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Polychlorinated biphenyls in sediments of the Tam Giang-Cau Hai Lagoon, Central Vietnam

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Abstract

The Tam Giang-Cau Hai Lagoon, the largest in south east Asia, suffers from a great anthropogenic pressure and appears subject to a process of progressive environmental deterioration. To establish causes, history and trends of lagoon contamination, sediment samples representing three major parts of the systems were sampled and analysed for porosity, organic carbon, total nitrogen, $\delta^{13}C$, grain size, radiotracers and PCBs. PCB surficial concentrations range from 10.2 to 24.5 μg kg^{-1}, and maximum values are close to NOAA ERL guidelines. The downcore distribution of PCBs shows two well defined peaks in the core from Tam Giang (25.5 and 16.2 μg kg^{-1}), whereas the sediment from Cau Hai is characterised by a maximum at the surface. Therefore, present trends indicate that contamination is still increasing or slightly decreasing. The prevailing congeners are 3-CB and 4-CB, hence recent PCBs appear to be mostly originated by Aroclor 1016- and 1242-like mixtures, with some samples influenced also by Aroclor 1248. Present sources seem to be similar everywhere and probably they are widespread all over the territory of the Thua Thien-Hue province.

Keywords: PCBs; Sediments; Chronology; Tam Giang-Cau Hai Lagoon; Vietnam

1. Introduction

Polychlorinated biphenyls (PCBs) are mixtures of synthetic organic chemicals that due to their non-flammability, chemical stability, high boiling point and electrical insulating properties, were used in hundreds of industrial and commercial applications. According to the US EPA, PCBs have been demonstrated to cause a variety of adverse health effects. In particular, PCBs have been shown to cause cancer in animals, and a series of non-cancer effects such as reproductive and developmental problems, immune deficiency, nervous system alterations, endocrine disruption, gastrointestinal system bleeding and liver damage. Dermal and ocular effects are also very common. Furthermore, studies in humans provide supportive evidence for potential carcinogenic and non-carcinogenic effects of PCBs.

In spite of the use ban in open systems, PCBs are still used in enclosed transformers, and therefore constitute a class of compounds of major concern for both the environment and the human population. It follows that concentrations in soils and sediments should be well known because high contamination levels can represent a major problem of environmental management.

From the point of view of PCB contamination, Vietnam makes no difference with respect to more industrialised countries though the concentrations in the environment reported so far are not high (e.g. Iwata et al., 1994; Nhan

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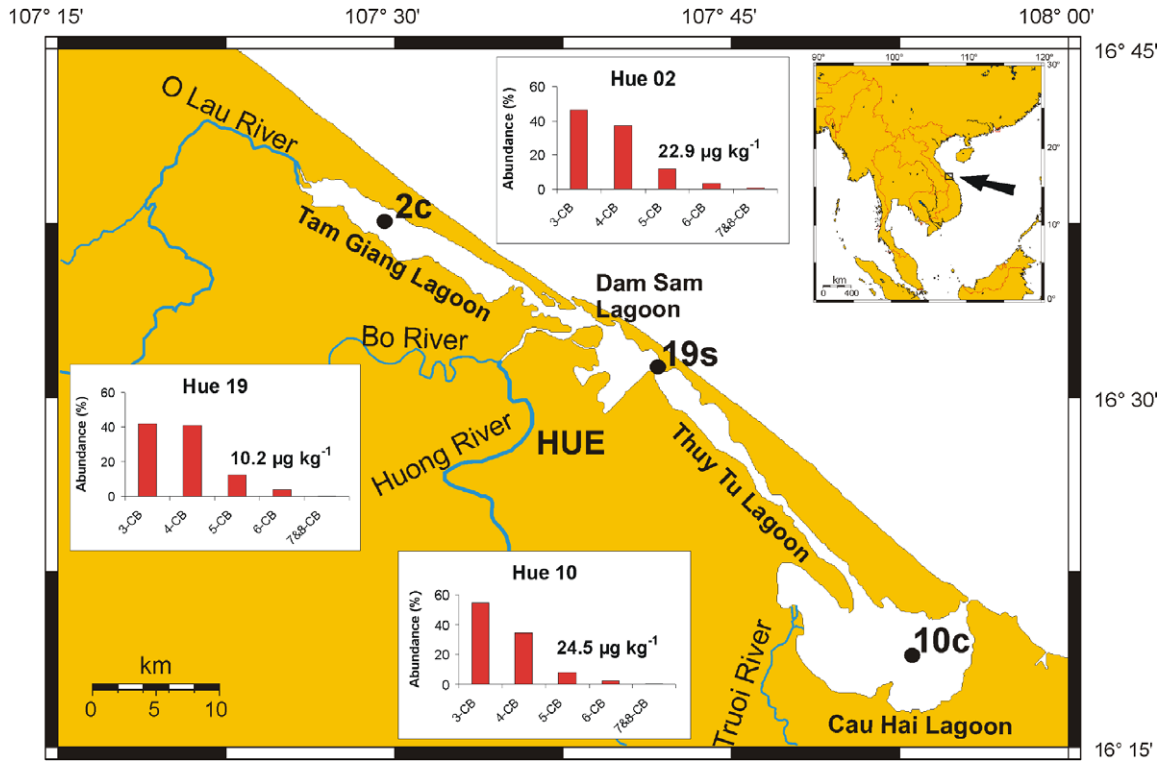

Fig. 1. Map of the Tam Giang-Cau Hai Lagoon, sampling sites, PCB concentrations and homologue profiles in surficial samples. C and S mean "core" and "surficial", respectively.

et al., 1998, 1999). In fact, due to the rapid industrial development, the risks that pollution levels reach high values in the near future is real (Thanh et al., 2004). Therefore, Vietnamese authorities need to set up environmental regulations and management capability to reverse environmental degradation trends.

It is quite clear that any effective environmental damage prevention and recovery of contaminated areas should be based on sound scientific information. However, so far, in Vietnam the research-based knowledge of key environmental processes is rather poor, and not many data are available about levels of contamination, sources and trends, in many areas of the country. This lack of information is especially problematic for some of the most toxic organic pollutants. Furthermore, since the coastal zones are undergoing an ever increasing anthropogenic pressure, the lagoons of Central Vietnam should be considered as priority environments.

The Tam Giang-Cau Hai (TG-CH) Lagoon, a typical tropical coastal wetland, is the largest in southeast Asia, being 70 km long and covering an area of about 216 km^2. This system (Fig. 1) is quite complex because it is composed of four parts with different shapes and extension referred to as Pha Tam Giang (27 km long, 0.6–3.5 km wide, up to 2 m deep), Dam Sam (5 × 5.5 km, 2 m deep), Dam Thuy Tu (24.5 km long, 0.8–2.6 km wide, up to 4 m deep) and Dam Cau Hai (a southernmost, semi circular basin 17 km wide and 1–1.5 m deep).

Characteristics and problems of the TG-CH lagoon were extensively described by Frignani et al. (2003). In particular, it is said that this environment is now facing degradation due to oil pollution, coliform contamination, eutrophication and presence of anthropogenic chemicals. The purpose of this work was to provide a first insight on the presence of PCBs in lagoon sediments with a particular attention to composition, possible sources, history and trends.

2. Materials and methods

Sediment samples were collected from three basins of the TG-CH Lagoon in December 2002. A manual piston core was used to retrieve both surficial samples (e.g., 19S) and short cores (02C and 10C). Sampling locations are shown in Fig. 1. After collection, the cores were X-radiographed at the hospital of the city of Hue, and then extruded and sectioned at intervals of 2–4 cm, with higher resolution at the top. Sediment slabs were then divided into two parts for the different analyses, put in polyethylene vessels and stored in a refrigerator at 0 °C until the arrival to the lab. Afterwards, they were conserved at −18 °C until the analyses.

For PCB analyses, sediment samples were lyophilised and then homogenized and extracted in a sonication bath with 50 ml of a mixture of pesticide grade *n*-hexane–dichloromethane (4:1, v/v). The solutions were dried by

anhydrous Na_2SO_4 and reduced to 25 ml under a gentle stream of nitrogen. After sulphur removal by several treatments with 2 ml of mercury, extracts were purified by solid–liquid chromatography (stationary phase alumina/florisil, eluent *n*-hexane) and then reduced to 0.5 ml. PCB analyses were carried out by HRGC–LRMS following the method described by Moret et al. (2001). Five ^{13}C labeled PCBs were added to the samples before extraction and used as internal standards in the quantification: ^{13}C-CB28 for 2- and 3-CBs, ^{13}C-CB52 for 4-CBs, ^{13}C-CB101 for 5-CBs, ^{13}C-CB153 for 6-CBs, ^{13}C-CB180 for 7- and 8-CBs. Crude concentration values were corrected with congener-specific instrumental response factors.

PCB concentrations were calculated as the sum of 53 congeners (all those present in a detectable concentration) correspondent to 44 chromatographic peaks (35 as single congeners and 9 as pairs of congeners). Accuracy was checked with respect to a certified standard (NIST®, Standard Reference Material 1941b). Precisions, calculated using the same certified standard, are typically 10% on the sum of congeners and, for the latters, span the interval 5–20%.

For ^{137}Cs determinations, freeze-dried sediments were put in standard vessels of suitable geometries and gamma counted. ^{210}Pb analyses were carried out by leaching dry samples with hot HNO_3 and H_2O_2 followed by elimination of the acid, dissolution in HCl and plating of ^{210}Po on silver planchets. ^{210}Po, considered in secular equilibrium with its parent ^{210}Pb, was alpha counted by a surficial barrier detector coupled with a multichannel analyser.

Porosities were determined by drying sediments at 60 °C, and calculated assuming a particle density of 2.5 g cm^{-3}. Organic carbon (OC) and total nitrogen (N) contents plus $\delta^{13}C$ were obtained using a CHN analyzer by Fison Instruments, coupled with a Finnigan Delta Plus mass spectrometer, after elimination of the carbonate fraction with HCl directly in a silver capsule. Grain size analyses were carried out by wet sieving, to separate sands, after a pretreatment with H_2O_2. Silt and clay contents were determined by a X-ray Micromeritics SediGraph.

All concentrations and activities were calculated on a dry weight basis.

Chemometric multivariate analysis of the data was performed through Principal Component Analysis (PCA) using Statgraphics Plus 5.1 (Manugistics, Inc. Rockville, MD, USA) software package.

3. Results and discussion

3.1. Sediment features

Table 1 summarises the results regarding sediment characteristics. X-radiographs are shown in Fig. 2, together with the depth distributions of sediment porosity and grain size. X-radiographs can reveal both major physical discontinuities and the presence of disturbance of the record throughout the sediment column. In these cases, sediments appear fairly homogeneous, with traces of bioturbation that are more evident at depth in 10C.

Table 1
Organic carbon and total nitrogen concentrations, C/N ratios, $\delta^{13}C$ and % content of grain size fractions in selected samples

Sample	OC %	N %	C/N	$\delta^{13}C$	Sand %	Silt %	Clay %
02C 0–2	1.28	0.13	10.10	−24.63	7.51	62.02	30.47
02C 10–12	1.11	0.11	10.28	−24.18	12.17	56.44	31.39
02C 23–26	1.24	0.12	11.63	−24.99	9.43	62.24	28.33
02C 50–54	1.15	0.10	11.32	−25.02	n.d.	n.d.	n.d.
02C 70–74	1.66	0.14	12.18	−25.73	n.d.	n.d.	n.d.
19S 0–2	0.53	0.06	8.67	−23.77	54.80	29.47	25.73
10C 0–2	1.09	0.13	8.58	−21.13	11.71	78.15	30.14
10C 6–8	0.89	0.09	9.47	−21.19	15.45	55.95	28.60
10C 14–16	0.72	0.08	9.51	−21.64	18.07	52.6	29.33
10C 26–29	0.88	0.09	10.25	−23.51	4.56	61.65	33.79

n.d. = not determined.

According to the grain size distribution map of Lan et al. (1995), 10C, 02C, and 19S were collected in areas characterized by rather fine sediment composition: fine silt, silt, and very fine sand, respectively. Our results show that all samples can be classified as clayey silts, except 19S which is a silty sand. This sample well represents the dynamic environment of the area, deeply influenced by both the mouth of the Huong river and the principal lagoon inlet.

Depth profiles of both porosity and sand contents (Fig. 2) account for a major environmental change. The increase of coarse sediment concentrations at 40–32 and 23–17 cm depth in cores 02C and 10C, respectively, suggests that the lagoon environment was subjected to higher hydrodynamics, with the removal of fine sediment due to both more efficient transport and resuspension. In recent times the water dynamics is decreasing again and this is recorded by sediments as an increase of porosity and a decrease of the content of sand close to the sediment–water interface. The patterns are quite similar in the two basins, even though variations seem more regular in core 10C. The correlations of depth profiles of Fig. 2 suggest that the sediment accumulation rate at 02C is nearly double than at 10C, which than spans a longer time interval.

The OC contents (Table 1) range from 1.11–1.66% to 0.72–1.09% in cores 02C and 10C, respectively. In sediment 19s, which is coarser, the OC concentration is lower (0.53%). The generally low values could be due to a number of factors, such as low production, high rate of decomposition of autochthonous organic matter, reduced input from the rivers and/or dilution by mineral particles. C/N ratios, calculated assuming that N is mostly organic, are higher than 10 (10.10–12.18) in core 02C, whereas values in core 10C range between 8.58 and 10.25. Values higher than 10 account for a terrestrial origin of the organic matter, prevalent at site 02C, whereas a mixed source influences 10C. The changes of both C/N ratios and $\delta^{13}C$ within both cores suggest a shift from a sediment at depth dominated by terrestrial OC sources to a present situation

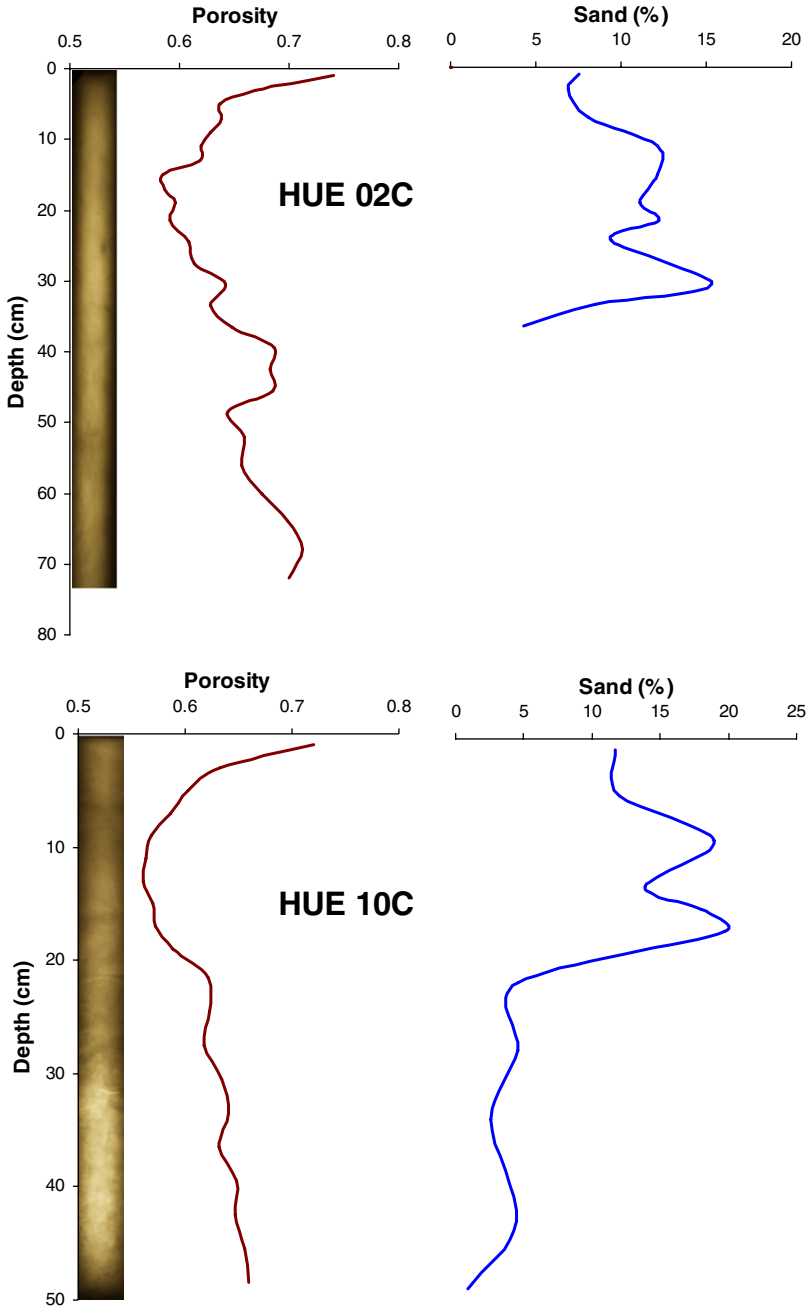

Fig. 2. X-radiographs of cores 02C and 10C and depth distributions of porosities and sand contents.

where the marine component has a higher relative importance (Goñi et al., 2003).

3.2. PCB concentrations and distributions

Table 2 lists both total PCB concentrations and the sums of homologues. Since data on the occurrence of non dioxin like PCBs in food and feed are often reported as the sum of six PCB congeners (CB-28, CB-52, CB-101, CB-138, CB-153 and CB-180) referred to as indicator PCBs (EFSA, 2005), Table 3 shows the concentrations in sediments of these compounds. They represent 28.6–40.2% of the total PCBs. In general, congeners 31 + 28 (2,4′,5- and 2,4,4′-CBs) were prevalent in most samples, whereas none of the non-*ortho* PCB congeners was detected.

In Table 4, the concentrations found in samples from the TG-CH are compared with literature data. Our results are close to those reported by Nhan et al. (1999) for the northern coast of Vietnam (0.5–28.1 μg kg^{-1}) and lower than those found by Iwata et al. (1994) near Vietnamese cities. These data also demonstrate that the contamination of the TG-CH lagoon is relatively low, not comparable with that of the most industrialised areas. However, the

Table 2
PCB concentrations in sediments of the Tam Giang-Cau Hai Lagoon

Sample	3-CBs	4-CBs	5-CBs	6-CBs	7&8-CBs	Total
02C 0–2	10.7	8.53	2.72	0.80	0.15	22.8
02C 8–10	13.5	8.40	2.07	0.63	0.08	24.7
02C 20–23	1.05	0.74	0.43	0.09	l.d.l.	2.31
02C 32–35	8.74	4.61	1.51	0.30	l.d.l.	15.4
02C 47–50	1.07	0.57	0.32	0.07	l.d.l.	2.03
19S (0–2)	4.31	4.18	1.26	0.41	0.06	10.2
10C 0–2	13.4	8.50	1.97	0.55	0.97	24.5
10C 2–4	15.7	7.05	0.96	0.17	l.d.l.	23.8
10C 8–10	1.93	0.19	0.11	0.05	l.d.l.	2.28
10C 12–14	2.27	3.34	0.78	0.16	l.d.l.	6.55
10C 18–20	1.22	1.01	0.83	0.14	l.d.l.	3.20
10C 23–26	1.09	1.45	1.06	0.16	l.d.l.	3.75
10C 38–41	0.83	1.06	0.85	0.11	l.d.l.	2.85

Values are in $\mu g\,kg^{-1}$. l.d.l. = lower than detection limits.

maximum values in both cores (the most recent ones) are very close to the NOAA effect range low (ERL) guidelines (Long et al., 1995) and can cause adverse effects only occasionally.

The highest contamination, among surficial samples, characterises site 10C ($24.5\,\mu g\,kg^{-1}$), followed by 02C ($22.9\,\mu g\,kg^{-1}$) and 19S ($10.2\,\mu g\,kg^{-1}$). This pattern probably depends on the different inputs to the various sections of the lagoon and the relative mechanisms of dispersion and accumulation, hence it may be influenced by both the relative importance of the river sources and the sediment grain size. In fact, the lowest surficial value seems to be caused by the prevalence of sand at site 19S. Actually, the input of solid material from the Huong River system, being close to the Thuan An lagoon inlet, can be more easily lost to the sea, especially the finest particle fraction, which is the most efficient pollutant carrier. In turn, the northern and the southern basins should act more efficiently as fine sediment traps. On the other hand, the homologue profiles of Fig. 1 show that PCBs in surficial sediments have a similar composition, especially 02C and 10C, thus suggesting a unique type of source. The PCB profile of sample 19S could have been modified by the particular site conditions already mentioned. Therefore, present sources seems to be similar everywhere and probably widespread all over the inland territory. Actually, the presence of electrical devices is very visible all over the Thua Thien-Hue province and their PCB content can represent the most important source of contamination.

Table 4
Comparison of PCB concentrations in sediments of the Tam Giang-Cau Hai Lagoon with literature data regarding places in Asia, Australia and Europe

Location	PCBs ($\mu g\,kg^{-1}$)	Reference
Tam Giang	2.03–24.7	This paper
19S	10.2	This paper
Cau Hai	2.85–24.5	This paper
India	4.8–1000	Iwata et al. (1994)
Thailand	11–520	Iwata et al. (1994)
Vietnam	0.18–630	Iwata et al. (1994)
Paddy field	0.65–140	Iwata et al. (1994)
Paddy field and mangroves	2.1–9.7	Iwata et al. (1994)
Urban area (municipal sawage)	0.10–630	Iwata et al. (1994)
Coastal sediments (north Vietnam)	0.18–6.13	Nhan et al. (1999)
Surficial sediments, Red River delta	1.01–10.5[a]	Nhan et al. (1998)
Soils	0.61–320	Thao et al. (1993)
Indonesia	5.9–220	Iwata et al. (1994)
Japan	63–240	Iwata et al. (1994)
Australia	0.49–790	Iwata et al. (1994)
Italy–Venice Lagoon	0.56–18.4	Frignani et al. (2001)
Italy–Venice, industrial canals	10.8–5245	Frignani et al. (2004)
Black Sea	<0.06–5.8[a]	Fillman et al. (2002)
Noethwestern Mediterranean	1.1–311	Eljarrat et al. (2001)
Adriatic Sea	l.d.l.–2203	Picer (2000)
Port of Trieste	68–2368	Adami et al. (2000)
Ligurian Sea	l.d.l.–227	Bertolotto et al. (2004)
Aquatic areas of Morocco	0.80–164[b]	Piazza et al. (2004)

[a] As Aroclor 1254; 0.11–6.02 $\mu g\,kg^{-1}$ as Aroclor 1260.
[b] Surficial sediment from the port of Tangier.

Table 3
Concentrations ($\mu g\,kg^{-1}$) of the PCB congeners known as indicators in sediments of the Tam Giang-Cau Hai Lagoon

Sample	31 + 28-CB	52-CB	101 + 90-CB	138 + 158-CB	153-CB	180-CB	Sum	%
02C 0–2	5.92	2.07	0.52	0.13	0.13	0.05	8.81	38.56
02C 8–10	7.73	1.69	0.36	0.07	0.07	0.02	9.94	40.17
02C 20–23	0.51	0.26	0.08	0.02	0.02	l.d.l	0.89	38.40
02C 32–35	4.23	1.08	0.26	0.04	0.05	l.d.l	5.66	36.88
02C 47–50	0.44	0.18	0.05	0.01	0.01	l.d.l	0.70	34.65
19S (0–2)	2.08	0.75	0.21	0.06	0.08	l.d.l	3.17	31.02
10C 0–2	7.46	1.63	0.33	0.07	0.10	0.04	9.64	39.29
10C 2–4	7.52	1.21	0.13	l.d.l	0.05	l.d.l	8.92	37.42
10C 8–10	0.55	0.05	0.03	0.01	0.02	l.d.l	0.66	28.76
10C 12–14	1.26	0.64	0.11	0.02	0.03	l.d.l	2.05	31.34
10C 18–20	0.58	0.24	0.14	0.02	0.03	l.d.l	1.00	31.19
10C 23–26	0.84	0.30	0.17	0.02	0.02	l.d.l	1.35	35.91
10C 38–41	0.41	0.24	0.14	0.02	0.02	l.d.l	0.82	28.61

The values of both the sum and percent contribution to the total are also reported. l.d.l. = lower than detection limits.

Fig. 3 shows the concentration-depth profiles of PCBs in sediments of sites 02 and 10. In both cores the concentrations are maximum in the surface (10C) or near-surface (02C) sediment. This means that different to many other places where controls on the manufacture, use, and disposals of PCBs resulted in their decreased input, the situation in the TG-CH lagoon is not improving. The pattern at 02C shows also a very significant peak recorded between 20 and 48 cm depth, and the surficial value suggests a present slightly decreasing trend. In turn, the PCB concentration-depth profile of core 10C shows a slight increase from 9 to 18 cm depth, thus accounting for a very minor effect of the same older episode of contamination that was well recorded in the Tam Giang basin.

The depth distributions of PCBs in the two cores seem not to be significantly influenced by the changes of the

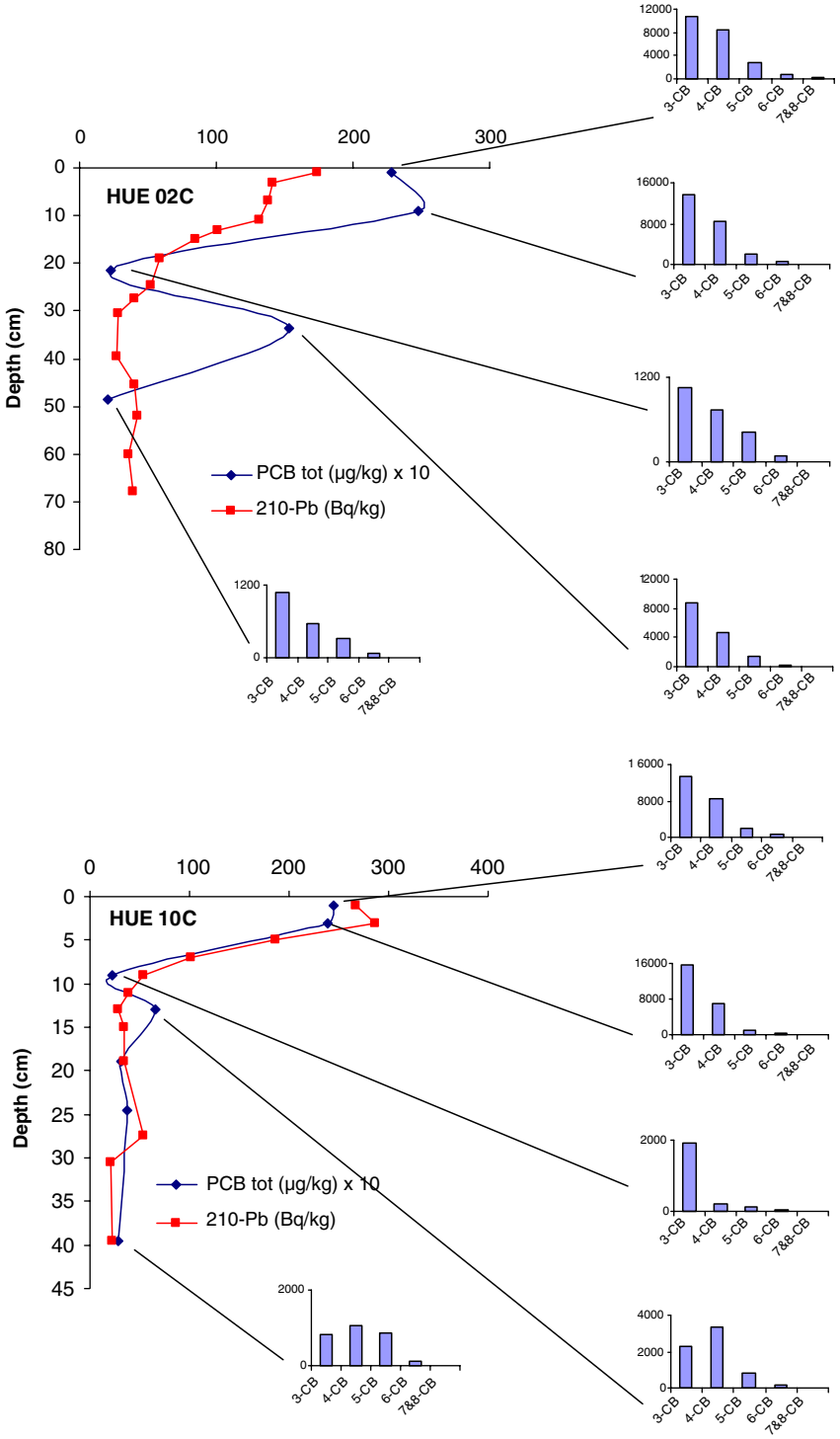

Fig. 3. Activity-depth profiles of total ^{210}Pb and depth distributions of PCBs in cores 02C and 10C. PCB homologue profiles are also shown.

sediment characteristics. However, the congener profiles of Fig. 3 show that while the PCB composition remain unchanged during the time interval represented by core 02C, there are differences at depth in core 10C. These evidences suggest that while the more recent contamination of the Tam Giang and Cau Hai basins has probably a common source, the first PCB arrived into the Southernmost part of the lagoon were slightly different, assuming that the composition was not modified by selective *in situ* processes.

A mean ^{137}Cs inventory of 643 Bq m^{-2} in soils was reported for the Vietnam territory (Hien et al., 2002) and ^{137}Cs activities of 2.80–3.78 Bq kg^{-1} were measured in surficial soils close to the study area (Quang et al., 2004). The ^{137}Cs activities found in lagoon sediments were similar or lower than detection limits, thus providing inventories much lower than expected, especially for core 10C. It is possible that the input of ^{137}Cs to the mainland is very diluted by the huge amount of sediment delivered to the lagoon by rivers, and part is lost to the sea with the finest particles.

Due to the absence of an useful ^{137}Cs signal, inferences on sediment chronologies can be obtained only from the activity-depth profiles of ^{210}Pb (Fig. 3). As a first approximation, we can assume that the depth of ca. 31 cm in core 10C, where ^{210}Pb reaches the background value (that supported by *in situ* decay of ^{226}Ra), corresponds to 100 years. This provides a mean sediment accumulation rate of 0.31 cm y^{-1}. Since the ^{210}Pb concentration changes over time, we can assume that this is mainly due to variations of particle accumulation. According to core correlation, based on the major shifts in grain size composition (Fig. 2) the mean sediment accumulation rate at site 02C is ca. 0.6 cm y^{-1}. Since neither physical mixing nor bioturbation were taken into account, these rates should be considered as upper limits. According to these rates and dates, PCBs were found below 30 cm depth in core 02C, corresponding to the early 1900s. Similar observations of the presence of PCBs in sediments older than the period of first production and commercialisation have been made in other cases (e.g., Gevao et al., 2000; Yamashita et al., 2000; Frignani et al., 2004) and can be ascribed to different factors. Smearing is impossible, since we excluded the outer part of sediment sections, whereas physical mixing and bioturbation can have played a significant role.

3.3. PCB composition

The homologue composition of PCBs in most samples is characterized by the prevalence of 3-CB (Table 2; Figs. 1 and 3). Overall, 3-, 4- and 5-CBs collectively accounted for 95.6–99.6% of total PCB concentrations, whereas 3-CBs and 4-CBs together represent 66.3–95.6%.

A PCA was used to obtain further information on PCB compositions and sources by comparing samples and commercial mixtures (Aroclor 1016, 1242, 1248, 1254, 1260; Erickson, 2001). Homologue percent concentrations were used.

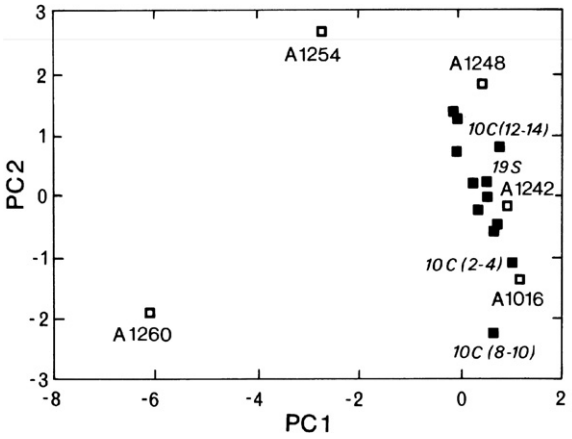

Fig. 4. Comparison of PCB compositions of both surficial and core sediments with those of commercial mixtures. Results of a PCA (see text for details).

The score plot obtained from the first two principal components (which together explain 88.3% of the total variance) is shown in Fig. 4. Most samples are grouped around Aroclor 1242 (where 3-CBs are the most abundant components, with a significant contribution of 4-CBs) and 1016 (with prevalence of 3-CBs and very little 5-CBs). In particular, this latter is very close to sample 10C (2–4 cm). Some samples show a similarity also with Aroclor 1248 (abundant 4-CBs with significant contributions of both 3-CBs and 5-CBs). The most evident exception is represented by sample 10C (8–10 cm) that has an absolute prevalence of 3-CBs over the other homologues. Minor differences are relative to samples 10C (12–14 cm), which is characterised by a slight prevalence of 4-CBs, and the surficial sample 19S, whose homologue profile shows similar abundances of 3-CBs and 4-CBs. These differences may be attributable either to shift in the sources or to a different fractionation during the transport to and within the lagoon. However, at least in the case of site 19S, the different grain size composition could play a role.

It would be important, at this stage, to obtain information on the activities responsible for the PCB contamination of the lagoon and its mainland, the transport from the different rivers and the mechanisms that drive particle and contaminant distribution and accumulation in sediments.

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