Polychlorinated Biphenyls in Sediments of the Tam Giang-Cau Hai Lagoon (Central Vietnam): First Results

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Introduction

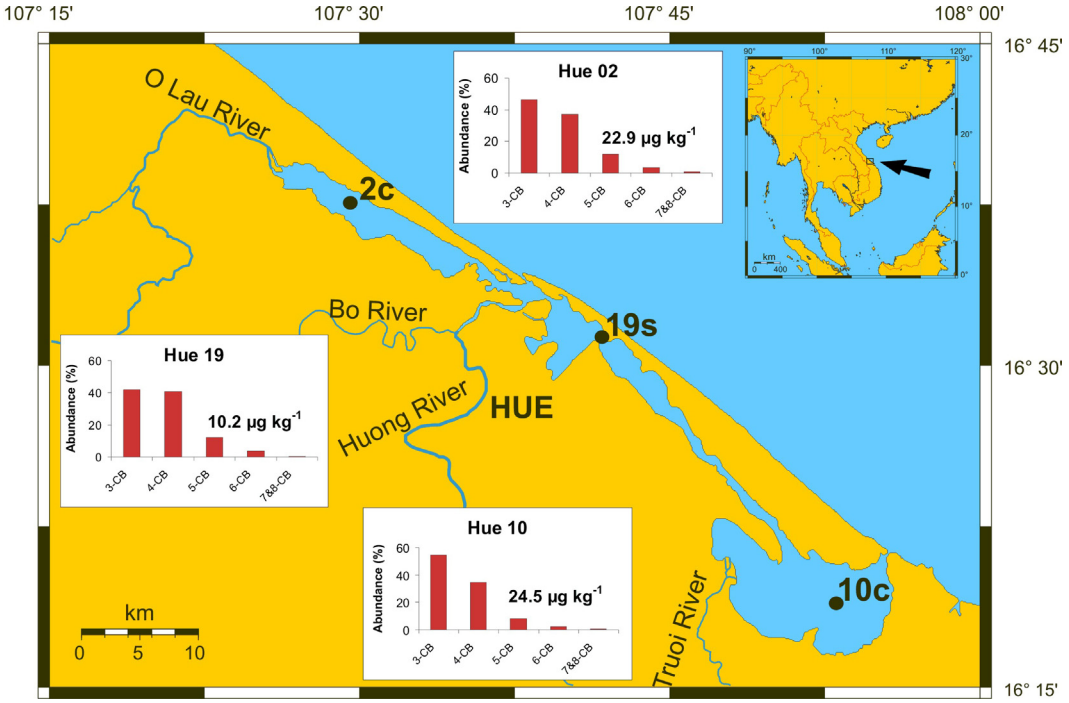
The Tam Giang-Cau Hai (TG-CH) Lagoon, a typical tropical coastal wetland, is the largest in southeast Asia, being 70 km long with a surface of 216 km^2. This system (Fig. 1) is quite complex because it is composed of four parts with different shapes and extension called Pha Tam Giang (27 km long, 0.6-3.5 km wide, up to 2m deep), Dam Sam-An Truyen (5x5.5 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.[1]. In particular, this environment is now facing many critical problems such as oil pollution, coliform contamination, eutrophication and presence of anthropogenic chemicals. This situation requires a management, capable to reverse the negative trend, that should be based on a sound scientific background. However, so far, the research-based knowledge on the key environmental processes and the levels, sources and trends of contamination is rather poor, especially regarding the most dangerous organic species. The purpose of this work was to provide a first insight on the presence of PCBs in lagoon sediments with a particular attention to the history and trends of the inputs.

Materials and Methods

Sediment samples were collected from the three basins of the TG-CH Lagoon in December 2002. A manual piston core was used to retrieve both surficial samples (02S and 19S, 4 and 2 cm thick, respectively) 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 2-3 cm, with higher resolution at the top. Sediment slabs were then divided

in two parts for the different analyses, put in polyethylene vessels and stored in a refrigerator at 0°C until the arrival at the lab. Afterwards, they were conserved at –18°C until the analyses.
For ^{137}Cs determinations lyophilised 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 a silver planchet. The ^{210}Po, considered in secular equilibrium with its parent ^{210}Pb, was alpha counted by a surficial barrier detector coupled with a multichannel analyser. Excess ^{210}Pb ($^{210}Pb_{ex}$) was calculated by subtracting the background value, i.e. the constant activity below the base of the profile, from the total.
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 solution, was dried by anhydrous Na_2SO_4 and reduced to 25 ml under a gentle stream of nitrogen. After sulfur removal by several treatment with 2 ml of mercury, the extracts were purified by solid-liquid chromatography (stationary phase Alumina Oxyde/Florisil, eluent n-exane) and then reduced to 0.5 ml. PCB analyses were carried out by HRGC-LRMS (mass spectrometer: HP 5973) following Moret et al.[2]. Five ^{13}C labeled PCBs were added to the samples before extraction, as internal standards. Crude concentration values were corrected with congener-specific instrumental response factors.

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.

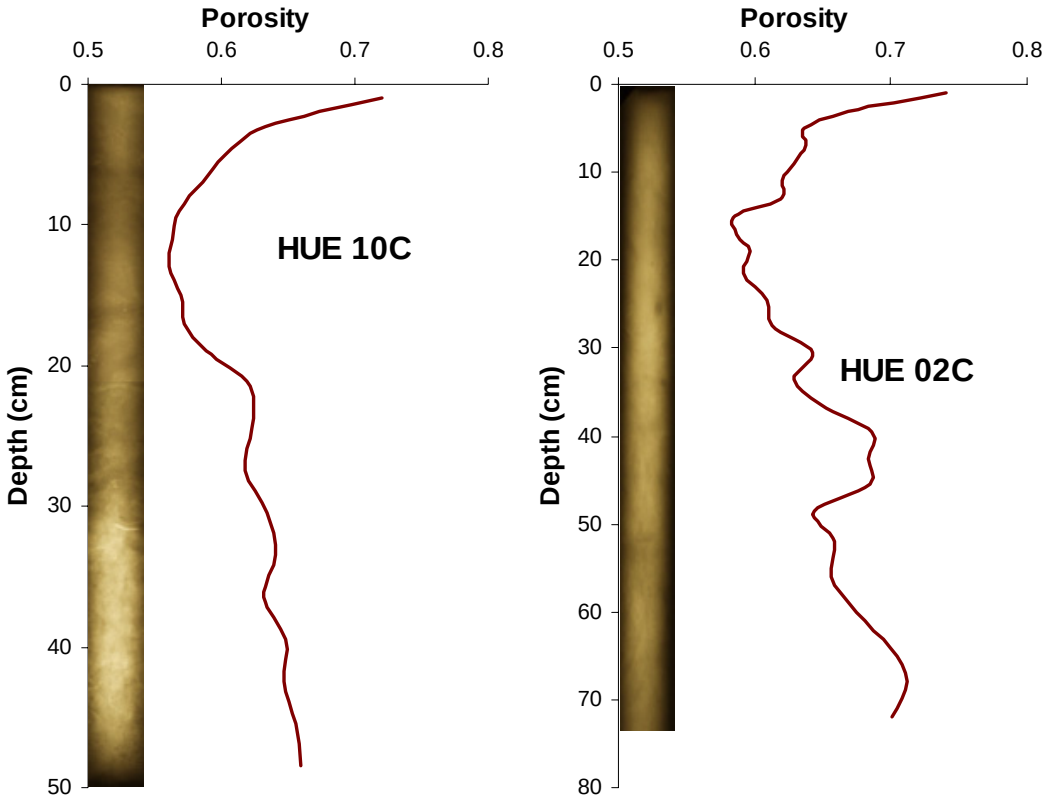


PCB concentrations were calculated as a sum of 54 congeners (45 chromatographic peaks). Accuracy was checked with respect to a certified standard (NIST®, Standard Reference Material 1941b). Precision, calculated using the same certified standard, are typically 10% on the sum of congeners and, for the homologues, span the interval 5-20%. PCB concentrations and radiotracer activities were calculated with respect to dry weight.

Results and Discussion

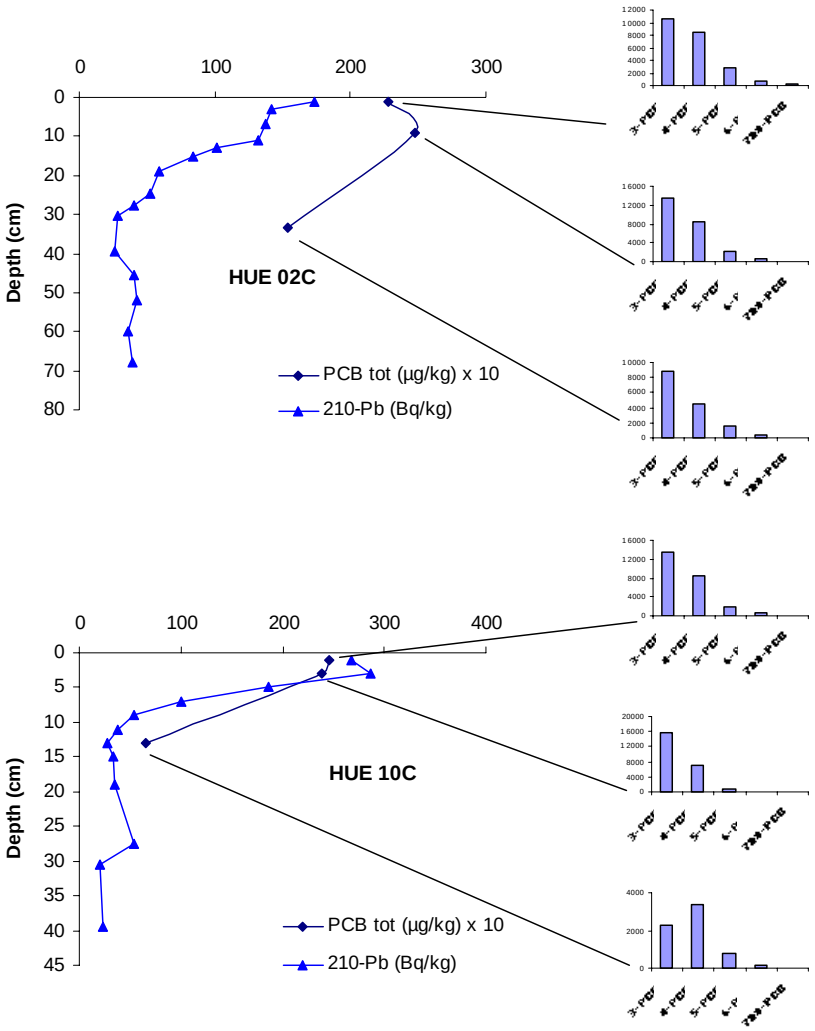
Sediment features: The X-radiographs of cores 02C and 10C are shown in Fig. 2, together with the depth distributions of sediment porosities. These sediments appear fairly homogeneous, with traces of bioturbation more evident at depth in 10C. According to the map of grain size distribution[3], 10C, 02C, and 19S are a fine silt, a silt, and a very fine sand, respectively. The patterns of porosities, first decreasing and then increasing downcore, account for environmental changes that are quite similar in the two basins, even though they seem more regular in core 10C. Tentative correlations suggest that the accumulation rate at 02C is higher than at 10C.

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



PCB surficial concentrations: PCB concentrations in surficial sediments, together with their homologue profiles, are shown in Fig. 1. Among these samples, the highest value characterizes site 10C (24.5 μg kg^{-1}), followed by 02S and 02C (18.1 and 22.9 μg kg^{-1}), and 19S (10.2 μg kg^{-1}).
This pattern probably depends on both the different inputs to the various parts of the lagoon and the relative mechanisms of dispersion and accumulation, hence it may be influenced by the relative importance of the river sources and the sediment grain size. In particular the southern basin receives the contribution of both the Truoi River and the Dai Giang River, whereas the northern part is alimented by the O Lau River. In turn, the central lagoon is fed by the Huong river and tributary Bo River. 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.

Fig. 3. Activity-depth profiles of $^{210}Pb_{ex}$ and depth distributions of PCBs in cores 02C and 10C. The homologue profiles of PCBs are also shown.



In turn, the northern and the southern basins should act as sediment traps where much of the suspended load settles to the bottom before going out to the sea. On the other hand, the homologue profiles of Fig. 1 suggest that PCBs in surficial sediments have a similar composition, especially 02C and 10C, thus suggesting a unique type of source, widely diffused in the mainland.

The concentrations are comparable with those reported by Nhan et al.[4] for the northern coast of Vietnam (0.5-28.1 µg kg^{-1}) and lower than the values found by Iwata et al.[5] in Vietnamese cities.

The two surficial samples taken from site 02, a surficial sediment and the top of core 02C, provided similar results, in spite of their different thickness: 4 and 2 cm, respectively.

PCB depth distributions: Figure 3 shows the concentration-depth profiles of PCBs in sediments of sites 2 and 10. In both cores the concentrations are maximum in the surface (10C) or near-surface (02C) sediments and decrease downwards. The decline is more fast and significant at 10C, where the concentration at 13 cm depth is 3.7 times lower than at the surface. On the contrary, in core 02C the concentration at 35 cm depth has decreased only ca. 33 % with respect to the peak value.

Chronology: Activities of ^{137}Cs in lagoon sediments are very low, close or below detection limits. Because of this, inferences on sediment chronologies can be obtained only from the activity-depth profiles of $^{210}Pb_{ex}$ (Fig. 3). Therefore, apparent accumulation rates were calculated using $^{210}Pb_{ex}$ depth distributions in cores 02C and 10C through the linear regression of the log normal activities of the decreasing portion of the curve (CF-CS model). In this way, rates of 0.36 and 0.10 cm y^{-1} could be calculated for sites 02C and 10C, respectively. Since we did not account for the role of bioturbation and physical disturbances in modifying the profile, these rates should be considered as apparent and probably overestimated. Furthermore, such values appear rather low and bring about a chronology that does not match with the presence of PCBs, first commercialized in the early 1930s, at depth in the two cores. It is possible that the CF-CS model, which assumes a constant flux of $^{210}Pb_{ex}$ and a constant sedimentation, is inadequate to represent the actual mechanism of accumulation within the TG-CH lagoon. We should consider significant also the penetration of $^{210}Pb_{ex}$ below the tract of the curve used for the model. This means that the presence of both $^{210}Pb_{ex}$ and PCBs below 29-32 and 13-14 cm depth in cores 02C and 10C, respectively, can be due either to a mechanism of bioturbation or to variations of the rate of accumulation of $^{210}Pb_{ex}$ and/or sediment particles. In any case, the $^{210}Pb_{ex}$ activity-depth profile alone is not suitable to obtain a chronology of the cores.

Sources: The homologue composition of PCBs in both surficial and deep samples is characterized by the prevalence of three-chlorinated biphenyls. A Principal Component Analysis (PCA) on the homologue concentrations suggests a similarity of the patterns with that of Aroclor 1242 (in that it has a prevalence of 3-CB and a significant contribution of 5-CB) and 1016 (which has a great prevalence of 3-CB but very little 5-CB). The only two exceptions are the sample at 13 cm depth in core 10C, which is characterised by a slight prevalence of 4-CB, and the surficial sample 19S, whose homologue profile shows similar abundances of 3-CB and 4-CB. These differences may be attributable either to shift in the sources or, at least in the case of site 19S, to the slightly different grain size. It is important, at this stage, to obtain information on the activities responsible for the PCB contamination of the TG-CH lagoon and its mainland, and better understand the processes that drive particle and contaminant distribution and accumulation in sediments.

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