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Occurence and Toxicity Assessment of Polycyclic Aromatic Hydrocarbons (PAHs) in Mangrove Soil of Dong Rui area, Vietnam

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***Abstract*- An evaluation of the polycyclic aromatic hydrocarbons (PAHs) contamination in the mangrove soil from Dong Rui area, Vietnam was carried out. Twelve representative soil samples were collected in July 2016. The analyzed results indicated the wide occurrence of contamination of PAHs in Dong Rui. In sampling sites along BaChe River, total concentrations of Σ16PAHs ranged from 522.1 to 1405.1 µg kg-1 dw (from DR1 to DR4, mean 881.7 ± 392.1 µg kg-1 dw), while those along Voi Lon River and Voi Be River ranged from 432.5 to 1873.4 µg kg-1 dw (from DR5 to DR12, mean 928.4 ± 523.2 µg kg-1 dw). The total toxic equivalence (TEQ) value of PAHs in the soil samples from Dong Rui range from 52.2 to 310.3 µg TEQ kg−1 dw. The values of ecological risk quotient (RQ) of BaP ranged from 1.98 to 13.71 whereas those of other RQ were less than 1. Due to the propensity of PAHs to accumulate in various compartments of environment, further evaluation of ecotoxicological should be undertaken as a high priority.**

Keywords: PAHs; mangrove soil; toxic equivalence; ecological risk quotient.

I. INTRODUCTION

Polycyclic Aromatic Hydrocarbons (PAHs) are contained in the group of persistent organics; since they have high toxicity and ability to bioaccumulate, they can result in pollution with long-term effects on the human body and the environment. The main sources of PAHs are from industrial waste (thermal power plant), transport (combustion of fuels such as gasoline and oil), and daily activities (combustion of coal and wood...). PAHs are also present in the components of petroleum and thus, can also penetrate into the environmental components in case of oil leaks or oil spills. A number of PAHs are capable of causing cancer and dominant gene mutation; in addition, they can exist with considerable concentration in the environment. According to the classification of the U.S Environmental Protection Agency, there are 16 typical PAH including: naphthalene (Nap), acenaphthylene (Acey), acenaphthene (Ace), fluorene (Flu), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), pyrene (Pyr), chrysene (Chr), benzo[a]anthracene (BaA), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno [1,2,3-cd]pyrene (IcdP), dibenzo[a,h]anthracene (DahA), and benzo [g,h,i]pyrene (BghiP). There are a number of studies about these 16 PAHs in Vietnam; most of them focus on the air and dust environment, PAHs spread in the air and water, then deposit in the sediment of urban rivers; as a result, they continue to cause long-term impact on the environment. However, there is a little information on PAHs residue on mangrove soil in Vietnam.

Dong Rui area, the large land mangroves in Tien Yen district, Quang Ninh province, has high ecosystem diversity. To our knowledge, no data are available for the contamination of PAHs in this area as well as the toxicological risk assessment of PAHs residues on human. The objectives of this research are to assess occurence of selected PAHs in mangrove soil of Dong Rui area, Vietnam and their potential health risk assessment to fill this gap.

II. EXPERIMENTAL

The sampling was carried out in July 2016, during the dry season. Twelve representative sites were selected to investigate. Four surficial soil samples (marked from DR1 to DR4) were collected along BaChe River which is situated in the right side of Dong Rui' mangrove land. Meanwhile, eight surficial soil sample (marked from DR5 to DR12) were collected along VoiLon River and VoiBe River which is situated in the left side of Dong Rui' mangrove land (Fig.1). On the site DR4, DR5, DR6, several core soil samples were taken at the depth from 0 – 5 cm, 5 – 10 cm, 10 – 15 cm, 15 – 20 cm. The soil samples were collected with a stainless steel grab. All the samplers were immediately transferred to the laboratory. The samples were freeze-dried and homogenized. All the equipments used for sample collection, transportation, and preparation, were free from PAHs contamination.

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The PAHs concentration in the samples were determined using the method 8270D provided by US EPA with sightly modification [1]. Firstly, 10 grams of soil sample added with 10 g of anhydrous Na2SO4 and extract with 150 ml mixture of dichloromethane and acetone (ratio 1:1 v) for 24h using a Soxhlet apparatus. Collect the extract, reduce volume to about 1 ml. Add 10 ml of cyclohexane and then reduce to about 2 ml. Clean the extract through a column containing 10 g silicagel and 2 g Na2SO4 on top. Wash column with 25 ml of pentane and discard. Elute the column with 25 ml mixture of dichloromethane:pentane (ratio 4:6 in volume). Concentrate the extract to final volume 1ml. The samples were analyzed by gas chromatography (GC, Agilent Technologies 7890A)/mass spectrometry (MS, Agilent Technologies 5975C). The average recoveries were 74.0-113.9%. The method detection limits were 1 µg kg−1 dw for all selected PAHs. Duplicates of soil samples were also performed and relative standard deviations were less than 10%. All concentrations were calculated with respect to the dry weight (dw) of soil samples. Mixture of PAHs standards have concentration of 10 ng/ml for each selected PAHs and were purchased from the Laboratories of Dr.Ehrenstorfer, Germany.

III. DISCUSSION RESULT

The PAHs concentrations in the surficial soil samples are shown in Table 1. In sampling sites along BaChe River, total concentrations of Σ16PAHs ranged from 522.1 to 1405.1 µg kg-1 dw (from DR1 to DR4, mean 881.7 ± 392.1 µg kg-1 dw), while those along VoiLon River and VoiBe River ranged from 432.5 to 1873.4 µg kg-1 dw (from DR5 to DR12, mean 928.4 ± 523.2 µg kg-1 dw). It is observed that the highest value corresponded with site near estuary (site DR5, 1873.4 µg kg-1 dw) where interference between three rivers BaChe, VoiLon và VoiBe. PAHs were detected in all samples at the remarkable levels. The obtain results point out the wide occurrence of PAHs in the DongRui area. The concentrations of eight carcinogenic PAHs (Σ8C-PAHs: BaA, Chr, BbF, BkF, BaP, Ind, BghiP and DahA) range from 290.5 to 1058.3 µg kg-1 (mean 527.7 µg kg-1). The Pearson correlation coefficient between concentration of Σ16PAHs and Σ8C-PAHs in soils of Dong Rui is 0.99.

In Vietnam, no environmental standards have been established for PAHs in mangrove soil. When compared with other regions, the levels of PAHs in surficial soil of Dong Rui are higher than residues found in coastal soil of the Russian Arctic (20 – 1380 µg kg-1 dw), surficial soils collected from reed wetland of Liaohe estuary, China (235 – 374 µg kg-1 dw) [2,3]. Meanwhile, the levels of PAHs in Dong Rui are lower than residue in soils of the coastal and estuarine areas of the northern Bohai and Yellow Seas, China (66 - 920 µg kg-1 dw) as well as in soil collected from the Northern coast of the Persian Gulf, Iran (42.76 - 5596.42 µg kg-1 dw) [4,5].

In the core soil samples (Fig.2), total concentrations of Σ16PAHs ranged from 881.9 to 2236.1 µg kg-1 dw. The highest concentration is found at the depth from 5 to 10 cm. This results indicated that residues of PAHs were not only found in surface soil, but also could penetrate deeper below the soil layers.

Significant PAHs residue raised question on the level of toxicity of PAHs in soil of Dong Rui. This requirement can be done by using toxic equivalence (TEQ) and ecological risk quotient (RQ). The TEQ value for each sample was obtained from the concentrations of PAHs using their toxic equivalency factors (TEFs) proposed by World Health Organization. TEQ of ∑8C-PAHs and ∑16PAHs range from 51.8 to 309.2 µg TEQ kg−1 dw (mean 141.6 ± 76.2 µg TEQ kg−1 dw) and from 52.2 to 310.3 µg TEQ kg−1 dw (mean 142.1 ± 76.4 µg TEQ kg−1 dw), respectively. Eight carcinogenic PAHs have higher toxic potential than other PAHs indicators. Regarding to calculated TEQ values above, toxic effect of PAHs in soil of Dong Rui mainly contributed by ∑8C-PAHs. The mean value of TEQ of ∑16PAHs in Dong Rui were higher than those in Liaohe estuary, China (0.1 µg TEQ kg−1 dw), but lower than those in northern Bohai and Yellow Seas, China (32.6 µg TEQ kg−1 dw) and Northern coast of the Persian Gulf, Iran [3,4,5].
RQ is calculated by ratio of analyzed concentration and allowable concentration of selected PAHs in soil. The used allowable concentrations of PAHs are recommended by USEPA [6]. The values of RQ for individual PAHs are showed in table 2. Incase of BaP, the values of RQ ranged from 1.98 to 13.71 whereas almost those of other RQ were less than 1. Thus, individual PAHs might have little adverse effects on ecosystem. Due to the propensity of PAHs to accumulate in various compartments of environment, further evaluation of ecotoxicological should be undertaken as a high priority.

IV. CONCLUSION

This work investigated the contamination status of PAHs in mangrove soil from Dong Rui area, Vietnam. Wide occurrence and high residue levels of PAHs has been found in the study area (from 432.5 to 1873.4 µg kg−1 dw). Compositions analyses show that PAHs with 4–6 ring compounds (74.5% of the ∑16PAHs) were most dominant compounds in analyzed soil. The total TEQ values of ∑16PAHs in the soil samples from Dong Rui range from 52.2 to 310.3 µg TEQ kg−1 dw. The calculation of RQ of individual PAHs show that PAHs might have little adverse effects on ecosystem. The main source of PAHs in Dong Rui are attributable to both petroleum conbustion and biomass burning.

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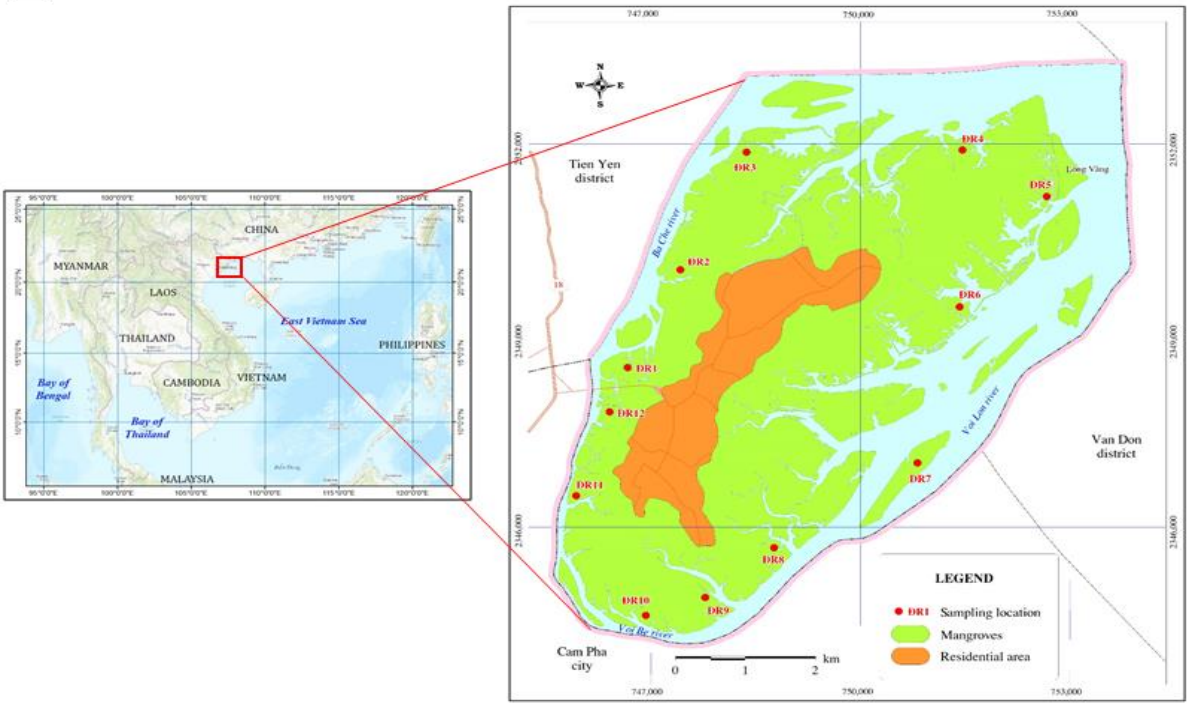

Fig. 1. Study area and sampling locations

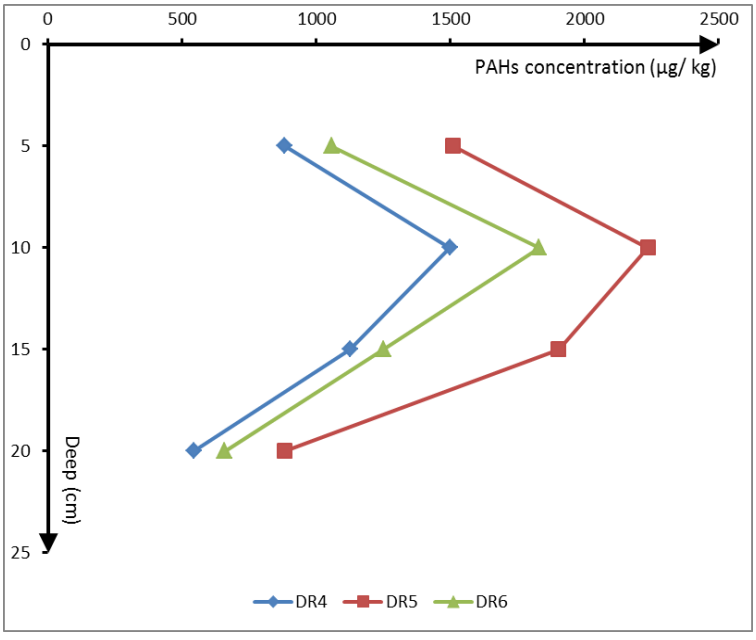

Fig. 2. Depth profile of PAHs concentration in soils of DongRui area.

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Table 1. TOC (%) and concentrations of PAHs (µg kg^{-1} dw) in the soil samples

Compound	TEF	Soil sample	
		Sampling sites along BaChe River	Sampling sites along VoiLon, VoiBe River
Nap	0.001	9 - 72.5 (29.45 ± 29.30) [c]	8.2 - 64.6 (27.87 ± 22.07)
Acy	0.001	12.4 - 101.6 (47.55 ± 38.13)	10.7 - 128.9 (53.85 ± 41.85)
Ace	0.001	18.5 - 53.4 (35.15 ± 17.72)	11.4 - 57.5 (38.46 ± 18.48)
Flu	0.001	6.9 - 37.9 (22.4 ± 13.92)	5.9 - 118.3 (30.97 ± 36.99)
Phe	0.001	30.4 - 103.5 (68.22 ± 29.9)	15.8 - 145.6 (75.14 ± 48.94)
Ant	0.01	7.3 - 21.9 (13.82 ± 6.33)	4.8 - 35.1 (16.48 ± 10.54)
Py	0.001	26.8 - 135.5 (67.85 ± 47.95)	19.6 - 125.6 (62.66 ± 37.01)
Flt	0.001	21.3 - 210.7 (95.5 ± 81.41)	21.7 - 171.5 (82.32 ± 59.98)
BaA	0.1	21.5 - 75.7 (53.8 ± 26.55)	12.3 - 165.4 (91.77 ± 56.68)
Chr	0.01	25.7 - 73.6 (56.37 ± 21.26)	24.8 - 111.3 (61.82 ± 31.58)
BbF	0.1	22.7 - 150.7 (79.7 ± 56.65)	23.4 - 192.7 (77.15 ± 60.47)
BkF	0.1	17.2 - 63.8 (36.5 ± 19.74)	13.7 - 86.4 (39.05 ± 24.21)
BaP	1	42.3 - 162.6 (91.9 ± 54.58)	31.7 - 219.5 (102.32 ± 60.45)
Ind	0.1	80.8 - 109.7 (97.475 ± 14.14)	47.3 - 163.7 (90.75 ± 47.57)
BghiP	0.01	31.5 - 145.7 (75.475 ± 49.27)	20.7 - 112.5 (64.07 ± 34.34)
DahA	1	5.8 - 14.6 (10.525 ± 3.62)	6.3 - 26.8 (13.67 ± 6.67)
Σ_8C-PAHs [a]		293.7 - 712 (501.75 ± 178.53)	290.5 - 1058.3 (540.62 ± 280.13)
Σ_{16}PAHs [b]		522.1 - 1405.1 (881.7 ± 392.06)	432.5 - 1873.4 (928.38 ± 523.23)

a: sum of 8 carcinogenic PAHs compounds (BaA. Chr. BbF. BkF. BaP. Ind. BghiP. DahA); b: sum of all 16 selected PAHs; c: Min - Max (mean ± standard deviation).

Table 2. The values of RQ for individual PAHs in soil samples in Dong Rui area

Compound	Allowable concentrations of PAHs	RQ		
		Min	Max	Mean
Nap	3.8	$2.16x10^{-3}$	$1.9x10^{-2}$	$7.47x10^{-3}$
Acy	- [a]	-	-	-
Ace	360	$3.17x10^{-5}$	$1.6x10^{-4}$	$1.04 x10^{-4}$
Flu	240	$2.46x10^{-5}$	$4.93x10^{-4}$	$1.17 x10^{-4}$
Phe	-	-	-	-
Ant	1800	$2.67x10^{-6}$	$1.95x10^{-5}$	$8.66x10^{-6}$
Py	180	$1.09x10^{-4}$	$7.53x10^{-4}$	$3.58x10^{-4}$
Flt	240	$8.88x10^{-5}$	$8.78x10^{-4}$	$3.61x10^{-4}$
BaA	0.16	$7,68x10^{-2}$	1.03	0.49
Chr	16	$1.55x10^{-3}$	$6.95 x10^{-3}$	$3.75 x10^{-3}$
BbF	0.16	0.14	1.2	0.49
BkF	1.6	$8,56x10^{-3}$	0.05	$2.38x10^{-2}$
BaP	0.016	1.98	13.71	6.17
Ind	0.16	0.29	1.02	0.58
BghiP	-	-	-	-
DahA	0.016	0.36	1.68	0.79

a: no data