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Residue and Temporal Trend of Polychlorinated Biphenyls (PCBs) in Surface Soils from Bacninh, Vietnam

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***Abstract*—**An evaluation of the PCBs residues in the surface soils from Bacninh, Vietnam was carried out. Sixty representative soil samples were collected from the centre of Bacninh and three surrounding districts. The analyzed results indicated the wide extent of contamination of total PCBs in Bacninh. In industrial and urban zones, total PCBs concentrations ranged from ranged from <0.02 to 32.68ng g^{-1} (mean 19.89 ±15.64ng g^{-1}) dry weight, while those in agricultural zones ranged from <0.02 to 13.26ng g^{-1} (mean 8.14 ± 4.89ng g^{-1}) dry weight. The mean percentages of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180 compared with Σ6PCBs in the analyzed soil samples are 3.1%, 13.9%, 21.7%, 30.7%, 25.8% and 4.8%, respectively. These values can be explained by the chemical properties as well as the compositions of PCBs mixture which probably escaped from dielectric oil. An increasing trend and the long-time release of PCBs are observed.

***Keywords*—**Contamination, PCBs, soil, temporal trend.

I. Introduction

POLYCHLORINATED Biphenyls (PCBs) are industrial products which constitute a global environmental health hazard of a solely anthropogenic origin. Theoretically, there are 209 PCB isomers and congeners with one to ten chlorine atoms attached to the biphenyl molecule. They are very resistant to decomposition and have an excellent insulating property as well as a high heat capacity. Their properties have led to many industrial applications but also make PCBs a major environmental pollutant. Studies in humans provide supporting evidence for potential carcinogenic and non carcinogenic effects of PCBs (neurological, immune, endocrine and reproductive effects). The most serious cases of PCB effects on human health were the accidental leakages of PCBs containing industrial fluids into rice oils that resulted in the exposure of several thousand individuals in two separate incidents, one in northern Kyushu Island in Japan in 1986 (Yusho) and the other in Tai Chung in central Taiwan in 1979 (Yu-Cheng).

PCBs have never been manufactured in Vietnam. PCBs were imported into Vietnam as industrial fluids such as hydraulic and heat transfer fluids, in gas turbines; as lubricating oils; and as plasticizers. There is no official information on the first import of PCBs into Vietnam but it was probably sometime around 1950. They have been mainly used as dielectric oil in transformers and in capacitors. Bacninh province, located in the Red River delta, is a typical centre of industrial, agricultural and traditional village in Viet Nam. Bacninh is composed of the city and 3 three surrounding districts including TuSon, TienDu and YenPhong with many factories and urban zones. One recent research has been conducted in order to determine the concentrations of PCBs in street dust from Bacninh [7]. According to that research, PCBs were detected in all dust samples and thus, clearly indicated their long-term usage in this province. However, to our knowledge, little data is available concerning PCB contamination in the surface soils of Bacninh. The present study aims to fill this gap by assessing the contamination of PCBs in the surface soil of Bacninh.

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II. Materials and Methods

The sampling was carried out in February 2012, during the dry season. The sampling locations were chosen at random with an attempt to get them evenly distributed over selected region of Bacninh (approximately 800km^2, Fig. 1). Sixty samples from the upper 5cm of the soil were collected from industrial, agricultural, and urban zones in Bacninh City, as well as from three surrounding districts (TuSon, TienDu and YenPhong).

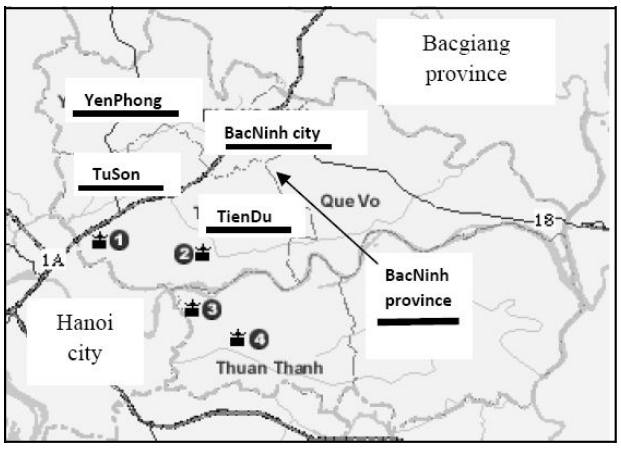

Fig. 1 Map of the study area

Each sample was a mixture of five sub-samples (four in the corners of a rectangle and one in the crossing point of two diagonals). The samples were taken with solvent-rinsed

stainless steel scoops from the upper 5cm of the soil and then transferred to pre-cleaned polyethylene bags.

In short, about 20 grams of soil sample was wetted with distilled water and then extracted 3 times in a separator funnel, each time with 100ml acetone for 30 minutes. 100ml distilled water were added to the acetone extract and the resulting solution was extracted again 3 times, each time with 100ml *n*-hexane for 30 minutes. After that, the *n*-hexane extract was combined, dried by passing through anhydrous sodium sulphate and concentrated to around 5ml. The concentrated extract was filtered through 8 grams of activated Florisil packed in a glass column, which was then eluted by 45ml of *n*-hexane. The resulting solution was de-sulphurated using activated copper chips and finally treated with concentrated sulphuric acid. Final extracts were concentrated to 5ml using a rotary vacuum evaporator and then to around 300µl under a gentle stream of purified nitrogen. All chemicals used were of analytical grade purchased from Fluka Chemical Co., Switzerland. The samples were analyzed with the gas chromatograph (Varian Star 3400Cx) equipped with SPB-5 column (30m length x 0.32 i.d. mm x 0.25µm film thickness, Supelco, Darmstadt, Germany), mass spectrometer (Varian Saturn 2000) and autosampler (Varian 8200Cx). The mixture of Aroclor (Aroclor1016, Aroclor1232, Aroclor1248 and Aroclor 1260) and the mixture of six selected PCB congeners were used as external standards to determine the concentration of total PCB and Σ6PCB, respectively. These mixtures were purchased from Fluka Chemical Co. (Switzerland) and Supelco Chemical Co. (Germany). PCB recoveries were examined by using fortified soil samples from Bacninh which had been prepared according to the method of Aydin et al. [2], [5]. The fortified soil samples were spiked with 40ng g^{-1} dry weight of each of the six selected PCB. The recoveries ranged from 90 - 96%. The method detection limits were calculated from real soil samples as being 3 times the signal/noise ratio and were 0.02ng g^{-1} dry weight for each PCB. One blank sample was run for every set of five soil samples to check for secondary contamination. The PCB concentrations were not corrected for recoveries. Duplicates of soil samples were performed and relative standard deviations were less than 15%. All concentrations were calculated with respect to dry weight.

III. Results and Discussion

The PCBs concentrations in the collected soil samples from Bacninh province are all shown in Tables I and II. The total PCBs concentrations in industrial and urban areas ranged from <0.02 to 32.68ng g^{-1} (mean 19.89 ±15.64ng g^{-1}). It is observed that the highest value corresponded with site in the centre of Bacninh city (32.68ng g^{-1}), which is located in a densely populated Bacninh downtown area. Close to this site, there are several small old transformers and the area is also influenced by heavy traffic. The other significant levels of total PCBs were found at sampling sites close to industrial parks in the TuSon and TienDu district which have concentrations are 29.12ng g^{-1} and 32.47ng g^{-1}, respectively (Table I).

TABLE I
PCBs Concentrations (ng/g Dry Weight) in the Surface Soil of Industral and Urban Zones from Bacninh

Location	Number of soil samples	Industrial and Urban zones	
		Σ6PCBs [a]	*Total PCBs* [b]
TuSon district	10	<0.02 - 3.34 (2.19) [c]	<0.02 - 29.12 (19.07)
TienDu district	10	2.07 - 4.38 (3.19)	15.38 - 32.47 (23.74)
Bacninh City	10	<0.02 - 4.08 (2.73)	<0.02 - 32.68 (21.87)
YenPhong district	10	<0.02 - 2.67 (1.34)	<0.02 - 18.44 (9.22)

(a) Σ6PCBs – sum of six selected PCB congeners; (b) Total PCB – sum of all PCB isomers and congeners in soil sample; (c) min – max (mean) value

TABLE II
PCBs Concentrations (ng/g Dry Weight) in the Surface Soil of Agricultural Zones from Bacninh

Location	Number of soil samples	Agricultural zones	
		Σ6PCBs [a]	*Total PCBs* [b]
TuSon district	5	0.93 – 1.53 (0.92)	<0.02 – 13.26 (8.19)
TienDu district	5	<0.02 - 1.45 (1.08)	<0.02 - 10.82 (8.17)
Bacninh City	5	<0.02 - 1.62 (0.99)	<0.02 - 12.53 (7.52)
YenPhong district	5	<0.02 - 1.47 (0.66)	<0.02 - 10.57 (4.32)

(a) Σ6PCBs – sum of six selected PCB congeners; (b) Total PCB – sum of all PCB isomers and congeners in soil sample; (c) min – max (mean) value

Due to the historical use of PCBs in Vietnam, its main source of contamination in industrial and urban areas could originate from the dielectric oil used in old hanging transformers and capacitors which were widely used in Bacninh province. From these installations, PCBs could have penetrated into the environment by mechanical damage, electrical accidents and fire. During the retro-filling of dielectric oil containing PCBs, there is a risk of PCBs escaping into the environment.

At present, there is no Vietnamese standard on the maximum allowable concentration of total PCBs in surface soil, nor is there official quantitative information on the cumulative use of PCBs in Bacninh. The management of old hanging transformers and capacitors in the Bacninh province belong to Electricity of Vietnam and the surveys of possible PCBs concentrations from those equipments is only allowed with official environmental agency. Because these results are not published, it is impossible to calculate PCBs quantity used in Bacninh. It has been reported that the total amount of dielectric oil contaminated with PCBs in the entire country is approximately 19000 tonnes, mainly from old transformers [6]. This clearly indicates a huge contaminative source of PCBs. According to the survey carried out by Electricity of Vietnam [3], insulating oils in the transformers installed on the electric grid in Northern provinces of Vietnam were selected for analysis with regards to PCBs and some types of those oils are supplied with non PCBs labels but their PCBs levels still exceed standards. Therefore, it is not sure that insulating oils which were imported into Vietnam are free from PCBs in any case. This could account for PCBs contamination in the environment in Vietnam. The other possible source of PCBs in

Bacninh could be from traffic-related activities. PCBs could be used as one component of lubricating oils of motor vehicles [9]. With regard to the soil samples from agriculture areas, total PCBs concentrations ranged from <0.02 to 13.26ng g^{-1} (mean: 8.14 ± 4.89ng g^{-1}). These sites are not far from densely populated villages as well as the urban zones of 3 surrounding districts. Therefore, total PCBs were probably deposited into the agriculture sites by atmospheric transport from industrial and urban areas. In general, the total PCBs concentrations were highest in industrial soil samples, followed by those in urban soils and in agricultural soil. This also applies for the usage of PCBs in Vietnam.

When compared with some regions in the world, the concentrations of Σ6PCBs in soil samples from Bacninh are comparable to those in soil samples of the Moscow region (2.0 – 34ng g^{-1}), but higher than in the mineral topsoil of mainly rural areas in the United Kingdom and in Switzerland (3.5 – 17ng g^{-1}) and in remote areas in Germany (0.9 - 4.8ng g^{-1}) [1]-[10]. The comparison of total PCBs concentration with those described in other studies was not possible due to the difference in the PCB standards used for quantification.

Concerning the composition analyses, PCBs congeners could be detected from tri-CB to octa-CB in the collected soil samples. The mean percentages of 6 selected PCB congeners in the analyzed soil samples from Bacninh and Bacninh followed the order PCB138 > PCB153 > PCB101 > PCB52 > PCB180 > PCB28 (Fig. 2). This order can be explained by the fact that lightly chlorinated PCBs are less persistent, have lower log K_{ow} and are more volatile than heavily chlorinated PCBs. Therefore, heavily chlorinated PCBs are more accumulative in the soil, whereas lightly chlorinated PCBs are degraded and volatilized faster than the other PCBs. Another explanation could be related to the composition of PCBs mixtures which probably escaped from dielectric oil. According to Electricity of Vietnam, up to April 1998, 48.3% of the total quantity of dielectric oils was imported from the Soviet Union.

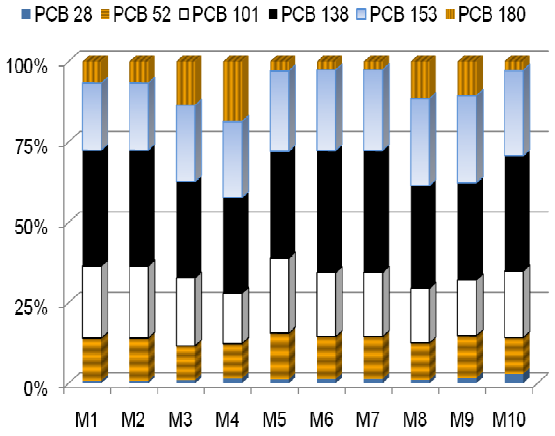

Fig. 2 Mean percentages of PCB isomers in soil samples

Japan and China contributed with much smaller percentages which are 7.5% and 3.6%, respectively. It has been reported that there are 2 typical models of Chinese commercial PCBs and their principal technical formulations are similar to Aroclor 1242 and Aroclor 1254 [4]. Regarding the composition of PCBs congeners in imported dielectric oil, the percentages of PCB138, PCB153, PCB101 in Sovol and Aroclor 1254 are significantly higher than the others, while the percentages of PCB28 and PCB52 are predominant in Aroclor1242, KC300 and KC400. Here, the mean percentages of PCB28, PCB52, PCB101, PCB138, PCB153 and PCB180 compared with Σ6PCBs in the analyzed soil samples are 3.1%, 13.9%, 21.7%, 30.7%, 25.8% and 4.8%, respectively. It is also evident that the predominance of heavily chlorinated PCBs, PCB138 and PCB153, are still remained when they penetrated the soil from Bacninh. In general, low percentages of lightly chlorinated PCBs and a high percentage of heavily chlorinated PCBs in the analysed soil samples reflect their long-time release. The statistic coefficient between concentrations of Σ6PCBs and total PCBs in this study are 5.31 (n=60 soil samples; R^2=0.95). It is the preliminary result and could be used for assessing the relation between the level of 6 PCBs indicators and total PCBs in the soil environment of Bacninh.

With regards to ΣPCB concentrations in soil samples from Bacninh reported in the other studies, the temporal trend of PCB levels could be shown. It was reported that the mean ΣPCB concentration in soil samples from Bacninh in 1993, in 2000 and in 2012 (60 soil samples) range from <0.02 to 22.14 ng g^{-1}, from <0.02 to 26.43ng g^{-1} and from <0.02 to 32.68ng g^{-1}, respectively ([7], [8], this study). An increasing trend for ΣPCB concentration can clearly be seen, which is further confirmed by high residues of PCBs detected in 2012. There is a high possibility that this trend may continue in the future since Vietnam has not banned the use of PCBs yet and a huge amount of dielectric oil containing PCB still exists throughout the country. From this source, PCBs can be further released into the environment.

ACKNOWLEDGMENT

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