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Polychlorinated Biphenyl Congeners in the Aquatic Environment of the Mekong River, South of Vietnam

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Abstract Polychlorinated biphenyl compounds (PCBs) were analyzed in sediments and clams' soft tissues from sampling sites in the Mekong River delta from the border with Cambodia to the coast of South China Sea. Concentrations of 13 individual PCB congeners are reported. Median concentration of ΣPCB congeners was 0.279 ng g^{-1} dry weight (range 0.106–2.016 ng g^{-1} dry weight) in sediments, and 5.20 ng g^{-1} dry weight (range 1.89–19.37 ng g^{-1}) in clams. Distribution and bioaccumulation of PCBs in the delta are discussed. It is concluded that in the Mekong River delta PCB concentrations were generally lower than in other regions of Vietnam and their likely sources have been waste discharges from repair workshops and other facilities in the delta cities.

Keywords PCB congeners · PCBs in sediments · PCBs in bivalve molluscs · Bioaccumulation

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The Mekong River, one amongst the ten largest rivers of Asia, flows through six countries and directly influences the living of more than 60 million human beings. The economic and social importance of this river has brought together four riparian countries to coordinate efforts through the International Mekong Commission in order to improve the management of common interest issues, such as water management, water quality, fisheries, irrigation, and rice production (MRC 2008).

In the South of Vietnam, the Mekong River delta represents a vast area estimated at about 39,000 km^2 that accounts for 12% of the Vietnam territory (325,000 km^2) and for 5% of the Mekong watershed (795,000 km^2). The delta encompasses highly productive lowlands of alluvial origin, producing mainly rice, vegetables, and fruits. Heavy industries are scarce in this region and most of the 16.5 million inhabitants of the delta population work in agriculture activities. Several major cities in the delta, such as Can Tho, Vinh Long, Soc Trang, and My Tho, have electric power plants, mechanical repair workshops, shipyards, and food industries. Agrochemicals and industrial organic chemicals are thus used in the delta region and chemical residues once released readily end up in the aquatic environment. Recently, results of a survey on agrochemical residues, including herbicides and organochlorine and organophophorous pesticides were reported in the aquatic environment of the Mekong River delta (Carvalho et al. 2008).

Polychlorinated biphenyl compounds are common industrial chlorinated organic chemicals with 209 possible congeners. The physical chemical properties of these compounds have made them attractive as insulators in transformers, hydraulic fluids, paint additives, fire retardants, and pesticide extenders (Waid 1987; US EPA 2008). In many countries, the leakage of old electric transformers

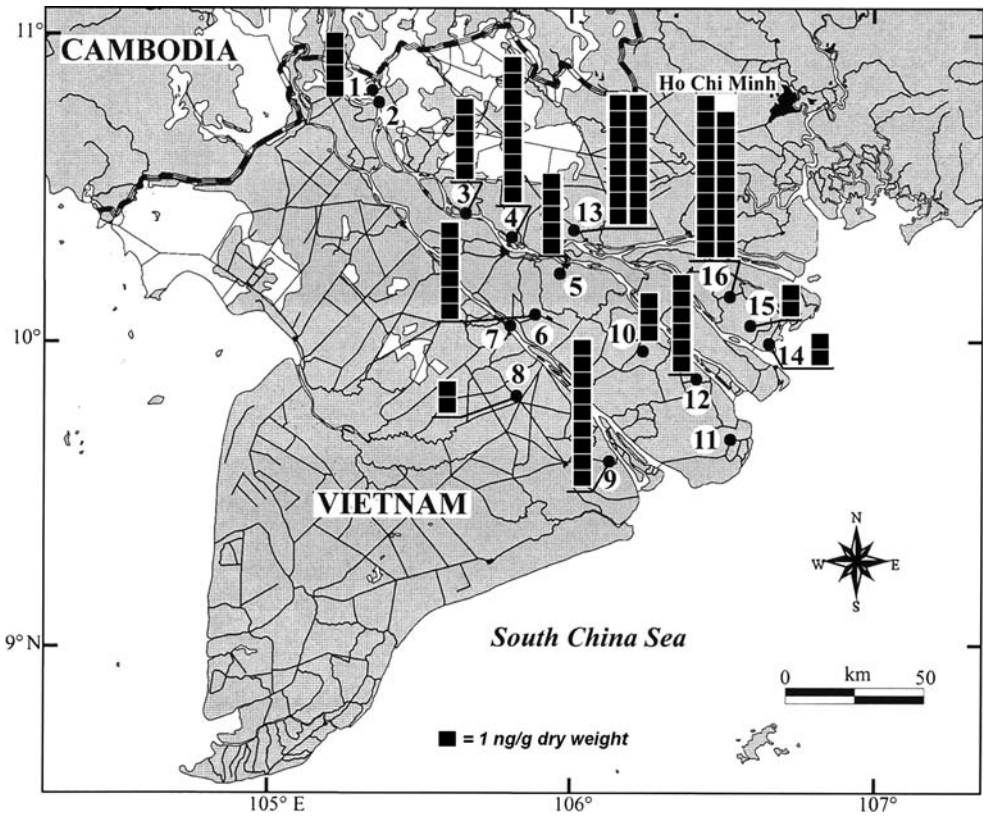

Fig. 1 The Mekong River delta and sampling stations indicated with *numbers*. Concentrations of ΣPCB in clams, rounded up to the nearest unit value, are indicated by *columns*

was the source of PCB releases into the environment where they persist for many years. In the aquatic environment PCBs are bioconcentrated and transferred in the food chains and may return to humans with the sea food (Albaigès 1993; De Boer 1995). Due to confirmed deleterious effects on biota and on human health, PCBs in the environment and in the food are under close monitoring worldwide (Colborn et al. 1993; UNEP 1997; Reyes et al. 2003; Taylor et al. 2003).

Polychlorinated biphenyl compound sources in the delta region are not fully identified. As this region is often impacted by typhoons and floods, the environmental dispersion and concentrations of these contaminants in the aquatic environment is also not known. This paper reports for the first time concentrations of individual chlorobiphenyl congeners in sediments and molluscs from the Mekong River delta and assesses the distribution and origin of these compounds in this region.

Materials and Methods

Collection of samples was performed in the Mekong River delta, from the border of Vietnam with Cambodia to the coast of South China Sea, just before the rainy season (May 1998) (Fig. 1). Approximately 0–5 cm surface sediment samples were collected into hexane rinsed glass jars and kept frozen until processing. These samples were freeze dried and sieved through a 250 μm metal sieve in order to remove the coarse materials (less than 2% in weight of sediment collected). Sediment-burrowing bivalve molluscs (clams) were collected from the bottom sediment of Mekong River at 13 out of the 16 sampling sites. Names of biological species are reported elsewhere (Carvalho et al. 2008). From each site, soft tissues of more than 20 clams were dissected and combined in one sample. These samples were frozen, freeze dried, and the resulting powder homogenized.

About 5 g of dry sediment, and a similar amount of bivalve tissues, were used for analysis of chlorinated hydrocarbons, according to techniques described in detail elsewhere (Villeneuve and Cattini 1986; Villeneuve et al. 1999). Briefly, at the beginning of the analysis 25 ng of 2, 4, 5 trichlorobiphenyl were added to each sample as an internal standard for individual sample control of the chemical recovery yield. Chemical yields averaged 71% (range 62–83%) for sediment samples and 76% (range 69–87%) for biota samples. Samples were extracted for 8 h in a Soxhlet apparatus with a mixture of hexane and dichloromethane (1:1) for sediments and with hexane only for biological samples. Extracts were clean up in Florisil

columns pre cleaned with methanol and hexane. From Florisil columns three fractions were eluted for separation of PCBs, HCB, DDE, and aldrin, obtained in the 1st fraction, from other chlorinated pesticides removed in the second and third eluted fractions (not reported herein). Hexane was used as elution solvent for the PCB fraction. Chromatographic analyses were performed with a gas chromatograph HP 5880A, equipped with a capillary column 25 m long, 0.2 mm internal diameter, coated with SE 54 silica phase, and an electron capture detector (ECD). Chromatographic conditions used were the following: splitless injector 250°C, detector 300°C, oven 70°C (2 min) with a ramp of 3°C/min to 260°C (20 min). Confirmatory analyses were performed with a mass spectrometer HP5889 "Engine B" using negative ion chemical ionisation (NICI).

Solvents used were of high purity pesticide quality (Burdick and Jackson Labs, Muskegon, MI, USA). Authentic standards of organochlorine compounds, purity >99%, were purchased from Interchim and Promochem (France). Analytical quality control for chlorinated hydrocarbons was made using reference materials IAEA-142 (mussel tissue homogenate) and IAEA-357 (estuarine sediment), co-analysed with each batch of samples.

Thirteen polychlorobyphenyl congeners or isomers (IUPAC nr. 44, 49, 52, 101, 105, 118, 128, 138, 149, 153, 170, 180, 201) were individually quantified in sediment and biota samples. This selection encompasses the seven congeners recommended by the World Health Organization for environmental monitoring (WHO 1999). PCB results are shown as concentrations for individual congeners, as ΣPCB (the sum of 13 congeners), and as equivalent to the commercial mixtures aroclor 1254 and aroclor 1260. Aroclors are widely used commercial mixtures of PCBs with known congener composition. Concentrations of PCBs are reported in ng g^{-1} (dry weight). Conversion of concentrations on dry weight to lipid weight can be made using the concentration of hexane extracted lipids in sample materials (HEOM) provided in Tables. Whenever a compound was not detected, the result is reported as <LOD value (below the limit of detection for that compound).

Results and Discussion

Concentrations of PCB congeners in sediments and in mollusc soft tissues are shown in Tables 1 and 2, respectively. Tables also include values for PCBs as ΣPCB, the sum of all 13 congeners quantified, and as aroclor 1254 and aroclor 1260. Concentrations of ΣPCB in sediments display a median value of 0.279 ng g^{-1} dry weight (range 0.106–2.016 ng g^{-1}, n = 16), while in molluscs the median value was 5.20 ng g^{-1} dry weight (range 1.89–19.37 ng g^{-1}, n = 13). Using aroclor equivalent concentrations instead of ΣPCB, average results would not be very different.

Based on the median of ΣPCB values, concentration of these compounds in molluscs is about 24 times higher than concentrations in sediments. Actually, accumulation of PCBs in molluscs' soft tissues is significantly correlated with concentrations of these compounds in sediments at the same sampling stations. The equation of the regression equation that can be obtained using ΣPCB paired data in Tables 1 and 2, is

$$\text{Log } y = 0.64(\text{Log } x) + 1.00$$

with x = concentration in sediment, ng g^{-1} dry weight, y = concentration in biota, ng g^{-1} dry weight (n = 13, $R^2 = 0.61$; $p = 0.002$).

Accumulation of individual PCB congeners in sediments and in molluscs is not the same for every congener and depends upon the lipophilic properties of each isomer (Spacie et al. 1995; Lu et al. 2007). Using the octanol-water partitioning coefficient (Kow) as a measure of lipophilic properties, and plotting concentrations of PCB congeners in biota and sediments as a function of Log Kow, the graphic plot highlights differences in bioconcentration amongst congeners with the same chlorination degree, i.e., with the same number of chlorine atoms in their molecular composition (Fig. 2). Furthermore, it suggests that there are similar bioconcentration trends amongst congener groups as reported before (De Boer 1995). Congener specific bioaccumulation kinetics was experimentally demonstrated in sea stars (Danis et al. 2006).

The relative contribution of congeners to ΣPCBs in sediments and in molluscs' tissues is shown in Fig. 3, showing a major accumulation of isomers with four and five chlorine atoms. As the number of chlorine atoms in congener's molecules is different, and the properties of congeners and their bioaccumulation also vary with it, the fate of PCB congeners in the aquatic environment might vary as a function of the number and position of chlorine atoms (De Boer 1995; Lu et al. 2007).

The geographic distribution of PCBs in the delta region is depicted in Fig. 1 using ΣPCB values determined in bivalve molluscs. It is apparent that ΣPCB concentrations are not uniformly distributed throughout the aquatic environment of the delta. Higher values were generally measured near the largest cities of this region and likely originate in leakage from old electric transformers and waste discharges from repair workshops. Furthermore, it may be noticed that there is a widespread presence of PCBs in the delta region. Dispersal of PCBs into rural areas may occur by tidal currents in the water canals, by storms and typhoons, and direct leakage from boat engines into the water. Atmospheric deposition of PCBs transported from more industrialized regions to the Mekong River delta is

Table 1 Concentrations (ng g^{-1} dry weight) of chlorinated hydrocarbons in sediment samples of the Mekong River delta, Vietnam

	Station 1	Station 2	Station 3	Station 4	Station 5	Station 6	Station 7	Station 8	Station 9	Station 10	Station 11	Station 12	Station 13	Station 14	Station 15	Station 16
	Hong Ngu Mekong river	Hong Ngu Canal	Cao Lanh River	Nav. Canal Near houses	Vinh Long Canal	Can Tho Canal	Can Tho River mudflat	Phung Hiep Canal	Long Phu Canal	Tra Vinh Canal	Duyen Hai Mouth Mekong	S Tra Vinh Canal	Canal	Ba Tri At the coast	Canal Rice zone	Giong Trom Canal
Latitude (N)	10° 48.137′	10° 46.550′	10° 25.059′	10° 20.397′	10° 13.316′	10° 05.380′	10° 03.132′	09° 49.431′	09° 36.732′	09° 58.186′	09° 41.052′	09° 52.726′	10° 21.772′	09° 59.640′	10° 03.189′	10° 08.887′
Longitude (E)	105° 20.484′	105° 21.509′	105° 38.630′	105° 47.645′	105° 57.101′	105° 52.219′	105° 47.396′	105° 48.597′	106° 06.818′	106° 13.319′	106° 30.477′	106° 23.802′	105° 59.743′	106° 37.976′	106° 34.337′	106° 30.344′
Compounds																
Dry/wet weight	0.75	0.57	0.64	0.56	0.69	0.56	0.62	0.50	0.42	0.41	0.44	0.51	0.46	0.56	0.4	0.37
HEOM (mg/g)	0.003	0.13	0.11	0.11	0.082	0.22	0.063	0.21	0.16	0.21	0.17	0.29	0.31	0.13	0.12	0.30
PCB 44	<0.003	0.004	<0.003	<0.003	0.014	<0.003	<0.003	<0.003	<0.003	<0.003	<0.003	0.032	0.037	<0.003	<0.003	0.020
PCB 49	0.010	0.021	0.009	0.015	0.039	0.025	0.013	0.018	0.025	0.030	0.028	0.095	0.050	0.016	0.020	0.054
PCB 52	<0.006	<0.006	<0.006	<0.006	0.021	0.009	0.006	0.015	0.006	<0.006	<0.006	0.054	0.10	<0.006	<0.006	0.028
PCB 101	<0.004	0.007	0.007	0.022	0.059	0.015	0.017	0.030	0.034	0.030	<0.004	0.17	0.28	0.005	0.019	0.088
PCB 105	0.007	<0.003	<0.003	<0.003	0.018	<0.003	<0.003	<0.003	0.010	<0.003	<0.003	0.029	0.067	<0.003	<0.003	<0.003
PCB 118	0.005	0.007	0.007	0.008	0.041	0.080	0.007	0.015	0.008	0.011	<0.003	0.034	0.18	<0.003	0.004	0.023
PCB 128	0.005	0.008	0.004	0.009	0.020	<0.003	0.010	0.017	0.007	<0.003	<0.003	0.023	0.063	0.005	0.006	0.028
PCB 138	0.018	0.074	0.044	0.088	0.11	0.045	0.059	0.058	0.059	0.035	0.019	0.14	0.38	0.021	0.028	0.25
PCB 149	0.010	0.036	0.023	0.044	0.059	0.020	0.027	0.026	0.039	0.017	0.007	0.071	0.23	0.008	0.017	0.20
PCB 153	0.012	0.068	0.037	0.077	0.092	0.033	0.044	0.044	0.057	0.030	0.015	0.12	0.29	0.016	0.025	0.26
PCB 170	0.029	0.021	0.017	0.048	0.049	0.012	0.030	0.017	0.032	0.016	0.018	0.082	0.12	0.016	0.019	0.13
PCB 180	0.036	0.052	0.036	0.093	0.092	0.028	0.063	0.042	0.077	0.047	0.021	0.13	0.20	0.019	0.031	0.25
PCB 201	<0.004	0.005	<0.004	<0.004	0.007	0.008	<0.004	<0.004	<0.004	0.007	0.010	0.005	0.019	<0.004	<0.004	0.017
ΣPCB	0.132	0.303	0.184	0.404	0.621	0.275	0.276	0.282	0.354	0.223	0.118	0.985	2.016	0.106	0.169	1.348
Aroclor 1254	0.088	0.13	0.15	0.19	0.85	0.35	0.19	0.27	0.25	0.094	0.073	1.3	2.0	0.10	0.11	0.83
Aroclor 1260	0.28	0.40	0.31	0.81	0.81	0.43	0.56	0.34	0.70	0.25	0.19	1.2	1.8	0.20	0.28	2.3

HEOM Hexane extractable organic matter. Values indicated as <0.00*x*: <limit of detection

Table 2 Concentrations (ng g^{-1} dry weight) of chlorinated hydrocarbons in biota samples (clams) of the Mekong River delta, Vietnam

	Station 1 Hong Ngu Mekong river	Station 3 Cao Lanh River	Station 4 Nav.Canal Near houses	Station 5 Vinh Long Canal	Station 6 Can Tho Canal	Station 8 Phung Hiep Canal	Station 9 Long Phu Canal	Station 10 Tra Vinh Canal	Station 12 S Tra Vinh Canal	Station 13 Canal	Station 14 Ba Tri At the coast	Station 15 Canal Rice zone	Station 16 Giong Trom Canal
Compounds													
Dry/wet weight	0.24	0.22	0.30	0.26	0.17	0.19	0.22	0.13	0.19	0.21	0.23	0.25	0.18
HEOM (mg/g)	82	72	79	45	82	49	70	77	78	37	59	64	89
PCB 44	0.072	<0.012	0.11	0.051	<0.012	<0.012	0.30	<0.012	0.31	0.21	<0.012	<0.012	1.1
PCB 49	0.69	0.84	0.52	0.30	1.1	0.29	0.79	2.0	0.58	0.47	0.17	0.20	1.4
PCB 52	0.16	0.079	0.13	0.075	0.22	0.090	0.49	0.061	0.42	0.69	0.056	0.050	1.1
PCB 101	0.44	0.39	0.59	0.34	0.62	0.18	1.3	0.33	0.83	2.9	0.12	0.055	1.8
PCB 105	0.065	0.17	<0.010	0.045	0.14	0.064	0.14	0.087	0.087	0.51	0.024	<0.010	0.33
PCB 118	0.27	0.39	0.24	0.17	0.45	0.11	0.56	0.034	0.34	1.8	0.054	0.12	0.68
PCB 128	0.12	0.21	0.19	0.15	0.17	0.032	0.28	<0.011	0.16	0.50	0.15	0.10	0.64
PCB 138	0.75	0.92	2.0	1.0	0.96	0.34	1.6	0.16	0.74	3.3	0.32	0.20	3.9
PCB 149	0.38	0.48	1.4	0.64	0.62	0.23	1.5	0.23	0.67	1.9	0.26	0.92	2.6
PCB 153	0.6	0.88	2.0	0.96	1.5	0.34	1.8	0.28	1.2	2.6	0.47	0.19	4.9
PCB 170	0.17	0.23	0.45	0.45	0.11	0.096	0.20	0.11	0.053	0.33	0.11	0.018	0.27
PCB 180	0.32	0.34	0.82	0.99	0.31	0.25	0.27	0.15	0.11	0.79	0.31	0.040	0.54
PCB 201	<0.016	0.082	0.042	0.029	0.026	0.010	0.038	<0.016	<0.016	0.098	<0.016	<0.016	0.11
ΣPCB	4.037	5.011	8.492	5.2	6.226	2.032	9.268	3.442	5.5	16.098	2.044	1.893	19.37
Aroclor 1254	5.1	5.1	5.6	3.1	7.0	2.8	13	6.1	9.7	24	1.6	5.1	22
Aroclor 1260	2.3	2.7	8.0	7.1	2.6	2.0	5.4	1.7	1.5	5.8	1.5	0.38	9.1

HEOM Hexane extractable organic matter. Values indicated as <0.00*x*: <limit of detection

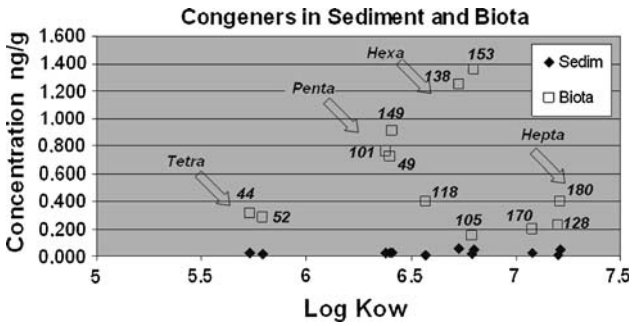

Fig. 2 Concentrations of PCB congeners plotted against Log Kow of congeners. *Congener numbers* are indicated close to concentration values in biota

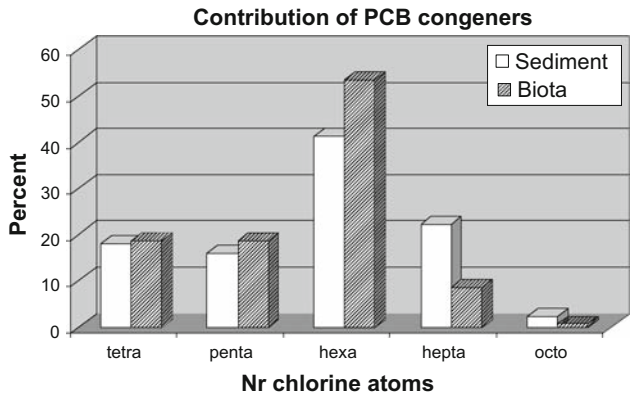

Fig. 3 Percent contribution of congener groups to ΣPCB in biota and sediments. Congeners in each group are: tetraCB 44, 49, 52; pentaCB 101,105,118; hexaCB 128, 138, 149, 153; heptaCB 170, 180; octoCB 201

not unlikely either. Actually, it was documented that concentrations of PCBs in surface air at South-East Asia and in surface water of the Indian Ocean are deposited following atmospheric transport from industrial regions (Iwata et al. 1994).

Concentrations of PCBs in the aquatic environment of the Mekong River delta are lower than concentrations reported in freshwater canals near Ho Chi Minh City, and in the Sai Gon-Dong Nai River in South Vietnam, a region more industrialized than the Mekong delta (Phuong et al. 1998; Minh et al. 2007). Concentrations reported herein are also lower than PCB concentrations measured in the water canals near Hanoi, and on the coast of North of Vietnam (Nhan et al. 1998, 1999, 2001). For example, PCBs in sediments of Ho Chi Minh City canals ranged from 46 to 150 ng g^{-1} dry weight (Minh et al. 2007) while in sediments of freshwater canals near Hanoi ΣPCB ranged from 0.74 to 33.68 ng g^{-1} dry weight (Nhan et al. 2001). These differences in PCB levels amongst regions underline the agricultural characteristics of the Mekong delta region in contrast with other more industrialized regions of Vietnam.

From the ecotoxicological point of view, PCB concentrations in the Mekong river delta sediments (maximum measured 2.016 ng g^{-1} dry weight) are well below the threshold effects level for PCBs, 21.55 ng g^{-1} dry weight, adopted in some countries as a guideline for marine sediment quality and considered to afford sufficient protection to aquatic fauna (Gómez-Gutierrez et al. 2007).

In brief, PCBs were present in sediments and sediment burrowing bivalve molluscs of the Mekong River delta. PCB concentrations measured in this region were lower than in more industrialized regions of Vietnam such as, for example, Ho Chi Minh City and Hanoi regions, which was not surprising due to the rural characteristics of the Mekong delta area. Despite the generally low levels of PCBs in the Mekong delta, still relatively higher concentrations were measured near the main cities of the delta. These higher concentrations probably originated in waste discharges from repair workshops and leakage from old electrical transformers. In the most remote and rural areas of the delta, lower PCB concentrations may have originated, at least partially, in atmospheric depositions as well as in the environmental redistribution of PCBs by floods and storms that are frequent in the South-East Asia. The highest PCB concentrations currently measured in sediments of the Mekong delta were below the threshold effects level for marine fauna. Nevertheless, measures for better control of PCB discharges into the environment should be implemented in order to keep environmental concentrations of PCBs low and to prevent their ecotoxicological effects.

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