Ecotoxicology and Environmental Safety 145 (2017) 640–652

Contents lists available at ScienceDirect

Ecotoxicology and Environmental Safety

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

Comprehensive and comparative ecotoxicological and human risk assessment of polycyclic aromatic hydrocarbons (PAHs) in reef surface sediments and coastal seawaters of Iranian Coral Islands, Persian Gulf

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ARTICLE INFO

Keywords:
PAHs
Ecotoxicological risk assessment
Human risk assessment
PCA
Coral reef
Persian Gulf

ABSTRACT

The concentration and spatial distribution along with ecotoxicological risk of 30 polycyclic aromatic hydrocarbons were investigated in the reef surface sediments (RSSs) and coastal seawater (CSW) of ten coral Islands from the Persian Gulf, Iran, in January 2015. For all sampling sites, assessment of ecological risk was undertaken using several approaches. Mean concentration of Σ30PAHs varied between 70 and 884 ng L^{-1} with an overall mean value of 464 ng L^{-1} in the CSW, while the RSS ranged from 274 to 1098 ng g^{-1} dw with a total average of 619 ng g^{-1} dw. The results showed a gradient in PAH concentration and toxicity estimates from the northern Hormoz site increasing to the eastern Kharg site. Most of the toxicity estimates were in the moderate range or less than risk values for damage to the marine environment. The calculated Dermal Hazard Quotient (HQs), the sum of HQs (HI) and other cancer risk values of most compounds were less than safety values at most sites. It means that the possibility of negative effects of PAHs via dermal absorption from sediments for children and adults is low. Some sampling sites studied have already been impacted with hazardous pollutants for an extended period of time and evidence from this investigation demonstrates that mixtures of PAHs may be carcinogenic to humans, especially in the western part of the Gulf.

1. Introduction

It is well documented that assessing concentration of PAHs in particles and sediments were broadly studied all over the world (Ranjbar Jafarabadi et al., 2017). The water column above sediments in the marine environment could also contain dissolved and suspended hydrocarbons, which are more bioavailable and therefore more toxic for marine biota than contaminants in surface sediments (Ranjbar Jafarabadi et al., 2017). Organisms can accumulate the biologically available forms of the pollutants from their environmental surroundings. In aquatic environments, the re-suspension and bioturbation of immobile contaminants in the water column could affect bioaccumulation via the food chain (Akhbarizadeh et al., 2016). PAHs in seawater transported in water currents may produce direct and irreversible toxic effects on organisms (Mirza et al., 2012). The key factors controlling the presence and deposition of pollutants in sediments includes biological and chemical characteristics of marine sediments, textural, geochemical characteristics of pollutants and environmental conditions (pH, Eh etc.) (Chakraborty et al., 2015).

The concentrations of organic contaminants in this aquatic system have already been extensively monitored throughout the last decades with the aim of screening for persistent organic pollutants (POPs) (Pintado-Herrera et al., 2016). For humans and marine organisms exposed to mixtures of PAHs a series of health issues (i.e. increased risk of skin cancers) have already been reported (Diggs et al., 2011). The health effects of these compounds are in practically all cases related to their carcinogenic potential, predicated on evidence from numerous epidemiological studies (Garcia-Suastegui et al., 2011; Kim et al., 2013). Long-term exposure of PAHs is suspected to increase the risks of cell impairment via gene mutation and cardiopulmonary mortality (Kuo et al., 2003).

Oil extraction, transportation, and the effluent discharges from the coastal cities and oil refineries are the major sources of oil pollution in the Persian Gulf (Ranjbar Jafarabadi et al., 2017). The Persian Gulf is one of the most important industrial development zone with several large seaports. In recent years, the Gulf's ecology has been destroyed by industrialization and spills (Aghadadashi et al., 2017). Mobile pollutants in these water columns could bioaccumulate through the food chain. Therefore, maintaining the ecosystem health and quality is extremely important. Because of the potential impact of the pollution

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http://dx.doi.org/10.1016/j.ecoenv.2017.08.016
Received 2 May 2017; Received in revised form 1 August 2017; Accepted 4 August 2017

from anthropogenic sources on marine organisms, it is essential to recognize and assess the risk brought on by anthropogenic pollution on the ecosystem of the Persian Gulf. There is little data about PAH contaminations in seawater and reef sediments in this area. Thus, this study is the first comprehensive investigation on the status of ecotoxicological risk of PAHs in the coral reef seawater and sediments of the Persian Gulf area. This study was designed to: 1) Evaluate the concentration and spatial distribution of PAHs in seawater and reef sediments. 2) Calculate different pollution indices such as: Threshold Effects Level to Probable Effects Level (TEL/PEL), the Mean Toxicity Quotients (*m*-ERM-Q) and Mean Probable Toxicity Quotients (*m*-PEL-Q) approaches, the Toxicity Equivalent (TEQ) and Human Risk including cancer indication (TEQcarc), Mutagenic Equivalents (MEQ), Risk Quotient (RQ) and HQs methods. 3) Demonstrate the spatial variations of ecotoxicological and human risks of PAHs. 4) Provide data on the source and potential ecological and human health risk of PAHs to the local ecosystem of the Persian Gulf.

2. Materials and methods

2.1. Study area and sampling

Coral reefs (stations) were chosen based on the severity of human activities, especially oil pollution (mostly PAHs compounds) (Ranjbar Jafarabadi et al., 2017). Ten coral Islands from the east to the west of the Persian Gulf have been chosen. The major economical parts of the Gulf are limited to the west and center part of the Gulf where Kharg, Lavan and Siri Islands in vicinity of many major point source of pollution sources (petrochemical and gas stations, oil wells and industrial wastes as well as untreated waste waters etc.). Qeshm, Hengam, Hormoz are far from dense human activates and contamination source which is located in the east part of the Persian Gulf. Shidvar, Lark, Kish and to some extent Hendurabi is located in the center toward east of the Gulf with seemingly known point and non-point sources of pollution in its vicinity. And finally the Lark Island which most activity in this area is fishing and military activities. In each Island, 12 sampling zone with 3 repetitions were sampled during January 2015. Point selection was on the base of having the sites with relatively discernible applications to assess the possible importance of local sources and human made pressures in the term of PAHs levels. Regarding to that, the samples were collected from close to coast (6 samples) and far from the coast (6 samples) of the coral Islands of the Persian Gulf. A total of 360 samples of CSW and 360 samples of RSSs were collected. More detailed information about the sampling sites were drawn in Fig. 1. RSSs (above 5-cm depth) and CSW samples, 0.5 m below the air-water interface, were collected with pre-cleaned 25 L containers utilizing cylinder samplers and pre-cleaned glass bottles with polytetrafluoroethylene screw caps, respectively during the period of January 2015. All samples were sent to the central Laboratory of the environmental science of the Tarbiyat Modares University (TMW), Tehran, Iran.

2.2. Sample pretreatment

PAH analyses for RSSs were performed by the method described by (Bakhtiari et al., 2009) comprising the preparation of samples, extraction, and biomarkers analysis which have been used by many researches (Ranjbar Jafarabadi et al., 2017; Shirneshan et al., 2016). As a summary, about 5 g of each freeze-dried sample were purified and fractionated. The extraction of PAHs was carried out by a Soxhlet apparatus using 100 mL of dichloromethane for a period of 12 h for each RSS. 100 μL of PAH-SISM (surrogate standard mix) comprising naphthalene-d_8, anthracene-d_{10}, chrysene-d_{12} and perylene d_{12} (200 ppb of each component) was added for quality control of PAH analyses. After rotary evaporation, the extracts were decreased to 4–5 mL. After further volume reduction with nitrogen gas, each extract was transferred to the top of a fully activated silica gel chromatography column. The PAH fraction was eluted with 14 mL of dichloromethane/hexane (1:3, v/v). After that, each obtained PAH fraction was evaporated to about 1 mL, and was transferred to a 1.5 mL amber ampoule. After further reduction with nitrogen gas to dryness, extracts were re-dissolved in 100 μL of isooctane containing p-terphenyl-d_{14} as an IIS (internal standard) for PAH analysis.

For seawater samples the dissolved PAH analysis was conducted based on the approach of (Liu et al., 2016). Dissolved PAHs in CSW samples were extracted by SPE10 using Oasis HLB cartridges (Waters, Milford, MA, USA). Briefly, before used in extraction, the HLB cartridges were consecutively preconditioned with 20 mL methanol, 20 mL ultra-pure water and 20 mL dichloromethane. Then 2 L filtered seawater in each sampling site was passed through a SPE cartridge at a rate of 5 mL/min. All the cartridges were subjected to freeze drying for 96 h to eliminate moisture. Cartridges were eluted five times with 6 mL ethyl acetate at each time. The eluents for individual samples were combined, concentrated, and solvent-exchanged to1 mL methanol using a rotary evaporator prior to GC-MS analysis.

2.3. Analytical procedures

The PAHs were analyzed on an Agilent 7890A gas chromatograph

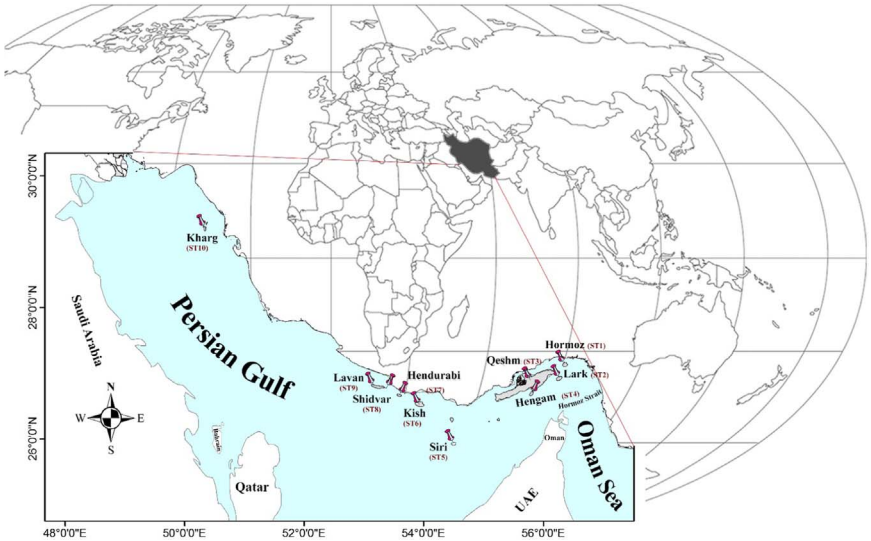

Fig. 1. Study area and sampling sites in the Persian Gulf.