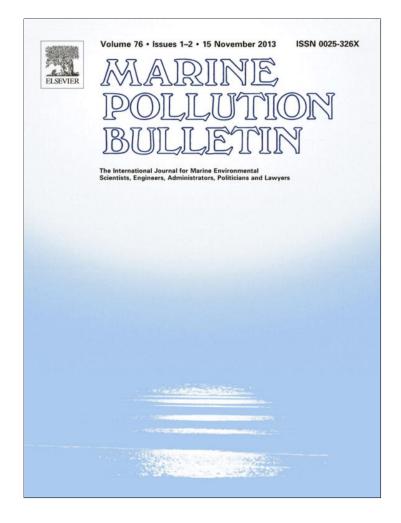
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Marine Pollution Bulletin 76 (2013) 370-378



Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Baseline

Risk assessment of the presence of polycyclic aromatic hydrocarbons (PAHs) in coastal areas of Thailand affected by the 2004 tsunami



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ARTICLE INFO

Keywords: Tsunami Polycyclic aromatic hydrocarbons (PAHs) Risk assessment Total toxic benzo[a]pyrene equivalent Soil cleanup target level (SCTL)

ABSTRACT

The total concentrations of twelve, likely carcinogenic, polycyclic aromatic hydrocarbons (PAHs) (i.e., phenanthrene (Phe), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), benz[a]anthracene (B[a]A), chrysene (Chry), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), benzo[a]pyrene (B[a]P), indeno[1,2,3-cd]pyrene (Ind), dibenz[a,h]anthracene (D[a,h]A), and benzo[g,h,i]perylene (B[g,h]P) in backwash deposits of the 2004 Khao Lak tsunami were carefully investigated and compared with the concentrations of world marine sediments (WMS). In general, \sum_{12} PAHs in this study (i.e., 69.43 ± 70.67 ng g⁻¹) were considerably lower than those values observed in marine sediments from Boston (54,253 ng g^{-1}), coastal sediments from Barcelona Harbour (15,069 ng g^{-1}), and riverine sediment from Guangzhou Channel (12,525 ng g^{-1}), but were greater than values from coastal sediments in Rosas Bay (12 ng g^{-1}), Santa Ponsa Bay (26 ng g^{-1}) and Le Planier (34 ng g^{-1}). The total toxic benzo[a]pyrene equivalent (TEQ^{Carc}) values calculated for Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS), and Songkhla Lake sediments (SLS) were $10.3 \pm 12.2 \text{ ng g}^{-1}$, $16.0 \pm 47.7 \text{ ng g}^{-1}$, and 5.67 ± 5.39 ng g⁻¹, respectively. Concentrations of PAHs at all study sites resulted in risk levels that fell into the "acceptable" range of the US EPA model and were much lower than those of other WMS. The cancer risk levels of PAH content in KCS ranged from 7.44×10^{-8} to 2.90×10^{-7} , with an average of $1.64 \times 10^{-7} \pm 8.01 \times 10^{-8}$; this value is 119 times lower than that of WMS. In addition, soil cleanup target levels (SCTLs) for both non-carcinogens (i.e., Phe, An, Fluo and Pyr) and carcinogens (i.e., B[a]A, Chry, B[b]F, B[k]F, B[a]P, Ind, D[a,h]A and B[g,h,i]P) in the KTS samples were estimated for all target groups, with an average value of 115,902 \pm 197,229 ng g⁻¹

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The massive tsunami generated by an earthquake on December 26th, 2004 was responsible for a wide range of negative effects, such as post-traumatic stress symptoms among Norwegian tsunami survivors, malaria outbreaks in Andaman & Nicobar Islands, damage to coastal ecosystems and other adverse geological impacts. Further difficulties in establishing post-disaster recovery policies, reconstruction of transportation systems and other environmental management problems were also exacerbated (Cochard et al., 2008; Danielsen et al., 2005; Ghobarah et al., 2006; Goff et al., 2006; Heir et al., 2010; Kumari et al., 2009; Matsumaru et al., 2012; Rigg et al., 2005; Thanawood et al., 2006). Apart from these adverse effects, the deposition of a layer of sediment rich in salts and other pollutants on the inundated coastal land areas raised serious concerns for geoscientists (Chaudhary et al., 2006; Szczuciński et al.,

2005, 2006; UNEP, 2005). Recent investigations examined standing brackish or saline water in numerous paddy fields on the Sendai plain caused by saltwater inundation triggered by the 2011 Tohoku-oki tsunami (Chagué-Goff, Niedzielski et al., 2011; Chagué-Goff, Andrew et al., 2011). By using geochemical marine markers (S and Cl) 2 months after the 2011 Tohoku-oki tsunami, the extent of tsunami inundation was identified as being 4.85 km from the shore (Chagué-Goff et al., 2012).

Thailand was one of the countries that was most affected by the catastrophic tsunami of December 26th, 2004, particularly in terms of coastal contamination from tsunami deposits. Pongpiachan et al. (2013a) applied Fourier Transform Infrared (FTIR) Spectroscopy followed by analysis of variance (ANOVA), Gaussian distribution, hierarchical cluster analysis (HCA) and principal component analysis (PCA) to discriminate typical marine sediments from tsunami backwash deposits. The same research group also used other environmental proxies, such as sulphur K-edge X-ray Absorption Near Edge Structure (XANES) spectroscopy and micro-beam

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synchrotron X-ray fluorescence (μ -SXRF), to identify tsunami backwash deposits in Khao Lak coastal areas (Pongpiachan et al., 2012, 2013b). Despite the various adverse effects and coastal contamination reported to date, there has been no environmental risk assessment on the presence of carcinogenic substances, such as polycyclic aromatic hydrocarbons (PAHs), in the areas of Thailand affected by the tsunami.

Polycyclic aromatic hydrocarbons (PAHs) have several characteristics that make them more long-term pollutants than water soluble ionic species and/or acid-leached heavy metals: (i) they contain benzene rings, whose structure is especially stable and difficult to break down, so they persist over the long-term (Beck et al., 1995; Wild et al., 1991; Wild and Jones, 1995); (*ii*) they are widely distributed in aerosols, terrestrial soils and marine sediments (Mostafa et al., 2009; Neff, 2002; Pongpiachan et al., 2013c; Pongpiachan, 2013d,e; Perra et al., 2011; Tipmanee et al., 2012; Wild et al., 1991; Wild and Jones, 1995); (iii) they are hydrophobic compounds so rain may not effectively dilute or remove these chemical substances (Meador, 2008) and (iv) they have to potential to bioaccumulate and to be carcinogenic (Luch and Baird, 2010; Rosenfeld and Feng, 2011). The main sources of PAHs are traffic exhaust, emissions from the iron, steel and coke industries, accidental oil spills and biomass burning (Rajput et al., 2011; Rosenfeld and Feng, 2011; Slezakova et al., 2012; Yim et al., 2002). Without abating these root causes, PAHs are likely to remain present in depositional sedimentary environments, including the tsunami inundation zone (see Figs. 1 and 2).

Overall, the objectives of this study were (*i*) to analyse PAH content in the marine sediments and terrestrial soils affected by the 2004 tsunami, and (*ii*) to employ various analogous risk assessment models to gain a better understanding of the potential carcinogenic risk in the tsunami-affected area. This is the first study to produce such a risk assessment for this area. The purpose of this paper to illustrate the general principle of using PAHs as proxies for cancer risk assessment in tsunami inundation zones rather than to determine the source apportionment or to produce an analysis of spatial variation in PAHs. In addition, soil cleanup target levels (SCTLs) for carcinogens and non-carcinogens in affected soils were estimated and discussed.

This study was carried out offshore along the west coast of Phang Nga province, in the southwest region of Thailand, which was heavily affected by the 2004 tsunami. Climate in this area is governed by two seasonal monsoons: the northeast monsoon from mid October until March and the southwest monsoon from May to September. The research area covered approximately 1000 km² between Thap Lamu and Pakarang Cape, up to a water depth of 70 m. Our research was carried out on cruises taken 1 year apart, from November–December 2007 with RV CHAKRATONG TONGYAI and from November–December 2008 with RV BOONLERT PASOOK. Hydroacoustic profiles (side scan sonar, multibeam echo sounder and shallow reflection seismic scan with a boomer system) covering approximately 1500 nautical miles were recorded near Pakarang Cape.

A side scan sonar system (conventionally used for sea floor acoustic backscatter measurements) was used in conjunction with characteristics of sediment samples to create sediment distribution maps for the area. Finer sediments appeared in lighter colours, while coarser sediments were displayed in darker colours. Multibeam echo sounders were employed to create detailed bathymetric maps, and the boomer, a high-resolution shallow water seismic system, provided some information related to the sediment structures in the subsurface. The sediment samples discussed in this study were taken with a Van-Veen-type grab sampler. The grab sampling stations were located on a transect line corresponding to surface sediment sampling stations. Twenty-two soil sampling stations were selected to represent the areas affected by the 2004 tsunami (Szczuciński et al., 2005). Samples were collected from 18 to 22 July 2009 from the surface layer (approximately the first 10 cm) using a clean shovel. Approximately half a kilogram of composite sample was taken from a 2 m^2 area at each station and pre-treated as surface sediment samples.

All solvents used were HPLC grade, purchased from Fisher Scientific. A cocktail of 12 PAH Norwegian Standards (NS 9815: S-4008-100-T) (phenanthrene (Phe), anthracene (An), fluoranthene (Fluo), pyrene (Pyr), benz[a]anthracene (B[a]A), chrysene (Chry), benzo[b]fluoranthene (B[b]F), benzo[k]fluoranthene (B[k]F), benzo [a]pyrene (B[a]P), indeno[1,2,3-cd]pyrene (Ind), dibenz[a,h]anthracene (D[a,h]A), and benzo[g,h,i]perylene (B[g,h]P); each 100 μ g mL⁻¹ in toluene; unit 1 \times 1 mL) and a mix of recovery Internal Standard PAHs (d_{10} -fluorene; d_{10} -Fl, d_{12} -perylene; d_{12} -Per); each 100 μ g mL⁻¹ in xylene; unit 1 \times 1 mL) were supplied by Chiron AS (Stiklestadveine 1, N-7041 Trondheim, Norway). Standard stock solutions of $4 \mu g m L^{-1}$ deuterated PAHs (used as internal standards) and 100 μ g mL⁻¹ native PAHs were prepared in nonane. Working solutions were obtained by appropriate dilution in n-cyclohexane. All solutions were stored in amber coloured vials at −20 °C.

After the freeze-drying process was completed, samples were weighed and placed in pre-cleaned Whatman cellulose extraction thimbles. The extraction of PAHs was conducted using 250 mL of Soxhlet extractor, spiked with a known amount of internal standard (d_{10} -Fl: Phe, An, Fluo, Pyr, B[a]A, and Chry; d_{12} -Per: B[b]F, B[k]F, B[a]P, Ind, D[a,h]A, and B[g,h,i]P) and extracted with DCM containing 1 g of copper powder for 8 h. The fractionation/cleanup process followed the conventional method using the difference in solvent polarity (Gogou et al., 1996, 1998). After the extraction, the DCM solvent was concentrated to incipient dryness by a combination of rotary evaporation and blowing under a gentle nitrogen stream. The concentrated extract was then diluted in 10 mL of *n*-hexane before being applied to the top of a column of disposable silica gel. The extract was then fractionated into individual compound classes by flash chromatography by applying the concentrate to the top of a 30×0.7 cm diameter column containing 1.5 g of silica gel (activated at 150 °C for 3 h).

Nitrogen pressure was used to obtain a flow of 1.4 mL min⁻¹ at the bottom of the column. The following solvents were used to elute the different compound classes: (*i*) 15 mL *n*-hexane (fraction 1, light molecular weight PAHs); and (*ii*) 15 mL toluene-*n*-hexane (5.6:9.4) (fraction 2, heavy molecular weight PAHs). In consideration of the toxicity of the solvent and the solubility of PAHs, toluene was selected for the study. After fractionation, the eluates were concentrated using a rotary evaporator followed by evaporation under a gentle nitrogen stream (flow rate set at 1.0 mbar). Because of the low dissipation capability of toluene, a percentage (5–25%) of acetone was added to increase the volatility. The sample was further reduced to incipient dryness prior and then brought to volume with cyclohexane (exactly 100 µL in a GC/MS vial insert) prior to GC/MS analysis. The recovery of the internal standards d_{10} -FI and d_{12} -Per were 57% and 53%, respectively.

GC/MS analyses were performed using a Varian GC/MS–MS system comprised of a CP-3900 gas chromatograph (Walnut Creek, CA, USA) with a 1077 universal injector and a three-dimensional quadrupole ion-trap selected ion storage mass spectrometer (Varian Saturn 2200). The target compounds were separated on a 60 m length \times 0.25 mm i.d. capillary column coated with a 0.25 µm thick film (phase composition: cross-linked/surface bonded 5% phenyl, 95% methylpolysiloxane, as specified by EPA methods 207, 508, 515, 515.2, 524.2, 525, 548.1, 680, 1625, 1653, 8081, 8141, 8270 and 8280) stationary phase (Agilent JW Scientific DB-5 GC columns). Helium (99.999%) was employed as carrier gas at a constant column flow of 1.0 mL min⁻¹ and a pressure pulse of 25 psi with a duration of 0.50 min. All injections (1 µL) were performed through

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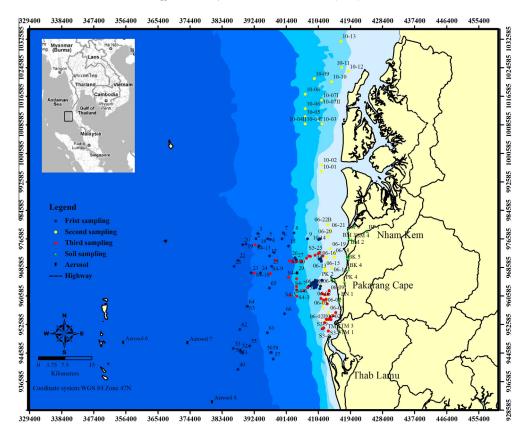


Fig. 1. Sampling sites for coastal sediments and terrestrial soils adjacent to the Khao Lak area affected by the 2004 tsunami.

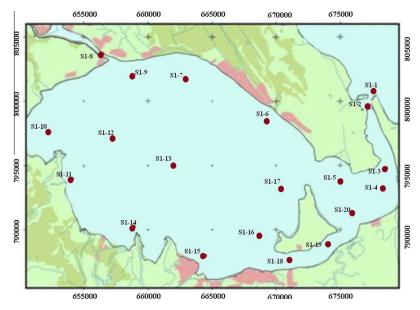


Fig. 2. Sampling sites for the Songkhla Lake sediments.

a universal injector (Varian 1077) in the splitless mode, and the standards were introduced using a 10 μ L Hamilton syringe. The GC oven temperature was programmed as follows: 1 min at 40 °C increased by 8 °C min⁻¹ to 300 °C and held for 45 min. By employing these chromatographic conditions it was possible to qualitatively distinguish between Ph/An and B[a]A/Chry, two pairs of isomers that are commonly co-eluted by gas chromatographic systems. All details of the GC/MS–MS ion trap operational parameters for the analytical method are given in Table 1.

The molecular markers were identified by first comparing retention times with authentic standards (within a range of $\pm 0.2 \text{ min}$) and secondly by quantification of ions of each individual congener (Pongpiachan et al., 2009). The relative response factor (*RRF*), used to quantify the target congeners, was calculated as follows:

$$RRF = \frac{A_{nat}}{A_{is}} \times \frac{C_{is}}{C_{nat}} \tag{1}$$

where A_{nat} = the peak area of the native compound in the standard; C_{nat} = the concentration of the native compound in the standard; A_{is} = the peak area of the internal standard; and C_{is} = the concentration of the internal standard. The *RRF*_{STD} used for quantifying samples was the mean of the *RRF* values calculated for the two quantification standards run on the same day. Concentration (*C*) of analytes in sample extracts was calculated using the following formula:

$$C = \frac{A_{nat}}{A_{is}} \times \frac{1}{RRF_{STD}} \times \frac{W_s}{W_{is}}$$
⁽²⁾

where W_{is} = weight of internal standard (*IS*) added to the sample, and W_s = the weight or volume of the sample analysed. A recovery determination standard (*RDS*) was used for the calculation of *IS* and the sampling efficiency standard (*SES*) of recoveries during sample preparation and extraction/purification. A known amount of *RDS* was added at the final stage prior to GC/MS analysis and was assumed to suffer zero loss. The percent recovery was calculated as follows:

$$\% \text{Recovery} = \left[\left(\frac{A_{is}}{A_{RDS}} \right)_{S} \times \left(\frac{A_{RDS}}{A_{is}} \right)_{STD} \right] \\ \times \left[\left(\frac{C_{is}}{C_{RDS}} \right)_{STD} \times \left(\frac{C_{RDS}}{C_{is}} \right)_{S} \right] \times 100\%$$
(3)

where A_{RDS} = the peak area of the recovery determination standard; and C_{RDS} = the concentration of the recovery determination standard. Recoveries of *IS* were used as an indication of the analytical losses during the extraction, pre-concentration, cleanup/fractionation and blow down stages. Standard reference materials (SRM-NIST1941b-Marine Sediment) were used to perform both quality control and quality assurance of analytical results. Mean recovery (based on extraction of matrix-matched certified reference materials SRM-NIST1941b-Marine Sediment, n = 8) ranged from 78% to 107%. The precision of the procedure, calculated as the relative standard deviation of duplicate samples, was less than 15%. Sample concentrations were calculated using *RRFs* run in between each batch.

Table 1

GC-EI-MS-MS ion trap operational parameters for PAHs analyses.

I. Ion preparation parameters	
CID waveform	Resonant and non-resonant
Mass isolation window	1
Isolation time	5 ms
Excitation time	5 ms
Ejection amplitude	20 V
Broadband amplitude	30 V
Modulation rate	30 μ s step ⁻¹
II. Ionisation parameters	
Ion trap temperature	200 °C
Transfer temperature	280 °C
Manifold temperature	120 °C
Axial modulating voltage	4.0 V
Emission current	90 μA
Electron multiplier voltage	1550 V
Scan rate	0.58 s scan^{-1}
Pre-scan ionisation time	1500 μs
Target TIC	20,000
Maximum ionisation time	25,000
RF pump value	650 m z^{-1}
Background mass	45 m z^{-1}
Count threshold	1 count
Isolation window	3.0 m z^{-1}
III. Injection port parameters	
Injection volume	1 μL
Injection mode	Splitless
Injector type	Glass liner
Carrier gas type	Helium
Septum purge flow rate	1 mL min^{-1}

Instrumental detection limit (*IDL*) and limit of quantification (*LOQ*) values ranged from 0.77 to $61.25 \,\mu g \,m L^{-1}$ and 2.57 to 204.17 $\mu g \,m L^{-1}$, respectively (Table 2). The range of the concentrations was comparable to previous studies (Helaleh et al., 2005; Ströher et al., 2007).

Table 3 lists the average, minimum, maximum and standard deviation of PAH concentrations in the marine sediments of Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS), Songkhla Lake sediments (SLS) and world marine sediments (WMS). The total concentrations of twelve potentially carcinogenic PAHs in KCS ranged from 12.58 ng g^{-1} to 278.10 ng g^{-1} , with an arithmetic mean of 69.43 ± 70.67 ng g⁻¹. \sum_{12} PAHs refers to the sum of analysed Phe, An, Fluo, Pyr, B[a]A, Chry, B[b]F, B[k]F, B[a]P, Ind, D[a,h]A and B[g,h,i]P. The observed \sum_{12} PAHs were much lower than those values reported in harbour sediments from Boston (54,253 ng g^{-1} '), coastal sediments from Barcelona Harbour (15,069 ng g^{-1}), riverine sediments from Guangzhou Channel (12,525 ng g^{-1}), mangrove sediments from Hong Kong (3714 ng g^{-1}), coastal sediments from Cotonou (1189 ng g⁻¹) and coastal sediments from Carteau (210 ng g⁻¹), but were higher than values from coastal sediments in Rosas Bay (12 ng g^{-1}), Santa Ponsa Bay (26 ng g^{-1}) and Le Planier (34 ng g^{-1}) (Baumard et al., 1998; Bixian et al., 2001; Deshmukh et al., 2001; Mielke et al., 2001; Soclo et al., 2000; Tam et al., 2001; Wang et al., 2001).

It is generally accepted that B[a]P can be considered a representative member of PAHs due to its relatively high carcinogenic potency factor in comparison with other potentially carcinogenic PAHs (Peters et al., 1999). Despite a relatively high diversity in molecular structure, only B[a]A, B[a]P, B[b]F, B[k]F, Chry, D[a,h]A and Ind have been classified as potentially carcinogenic in humans PAH compounds (CPAHs) by the US EPA. Percentage contribution of CPAHs to \sum_{12} PAHs varied from 10.19% to 64.78%, with an average of 51.54 ± 66.05%; from 2.82% to 39.40%, with an average of 38.03 ± 39.85%; and from 35.21% to 44.79%, with an average of 39.86 ± 46.81% for KCS, KTS and SLS, respectively.

Because the toxicity of PAHs is extremely structurally dependent, it was therefore crucial to adopt the concept of toxic equivalency factors (*TEFs^{Carc}*) for the evaluation of total effects caused by all probable human carcinogenic PAHs. In this study, the default *TEFs^{Carc}* values for B[a]A, B[a]P, B[b]F, B[k]F, Chry, D[a,h]A and Ind were 0.1, 1, 0.1, 0.01, 0.001, 1 and 0.1, respectively, as defined by the US EPA. Further attempts to convert the above-mentioned seven PAHs into one toxic concentration for each observatory site were conducted using the corresponding *TEFs^{Carc}*. The evaluation of total toxic B[a]P equivalent (*TEQ^{Carc}*) for all seven PAHs was calculated as described below:

$$TEQ^{Carc} = \Sigma_i C_i \times TEF_i^{Carc} \tag{4}$$

Total *TEQ^{Carc}* values computed for samples collected at WMS, KCS, KTS and SLS varied from 1.68 to 9099 ng g^{-1} dry weight, with an average of 763 ± 2018 ng g^{-1} ; from 0.0345 to 55.7 ng g^{-1} dry weight, with an average of 10.3 ± 12.2 ng g^{-1} ; from 0.012 to 222 ng g^{-1} dry weight, with an average of 16.0 ± 47.7 ng g^{-1} ; and from 0.869 to 18.5 ng g^{-1} dry weight, with an average of 5.67 ± 5.39 ng g^{-1} , respectively.

Notably, the average total TEQ^{Carc} values for KCS, KTS and SLS were 74, 48 and 135 times lower than for WMS, indicating a relatively low CPAH content at the study sites. Interestingly, the average CPAH content of KCS was approximately two times higher than that from SLS, possibly reflecting the influence of CPAH-contaminated terrestrial input triggered by tsunami backwash in 2004. Contribution to the total TEQ^{Carc} of KCS of the different PAH congeners was as follows, in decreasing order: D[a,h]A (68.83%) > B[a]P (20.05%) > Ind (6.27%) > B[b]F (2.63%) > B[a]A (1.94%) > B[k]F (0.26%) > Chry (0.02%). A similar der was observed in SLS, with

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Table 2

Retention time (RT), instrumental detection limit (IDL), limit of quantification (LOQ), peak resolution (R), theoretical plate number (N) and height equivalent of theoretical plate (HETP) for each PAH.

РАН	RT (min)	$_{(\text{pg}\mu l^{-1})}^{\text{IDL}}$	$\begin{array}{c} \text{LOQ} \\ (\text{pg} \ \mu l^{-1}) \end{array}$	R	Ν	HETP (mm)	Low mass $(m z^{-1})$	High mass (m z ⁻¹)	Quantification ion $(m z^{-1})$	Excitation amplitude (V)
Phe	26.53	2.63	8.77	1.44	931693	0.064	177	179	178	0.50
An	26.63	2.80	9.33	1.44	936934	0.067	177	179	178	0.50
Fluo	30.13	1.47	4.90	7.62	2136956	0.029	201	203	202	0.40
Pyr	30.83	1.35	4.50	7.62	1624902	0.037	201	203	202	0.40
B[a]A	34.45	0.86	2.87	0.94	2418061	0.026	227	229	228	0.40
Chry	34.58	0.84	2.80	0.94	1962729	0.032	227	229	228	0.40
B[b]F	38.45	48.66	162.20	1.35	2102556	0.034	251	253	252	0.40
B[k]F	38.65	43.42	144.73	1.35	2112533	0.038	251	253	252	0.40
B[a]P	40.05	61.25	204.17	2.40	2176625	0.036	251	253	252	0.40
Ind	46.89	61.25	204.17	2.45	3913510	0.017	275	277	276	0.40
D[a,h]A	47.11	45.39	151.30	2.45	2244480	0.489	277	279	278	0.40
B[g,h,i]P	48.93	2.21	7.37	-	403043	3.930	275	277	276	0.40

Table 3

Summary statistics for PAH concentrations (ng g⁻¹) in Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS), Songkhla Lake sediments (SLS) and world marine sediments (WMS).

	KCS			KTS			SLS			WMS						
	Aver ^a	Stdev ^b	Min ^c	Max ^d	Aver	Stdev	Min	Max	Aver	Stdev	Min	Max	Aver	Stdev	Min	Max
Phe	6.81	3.40	1.28	20.0	16.4	39.1	3.72	194	10.0	9.22	1.12	36.5	673	1621	1.09	7770
An	2.95	1.66	0.44	8.94	2.60	5.77	0.06	28.4	1.21	1.28	0.18	5.03	181	447	0.16	2230
Fluo	3.76	2.22	0.74	11.7	31.4	113	2.27	545	2.65	2.54	0.75	10.8	802	2285	1.53	11,610
Pyr	4.78	2.67	0.94	16.9	25.8	71.4	3.74	350	3.05	1.79	0.89	6.66	776	2095	1.30	10,548
B[a]A	2.00	1.89	ND ^e	9.91	7.97	30.8	ND	149	0.98	1.32	0.14	4.68	483	1092	0.75	4924
Chry	1.82	2.59	ND	15.3	6.95	21.5	0.28	105	1.05	1.61	0.09	6.11	407	840	1.11	3915
B[b]F	2.71	2.69	0.14	10.7	12.4	44.0	ND	213	3.05	3.76	0.42	15.1	663	1911	1.85	8130
B[k]F	2.67	3.00	0.07	18.7	3.93	12.8	ND	62.0	1.64	2.68	0.23	10.7	391	623	11.0	1910
B[a]P	2.06	2.17	ND	11.7	8.80	33.1	ND	160	1.89	1.51	0.16	3.52	376	794	1.00	3714
Ind	6.45	7.39	0.20	38.4	5.76	13.9	0.11	67.8	2.87	2.88	0.41	11.6	427	841	0.95	3112
D[a,h]A	7.09	8.83	ND	37.9	4.53	5.58	ND	18.5	3.08	3.06	0.61	11.7	225	832	0.21	3745
B[g,h,i]P	5.02	4.75	0.15	20.0	5.77	15.0	ND	73.1	5.02	4.28	0.85	19.3	347	730	0.86	2650

Note that ^aAverage, ^bStandard Deviation, ^cMinimum, ^dMaximum and ^eNot Determined for WMS are averages for PAHs from marine sediments from Bayou St. John, New Orleans, USA (Mielke et al., 2001), harbour sediments form San Diego Harbour, California, USA (Deshmukh et al., 2001), marine sediments from the Northern Irish Sea, Northern Ireland, UK (Guinan et al., 2001), harbour sediments from Mystic River, Boston, USA (Wang et al., 2001), harbour sediments from Mangrove, Sai Keng, Hong Kong (Tam et al., 2001), marine sediments from Mangrove, Tolo, Hong Kong (Tam et al., 2001), Marine Sediments of Mangrove, Ho Chung, Hong Kong (Tam et al., 2001), Marine Sediments, Mangrove, Mai Po, Hong Kong (Tam et al., 2001), River Sediments of Pearl River, Guangzhou Channel, China (Bixian et al., 2001), river sediments from Pear River, Shiziyang Channel, China (Bixian et al., 2001), marine sediments from the northern Adriatic Sea, Italy (Notar et al., 2001), coastal sediments from Cotonou (Benin), Africa (Soclo et al., 2000), coastal sediments from the Mediterranean, France (Baumard et al., 1998), coastal sediments from Brazil (Medeiros and Bicego, 2004), and coastal sediments from Guba Pechenga, Russia (Savinov et al., 2003).

D[a,h]A (54.28%) > B[a]P (33.24%) > B[b]F (5.38%) > Ind (5.07%) > B[a]A (1.73%) > B[k]F (0.29%) > Chry (0.02%). This result highlights the importance of D[a,h]A in both coastal and lake sediments in the southern part of Thailand. The contribution of PAHs to the total *TEQ^{Carc}* of WMS was B[a]P (49.26%) > D[a,h]A (29.55%) > B[b]F (8.69%) > B[a]A (6.33%) > Ind (5.60%) > B[k]F (0.51%) > Chry (0.05%), which highlights the major contribution of B[a]P to the total *TEQ^{Carc}* of worldmarine sediments.

Environmental cancer risk analyses have been widely performed on marine deposits, lake sediments and terrestrial soils around the world (Bejarano and Michel, 2010; Cachada et al., 2012; Li et al., 2012; Mebarka et al., 2012; Mirsadeghi et al., 2011; Zhang et al., 2012). In this study, an assessment of both environmental cancer and non-cancer risks was conducted following the framework proposed by The Agency for Toxic Substances and Disease Registry (ATSDR), a federal public health agency of the US Department of Health and Human Services. In addition, an evaluation of environmental cancer risks was performed to assess occurrence of adverse health effects (carcinogenic and noncarcinogenic) resulting from ingestion or dermal contact with PAHcontaminated sediments. Cancer risk was calculated as follows:

$$Cancer \operatorname{Risk} = \operatorname{SF} \times \left(\frac{CS \times IR \times EF \times ED \times CF}{BW \times AT_{c}} + \frac{CS \times SA \times AF \times EF \times ED \times DAF \times CF}{BW \times AT_{c}}\right) \quad (5)$$

where SF = slope factor (1.5 mg kg⁻¹ body weight day⁻¹)⁻¹; CS = chemical concentration (ng g⁻¹); CF = a conversion factor of 10⁻⁶ kg mg⁻¹; EF = an exposure frequency, set to 40 days year⁻¹; ED = exposure duration, set to 10 years; IR = ingestion rate, set to 50 mg sediment day⁻¹; BW = body weight, set to 53 kg; $AT_c =$ averaging time for cancer risk, set to 25,550 days; AF = skin adherence factor, set to 0.3 mg sediment cm⁻¹; DAF = dermal absorption factor, set to 0.03 (unitless); and SA = skin surface area (available for contact), set to 4700 cm².

The evaluation of non-cancer risk was performed by calculating a Hazard Index (*HI*) using Eq. (6). The Hazard Index (*HI*) represents the non-cancer risk computed by dividing the sum of the average daily dose ingestion ($ADD_i = mg kg^{-1}$ body weight day⁻¹) and the average daily dose dermal ($ADD_d = mg kg^{-1}$ body weight day⁻¹) by the oral reference dose (RfD) (ATSDR, 2005) as follows:

$$HI = \frac{\frac{CS \times IR \times EF \times ED \times CF}{BW \times AT_{NC}} + \frac{CS \times SA \times AF \times EF \times ED \times DAF \times CF}{BW \times AT_{NC}}}{RfD}$$
(6)

Table 4

Levels of cancer risk from PAHs in Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS), Songkhla Lake sediments (SLS) and world marine sediments (WMS) (ATSDR, 2005).

	KCS		KTS		SLS		WMS		
	Aver ^a	Stdev ^b	Aver	Stdev	Aver	Stdev	Aver	Stdev	
Phe	2.785E-07	1.389E-07	6.720E-07	1.598E-06	4.100E-07	3.769E-07	2.751E-05	6.630E-05	
An	1.208E-07	6.796E-08	1.065E-07	2.361E-07	4.963E-08	5.234E-08	7.367E-06	1.830E-05	
Fluo	1.536E-07	9.062E-08	1.283E-06	4.606E-06	1.082E-07	1.038E-07	3.279E-05	9.345E-05	
Pyr	1.953E-07	1.092E-07	1.054E-06	2.919E-06	1.248E-07	7.302E-08	3.175E-05	8.567E-05	
B[a]A	8.159E-08	7.746E-08	3.258E-07	1.259E-06	4.022E-08	5.392E-08	1.973E-05	4.465E-05	
Chry	7.439E-08	1.061E-07	2.844E-07	8.778E-07	4.274E-08	6.572E-08	1.665E-05	3.435E-05	
B[b]F	1.109E-07	1.101E-07	5.057E-07	1.799E-06	1.247E-07	1.536E-07	2.711E-05	7.817E-05	
B[k]F	1.090E-07	1.229E-07	1.605E-07	5.216E-07	6.695E-08	1.096E-07	1.598E-05	2.550E-05	
B[a]P	8.442E-08	8.871E-08	3.599E-07	1.354E-06	7.709E-08	6.191E-08	1.536E-05	3.248E-05	
Ind	2.640E-07	3.024E-07	2.355E-07	5.682E-07	1.175E-07	1.178E-07	1.748E-05	3.440E-05	
D[a,h]A	2.899E-07	3.611E-07	1.853E-07	2.283E-07	1.259E-07	1.250E-07	9.215E-06	3.404E-05	
B[g,h,i]P	2.052E-07	1.941E-07	2.360E-07	6.148E-07	2.053E-07	1.750E-07	1.417E-05	2.986E-05	

^a Average.

^b Standard deviation.

where the abbreviations of incidental ingestion formula (Eq. (6)) can be described as follows:

HI: Hazard Index.

RfD: reference dose $(3.0 \times 10^{-4} \text{ mg kg}^{-1} \text{ body weight day}^{-1})$ *AT_{NC}*: averaging time for non-cancer effects = 3650 days.

Cancer risk levels from PAHs in KCS ranged from 7.44×10^{-8} to 2.90×10^{-7} , with an average of $1.64 \times 10^{-7} \pm 8.01 \times 10^{-8}$, which is 119 times lower than that of WMS (Table 4). All cancer risk levels for KCS, KTS and SLS were significantly lower (p < 0.001) than those for WMS. Interestingly, the average cancer risk level for KCS was 2.75 times lower than for KTS but only slightly (1.32 times) higher than for SLS. No statistically significant differences were observed between KCS and KTS or between KCS and SLS, implying that the distribution of PAHs in tsunami affected areas was relatively homogeneous (see Table 5).

According to the "Role of the Baseline Risk Assessment in Superfund Remedy Selection Decisions" memorandum provided by US EPA (1991), the non-carcinogenic Hazard Index (HI) can be used as a parameter to assess the cumulative, non-carcinogenic, site risk to an individual based on reasonable maximum exposure. If HI is less than one, action is not warranted unless there are adverse environmental impacts for both current and future land use. The Superfund Program uses a cancer risk level of 10^{-6} to indicate the point at which risk management decisions should be considered. Using the same standard, the possibility of adverse cancer-causing effects in tsunami-affected areas was considerably lower in comparison to world marine sediments. In this study, the HI for non-cancer risk from PAHs in KCS ranged from 1.16×10^{-3} to 4.51×10^{-3} , with an average of $2.55 \times 10^{-3} \pm 1.25 \times 10^{-3}$; this risk level is 392 times lower than that of US EPA (1991) baseline. Inaddition, the average HI of non-cancer risk from PAHs in WMS was 3.28 times lower than the US EPA baseline, further indicating that the HI falls into the "acceptable level" range.

In this study, the acceptable soil cleanup target levels (SCTLs) for both carcinogens and non-carcinogens for aggregate residents were calculated using the following equations developed by FDEP (2005):



where the abbreviations can be described as follows:

TR: target cancer risk (unitless; 0.000001). BW: body weight (Asian people BW 57.7 kg; Walpole et al., 2012). AT: averaging time (25,500 days). *EF*: exposure frequency (350 days yr^{-1}). ED: exposure duration (30 yr). RBA: relative bioavailability factor (unitless). FC: fraction from contaminated source (1.0 unitless). CSF_o : cancer slope factor for oral exposure (mg kg⁻¹ day⁻¹; chemical specific from WIDNR, 1997). IR_o : ingestion rate, oral (120 mg day⁻¹). *CSF_d*: cancer slope factor for dermal exposure (mg kg⁻¹ day⁻¹; chemical specific from WIDNR, 1997). SA: surface area of skin exposed ($4810 \text{ cm}^2 \text{ day}^{-1}$). AF: adherence factor (0.1 mg cm⁻²). DA: dermal absorption (0.01 unitless). CSF_i : cancer slope factor for inhalation (mg kg⁻¹ day⁻¹; chemical specific from WIDNR, 1997). IR_i : inhalation rate (12.2 m³ day⁻¹). PEF: particulate emission factor, calculated as $(Q \times 3600)/$ $(C \times 0.036 \times (1 - V) \times (U_m/U_t)^3 \times F(x))$ (2.23794 × 10⁸ m³ kg⁻¹; FDEP, 2005). Q/C: inverse of mean concentration at centre of a 0.5 acre² source (g m⁻²-s per kg m⁻³; 85.61; US EPA, 1996b). V: fraction of vegetative cover (dimensionless; 0.5; FDEP, 2005). U_t : equivalent threshold value of wind speed at altitude of 30 m (m s⁻¹; 6.0; DEDP,2001). U_m : mean annual wind speed (m s⁻¹; 4.4; DEDP, 2001). F(x): function dependent on U_m/U_t (unitless; 0.194; US EPA, 1985). *VF*: volatilisation factor (m³ kg⁻¹; chemical specific from ATSDR, 1995). CF: conversion factor $(10^{-4} \text{ m}^2 \text{ cm}^{-2})$. D_A : apparent diffusivity (cm² s⁻¹; chemical specific). *T*: exposure interval (*ED* (exposure duration) \times 3.1536 \times 10⁷ s).

SCTL: soil cleanup target level.

 ρ_b : dry soil bulk density (1.5 g cm⁻³; FDEP, 2005).

The values for U_m and U_t for the areas affected by the 2004 tsunami were adopted from the Department of Energy Development and Promotion, Ministry of Energy, Thailand. The calculated value of *PEF* was 2.23794 × 10⁸, which was one order lower than of the values of FDEP (i.e., 1.24101 × 10⁹). It is also worth mentioning that other calculations of SCTL were performed using values of

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Table 5

Levels of non-cancer risk from PAHs in Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS), Songkhla Lake sediments (SLS) and world marine sediments (WMS) (ATSDR, 2005).

	KCS		KTS		SLS		WMS		
	Aver ^a	Stdev ^b	Aver	Stdev	Aver	Stdev	Aver	Stdev	
Phe	4.332E-03	2.161E-03	1.045E-02	2.486E-02	6.378E-03	5.863E-03	4.280E-01	1.031E+00	
An	1.879E-03	1.057E-03	1.657E-03	3.673E-03	7.720E-04	8.142E-04	1.146E-01	2.846E-01	
Fluo	2.389E-03	1.410E-03	1.996E-02	7.165E-02	1.684E-03	1.615E-03	5.100E-01	1.454E+00	
Pyr	3.039E-03	1.699E-03	1.640E-02	4.541E-02	1.942E-03	1.136E-03	4.939E-01	1.333E+00	
B[a]A	1.269E-03	1.205E-03	5.068E-03	1.959E-02	6.257E-04	8.387E-04	3.070E-01	6.946E-01	
Chry	1.157E-03	1.650E-03	4.424E-03	1.365E-02	6.648E-04	1.022E-03	2.589E-01	5.344E-01	
B[b]F	1.726E-03	1.712E-03	7.867E-03	2.799E-02	1.940E-03	2.389E-03	4.216E-01	1.216E+00	
B[k]F	1.696E-03	1.911E-03	2.497E-03	8.113E-03	1.041E-03	1.705E-03	2.485E-01	3.966E-01	
B[a]P	1.313E-03	1.380E-03	5.599E-03	2.106E-02	1.199E-03	9.631E-04	2.390E-01	5.053E-01	
Ind	4.106E-03	4.704E-03	3.664E-03	8.838E-03	1.828E-03	1.832E-03	2.719E-01	5.351E-0	
D[a,h]A	4.509E-03	5.618E-03	2.882E-03	3.551E-03	1.958E-03	1.944E-03	1.433E-01	5.296E-0	
B[g,h,i]P	3.193E-03	3.019E-03	3.671E-03	9.563E-03	3.193E-03	2.722E-03	2.204E-01	4.645E-01	

^a Average.

^b Standard deviation.

Table 6

Soil clean up target level (SCTL; mg kg⁻¹) for carcinogens and non-carcinogens in Khao Lak terrestrial soils (KTS) (FDEP, 2005).

PAH congener	SCTL carcinoge	ens		SCTL non-carcinogens					
	Worker	Child	Aggregate resident	Worker	Child	Aggregate resident			
Phe	NA ^a	NA	NA	66,655	2628	35,031			
An	NA	NA	NA	666,508	26,277	350,312			
Fluo	NA	NA	NA	88,877	3,504	46,709			
Pyr	NA	NA	NA	66,659	2628	35,032			
B[a]A	1.40	0.109	2.04	NA	NA	NA			
Chry	138	10.7	194	NA	NA	NA			
B[b]F	1.39	0.108	2.00	NA	NA	NA			
B[k]F	14.1	1.09	20.7	NA	NA	NA			
B[a]P	0.140	0.0109	0.210	NA	NA	NA			
Ind	1.41	0.109	2.07	NA	NA	NA			
D[a,h]A	0.140	0.0109	0.210	NA	NA	NA			
B[g,h,i]P	14.1	1.09	20.8	NA	NA	NA			

^a Not Available.

BW, *AF*, *AT*, *EF*, *ED*, *IR*₀, *IR*₁ and *SA* of 16.8 kg, 0.2 mg cm⁻², 2190 days, 350 days yr⁻¹, 6 yrs, 200 mg day⁻¹, 8.1 m³ day⁻¹, and 2960 cm² - day⁻¹, respectively, for children and 76.1 kg, 0.2 mg cm⁻², 9125 days, 250 days yr⁻¹, 25 yrs, 50 mg day⁻¹, 20 m³ day⁻¹, 3500 cm² day⁻¹, respectively, for workers (FDEP, 2005).

A similar model was developed for calculating acceptable soil cleanup target levels for non-carcinogens as follows (FDEP, 2005):

$$SCTL = \frac{THI \times BW \times AT \times RBA}{EF \times ED \times FC \times \left[\left(\frac{1}{R_{D_{2}}} \times R_{0} \times 10^{-6} \frac{k_{T}}{m_{0}} \right) + \left(\frac{1}{R_{D_{1}}} \times SA \times AF \times DA \times 10^{-6} \frac{k_{T}}{m_{0}} \right) + \left(\frac{1}{R_{D_{1}}} \times R_{1} \times \left(\frac{1}{\frac{1}{2} - \frac{1}{m_{0}} \frac{1}{m_{0}}} \right)^{\frac{1}{2} + CF_{1} \times \frac{1}{2} + C$$

where *THI*, RfD_o , RfD_d and RfD_i are the target *HI* (unitless), reference dose for oral exposure (mg kg⁻¹ day⁻¹; chemical specific), reference dose for dermal exposure (mg kg⁻¹ day⁻¹; chemical specific) and reference dose for inhalation (mg kg⁻¹ day⁻¹; chemical specific), respectively.

In addition, D_A can be calculated as follows:

$$D_{A} = \frac{\left[\frac{\left(\theta_{a}^{10/3}D_{i}H' + \theta_{w}^{10/3}D_{W}\right)}{n^{2}}\right]}{\rho_{b}K_{d} + \theta_{w} + \theta_{a}H'}$$
(9)

where θ_a , θ_w , D_i , D_w , H' and K_d are the air-filled soil porosity (g cm⁻³), water-filled soil porosity (g cm⁻³), diffusivity in air (cm² - s⁻¹), diffusivity in water (cm² s⁻¹), Henry's law constant (unitless) and soil–water partitioning coefficient (L kg⁻¹), respectively (FDEP, 2005).

Results obtained from the estimations of soil cleanup target levels (SCTLs) for carcinogens and non-carcinogens are shown in Table 6. These results were based on the concentrations recorded in KTS samples, which showed a maximum SCTL for the carcinogenic PAH Chry of 194 ng g⁻¹, 10.7 ng g⁻¹ and 138 ng g⁻¹ for aggregate residents, children and workers, respectively. B[a]P and D[a,h]A are two PAH congeners that require the minimum SCTL for all target groups. In addition, SCTL for the non-carcinogens Phe, An, Fluo and Pyr were estimated for KTS samples using Eq. (8). SCTL ranged from 2628 to 666,508 ng g⁻¹ for all target groups, with an average value of 115,902 ± 197,229 ng g⁻¹.

The assessment of sediment toxicity based on the total toxic benzo[a]pyrene equivalent (TEQ_{carc}), evaluations of cancer and non-cancer risks, coupled with the estimation of soil cleanup target levels (SCTLs) from the PAHs data set produced by this study of the Khao Lak area affected by the 2004 tsunami, underscore two principal challenges. Firstly, the lack of knowledge related to the distribution of PAHs in tsunami-affected areas limits the ability to interpret data in comparison with other tsunami inundation zones. Secondly, the majority of default values used in the risk evaluation for both carcinogens and non-carcinogens, including calculations of SCTL, were developed for US citizens, and thus no effective Soil/Sediment Quality Guidelines (SQGs) are available for the southern part of Thailand or even the Andaman Sea. In spite of these research constraints, the approach adopted in this study emphasised that (i) the average concentrations of PAHs in all study sites were much lower than the average for world marine sediments, (ii) no adverse environmental and human health effects S. Pongpiachan et al./Marine Pollution Bulletin 76 (2013) 370-378

have been connected with exposure to PAHs in Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS) and Songkhla Lake sediments (SLS) and (iii) more comprehensive investigation based on site specific studies is necessary.

Polycyclic aromatic hydrocarbons are ubiquitous in Khao Lak coastal sediments (KCS), Khao Lak terrestrial soils (KTS) and Songkhla Lake sediments (SLS). The total concentrations of twelve potentially carcinogenic PAHs in all samples varied by three orders of magnitude, ranging from 3.97 ng g^{-1} to 1965.47 ng g^{-1} , with an arithmetic mean of $72.29 \pm 161.55 \text{ ng g}^{-1}$. The percentage contribution of CPAHs to $\sum_{12} \text{PAHs}$ in all samples varied from 2.82% to 64.78%, with an average value of 41.59 ± 50.90%. Remarkably, the average total TEQ^{Carc} values for KCS, KTS and SLS were 74, 48 and 135 times lower than those of WMS, respectively, signifying a comparatively low CPAH concentration at the study sites. It is also worth mentioning that the average HI for non-cancer risk from PAHs at KCS was 392 times lower than that of the US EPA (1991) baseline, emphasising the fact that the degree of HI falls into the "acceptable level" range. In addition, SCTL for the non-carcinogens Phe, An, Fluo and Pyr in KTS samples ranged from 2628 to 666,508 ng g⁻¹ for workers, children and aggregate residents, with an average value of $115,902 \pm 197,229 \text{ ng g}^{-1}$.

Acknowledgements

This work was performed with the approval of the National Institute of Development Administration (NIDA) and financial support from the National Research Council of Thailand (NRCT). The author acknowledges Assist. Prof. Dr. Charnwit Kositanont, Ms. Teeta Intasaen and Assist. Prof. Dr. Penjai Sompongchaiyakul from Chulalongkorn University for their contributions to field sampling and laboratory work.

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