Science of the Total Environment* 523, 129–137.

Wang, Y. B., Liu, C. W., Kao, Y. H. & Jang, C. S. 2015b Characterization and risk assessment of PAH-contaminated river sediment by using advanced multivariate methods. *Science of the Total Environment* 524, 63–73.

Weaver, N., Gibson, R. & Smith, C. 1983 *Occupational Exposure to Benzene in the Petroleum and Petrochemical Industries. Carcinogenicity and Toxicity of Benzene*. Princeton Scientific Publishers, Princeton, NJ.

Williams, J. F. & Chang, C.-y. D. 2008 *Taiwan's Environmental Struggle: Toward a Green Silicon Island*. Routledge and Taylor & Francis, London, UK, p. 214.

Yamamoto, K., Fukushima, M., Kakutani, N. & Tsuruho, K. 2001 Contamination of vinyl chloride in Shallow Urban Rivers in Osaka, Japan. *Water Research* 35 (2), 561–566.

Yin, S. N., Li, Q., Liu, Y., Tian, F., Du, C. & Jin, C. 1987 Occupational exposure to benzene in China. *Br. J. Ind. Med.* 44 (3), 192–195.

Yu, J. J. & Chou, S. Y. 2000 Contaminated site remedial investigation and feasibility removal of chlorinated volatile organic compounds from groundwater by activated carbon fiber adsorption. *Chemosphere* 41 (3), 371–378.