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Spatial and vertical distributions of sedimentary halogenated polycyclic aromatic hydrocarbons in moderately polluted areas of Asia

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ABSTRACT

The sedimentary halogenated (chlorinated and brominated) polycyclic aromatic hydrocarbons (Cl/BrPAHs), PAHs, and elements were analyzed to investigate contamination processes and sources. Assessments were conducted in sediments from three sites: surface sediments from the Yellow Sea and sediment cores from Kandy Lake and Negombo Lagoon, Sri Lanka. Most of ClPAHs targeted were detected in all sediments. Spatial distributions of total ClPAH concentrations in the Yellow Sea showed the presence of multiple hot spots that differed from those of total PAHs. In Kandy and Negombo sediments, total ClPAH concentrations were slightly higher in surface layers than in bottom layers; the opposite trend was observed for PAHs. Principal component analysis showed that the clusters of most ClPAHs were similar to those of anthropogenically derived elements, but were far from those of PAHs. Consequently, ClPAHs in sediments appear to be persistent contaminants, which may make them appropriate as indicators of anthropogenic sources.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are of great environmental concern because of their potential toxicity; for example, they may have mutagenic, carcinogenic, and endocrine disrupting properties (Baek et al., 1991; Bostrom et al., 2002; Ball and Truskewycz, 2013). Because most biologically toxic PAHs are hydrophobic and non-biodegradable, they are highly persistent in the environment (Meador et al., 1995; Wild and Jones, 1995). The occurrence of PAHs in aquatic environments is affected by inputs from the surrounding environment, including from atmospheric deposition, riverine inputs, municipal runoff, and accidental oil spills (Arzayus et al., 2001; Lima et al., 2005; Boonyatumanond et al., 2006). PAHs have also been known to be produced by incomplete combustion processes of organic materials and fossil fuels; PAHs are therefore frequently used as a marker to evaluate anthropogenic pollution (Mastral and Callén, 2000; Ravindra et al., 2008; Kim et al., 2013). Highly populated regions with a high amount of anthropogenic activity are likely to be highly polluted with PAHs, and the extent of PAH pollution reflects the history of pollution associated with anthropogenic activities.

Halogenated PAHs, e.g., chlorinated PAHs (ClPAHs) and brominated PAHs (BrPAHs), are currently of interest in the study of PAH derivatives as new classes of environmental contaminants (Ohura, 2007; Sun et al., 2013). Studies on atmospheric ClPAHs have been conducted by a number of researchers; these studies have produced knowledge about the specific behavior of ClPAHs. For example, concentrations of relatively high-molecular-weight ClPAHs, i.e. particulate ClPAHs, in the air are significantly correlated with corresponding parent PAH concentrations (Ohura et al., 2008, 2013a). This finding indicates that they are produced from common emission sources. However, there are few reports on the occurrence of ClPAHs in sediments (Horii et al., 2009; Sun et al., 2011). Horii et al. (2009) reported that all target ClPAHs were detected in sediments from industrialized areas, total levels of which had a range of 580–8800 pg/g. The profiles of individual

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ClPAHs in sediments were quite different between sediment samples, while sedimentary PAH profiles were similar between samples. This suggests that differences in profiles reflect differences in emission sources.

The Yellow Sea contains semi-enclosed areas, the continental shelf, and shallow seas, and is surrounded by various industrial cities in China and Korea. The rapid industrialization and population increase in the coastal area has caused an increase in the inflow of hazardous compounds to the Yellow Sea, resulting in sedimentation and an increase in ecological risk. Indeed, historical surveys of PAHs and elements in sediment cores from the Yellow Sea reflect the economic development in China and the corresponding increase in consumption of fossil resources (Wu et al., 2001; Hu et al., 2008; Zhang et al., 2009; Li et al., 2012). Sri Lanka has also undergone rapid industrialization since the 1980s; as a result, a number of critical environmental problems and issues have recently come about. In particular, Sri Lanka has experienced increases in air pollution resulting from an increase in vehicle emissions (Pathiratne et al., 2007). The concentrations of PAHs in sediments from Beira Lake in downtown Colombo, the capital of Sri Lanka, were in the range of 152–569 ng/g with an average of 329 ng/g (Pathiratne et al., 2007). The mean concentration was somewhat consistent with those of the Yellow Sea (280 ng/g) (Hu et al., 2008), Masan Bay, Korea (353 ng/g) (Khim et al., 1999), the Yalujiang River, China (290 ng/g) (Wu et al., 2003), and Lake Manzala, Egypt (363 ng/g on median concentration) (Barakat et al., 2012); these levels could be classified as moderately contaminated compared with San Francisco Bay, USA (2653–27,680 ng/g) (Pereira et al., 1996), and Pialassa Baiona, Italy (3032–87,150 ng/g) (Guerra, 2012).

The aims of this study were to evaluate the spatial and vertical distributions of ClPAHs, BrPAHs, PAHs, and elements in sediments collected from the Yellow Sea and from two water bodies in Sri Lanka. To our knowledge, this is the first study in which these contaminants were investigated simultaneously in sediments. Furthermore, we evaluated the sedimentary behavior, potential sources, and ecological risks of ClPAHs in comparison with those of PAHs and elements, and discuss the specificity in the sediments.

2. Material and methods

2.1. Sample collection

Sixteen surface sediment samples in the Yellow Sea were collected from the Korean ocean dumping site which received the sewage and waste sludge. The top 2 cm of surface sediments were collected from 77 to 82 m below the surface of the Yellow Sea in September, 2004 using a van Veen grab sampler. The sampling sites were located near the center of the Yellow Sea (approximately 150 km off the coast of South Korea), and were distributed at regular intervals (Fig. 1). Two sediment cores were collected in 2004 using a HR columnar type bottom sampler (length 50 cm, inner diameter 11 cm; Rigo Co. Ltd., Saitama, Japan) from each sampling site in Sri Lanka: one site was located approximately 1 km inside the inlet of Negombo Lagoon, and the other was located at the center of lake in Kandy (Kandy Lake) (Fig. 1). Detailed information of the sampling sites was listed in Table S1. The Negombo Lagoon is a shallow estuarine lagoon linked to the sea by a narrow channel near the city of Negombo, on the West Coast of Sri Lanka. Kandy Lake is a freshwater man-made lake located in the city of Kandy,

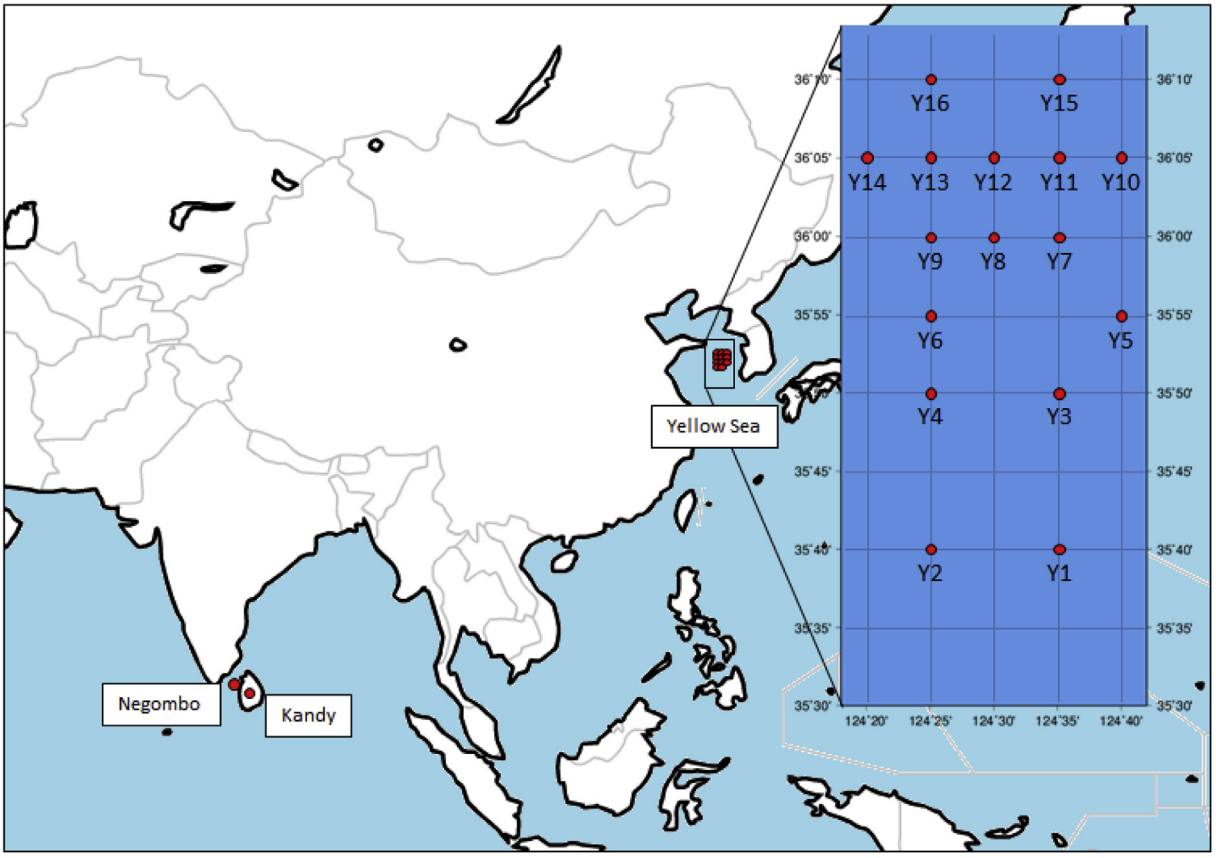

Fig. 1. Sampling sites in the study area.