



## Baseline

## Restoration and source identification of polycyclic aromatic hydrocarbons after the Wu Yi San oil spill, Korea



Yu Lee Jang<sup>a</sup>, Hyo Jin Lee<sup>a</sup>, He Jin Jeong<sup>a</sup>, Shin Yeong Park<sup>a</sup>, Won Ho Yang<sup>a</sup>, Heung-Yun Kim<sup>b</sup>, Gi Beum Kim<sup>a,\*</sup>

<sup>a</sup> Department of Marine Environmental Engineering, Gyeongsang National University, Republic of Korea

<sup>b</sup> Department of Aquatic Medicine, Chonnam National University, Republic of Korea

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## ABSTRACT

On January 31, 2014, an oil spill accident occurred in Yeosu, South Korea. A total 800–899 kl of oil from the pipeline was spilled into the sea. After the oil spill, the KIOST (Korea Institute of Ocean Science & Technology) researched PAHs (polycyclic aromatic hydrocarbons) in various media, but sedimentary PAHs were not analyzed despite their longer persistency than in other media. Therefore, this study examined PAH levels in intertidal sediments around Gwangyang Bay and identified PAH sources using oil fingerprinting. PAH residual levels showed a dramatic decrease during the four months after the accident and then remained at a relatively constant level. Analysis through regression equations indicate that this study area is likely to be restored to the PAH levels prior to the accident. Furthermore, the source analysis and oil fingerprinting analysis showed that PAH contamination in this study was unlikely to have originated from the spilled oil.

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On January 31, 2014, the oil tanker Wu Yi San collided with a pipeline of crude oil from a petrochemical plant in Yeosu, South Korea. According to a statement from South Korea's Coast Guard, a total 800–899 kl of oil was spilled into the sea and about 9.5 km of Gwangyang Bay was heavily affected by the spilled oil (KCG, 2015).

Polycyclic aromatic hydrocarbons (PAHs) are organic compounds with two or more fused aromatic rings. Although they represent <2% of an oil, these compounds are well known to be toxic substances (Randel et al., 2010). For example, low molecular weight PAHs may pose acute toxicity effects, while high molecular weight PAHs are genotoxic and carcinogenic to marine organisms (Varanasi, 1989; Saunders et al., 2002; White, 2002). Also, they are persistent in various environmental media for long periods of time, particularly in organisms and sediments (Hong et al., 2014; Nakata et al., 2014). Mackay et al. (2005) reported that the half-life of PAHs in marine sediments was 30 times higher than in seawater. Furthermore, residual PAHs in sediment may pose potential risks to marine organisms by the release of PAHs from sediment into the water (Peterson et al., 2003; Short et al., 2006; Michel et al., 2009). Therefore, it is essential to research PAHs in the surrounding coastal sediments when an oil spill accident occurs.

After the Wu Yi San oil spill accident, the Korea Institute of Ocean Science & Technology conducted researches of TPHs (total petroleum hydrocarbons) and PAHs in seawater, porewater, and organisms

(MOF, 2014). However, sedimentary PAHs were not analyzed despite their longer persistence compared to PAHs in seawater and marine organisms. Thus, the objective of this study was to examine PAH levels in intertidal sediments around Gwangyang Bay and to elucidate whether the PAH pollution was present in the surveyed area and what the source was using PAH source analysis and oil fingerprinting analysis.

Sediment samples were collected at 38 sites around the area of the oil spill and an additional two sites (sites 39 and 40) were selected as controls (Fig. 1). Surface sediments were collected at three-month intervals from May 2014 to May 2015. Approximately, the top 2 cm of sediment was taken using a stainless steel spatula and stored in a pre-combusted amber glass jar. The samples were immediately frozen in dry ice and stored in the freezer at  $-20^{\circ}\text{C}$  before laboratory analysis.

20 g of homogenized samples was mixed with 50 g of anhydrous sodium sulfate and extracted by soxhlet for 16 h using 200 ml of methylene chloride. Before the extraction, samples were spiked with 300 ng of deuterated surrogates (naphthalene- $d_8$ , acenaphthene- $d_{10}$ , phenanthrene- $d_{10}$ , chrysene- $d_{12}$ , and perylene- $d_{12}$ ) for examining the recovery. The extracts were purified with an alumina/silica gel column (300 mm  $\times$  10 mm i.d.). Eluted samples were concentrated, exchanged to hexane, and then re-concentrated to 0.5 ml for instrumental analysis. Before GC/MS analysis, samples were spiked with 300 ng of terphenyl- $d_{14}$  as the GC internal standard.

The PAHs were analyzed using an Agilent Model 7890 GC coupled with an Agilent Model 5975 MS. A DB-5MS capillary column (30 m  $\times$  0.25 mm i.d., 0.25  $\mu\text{m}$  thickness, J&W Scientific) was used and

\* Corresponding author.

E-mail address: [kgb@gnu.ac.kr](mailto:kgb@gnu.ac.kr) (G.B. Kim).

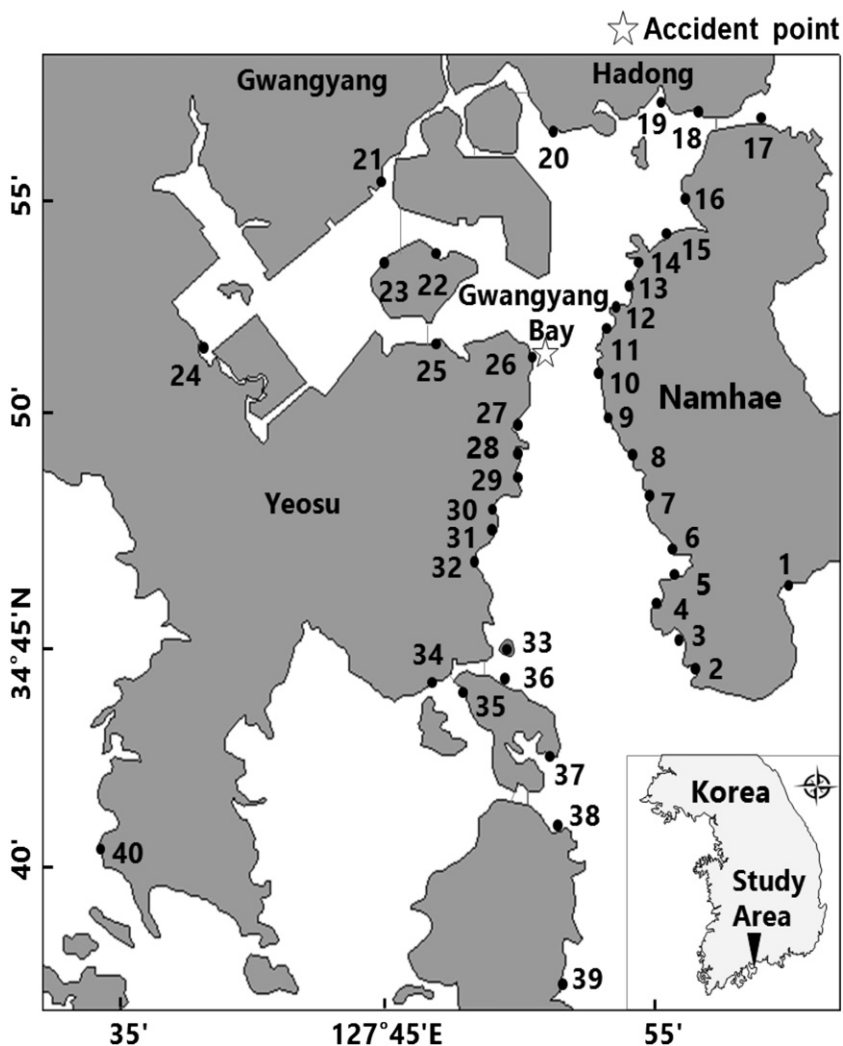


Fig. 1. Location map of sampling sites.

helium, the carrier gas, passed the column at the rate of 1 ml/min. The temperature program of the GC oven was as follows: initial temperature 2 min at 60 °C; first rate 6 °C/min to 300 °C; isothermal pause 13 min at 300 °C.

Sixteen US EPA priority PAHs were analyzed in this study: naphthalene, acenaphthylene, acenaphthene, fluorene, naphenanthrene, anthracene, fluoranthene, pyrene, benzo[*a*]anthracene, chrysene, benzo[*b*]fluoranthene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, indeno(1,2,3-*cd*)pyrene, dibenzo(*a, h*)anthracene, and benzo[*ghi*]perylene. And some of the alkylated PAHs that were found in high concentrations in the oil, 2-methylnaphthalene, 1-methylnaphthalene, 1,2-dimethylnaphthalene, 1,2,3-trimethylnaphthalene, 1-methyl fluorene, 1-methyl phenanthrene, 4-methyl dibenzothiophene and 1-methyl chrysene, were also analyzed to identify the input sources. After the alkyl PAHs (C1–C4-Na, C1–C3-Flu, C1–C4-Phe, C1–C3-DBA, and C1–C3-Chr) were identified by matching the retention times and the mass/charge of ion fragments with those obtained from alkyl PAH standard compounds, alkyl PAHs were calculated based on the integration of the peak area for individual homologues using the Agilent MSD Chemstation data analysis application.

A blank was used for each batch of eight samples to check for interfering substances in the analytical procedure. The surrogate standard recoveries ranged from 53.1–103% in all samples, which satisfied the acceptable range (40–120%). The method detection limits of 16PAHs and alkyl PAHs ranged from 0.001 (anthracene)–6.012 (pyrene) and 0.007 (C1-chrysene)–4.364 (C3-naphthalene) ng/g dry weight basis.

Fig. 2 presents 16PAH and alkyl PAH concentrations at 40 sites during five surveys. Concentrations of the 16PAHs and alkyl PAHs ranged from 216 (Aug 2014) to 137 (Feb 2015) ng/g (mean: 178 ng/g dw) and from 134 (May 2014) to 101 (Nov 2014) mg/g (mean: 119 ng/g dw), respectively. A positive correlation ( $p < 0.01$ ) between grain size and total PAH concentrations was obtained in sediment samples. There was no difference in PAH concentrations and also no temporal trend from control sites. Overall, relatively high PAH concentrations were found at site 34 in southern Yeosu in all surveys. Sixteen PAHs (489–2089 ng/g) were composed of 67% of total PAHs (727–3121 ng/g). As the 16PAH composition of Basrah light crude oil, known as spilled oil, was extremely low (0.3%), and the evaporation amount of volatile PAHs like naphthalene was very limited, site 34 was predicted to have been chronically contaminated from the combustion of fuels according to intense shipping activities. Elsewhere, sites 7–15 and 26–32, which were adjacent to the accident, had very low PAH levels except site 12. Due to practical reasons, sediment sampling at each collecting time point could not be performed at exactly the same location. So some sites showed in high variation of PAH concentrations. Therefore, physico-chemical properties, such as particle size and organic content, as well as the PAH concentration can vary greatly among samples collected at different time points within each site.

Hebei Spirit oil spill was the largest marine oil spill accident (10,800 tons) in Korean history (Hong et al., 2014). Both the Wu Yi San and Hebei Spirit oil spill showed a dramatic decrease of 16 PAH and alkyl PAH residual levels during the four months after the accidents, but

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