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Level and Distribution of Polychlorinated Biphenyls (PCBs) in Surface Soils from Hanoi, Vietnam

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Ploychlorinated biphenyls (PCBs) are industrial products that 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 noncarcinogenic 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).

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PCBs have never been manufactured in Vietnam. PCBs are 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 (Buxton 2001). They have been mainly used as dielectric oil in transformers and capacitors. Hanoi City, which is located in the Red River Delta in the North of Vietnam, is the center of culture, politics, economy and trade for the whole country. Hanoi is composed of the center and 5 suburban districts including SocSon, DongAnh, GiaLam, TuLiem and ThanhTri with many factories and agricultural areas. Some recent research has been conducted in order to determine the concentrations of PCBs in sediment, molluscs and human breast milk from Hanoi (Nhan et al. 2001; Minh et al. 2004). In these studies, PCBs were detected in all the environmental compartments listed above, which clearly indicated their long-term usage in this city. However, to our knowledge, little data is available concerning PCB contamination in the surface soils of Hanoi. The present study aims to fill this gap by assessing the level and distribution of PCBs in the surface soil of Hanoi.

Materials and Methods

The sampling was carried out in February 2006, during the dry season. Sixty soil samples were collected from both agricultural and industrial areas, from towns in all five suburban districts, as well as from the centre of Hanoi. The codes for the soil samples were marked from C1 to C60. The sampling locations were chosen at random yet were evenly distributed over Hanoi City (approximately 921 km^2, about 3 million people, Fig. 1).

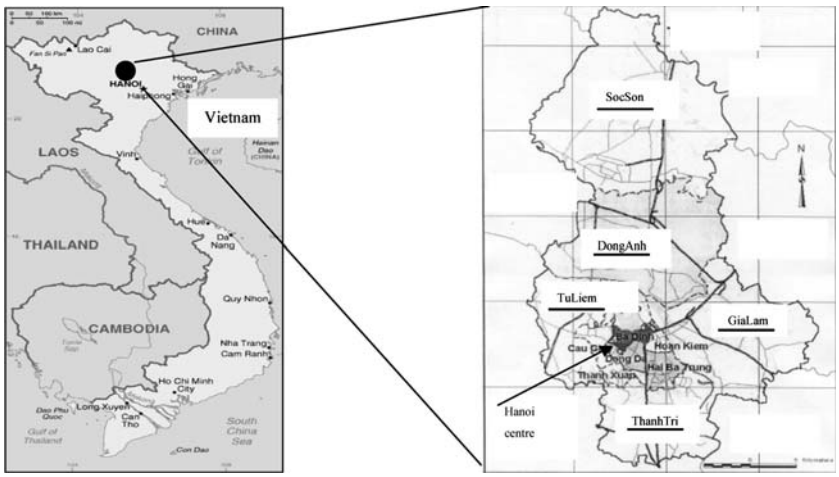

Fig. 1 Map of the study area

At each sample location, five samples were collected from a 100 m^2 square plot (located on the crossing diagonals: four in the corners and one in the crossing point), and then thoroughly mixed to form a composite sample. The samples were taken with solvent-rinsed stainless steel scoops from the upper 5 cm of the soil and then transferred to pre-cleaned polyethylene bags. The collected samples were homogenized, air dried at room temperature (22–25°C), ground and sieved through a steel mesh (1-mm grid size). All samples were maintained at 4°C until analysis.

The total concentration of PCBs (ΣPCB) and six selected PCB congeners (IUPAC number: PCB28, 52, 101, 138, 153, 180) were analyzed following the method described by Thao et al. (1993a). It has been reported that these PCB congeners were predominantly present in most commercial PCB mixtures and in environmental samples (Kim et al. 2004). Therefore, the collected soil samples have been analyzed to determine their composition with regard to PCB congeners. 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 100 ml acetone for 30 minutes. 100 ml distilled water was added to the acetone extract and the resulting solution was extracted again 3 times, each time with 100 ml *n*-hexane for 30 minutes. After that, the *n*-hexane extract was combined, dried by passing through anhydrous sodium sulphate and concentrated to around 5 ml. The concentrated extract was filtered through 8 grams activated Florisil packed in a glass column, which was then eluted by 45 ml *n*-hexane. The resulting solution was de-sulfurated using activated copper chips and finally treated with concentrated sulfuric acid. Final extracts were concentrated to 5 ml using a rotary vacuum evaporator and then to around 300 µl under a gentle stream of purified nitrogen. All chemicals used were of analytical grade and purchased from Fluka Chemical, Switzerland. The samples were analyzed with the gas chromatograph (Varian Star 3400Cx) equipped with SPB-5 column (30 m length x 0.32 i.d. mm x 0.25 µm 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 ΣPCB and Σ6PCB, respectively. These mixtures were purchased from Fluka Chemical (Switzerland) and Supelco Chemical (Germany). PCB recoveries were examined by using fortified soil samples from Hanoi that had been prepared according to the method of Aydin et al. (2006). The fortified soil samples were spiked with 40 ng g^{-1} dry weight of each of the six selected PCBs. The recoveries ranged from 81–96%. The method detection limits were calculated from real soil samples as being 3 times the signal/noise ratio and were 0.02 ng 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.

Results and Discussion

The PCB concentrations in the analyzed soil samples are all shown in Table 1. The ΣPCB concentrations in industrial and urban areas ranged from < 0.02 to 190.42 ng g^{-1} (mean 41.89 ± 35.64 ng g^{-1}). It was observed that the highest value corresponded with site C48 (190.42 ng g^{-1}), which is located in a densely populated Hanoi downtown area. Close to this site, there are several small old transformers and the area was also influenced by heavy traffic. The other significant PCB concentrations were found at sampling sites close to industrial parks in GiaLam district, TuLiem district and in individual factories in the centre of Hanoi such as C41 (79.44 ng g^{-1}), C42 (78.72 ng g^{-1}), C57

Table 1 PCB concentrations (ng g^{-1} dry weight) in the surface soil from Hanoi

Location	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	Σ6PCB [a]	ΣPCB[b]
A. Agricultural areas								
SocSon 1[c]								
C1	0.07	0.35	0.64	1.14	0.88	0.19	3.27	21.82
C2	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C3	0.07	0.32	0.59	1.06	0.82	0.18	3.04	20.28
C4	0.06	0.32	0.59	1.06	0.82	0.18	3.03	20.22
C5	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C6	0.07	0.29	0.53	0.96	0.74	0.16	2.75	18.36
C7	0.07	0.27	0.49	0.87	0.67	0.15	2.52	16.76
C8	0.06	0.26	0.47	0.85	0.66	0.14	2.44	16.32
DongAnh 1								
C9	0.06	0.28	0.52	1.36	1.12	0.28	3.62	21.18
C10	0.07	0.28	0.52	1.37	1.12	0.26	3.62	21.32
C11	0.06	0.32	0.53	1.44	1.18	0.29	3.82	22.44
C12	0.06	0.25	0.43	1.19	0.97	0.23	3.13	18.47
C13	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C14	0.06	0.26	0.46	1.26	1.03	0.26	3.33	19.63
C15	0.06	0.33	0.58	1.57	1.28	0.32	4.14	24.37
GiaLam 1								
C16	0.07	0.26	0.46	1.25	1.02	0.26	3.32	18.39
C17	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C18	0.12	0.25	0.43	1.19	0.95	0.19	3.13	17.43
Hanoi centre 1								
C19	0.08	0.18	0.32	0.85	0.72	0.12	2.27	11.38
C20	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C21	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
TuLiem 1								
C22	0.09	0.17	0.35	1.28	0.98	0.12	2.99	18.72
C23	0.06	0.15	0.32	1.13	0.85	0.12	2.63	16.48
C24	0.08	0.17	0.35	1.26	0.94	0.14	2.94	18.43
C25	0.06	0.15	0.32	1.08	0.85	0.12	2.58	16.14
C26	0.07	0.16	0.32	1.14	0.85	0.13	2.67	16.72
C27	0.09	0.18	0.37	1.35	1.03	0.12	3.14	19.65
C28	0.08	0.17	0.35	1.28	0.98	0.13	2.99	18.73
ThanhTri 1								
C29	0.07	0.38	0.54	1.63	1.02	0.25	3.89	21.65
C30	0.06	0.29	0.43	1.26	0.82	0.23	3.09	17.18
C31	0.07	0.26	0.42	1.32	0.82	0.23	3.12	17.23
B. Industrial and urban areas								
SocSon 2[d]								
C32	0.06	0.39	0.62	1.82	1.12	0.32	4.32	27.83
C33	0.06	0.43	0.62	1.79	1.12	0.32	4.33	27.94
C34	0.06	0.25	0.35	1.03	0.65	0.18	2.52	16.24
DongAnh 2								
C35	0.12	0.16	0.64	2.47	1.38	0.18	4.95	28.32
C36	0.13	0.34	0.85	3.28	1.83	0.13	6.56	37.54
C37	0.06	0.19	0.44	1.72	0.95	0.06	3.42	19.48
C38	0.12	0.35	0.82	3.18	1.76	0.14	6.37	36.45
C39	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02

Table 1 continued

Location	PCB 28	PCB 52	PCB 101	PCB 138	PCB 153	PCB 180	Σ6PCB [a]	ΣPCB[b]
GiaLam 2								
C40	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02	<0.02
C41	0.22	0.86	2.02	7.74	4.33	0.32	15.49	79.44
C42	0.23	0.85	1.99	7.67	4.29	0.32	15.35	78.72
C43	0.08	0.25	0.59	2.32	1.29	0.09	4.62	23.65
C44	0.08	0.24	0.57	2.22	1.23	0.08	4.42	22.64
C45	0.15	0.64	1.48	5.72	3.22	0.22	11.43	58.63
C46	0.17	0.64	1.49	5.73	3.22	0.22	11.47	58.84
Hanoi centre 2								
C47	0.06	0.42	0.68	0.97	0.84	0.15	3.12	14.82
C48	0.43	5.19	8.79	12.79	10.79	1.99	39.98	190.42
C49	0.12	1.52	2.62	3.78	3.19	0.59	11.82	56.29
C50	0.14	1.67	2.83	4.12	3.47	0.64	12.87	61.32
C51	0.06	0.45	0.77	1.13	0.95	0.17	3.53	16.82
C52	0.13	1.43	2.43	3.54	2.98	0.55	11.06	52.69
C53	0.12	1.22	2.06	3.02	2.53	0.47	9.42	44.78
TuLiem 2								
C54	0.07	0.34	0.62	1.65	1.35	0.32	4.35	23.56
C55	0.07	0.36	0.63	1.73	1.42	0.34	4.55	24.64
C56	0.37	0.93	1.62	3.53	4.33	0.87	11.65	61.63
C57	0.28	0.94	1.66	3.62	4.46	0.88	11.84	63.26
ThanhTri 2								
C58	0.12	0.45	0.79	2.17	1.77	0.42	5.72	34.62
C59	0.12	0.47	0.82	2.22	1.82	0.44	5.89	35.74
C60	0.08	0.24	0.43	1.18	0.96	0.23	3.12	18.92

[a] Σ6PCB, sum of 6 selected PCB congeners

[b] ΣPCB, sum of all PCB isomers and congeners

[c] SocSon 1, agricultural areas of SocSon

[d] SocSon 2, industrial and urban areas of SocSon

(63.26 ng g^{-1}) and C56 (61.63 ng g^{-1}), respectively. At present, there is no Vietnamese standard on the maximal allowable concentration of ΣPCB in surface soil, nor is there official quantitative information on the cumulative use of PCBs in Hanoi.

Due to the historical use of PCBs in Vietnam, the 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 Hanoi. From such equipment, PCBs could penetrate into the environment by mechanical damage, electrical accident and fire. During the re-filling of dielectric oil containing PCBs, there is a risk of PCBs escaping into the environment. It has been reported that the total amount of dielectric oil containing PCBs in the entire country is approximately 19 thousands tonnes, mainly from old transformers (NEA 2006). This clearly indicates a huge contaminative source of PCBs. At present, the import and use of dielectric oil containing PCBs in Vietnam is restricted, but it has not yet been banned yet (NEA 2006). According to the survey carried out by Electricity of Vietnam (EVN) in 1999, insulating oils in the transformers installed on the electric grid in northern provinces of Vietnam were selected for analyses with regard to PCBs, and some types of those oils are supplied with non-PCB labels but their PCB levels still exceed standards (EVN 2003). Therefore, it is not sure that insulating oils that are imported into Vietnam are free from PCBs in any case. This could account for the PCB contamination in the environment in Vietnam. The other possible source of PCBs in Hanoi could be from traffic-related activities. PCBs could be used as one component of lubricating oils of motor vehicles (Thao 1993b). With regard to the soil samples from agriculture areas, ΣPCB concentrations ranged from < 0.02 to 24.37 ng g^{-1} (mean 15.14 ± 7.89 ng g^{-1}). These sites are not far from densely populated villages as well as the towns of five surrounding suburban districts. Therefore, PCBs

were probably deposited into agricultural sites by atmospheric transport from industrial and urban areas. In general, the PCB concentrations were highest in industrial soil samples, followed by those in urban soil and agricultural soil. This also applies for the usage of PCBs in Vietnam. From the analyzed results in Table 1, it is evident that the distribution of low level contamination of ΣPCB occurs in agricultural areas, whereas the distribution of relatively high level contamination of ΣPCB occurs in industrial and urban areas.

When compared with some regions in the world, the concentrations of Σ6PCB in soil samples from Hanoi (< 0.02–39.98 ng g^{-1}, Table 1) are comparable to those in soil samples of the Moscow region (2.0–34 ng g^{-1}), but higher than in the mineral topsoil of mainly rural areas of the United Kingdom and Switzerland (3.5–17 ng g^{-1}) and in remote areas of Germany (0.9–4.8 ng g^{-1}) (Alcock et al. 1993; Wilcke et al. 2006). The comparison of ΣPCB concentrations with those described in other studies was not possible due to the difference in the PCB standards used for quantification.

Besides the contamination of soil, PCBs also found ways to penetrate into the human body and into other environmental compartments in Hanoi. According to Nhan et al. (2001), the PCB concentrations measured as Aroclor 1254 in sediments and molluscs from canals in the downtown areas and in the suburbs of Hanoi City in August 1997 were as high as 40 ng g^{-1} and 76 ng g^{-1}, respectively. In addition, the research by Minh et al. (2004) found that the mean ΣPCB concentrations in 42 human breast milk samples in Hanoi were 74 ng g^{-1} lipid weight. Those results, together with this research, highlight the wide extent of contamination of PCBs in Hanoi.

Concerning the PCB congeners, tri-chlorinated biphenyls (CB) to octa-CB were detected in the collected soil samples. The mean percentages of six selected PCB congeners in the analyzed soil samples from Hanoi followed the order PCB138 > PCB153 > PCB101 > PCB52 > PCB180 > PCB28. 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. Another explanation could be related to the compositions of PCB 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. Japan and China contributed with smaller percentages of 7.5% and 3.6%, respectively (EVN 2003). It has been determined that the percentages of PCB138, PCB153, PCB101, PCB52, PCB180, PCB28 in Sovol (trade name of Soviet Union dielectric oil) were 11.4, 7.0, 6.5, 3.6, 0.4 and 0.8%, respectively (Falandysz et al., 2006). Here, the mean percentages of each selected PCB congener compared with Σ6PCB in the analyzed soil samples are PCB138, 39.8%; PCB153, 29.3%; PCB101, 15.2%; PCB52, 8.3%; PCB180, 5.5%; PCB28, 1.9%. It is evident that the predominance of heavily chlorinated PCBs, PCB138 and PCB153, still remained when they penetrated the soil from Hanoi. In general, low percentages of lightly chlorinated PCBs and a high percentage of heavily chlorinated PCBs in the analyzed soil samples reflect their long-time release.

With regards to ΣPCB concentrations in soil samples from Hanoi 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 Hanoi in 1992 (6 soil samples), in 2000 (8 soil samples) and in 2006 (60 soil samples) range from 9.1 to 29 ng g^{-1} (mean 12.6 ± 8.9 ng g^{-1}), from 0.6 to 120 ng g^{-1} (mean 21.2 ± 25.2 ng g^{-1}) and from < 0.02 to 190.24 ng g^{-1} (mean 28.08 ± 28.57 ng g^{-1}), respectively (Thao 1993b; Viet et al. 2000; this study). An increasing trend for ΣPCB concentration can clearly be seen, which is further confirmed by high residues of PCBs detected in 2006. 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.

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