Marine Pollution Bulletin 58 (2009) 752–760

Contents lists available at ScienceDirect

Marine Pollution Bulletin

journal homepage: www.elsevier.com/locate/marpolbul

Polycyclic aromatic hydrocarbons in coastal sediments of southwest Taiwan: An appraisal of diagnostic ratios in source recognition

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ARTICLE INFO

Keywords:
PAHs
Sediment
Diagnostic ratio
Source recognition
Taiwan

ABSTRACT

Fifty-seven surface sediment samples were collected from the coast of southwest Taiwan and analyzed for polycyclic aromatic hydrocarbons (PAHs). Concentrations of total PAHs (28 PAH compounds) ranged from 15 to 907 ng g^{-1} dry weight. Diagnostic ratios showed that PAHs in the sediments of the Gaoping estuary were predominantly of petroleum origin, whereas sediments from the Kaohsiung coast contained principally combustion-derived PAHs. Principal component analysis indicated that emissions from automobiles and coal burning were the main sources of combustion-derived PAHs. The relatively high ratios of perylene/penta-aromatic PAH isomers in sediments from the Tainan coast and some off-shore stations on the Kaohsiung coast suggest a significant diagenetic PAH contribution. The study shows that certain diagnostic ratios are useful and sensitive in delineating the distribution of PAHs from specific sources in southwest Taiwan. The phenanthrene/anthracene ratio is a better indicator than the methylphenanthrenes/phenanthrene ratio for tracing petrogenic PAHs, and the benzo(*a*)anthracene/chrysene and indeno(1,2,3-*c*,*d*)pyrene/benzo(*g*,*h*,*i*)perylene ratios are more specific than the benzo(*a*)pyrene/benzo(*e*)-pyrene and benzo(*b*)fluoranthcene/benzo(*k*)fluoranthcene ratios in distinguishing PAHs from various pyrogenic sources.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are one of the largest groups of particle-reactive hydrophobic organic chemicals in coastal water. Because of their hydrophobic nature and their usefulness in source identification, the study of these tracer compounds in coastal sediments provides a good understanding of their land-to-sea transfer (Fang et al., in press), and also gives an integrated picture of their transport in the marine environment. Additionally, the bioavailability of these sediment-bound compounds, which are readily enhanced in food webs by resuspension or bioturbation of sediment into the water column, makes them an important area of study. Some PAHs including benzo(*a*)anthracene, benzo(*a*)pyrene, dibenzo(*a*,*h*)anthracene, and naphthalene have been demonstrated to be carcinogenic to mammals (IARC, 1991; USEPA, 1991). Therefore, the importance of studying PAHs in coastal sediments also relates to their bioaccumulation in shellfish and other benthic organisms, and possible entry to human food chains by this route.

In general, sources of PAHs in the marine environment can be classified into three main groups: petrogenic, pyrogenic, and diagenetic. Pyrogenic PAHs, which are common in aquatic environments, come from such things as natural fires as well as anthropogenic sources including waste incineration and combustion of fossil fuels, and give rise to complex mixtures of PAHs characterized by high molecular weight parental PAHs (Wang et al., 1999; Zeng and Vista, 1997). Petrogenic PAHs are derived from petroleum hydrocarbons, and their presence can be attributed to petroleum transportation/spills, off-shore oil exploitation, and natural oil seeps. The PAH composition of petroleum hydrocarbons is complex, and is characterized by a predominance of low molecular weight and alkylated PAHs (Lake et al., 1979; Laflamme and Hites, 1978). Diagenetic PAHs, such as perylene, are derived from biogenic precursors via short-term diagenetic processes (Venkatesan, 1988). Certain isomer ratios, such as phenanthrene/anthracene (Ph/An), methylphenanthrene/phenanthrene (MeP/Ph), and fluoranthene/pyrene (Flt/Py), have been widely used to identify sources of PAHs (Baumard et al., 1998; Budzinski et al., 1997). Some parent PAH diagnostic ratios including anthracene/anthracene + phenanthrene (An/178),

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0025-326X/$ - see front matter © 2008 Elsevier Ltd. All rights reserved.
doi:10.1016/j.marpolbul.2008.12.017

fluoranthene/fluoranthene + pyrene (Flt/Flt + Py), benz(*a*)anthracene/benz(*a*)anthracene + chrysene (BaA/228) and indeno(1,2,3-*c*,*d*)-pyrene/indeno(1,2,3-*c*,*d*)-pyrene + benzo(*g*,*h*,*i*)perylene (IP/IP + BghiP), have recently proven useful in PAH source identification (Fang et al., 2007; Sprovieri et al., 2007; Guo et al., 2006; Lee et al., 2005; Yunker et al., 2002). In addition, diagnostic ratios of benzo(*a*)anthracene/chrysene (BaA/Chry), benzo(*b*)fluoranthcene/benzo(*k*)fluoranthcene (BbFa/BkFa), benzo(*a*)pyrene/benzo(*e*)pyrene (BaPy/BePy) and indeno(1,2,3-*c*,*d*)pyrene/benzo(*g*,*h*,*i*)perylene (IP/BghiP), have been used to evaluate the relative contribution of specific types of combustion, such as vehicle exhaust, coal/coke combustion, forest fires, and smelters (Dickhut et al., 2000).

The coastal waters off southwest Taiwan receive contaminants from a wide range of anthropogenic activities. The densely populated cities of Kaohsiung (the largest industrial city in Taiwan; population 1.5 million including Kaohsiung Harbor and Zuoying Navy Base) and Tainan (population 1.1 million including Anping Harbor, and Sinda Harbor) are located in this area. Intensive maritime activities in the area may be responsible for the accumulation of significant amounts of petrogenic pollutants. The Kaohsiung coast also receives wastewater from three ocean outfalls (Zuoying, Jhongjhou, and Dalinpu), two of which receive effluent from several industrial parks (with petrochemical, steel, shipbuilding, and electro-chemical industries) and the Southern Taiwan Science Park (Yang, 1995). A significant addition of industrial waters to the Gaoping estuary from the Linyuan industrial park, via the Gaoping River and the Dalinpu ocean outfall pipe, is also expected. In addition, two coal-fired power plants, Sinda and Dalin, and several refuse incineration plants, probably also contribute contaminants to nearby waters through atmospheric deposition. Therefore, the anthropogenic pollution in this area is expected to be complex, and needs to be examined. However, only in Sinda Harbor and the estuary of the Gaoping River have the distributions of PAHs been studied and local sources delineated (Fang et al., 2007, 2003).

In this study we investigated diagnostic PAH ratios and the PAH pollution topography at a relatively large spatial scale in the

Table 1
PAHs analyzed in this study.

PAH compounds	Abbreviation
Fluorene	Fluo
Phenanthrene	Ph
Anthracene	An
1-Mefluorene	1MeF
Dibenzothiophene	DBT
4,5-Methylenephenanthrene	45MeP
3-Methylphenanthrene	3MeP
2-Methylphenanthrene	2MeP
2-Methylanthracene	2MeA
9/4-Methylphenanthrene	4 + 9MeP
1-Methylphenanthrene	1MeP
Fluoranthene	Flt
Pyrene	Py
4,6-Dimethyl-dibenzothiophene	46DMD
3,6-Dimethylphenanthrene	36DMP
Benzo[*a*]fluorene	BaFl
Benzo[*b*]fluorene	BbFl
Benzo[*a*]anthracene	BaA
Chrysene + triphenylene	Chry+TriPhe
benzo(*b*)fluoranthene	BbFa
Benzo[*k*]fluoranthene	BkFa
Benzo[*e*]pyrene	BePy
Benzo[*a*]pyrene	BaPy
Perylene	Pery
Indeno[1,2,3-*c*,*d*]pyrene	IP
Benzo[*g*,*h*,*i*]perylene	BghiP
Dibenz[*a*,*h* + *ac*]anthracene	DahacA
Coronene	Coro

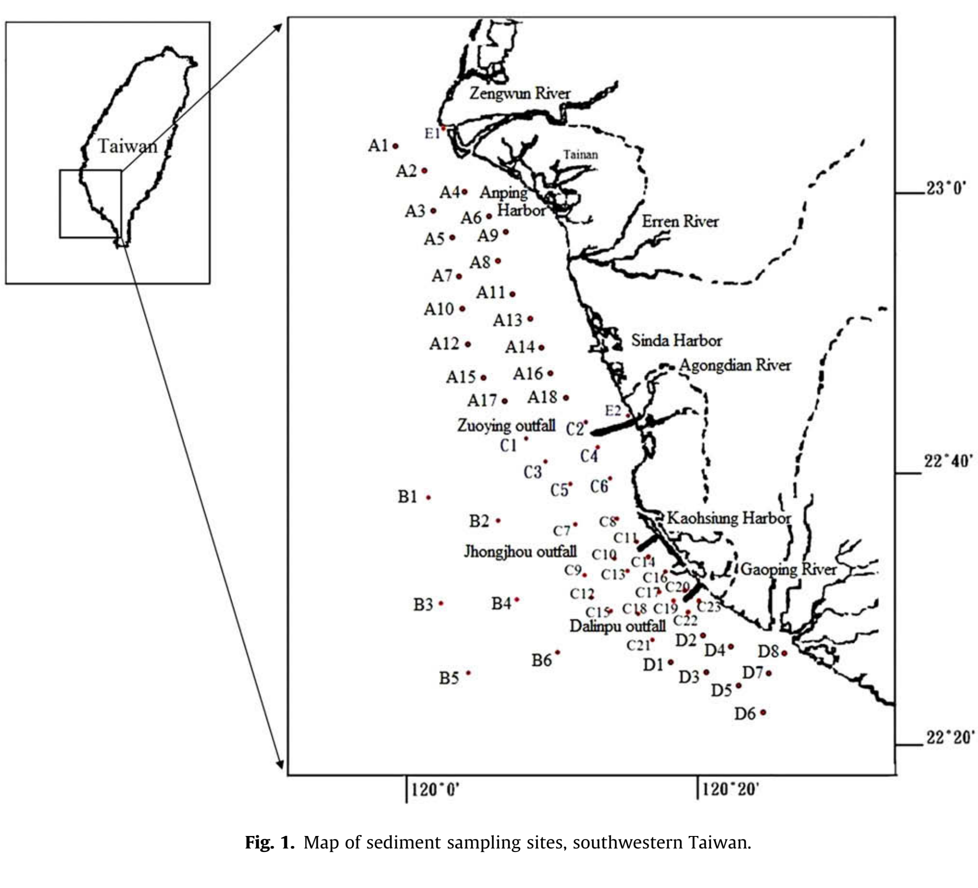

Fig. 1. Map of sediment sampling sites, southwestern Taiwan.

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