

The fate of fluvial PAHs in the Kao-ping River and Kao-ping Submarine Canyon (KPRSC) System

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計劃名稱：高屏河－海輸運系統中物質之形成、傳輸、變化和沉積的整合研究
子計畫二：高屏河－海系統中多環芳香烴化合物之來源和宿命研究

Abstract

Polycyclic Aromatic Hydrocarbons (PAHs) are ambiguous pollutants. Their sources in coastal areas can be differentiated into three categories: petrogenic, pyrogenic and biogenic, based upon diagnostic ratios or compositional distributions. PAHs tend to adsorb onto particles in aquatic environment regarding to their hydrophobicity and could be treated as source tracers of particles. Kaoping River, located in the southern part of Taiwan, has a drainage basin of about 3,300 km². Characterized as a mountainous river, Kaoping River discharges about 49 Mt of sediments annually. Located at one km seaward of Kaoping River mouth, Kaoping submarine canyon acts as a trap and conduit for both seaward and landward transport of sediment. Kaoping submarine canyon terminates in the northern part of South China Sea and plays an important role in transporting terrestrial substance to deep sea. In this study, surface sediment samples collected from Kaoping coast to off-shore region (about 2000 m deep) were analyzed for PAHs to probe possible PAH sources. In addition, down-core PAH profiles of a sediment core collected in down-canyon area was used to re-construct the historical record of PAHs contamination.

Sampling sites were roughly separated into two groups, coastal and off-shore sites, based on a virtually extended tangent line on the southern border of Liu-chiu Island (Figure 1). Total PAH concentrations of coastal and off-shore sediment samples ranged between 33.0~910 ng/g dw with an average concentrations of 270±150 ng/g dw (n=33). The highest concentration was found at station D2 (910 ng/g) which is two to thirty times of those of other stations. Total PAH concentrations correlated well with TOC and with percentages of <63% particles suggested that fine particles tend to adsorb organic carbon and therefore hydrophobic organic pollutants like PAHs (Figure 2). The outlier, D2, having highest total PAH concentration indicated a hot spot in sampling area (Figure 2). As shown in Figure 3, greater variations in total PAH concentrations were found at coastal sites than at off-shore sites. The reason for this observation is that in coastal area fine particles having higher PAH concentrations were diluted by large particles which can hardly be transported to off-shore area.

To differentiate PAH sources in the sediments, four isomer ratios were used in Figure 4a & b. For those sediments from coastal sites except F2 and D2, their isomer ratios suggest PAH sources of combustion of fossil fuel. Station F2 and D2, located at the northern shelf of Kaoping canyon, are recognized as grass, wood and coal combustion. The only difference of PAH source between coastal and off-shore sediments was the intrusion of PAH derived from grass, wood and coal combustion to

off-shore sediments which was suggested based on IP/IP+BghiP ratios. PCA result showed that sediments from Kaohsiung shelf (D2, D4, F2) exhibit higher PC1 scores (high positive loadings of fluoranthene and pyrene) suggest significant contribution of coal combustion (Larsen and Baker 2003; Fang et al. 2007). Higher PC2 scores (heavily weighted by perylene and indeno[1,2,3-c,d]pyrene) of off-shore sites attributed from diagenetic processes organic substances in marine environment and partially from atmospheric-deposited pyrolysis-origin sources.

Figures 6 & 7 showed cumulative sediment mass versus excess lead-210. (Huh et al. 2007) and down-core concentrations of total 16 PAHs in sediment core K38 (water depth about 1000 m). Along this core, total 16 PAH concentrations showed a trend of slightly increase after 1930s with an abrupt decrease peak on the top. This decrease of total 16 PAH concentrations was believed to be influenced by an episodic event, Haitang Typhoon (Huh et al. 2007). The intrusion of terrestrial substances was more clearly demonstrated by the concentration profile of a representative pyrolysis-origin PAH compound, indeno[1,2,3-c,d]pyrene (Figure 8). Indeno[1,2,3-c,d]pyrene is mainly from pyrogenic sources such as vehicle emissions. Gradual increase of indeno[1,2,3-c,d]pyrene after 1930s indicates an increasing usage of fossil fuel such as coal and/or petroleum which was also observed in literature (Guo et al. 2006). Figures 9 & 10 indicate that the PAHs in the samples are mainly from the petroleum combustion after 1900s with an obvious increase after 1940s. Similar results were also found in Pearl River Delta close to South China Sea? (Mai et al. 2003). From their results, petroleum combustion signals revealed from PAH isomer ratios in deposited sediment reflect the drastic increase in the consumption of petroleum products in China for the past decades. These observations lead to a conclusion that atmospheric deposition might be the main contribution of PAHs to this sedimentation setting with episodic intrusion.

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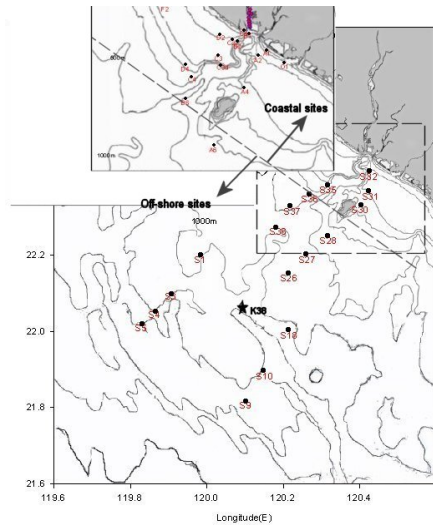


Figure 1. Map of sampling stations (ORIII-940 and ORI-732). Core sample K-38 is marked by asterisk

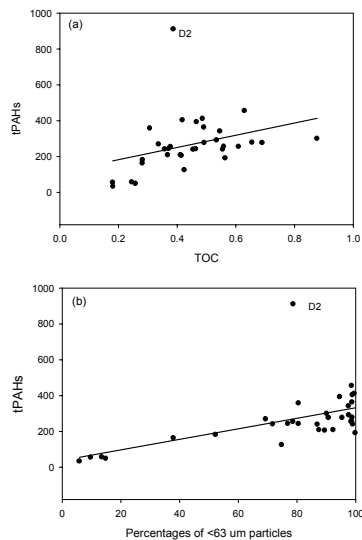


Figure 2. Correlations of total PAH concentrations versus (a) TOC and (b) portions of <63 um particles

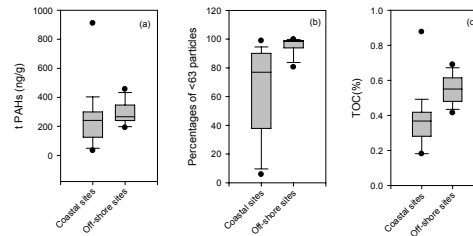


Figure 3. Box-plots of total PAH concentrations, TOC and portions of <63 um particles

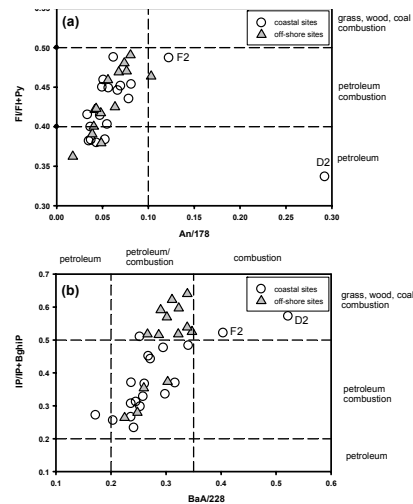


Figure 4. PAH isomer ratios of coastal and off-shore sediments (a) An/178 vs. Fl/F1+Py, (b) BaA/228 vs. IP/Ip+BghiP

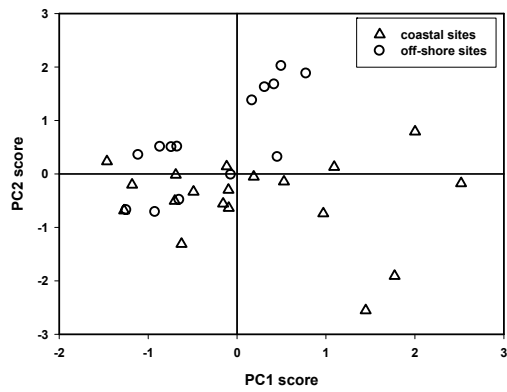


Figure 5. PC1 versus PC2 score plot of sediment samples from coastal and off-shore sites

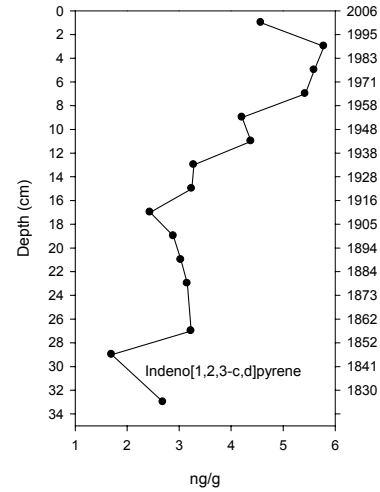


Figure 8. Concentrations profile of indeno(1,2,3-c,d)pyrene in sediment core K-38.

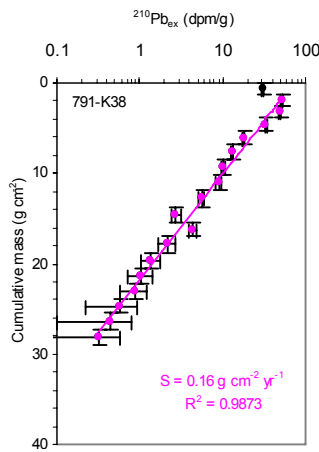


Figure 6. Cumulative sediment mass of core K-38 plotted versus excess lead-210. (Huh et al. 2007)

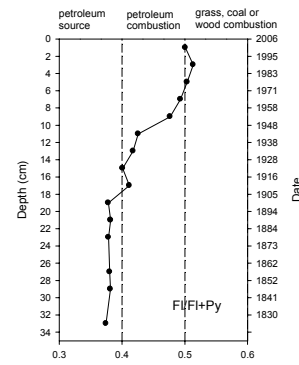


Figure 9. Down core profile of isomer ratios fluoranthene/ fluoranthene+pyrene in sediment core K-38.

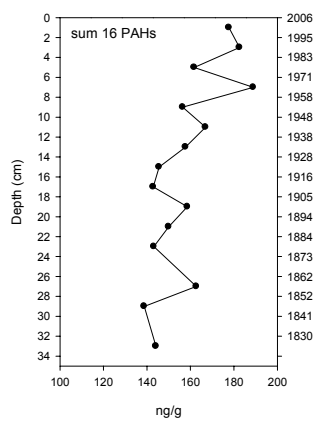


Figure 7. Concentrations profile of 16 PAHs in sediment core K-38.

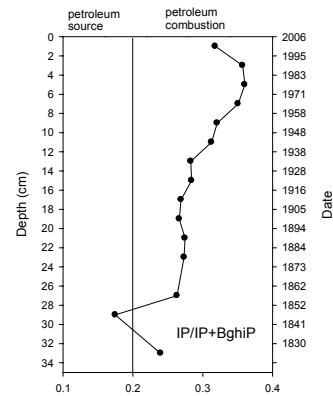


Figure 10. Down core profile of isomer ratios indeno [1,2,3-c,d]pyrene / indeno [1,2,3-c,d]pyrene + benzo(ghi)perylene in sediment core K-38.