Ecotoxicology and Environmental Safety 128 (2016) 11–20

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

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

Polycyclic aromatic hydrocarbons (PAHs) and alkylated PAHs in the coastal seawater, surface sediment and oyster from Dalian, Northeast China

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A R T I C L E I N F O

Article history:
Received 16 October 2015
Received in revised form
30 January 2016
Accepted 2 February 2016
Available online 11 February 2016

Keywords:
PAHs
Alkyl-PAHs
Coastal environment
Ecotoxicological risk
Water-sediment exchange

A B S T R A C T

A total of 46 polycyclic aromatic hydrocarbons (PAHs, 21 parent and 25 alkylated) were determined in seawater, surface sediment and oyster from coastal area of Dalian, North China. The concentration of Σ_{46}PAHs in seawater, sediment, and oyster were 136–621 ng/L, 172–4700 ng/g dry weight (dw) and 60.0–129 ng/g wet weight (ww) in winter, and 65.0–1130 ng/L, 71.1–1090 ng/g dw and 72.8–216 ng/g ww in summer, respectively. High PAH levels were found in industrial area both in winter and summer. Selected PAH levels in sediments were compared with Sediments Quality Guidelines (ERM–ERL, TEL–PEL indexes) for evaluation probable toxic effects on marine organism and the results indicate that surface sediment from all sampling sites have a low to medium ecotoxicological risk. Daily intake of PAHs via oyster as seafood by humans were estimated and the results indicated that oyster intake would not pose a health risk to humans even 30 days after a oil spill accident near by. Water-sediment exchange analysis showed that, both in winter and summer, the fluxes for most high molecular weight PAHs were from seawater to sediment, while for low molecular weight PAHs, an equilibrium was reached between seawater and sediment.

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1. Introduction

Dalian, with an area of 12574 km^2, lies on the south side of Liaodong Peninsula, Northeast China (E120°58′-E123°31′, N38°43′-N40°10′) (see Fig. SI-1, Supporting information, (SI)), a typical coastal city which is one of the metropolis in China with a population of 600 millions. During the last three decades, the coastal areas of this region have been influenced by a range of socio-economic activities, such as urbanization, industrialization and maritime transport that have resulted in widespread pollution by organic contaminants (Jia et al., 2011; Na et al., 2013; Hong et al., 2014). Besides, a fuel pipeline exploded and caught fire in the southwest of Dayao Bay in Dalian on July 16, 2010 (the location of oil spill place can be found in Fig. SI-1). About 1500 t of crude oil were spilled from the pipeline and a large extent of the shoreline in Dalian was significantly polluted (Liu et al., 2013; Zhang et al., 2013).

Polycyclic aromatic hydrocarbons (PAHs), defined as a group of aromatic hydrocarbons with two or more fused benzene rings, which are one of the most important classes of hydrophobicity organic contaminants. PAHs have proved to be the main components responsible for effects on organisms, because of their carcinogenic, mutagenic and toxic effects (Lotufo and Fleeger, 1997). PAHs in environment are mainly from two sources: petrogenic, composed by PAHs associated with spills of crude and refined oil; pyrogenic, which includes the compounds formed by fossil fuel combustion, waste incineration, biomass burning, and asphalt production. Petrogenic PAHs mainly consist of 2- and 3-rings, as well as alkylated PAHs with one to a few methyl group, while pyrogenic PAHs are often characterized by 4-6 aromatic rings. Alkylated PAHs are classified according to the number of parent rings and the carbon number of alkylated substituents, and they are peculiarly important in the case of crude oil, in which 90% PAH are alkylated (Lian et al., 2009b).

In coastal environments, PAHs come from atmosphere (Esen et al., 2008), urban runoff (McCready et al., 2000), municipal or

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http://dx.doi.org/10.1016/j.ecoenv.2016.02.003
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industrial effluents (Blanchard et al., 2001; Vogelsang et al., 2006), and spillage of fossil fuel (Pettersen et al., 1997). The inputs of PAHs would pose a high risk to the health of humans and wildlife, especially to aquatic organism (Barron et al., 1999). More importantly, alkylated PAHs have been shown to be more toxic (Fallahtafti et al., 2012) and contain higher concentrations than their parent compounds (Hawthorne et al., 2006).

Researches on PAHs in various environmental matrices have been investigated in Dalian, including surface seawater, sediment, soil, and atmosphere (Wan et al., 2006; Wang et al., 2007; Wang et al., 2008; Tian et al., 2009; Wang et al., 2011; Liu et al., 2013). On land, industrial and business/residential areas have been proved to contain higher levels of PAHs both in soil and air than rural and garden areas in Dalian (Wang et al., 2008). While factor analysis model results showed that gasline- and coal-combustion related residential emissions were the primary sources (Wang et.al., 2007). For the aquatic environment, Wang et al. (2011) presented 24 PAH compounds in seawater and sediment, and their water-sediment exchange. Recently, Liu et al. (2013) reported 25 PAH compounds in seawater and sediment sampled from Lingshui Bay to Xiaoyao Bay, Dalian, 50 days after the oil spill accident, and the results indicated that the PAHs in seawater likely originated from the spilt oil and PAHs in sediment were due to historical accumulation. However, these studies just focused on the parent PAHs, and little information of alkylated PAHs have been reported in environmental mediums from Dalian.

The current study investigates both the parent and alkylated PAHs concentrations in seawater, sediment and oyster in the coastal area of Dalian. The objectives of this study are 1) to investigate the pollution levels of parent and alkylated PAHs in multi-matrices, 2) to determine the potential risk caused by these chemicals to human and aquatic organisms, and 3) to analyze water-sediment exchange for each PAH compounds in a coastal marine environment.

2. Materials and methods

2.1. Sampling

Surface seawater, sediment, and oyster samples were collected at 15 sites (1 industrial, 2 urban, and 12 rural) in proximity to the shore around Dalian, Northeast China, in the January and August of 2010, respectively. Among the 12 rural sampling sites, 8 were from Bohai Sea (B01-R08) and 4 were from Huanghai Sea (H01-04). A map with locations of sampling sites can be found in Fig. SI-1, and detailed information is given in Table SI-1. We collected seawater, sediment, oyster samples from all sampling sites both in winter and summer, but no oyster at B01 in summer.

All samples (seawater, sediment and oyster) were packed in solvent-rinsed brown glass bottles with Teflon-lined caps. Surface sediment (0–5 cm) was collected using a bucket grab. An acetone rinsed bistoury was used to harvest edible parts from oyster shells and at least 15 individual samples were thus collected from each site Each water or sediment sample was composed of well-mixed five subsamples collected from different locations at each site. During seawater sampling, field blanks were performed for every 5 seawater samples consisting of 1 L MilliQ water sealed in glass bottles with Teflon-lined caps. After collection, all samples were sent to the laboratory of *the International Joint Research Center for Persistent Toxic Pollutants* (IJRC-PTS), Dalian Maritime University, Dalian, China and stored at −20 °C (sediment and oyster) and 4 °C (seawater) until the time for extraction.

2.2. Chemical and reagents

Organic solvents used in this study were of pesticide grade purity (J.T. Baker, USA). Silica gel (100–200 mesh) was purchased from Merck (Merck, Germany) and was activated at 130 °C for 16 h, then deactivated by MilliQ water (3.3%, w/w) and stored in a desiccator. Twenty-one parent PAHs and twenty-five alkylated PAHs standards were purchased from the Supelco, Inc. (Supelco, USA) (See Fig. SI-2, Fig. SI-3 and Table SI-2 for details). Naphthalene-D$_8$, fluorene-D$_{10}$, pyrene-D$_{10}$, and perylene-D$_{12}$ were used as the surrogate standards for all compounds (Ma et al., 2010; Ma et al., 2011; Liu et al., 2013).

2.3. Extraction and analyses

Samples were treated, extracted and analyzed according to the methods established at the National Laboratory for Environmental Testing (NLET), Environment Canada (Wang et al., 2008; Jia et al., 2011; Liu et al., 2013). After spiking with surrogated recovery standards (40 ng for naphthalene-D$_8$, 50 ng for fluorene-D$_{10}$, 20 ng for pyrene-D$_{10}$, and 40 ng for perylene-D$_{12}$), 1 L of seawater samples were extracted with 100 mL DCM in a separatory funnel with agitation followed by a 1 h setting time. Extraction was thrice repeated, followed by DCM collection and rotary-evaporation to 1 mL. Ten grams of sediment and 10 g anhydrous sodium sulfate were accurately measured into a pre-cleaned extraction thimble and spiked with surrogated recovery standards (40 ng for naphthalene-D$_8$, 50 ng for fluorene-D$_{10}$, 20 ng for pyrene-D$_{10}$, and 40 ng for perylene-D$_{12}$). After mixing, samples were Soxhlet extracted for 24 h with 200 mL mixed solvent (hexane/acetone, 1:1 v/v). The extraction methods for oyster samples were similar to sediment except using DCM/hexane (1:1 v/v) for extraction. For oyster sample, after Soxhlet extraction, gravimetric lipid determination was performed using 10% of the extract. Extracts were then rotary evaporated to 1 mL. The 1 mL extracts were fractionated using gel permeation chromatography (with 1:1 hexane/DCM as the mobile phase) and silica gel column chromatography. The extract was rotary evaporated to 2 mL, then solvent-exchanged into isooctane and reduced to 1 mL under nitrogen evaporation prior to gas chromatography–mass spectrometry (GC–MS) analysis.

All PAHs were identified and quantified using a Thermo Trace gas chromatograph (Thermo TRACE 2000) coupled with a Polaris Q mass spectrometry. Splitless injection was used (2 μL), along with a DB-5 column (HP 60 m × 0.25 mm i.d. × 0.25 μm film thickness). The mass spectrometer was operated in electron impact mode (70 eV). Helium gas was used as a carrier at constant flow 1.0 mL/min. For parent PAHs: the column oven temperature was programmed at a rate of 25 °C/min from an initial temperature of 60 °C to a temperature of 180 °C (1-min hold), 3 /min to 280 °C (30-min hold). For alkylated PAHs: the column oven temperature was set at 60 °C for 1 min, ramped at 6.5 °C/min to a temperature of 310 °C (5-min hold). The temperatures of injector, transfer line and ion source were held at 260, 270 and 250 C, respectively.

2.4. Organic matter fraction determination

Ten grams of sediment from each sample were used for organic matter fraction (φ_{OM}) determination. Sediment samples were first oven-dried at 105 °C for eight hours to a constant weight. After moisture elimination, the samples were placed in a muffle furnace and φ_{OM} determined by measuring their loss after baked at 550 °C for five hours.

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