

Risk assessment of polycyclic aromatic hydrocarbon infested sediments along the coast of Goa, India

Vasudha C. Bhatawadekar^{1,3}, Samir R. Damare^{1*}, Anita Garg²

¹Biological Oceanography Division, CSIR-National Institute of Oceanography,
Dona Paula, Goa-403004, India.

²Analytical Services Division, CSIR-National Institute of Oceanography,
Dona Paula, Goa-403004, India.

³Department of Microbiology, Goa University, Taleigao Plateau, Goa, India

*Corresponding author: samir@nio.org

*ORCID: 0000-0002-4201-4844

Abstract

Goa, a small state in India, is a tourist hotspot, known for its coastline and beaches. Anthropogenic activities lead to pollution in the environment. Amongst many other pollutants, polycyclic aromatic hydrocarbons represent a major class of contaminants owing to their widespread distribution and protracted environmental persistence. No information is available about the levels of PAH contamination in the sediments along the coastline of Goa. The study aimed to establish a baseline for PAH concentrations in the sediments along the shoreline to help comment on the pollution levels caused and thereby understand the risk and their impact on the marine life therein. The total concentration of PAHs along the selected sampling sites of the Goa coastline was 1.00 to 875 $\mu\text{g g}^{-1}$. Maximum PAH concentrations were detected in the Divar island mangrove (875 $\mu\text{g g}^{-1}$), least was observed at Galgibaga beach (365 $\mu\text{g g}^{-1}$). The results revealed that the sediment of the Goa coastline is heavily contaminated with PAH. Source apportionment of PAHs was analysed based on diagnostic ratios and results exhibited that petroleum products and their combustion were primarily responsible for their generation. The results of risk quotients showed that the values are way above ER-M (Effect range median), indicating these could pose a high risk to the ecosystem.

Keywords: Ecotoxicity; GC-MS; Goa; PAH; Petrogenic; Pyrogenic

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are priority organic pollutants (Behera et al. 2018). They are produced from coal, oil, natural gas combustion, and petroleum spillage (Ramzi et al. 2017; Hassan et al. 2018). They have two or more benzene rings fused in linear, angular, or cluster structural configurations (Liu et al. 2017). Based on the number of rings they possess, they are categorised as low molecular weight (LMW PAH) and high molecular weight (HMW PAH). As the molecular weight and number of rings in the PAH structure increase, the hydrophobic, chemical stability, and octanol-water coefficient increase while aqueous solubility decreases (Ghoshal et al. 2016). These characteristics cause PAH to adsorb on particulate matter in the air, water, soil, and sediment, reducing bioavailability and increasing its persistence in the environment ubiquitously.

In aquatic bodies such as rivers, estuaries, and marine ecosystems majorly, petroleum spillage, urban run-off, and atmospheric deposition can lead to elevated PAH levels (Hassan et al. 2018). Low affinity to water and high association with the solid phase leads to PAH adsorption on the suspended matter and sink to the bottom in the sediments (Hassan et al. 2018). The lipophilic nature of PAH makes their entry easy in living biota such as benthic organisms and fishes, and thereby enter the food chain (Udofia et al. 2021), leading to bioaccumulation (Ghoshal et al. 2016). PAHs are toxic, mutagenic, and carcinogenic, posing prolonged health hazards to aquatic and terrestrial ecosystems and human beings (Algebeleye et al. 2017). PAH concentration is comparatively higher in marine organisms, such as fish, jellyfish, etc. than in the sediment and water from where they are collected. This is due to bioaccumulation and biomagnification in the food chain, which could finally harm human health as they are the tertiary consumers in the food chain (Penko and Bajt 2019).

The state of Goa in India is a hotspot for tourists from all over the world. It is surrounded by the Arabian Sea, forming a stretch of 103 km coastline. Mormugao Port Trust (MPT) is India's one of the oldest and busiest port. Goa's majority population is pescatarian that is, they depend primarily on seafood as their staple food. The quality of seafood depends on the environment from where they are harvested. The riverine and estuarine streams in Goa are remarkably affected by the transportation of ferry boats, cargo ships, casinos, and various sports activities for tourists. Accidental oil spillage and fuel burning during transport may lead to the generation and accumulation of PAHs in the sediment. No details are available about the distribution and concentration of PAHs over the main streams and their possible ecological toxic effects from the coastline of Goa or estuaries. In this study, we investigated the levels of PAHs present in a few significant hot spots over the coastline of Goa, and the probable sources of these PAHs were

elucidated using accepted diagnostic ratios. Along with this, the possible toxic impacts due to the presence of PAHs in sediments were also assessed to present a holistic picture of the effect of PAHs on the environment.

2. Material and methods

2.1. Sampling sites

The sampling locations were selected based on the possible oil seepage during the movement of cargo ships, ferry boats, fishing trawlers, and anchored floating casinos at Zuari estuary and the mouth of Mandovi estuary. The Mormugao harbour is the common flush point of the Mandovi and Zuari estuaries. Sal River is one of the smallest rivers in Goa. Sal River's mouth is in Verna, flows about 16 km, and drains off at Betul in the Arabian Sea (Supplementary Figure 1). It also attracts tourists for river boating. There is a lot of fishing trawler movement. Divar Island is located across the Mandovi River, and is bordered by mangroves. Mangrove ecosystems have a distinct role in the dynamics of anthropogenic and biogenic carbon storage (Garcia and Martins 2021). They suffer pressure from urban run-off, boat movements, and fishing trawler transport, leading to oil and fuel pollution. Galgibaga is known as Turtle beach, as the beach is used by Olive turtles nestling. This site is pristine compared to other sites selected and sampled in this study. The sediment samples were collected using sediment grab from coastal regions of Goa (Mandovi estuary, Zuari estuary, Sal River, Galgibaga beach, and Divar mangrove) (Supplementary Figure 1). The data of sampling date and latitude-longitude details described in supplementary table 1.

2.2 Extraction and quantification of PAH from sediments samples

PAHs were extracted from the sediment samples using the modified 3550C method (US EPA 1996). Glassware was washed with acid, and ultrapure water, dried, and again washed with hexane and dried in an oven to remove any trace contaminant before extraction. The freeze-dried and homogenised sediment sample was weighed (10 g), mixed with anhydrous sodium sulfate to remove moisture, and subjected to sonication (probe number 6). Hexane and dichloromethane (1:1) mixture was used to extract PAHs. The sediment was sonicated for a total of 30 minutes, with a cyclic combination of pulse on for 5 seconds and pulse off for 15 seconds. This procedure was repeated thrice. Extracts were pooled together and concentrated using the rotary evaporator. The extracts were then purified using an activated silica gel. The PAH fractions were eluted using hexane and concentrated using a rotary evaporator. The extract was resuspended in 1 mL of acetonitrile. PAH concentration was determined using Gas chromatography-mass spectrometry (GC-MS Shimadzu, QP2010 plus). The separation of PAHs was achieved by using a Rtx-5MS capillary column (0.32

mm internal diameter, 0.25 mm film thickness, 30 m long) with a temperature programme of 50°C for 5 min hold, then increased by 4 °C min⁻¹ up to 310 °C, held for 10 min., while their identification was carried out by operating MS in full scan mode. And then, by applying retention time and m/z ratio, further PAH analysis was carried out by SIM (Selected Ion Monitoring). The carrier gas was helium (99.999% purity) with a flow rate of 1.5 mL min⁻¹. One µL sample was injected using split-less injection mode. Before GC-MS analysis, calibration and quantification were performed using a standard PAH mixture in acetonitrile procured from Sigma-Aldrich(2, 4, 6, 8, and 10 ppm) containing Naphthalene (Nap), Acenaphthylene (Acy), Acenaphthene (Ace), Fluorene (Flu), Phenanthrene (Phe), Anthracene (Ant), Fluoranthene (Flt), Pyrene (Pyr), Chrysene (Chr), Benzo(a)Pyrene (B(a)P), Benzo(b)Fluoranthene (B(b)F), Benzo(k)Fluoranthene (B(k)F), Benzo(a)Anthracene (B(a)A), Dibenzo(ah)Anthracene (D(ah)A), Benzo(ghi)Perylene (B(ghi)Per), Indeno (1,2,3-c,d)Pyrene (I(123-cd)P) (Each of 10 µg mL⁻¹). The total PAH concentration in the sediment samples was reported as µg g⁻¹. Spiked Phenanthrene (Phe) and Fluoranthene (Flt) were used to check their recovery from the sediment samples and the extraction procedure efficacy. Recoveries ranged up to 95%. Reproducibility was calculated by analysing the samples thrice.

2.3 Origin of PAHs in the sediment samples

The origin of PAH in the sediment depends mainly on the sources responsible for generating PAH (Penko and Bajt, 2019). There are three categories, namely; (i) petrogenic refers to the PAHs that emerged due to the influence of petroleum and crude oil or fossil fuel, (ii) pyrogenic indicates the origin of PAHs is due to incomplete combustion, and (iii) diagenesis which refers to the PAHs entering the sediment owing to biological processes (Udofia et al., 2021). The mathematical calculations derived in terms of ratios by Budzinski et al., 1997 and Yunker et al., 1999 were used for probable source apportionment of PAHs in the sediment. Phenanthrene/anthracene (Phe/Ant) >10 indicates petrogenic origin, while <10 shows its pyrogenic origin. The ratio of fluoranthene/pyrene (Flt/Pyr), if it is <1, shows petrogenic and >1 shows pyrogenic origin. Additionally, the ratio of LMW-PAH/HMW-PAH < 1 indicates pyrogenic origin, while >1 indicates petrogenic origin (Budzinski et al., 1997; Yunker et al., 1999). Furthermore, to confirm the origin of PAH in sediment, an isomer ratio such as Ant / (Ant+Phe) < 0.1 indicates petrogenic origin, while > 0.1 shows the pyrolytic source of PAH. Similarly, the ratio of Flt / (Flt+Pyr) < 0.4 indicates petrogenic, while > 0.4 shows the pyrolytic origin of PAH, and the ratio between 0.4 - 0.5 signifies the origin due to petroleum combustion (Yunker et al., 2001).

2.4 Assessment of the toxicity of PAH contamination

The quality of sediment in terms of PAH contamination is categorised into four types based on the total concentration of PAH in the following manner; $< 0.200 \mu\text{g g}^{-1}$ – not contaminated; $0.2-0.6 \mu\text{g g}^{-1}$ –slightly contaminated; $0.6-1.0 \mu\text{g g}^{-1}$ – contaminated; $>1.0 \mu\text{g g}^{-1}$ – heavily contaminated (Howard et al., 2021). Additionally, effect-based guideline values were utilised to evaluate the correlation between PAH concentration in the sediment and the toxic response expressed by an organism (Pradhap et al., 2021, Yu et al., 2018, Howard et al., 2021). The sediment quality can be ascertained based on the Effect Range-Low (ER-L) and Effect Range – Medium (ER-M) (Long et al., 1995). The values within ER-L suggest that the adverse biological effects due to contaminants are low. The values above ER-M suggest that contaminants can cause harmful biological effects. The concentration of individual PAH at all the sites in the different sampling locations was combined, and the mean values and standard deviation were compared with ER-L and ER-M limits (Table 3). Along with this, the integrated adverse effects due to PAH were evaluated by calculating the risk quotient using the below formula,

$$\text{ERM}Q_i = \frac{C_i}{C_{\text{ERM}i}}$$

$$\text{Risk Quotient (MERM}Q) = \sum_i n \frac{\text{ERM}Q_i}{n}$$

where $\text{ERM}Q_i$ is the ERM quotient of individual PAH, C_i is the concentration of individual PAH, $C_{\text{ERM}i}$ is the ERM value for discrete PAH, and MERM Q is the mean ERM quotient, while n is the total number of PAHs detected (Yu et al., 2018, Wang et al., 2018).

3. Results and discussion

3.1 Standardization of PAH on GCMS

The 10 ppm PAH standard was run initially in Full scan mode; PAH compounds were identified using the NIST library. Retention time (RT) and m/z were noted against each compound. The 10 ppm PAH standard was then re-run in SIM mode. The calibration curve of 2, 4, 6, 8, and 10 ppm PAH standard showed 0.99 r^2 for each compound (Supplementary Figure 3). The same calibrated method was used to quantify the PAH extracted from sediment.

3.2 Assessment of PAH concentration in the sediment samples

The concentrations of PAHs in the sediment from the sampling sites of the Mandovi estuary, Zuari estuary, Sal River, Galgibaga and Divar islands are presented in Table 1. The total

concentration of extractable PAHs along the selected hotspots of Goa coastline was in the range of 1.00 to 875 $\mu\text{g g}^{-1}$. In the Mandovi estuary at site M1 which is situated where the human interference due to casinos and ferry boat movement is maximum, compare to other sites in Mandovi estuary. The total concentration of extractable PAH was 273 $\mu\text{g g}^{-1}$ and 12 PAHs were detected (Table 1). Also, Only at M1, 5-ring PAHs, namely benzo (a)anthracene (B(a)Ant), benzo(a)pyrene(B(a)Pyr), benzo(k)fluoranthene(B(k)Flt), dibenzo (ah)anthracene (Db(ah)Ant), and benzo(ghi)perylene (B(ghi)P) were found. At almost all sites, acenaphthene (Ace) and acenaphthylene (Acy) were found in different sites of Mandovi estuary (Table 1). In samples of Zuari estuary, only 5 PAHs, namely, acenaphthylene (Acy), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flt), and pyrene (Pyr), were found, amongst which, Phe, Flt, and Pyr were present abundantly. The total PAH concentration ranged from 161 - 789 $\mu\text{g g}^{-1}$ (Table 1). 6 PAHs were found in the Sal River (Table 1). Amongst these, 4 were LMW, and 2 were HMW PAHs. Acenaphthylene (Acy), phenanthrene (Phe), fluoranthene (Flt), and pyrene (Pyr) were present abundantly. Total PAH concentrations ranged from 3.41 to 499 $\mu\text{g g}^{-1}$ (Table 1). Contamination due to LMW PAH is high (Fig. 2C). In Galgibaga samples, 7 PAHs were found. Amongst them, acenaphthylene (Acy), phenanthrene (Phe), and anthracene (Ant) were present in more concentrations (Table 1). Flt and Pyr were present only at five sites. Six-ring benzo (ghi) perylene (B (ghi)P) was found only at one site. Naphthalene (Nap) was detected at Galgibaga and Mandovi sampling locations. The total PAH concentration ranged from 1.0 – 365 $\mu\text{g g}^{-1}$ (Table 1). In the Divar mangrove, the total PAH concentration was 426 – 875 $\mu\text{g g}^{-1}$ (Table 1). Five PAHs were present abundantly with a maximum concentration of Flt. At two sites, 3 rings fluorene (Flu), and at the other two sites, 5 rings benzo (k) fluoranthene (B (k) F) were detected.

In Mandovi estuary, the total concentration of extractable PAHs was maximum at sampling site M1, compared to all other sites (Fig. 1). This could be due to accidental oil spillage and or fuel burning as a consequence of ferry boat transportation, fishing trawler movement, anchored floating casinos, and cruise boats for tourists within reach of site M1. The sites other than M1 were less perturbed by human activities; hence, the concentration of PAHs in the sediment from other sites of the Mandovi estuary was on the lower side. LMW-PAHs contribute more to PAH contamination than HMW-PAHs (Fig. 2A, 2B & Supplementary Figure 2).

The total concentration of extractable PAHs in the Zuari estuary at all sites is impartial. Maximum concentration is found at sites Z9 and Z10, located towards Mormugao harbour, where movement due to cargo ship is maximum. Phe, Flt, and Pyr were present abundantly at all sites

(Table 1). Together, at different sites within the Zuari estuary, concentration in increasing order of individual PAHs is as follows, $Acy < Pyr < Flt < Ant < Phe$ (Table 1).

Contamination due to LMW PAH was maximum compared to the HMW PAHs and also the abundance of LMW PAH was seen in all the sediment samples collected from Sal River (Fig. 2A, 2B & Supplementary Figure 2). The 3-ring PAHs, namely, Acy, Phe, and Ant, were present abundantly. The total concentration of individual PAHs in sediments collected from all sites of Sal River is as follows: $Flu < Acy < Pyr < Phe < Ant < Flt$ (Table 1). Similar to other sampling locations, Acy, Phe, and Ant were ubiquitously present at all the sites in Galgibaga. The level of PAH concentration is as follows: $Nap < Acy < Flt < Phe < Ant < Pyr$ at Galgibaga (Table 1). Amongst all other sampling locations, the Divar mangrove is highly contaminated with the total concentration of extractable PAHs (Fig. 1; Supplementary Figure 2). The order of PAH was as follows: $Flu < B(k) Flt < Acy < Phe < Ant < Flt < Pyr$ (Table 1). In increasing order, the total concentration of extractable PAHs at all sampling locations is Mandovi estuary < Galgibaga beach < Sal River < Divar island < Zuari estuary. While the LMW PAH contribution in Goa is maximum in Zuari estuary > Sal River > Divar island > Galgibaga beach > Mandovi estuary (Supplementary Figure 2). In Mandovi estuary and Galgibaga beach, the PAH contamination is equally distributed between LMW and HMW-PAHs (Supplementary Figure 2). In the Zuari estuary and Sal River, contamination is maximum due to LMW-PAH. While in the Divar mangrove samples, PAH contamination is maximum due to HMW-PAH. Divar mangrove and Zuari estuary are highly PAH-contaminated locations, while the least contaminated are Mandovi estuary and Galgibaga (Supplementary Figure 2). Table 2 gives an idea about the relative levels of PAH that have been reported from other parts of the world compared to the levels reported here.

3.3 Source apportionment of PAHs in the sediment

PAH origin in the sediments collected from the Goa coastline was depicted in the cross-plot of isomer ratios (Fig. 3). The isomer ratio of PAH in only highly contaminated sites (Divar 4; Zuari 9; Galgibaga 5; Sal 3) was selected to depict the cross plot. The probable source of PAHs in highly contaminated sites is due to petroleum combustion and incomplete combustion of wood, grass, or coal (Fig. 3). The sediments obtained from the Zuari estuary showed that the source of PAH is majorly due to petroleum combustion, and at some sites, it showed petrogenic. This area records a lot of shipping activities due to proximity to the Port area (MPT, Goa). The cross-plot indicates that the HMW PAH and LMW PAH originated from petroleum spillage and combustion in the Zuari estuary.

The Divar mangrove sediment samples showing a higher concentration of PAHs as compared to other sites from Goa, exhibited the pyrolytic origin of PAH (Fig. 3). Similarly, in Sal River, the PAH origin was pyrogenic (Fig. 3). This could be attributed to the fact that this site has residential interference where chances of coal burning for cooking could be routine practice, which could lead to incomplete combustion and hence generation of PAH. Also, this area sees a lot of ship movements, like ferry boats and fishing vessel movements. The concentration of PAH was comparatively low in Galgibaga samples, but the isomer ratios and individual ratios (Table 1) showed that the PAH source was petrogenic, and at only two sites, it was pyrogenic. This might be because Galgibaga is a tourist spot, leading to water sports activities, and also, fishing trawler movement may cause oil leakage leading to the generation of PAH. In the Mandovi estuary, PAH sources were Petrogenic (Fig. 3). Transportation of ferry boats, fishing trawlers, and refuelling of anchored floating casinos could lead to accidental oil seepage in the water, reaching the sediments. The ratio of LMW/HMW shows that the origin of PAH contamination was petrogenic (Table 1). However, only at M10, LMW/HMW shows that the PAH origin could be pyrogenic (Table 1). This result was on expected lines as the location is predominated by rural efflux, where coal burning for cooking or burning of other agricultural waste could be happening. The ratio of Phe/Ant, Flt/Pyr, and LMW/HMW indicates that the PAH origin was petrogenic (Table 1). High levels of PAH could be due to the movement of big ships to and fro from the Port. At this site, ratios of Phe/Ant, Flt/Pyr, and LMW/HMW indicated a petrogenic origin of PAH (Table 1).

3.4 Assessment of the toxicity of PAH contamination

As per the categorisation given by sediment quality guidelines, the results of the present study fall into the category of heavily PAH-contaminated (Table 1).

The risk quotient (MERMQ) was classified in four ways, < 0.1 indicates low risk; $0.11 - 0.5$ shows medium to low risk; $0.51 - 1.50$ shows medium to high risk; while > 1.50 indicates high priority risk (Yu et al., 2018). The risk quotient in the sediment collected from the Goa coastline is represented in Fig. 4. It reveals that PAH contamination in the sediments collected from the Goa coastline and rivers falls in the category of high-priority risk and can cause harmful effects to the ecosystem.

The lipophilic nature of PAH causes its accumulation and magnification in the fatty tissues of living biota which leads to adverse effects like disruption in physiological function, induction of antioxidant enzyme system which generates intermediates possessing a high affinity for the formation of stable DNA adduct having carcinogenic potency to distinct marine organism like sea

urchins, oyster, gastropods, sea snakes, fishes (Mote et al., 2015; Bhagat et al., 2017). PAH accumulation causes disturbance during the developmental stages of fish like teleost fish. It includes disruption during dorsoventral axis formation which leads to death before hatching, and hamper cardiac function which causes problems at any stage of the life cycle that includes premature death (Cherr et al., 2017; Incardona et al., 2016). The mass and strength of bone in fishes such as Pacific herring, pink salmon, and medaka have been reduced due to PAH (Barron et al., 2004; Billiard et al., 2006; Farwell et al., 2006). The liver and kidney are the major organs where bioaccumulation of PAH occurs (Mote et al., 2015). It has been observed that PAH concentration in fish causes a reduction in the level of enzymes responsible for cholesterol and fatty acid metabolism to a great extent (Souza et al., 2016). Along with this, it hampers the reproductive system, immune system and inflammatory response (Colli-Dula et al., 2018).

The concentration and toxicity impact of PAH in marine oyster *Saccostrea cucullata*, and gastropods *Cronia contracta*, *Nerita chaemeleon*, *Morula granulata* collected from PAH-contaminated sediments along the Goa coastline indicated that as PAH concentration increases, DNA integrity decreases revealing the genotoxic effect of PAH (Sarkar et al. 2008, 2014, 2018). Though the sampling sites and period of sample collection are different from the present study, it gives us an idea about the high concentration of PAHs from selected hotspots along the stretch of the Goa coastline that could lead to adverse toxic effects on the living biota.

4. Conclusions

The study gives us an idea about the PAH levels in the sediments from a few select representative coastal locations along the tropical rivers and estuaries and beach in the coastal state of Goa. Almost all the sediments collected in this study show a heavy PAH contamination. The diagnostic ratio analysis revealed that the origin of PAH contamination varies at different locations with equal contribution of petrogenic and pyrogenic-origin PAHs. The toxic effects of PAH contamination on the ecosystem in the Goa coastline are extremely high. The risk quotient values from all the samples call for the need for preventive protocols to be put in place to lower the PAH concentration. Further, it is important to study preventive and remediation approaches to reduce the PAH concentration and its toxic impact on the ecosystem.

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Statements & Declarations

Ethical approval

Not applicable

Consent to Participate

Not applicable

Consent to Publish

Not applicable

Authors contributions

The corresponding author planned the work. The first author carried out all the experimental work. The first and last authors carried out the GCMS analysis. All the authors contributed to writing the manuscript. The work is a part of the doctoral thesis of the first author.

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Competing Interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Availability of data and materials

Data will be made available on request.

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Legend to figures

Fig. 1. Spatial distribution of total PAH at different sampling locations.

Fig.2. Spatial distribution of Low Molecular Weight (LMW) and High Molecular Weight (HMW) PAH at the sampling locations.

Fig. 2A. Distribution of LMW-PAHs.

Fig. 2B. Distribution of HMW-PAHs

Fig. 3. Determination of PAH origin in sediment samples from the Goa coastline.

(P.C.: Petroleum combustion). $\text{Ant} / (\text{Ant} + \text{Phe}) < 0.1$ indicates contamination of Petrogenic origin, while the ratio of > 0.1 , indicates the Pyrolytic origin of LMW PAH. $\text{Flt} / (\text{Flt} + \text{Pyr}) < 0.4$ indicates contamination is Petrogenic, and the ratio of > 0.4 indicates Pyrolytic origin of HMW PAH.

Fig. 4. Correlation between PAH contamination in the sediment and adverse biological effects (< 0.1 – Low risk; $0.11 - 0.5$ – Medium-low risk; $0.51 - 1.50$ – Medium to high risk; > 1.50 – High priority risk).

Table 1. Concentrations of PAHs in the sediments collected from all the stations in the study

PAH	Concentration of PAHs ($\mu\text{g g}^{-1}$)																
	Nap	Ace	Acy	Flu	Phe	Ant	Flt	Pyr	B(A)Ant	B(A)Pyr	B(k)Flt	Db(ah)Ant	B(ghi)P	TPAH	Phe/Ant	Flt/Pyr	LMW/HMW
Stations	Mandovi Estuary sampling stations																
M1	20.5 \pm 2	27.5 \pm 2	21.87 \pm 3	24.52 \pm 2	24.46 \pm 1	---	22.62 \pm 1	21.76 \pm 1	22.62 \pm 1	20.95 \pm 0.4	22.15 \pm 1	23.03 \pm 0.89	21.28 \pm 1	273.33	---	1.0	0.76
M2	---	---	1.36 \pm 3	---	---	---	---	---	---	---	---	---	---	1.36	---	---	---
M3	---	---	2.4 \pm 1	---	---	---	---	---	---	---	---	---	---	2.42	---	---	---
M4	---	---	---	---	3.5 \pm 0.2	---	---	---	---	---	---	---	---	3.55	---	---	---
M5	---	2.1 \pm 0.6	1.8 \pm 0.09	---	---	---	---	---	---	---	---	---	---	3.96	---	---	---
M6	---	---	3.6 \pm 1	---	---	---	---	---	---	---	---	---	---	3.67	---	---	---
M7	---	0.8 \pm 0.01	---	---	---	---	---	---	---	---	---	---	---	0.84	---	---	---
M8	---	0.3 \pm 0.01	0.9 \pm 0.6	---	---	---	11.4 \pm 0.12	---	---	---	---	---	---	12.59	---	---	---
M9	4 \pm 0.23	2.5 \pm 1	3.3 \pm 1	---	1.8 \pm 1	3.1 \pm 1	---	---	---	---	---	---	---	15.07	---	---	0.1
M10	---	4.1 \pm 0.3	6.9 \pm 0.4	---	---	5.4 \pm 0.3	---	1.6 \pm 0.1	---	---	---	---	---	18.14	0.6	---	---
Total	24.84	30.51	42.25	24.52	29.86	8.52	23.41	22.62	22.62	20.95	22.15	23.03	21.28	334.93	---	---	9.9
	Zuari Estuary sampling stations																
Z1	---	---	1.3 \pm 0.1	---	65 \pm 6	51 \pm 5	15 \pm 1	165 \pm 2	---	---	---	---	---	298.21	1.3	0.09	0.6
Z2	---	---	1.2 \pm 0.8	---	47 \pm 6	37 \pm 4	---	76 \pm 3	---	---	---	---	---	161.00	1.2	---	1.1
Z3	---	---	1.5 \pm 0.1	---	41 \pm 8	34 \pm 6	13 \pm 2	187 \pm 8	---	---	---	---	---	277.74	1.2	0.07	0.4
Z4	---	---	1.9 \pm 1	---	302 \pm 10	224 \pm 10	16 \pm 1	9.8 \pm 0.5	---	---	---	---	---	552.80	1.3	1.6	20
Z5	---	---	1.6 \pm 1	---	36 \pm 5	30 \pm 3	394 \pm 10	12 \pm 0.6	---	---	---	---	---	473.68	1.2	33	0.16
Z6	---	---	---	---	213 \pm 10	158 \pm 10	15 \pm 0.9	14 \pm 0.5	---	---	---	---	---	399.96	1.3	1.04	13
Z7	---	---	1.6 \pm 0.9	---	43 \pm 7	36 \pm 5	21 \pm 2	222 \pm 12	---	---	---	---	---	325.23	1.1	0.09	0.33
Z8	---	---	1.5 \pm 0.09	---	49 \pm 4	40 \pm 3	398 \pm 10	14 \pm 0.4	---	---	---	---	---	501.61	1.2	29	0.21
Z9	---	---	1.7 \pm 0.07	---	423 \pm 10	312 \pm 10	45 \pm 2	7.7 \pm 4	---	---	---	---	---	789.71	1.3	5.9	13
Z10	---	---	1.7 \pm 1	---	205 \pm 8	154 \pm 6	26 \pm 1	186 \pm 7	---	---	---	---	---	572.37	1.3	0.13	1.4
Total	---	---	14.11	---	1425.33	1076.72	943	893	---	---	---	---	---	4352.365	---	---	---
	Sal River sampling stations																
SS1	---	---	15 \pm 5	---	18 \pm 5	19 \pm 5	---	---	---	---	---	---	---	52.51	0.97	---	---
SS2	---	---	9.8 \pm 0.64	3.3 \pm 0.09	---	---	---	---	---	---	---	---	---	13.16	---	---	---
SS3	---	---	2.2 \pm 0.04	---	33 \pm 2	34 \pm 2	14 \pm 0.9	98 \pm 2	---	---	---	---	---	181.23	0.97	0.14	0.6
SS4	---	---	2.4 \pm 0.2	---	33 \pm 3	36 \pm 5	181 \pm 10	10 \pm 0.89	---	---	---	---	---	262.92	0.92	17	0.37
SS5	---	---	1.5 \pm 1	---	18 \pm 0.8	19 \pm 0.9	---	178 \pm 4	---	---	---	---	---	216.28	0.91	---	0.22
SS6	---	---	---	---	4.2 \pm 0.35	4.2 \pm 0.3	89 \pm 4	4.3 \pm 0.2	---	---	---	---	---	101.66	1.01	20.66	0.09
SS7	---	---	---	---	25 \pm 3	26 \pm 3	156 \pm 9	8.4 \pm 0.7	---	---	---	---	---	215.53	0.96	18.51	0.31
SS8	---	---	---	---	226 \pm 10	228 \pm 10	18 \pm 2	26 \pm 3	---	---	---	---	---	499.26	0.99	0.71	10.17
SS9	---	---	1.2 \pm 0.1	---	91 \pm 12	92 \pm 10	13 \pm 2	15 \pm 2	---	---	---	---	---	213.40	0.99	0.86	6.57
SS10	---	---	1.4 \pm 1	1.9 \pm 0.005	---	---	---	---	---	---	---	---	---	3.41	---	---	---
Total	---	---	33.9	5.34	449.3	458.8	472.2	340.1	---	---	---	---	---	1759.37	---	---	---
	Galgibaga beach area sampling stations																

	Nap	Ace	Acy	Flu	Phe	Ant	Flt	Pyr	B(a)Ant	B(a)Pyr	B(k)Flt	Db(ah)Ant	B(ghi)P	TPAH	Phe/Ant	Flt/Pyr	LMW/HMW
GM1	0.16±0.007	---	0.9±0.6	---	---	---	---	---	---	---	---	---	---	1.1	---	---	---
GM2	---	---	4.6±0.009	---	---	---	---	---	---	---	---	---	---	4.60	---	---	---
GM3	0.38±0.04	---	6.7±0.3	---	21±1	21±1	---	---	---	---	---	---	---	49.50	1.01	---	---
GM4	---	---	1.06±0.75	---	2.2±0.1	2.1±0.1	---	---	---	---	---	---	---	5.62	1	---	---
GM5	---	---	0.9±0.6	---	---	---	---	---	---	---	---	---	---	0.93	---	---	---
GM6	---	---	2.3±0.2	---	13±1	14±0.9	3.9±0.1	145±2	---	---	---	---	---	179.19	0.92	0.02	0.19
GM7	---	---	4.4±0.8	---	64±10	66±10	32±4	28±3	---	---	---	---	---	193.99	0.95	1.14	2.26
GM8	---	---	4.5±0.9	---	58±4	59±5	23±1	15±0.8	---	---	---	---	---	159.84	0.97	1.46	3.17
GM9	---	---	3±0.1	---	48±0.6	50±0.9	13±0.9	252±10	---	---	---	---	---	365.46	0.96	0.05	0.38
GM10	---	---	2.5±0.1	---	22±0.6	25±0.8	16±0.6	13±0.2	---	---	---	---	2.0±0.1	80.66	0.90	1.12	1.60
Total	0.54	---	30.86	---	228.77	238.13	86.98	458.58	---	---	---	---	2.0	1040.87	---	---	---
Divar Island mangrove sampling stations																	
D1	---	---	6.9±1	---	124±9	129±10	464±10	22±1	---	---	---	---	---	748.39	0.96	20	0.53
D2	---	---	2.1±0.3	---	118±9	123±10	97±3	534±7	---	---	---	---	---	875.07	0.96	0.18	0.27
D3	---	---	4.4±1	4.6±3	86±10	88±9	32±10	452±10	---	---	5.8±2	---	---	674.54	0.97	0.07	0.27
D4	---	---	1.4±0.15	---	57±1	58±1	337±1	28±0.4	---	---	3.5±0.4	---	---	486.54	0.97	11.8	0.24
D5	---	---	0.98±0.6	0.89±0.6	48±10	45±4	173±10	156±10	---	---	---	---	---	426.80	1.05	1.11	0.22
Total	---	---	15.87	5.53	435.33	445.36	1104.9	1194.9	---	---	---	---	---	3211.36	---	---	---

Key: M1, M2, M3, M4, M5, M6, M7, M8, M9, M10 – Mandovi Estuary sampling stations

Z1, Z2, Z3, Z4, Z5, Z6, Z7, Z8, Z9, Z10 – Zuari Estuary sampling stations

SS1, SS2, SS3, SS4, SS5, SS6, SS7, SS8, SS9, SS10 – Sal River sampling stations

GM1, GM2, GM3, GM4, GM5, GM6, GM7, GM8, GM9, GM10 – Galgibaga beach area sampling stations

D1, D2, D3, D4, D5 – Divar Island mangrove sampling stations

--- Not detected

Nap – Naphthalene; Ace – Acenaphthene; Acy – acenaphthylene; Flu – Fluorene; Phe – Phenanthrene; Ant – Anthracene; Flt – Fluoranthene; Pyr – Pyrene; B(a)Ant - benzo (a)anthracene; B(a)Pyr - benzo(a)pyrene; B(k)Flt - benzo(k)fluoranthene; Db(ah)Ant - dibenzo (ah)anthracene; B(gh)P - benzo(ghi)perylene; Diagnostic ratios: Phe/Ant – Ratio of phenanthrene to Anthracene; Flt/Pyr ratio of Fluoranthene to Pyrene; LMW/HMW – the ratio of low molecular weight PAH to high molecular weight PAH

Supplementary Table 1: Details of sampling locations

Sampling date	Sampling station	Co-ordinates		Depth (m)
		Latitude	Longitude	
Mandovi estuary				
24-Aug-2018	M1	15.481111	73.788611	9.0
24-Aug-2018	M2	15.568889	74.020833	3.5
24-Aug-2018	M3	15.623333	74.002500	3.0
24-Aug-2018	M4	15.605000	74.060833	3.0
24-Aug-2018	M5	15.622222	74.125000	3.0
24-Aug-2018	M6	15.558056	74.125000	4.5
24-Aug-2018	M7	15.632778	74.206944	9.0
24-Aug-2018	M8	15.632850	74.201322	11.0
24-Aug-2018	M9	15.630398	74.201803	10.0
24-Aug-2018	M10	15.620049	74.209939	13.5
Zuari estuary				
3-Feb-2018	Z1	15.412056	73.902694	10.0
3-Feb-2018	Z2	15.410778	73.904111	5.0
3-Feb-2018	Z3	15.410722	73.898417	4.0
3-Feb-2018	Z4	15.418306	73.879667	4.0
3-Feb-2018	Z5	15.412556	73.833389	5.0
3-Feb-2018	Z6	15.410556	73.812139	7.0
3-Feb-2018	Z7	15.443028	73.805028	8.0
3-Feb-2018	Z8	15.420167	73.803806	9.0
3-Feb-2018	Z9	15.439444	73.835472	4.0
3-Feb-2018	Z10	15.424278	73.837944	4.0
Sal River				
8-May-2018	SS1	15.196050	73.970317	2.0

8-May-2018	SS2	15.194283	73.960933	3
8-May-2018	SS3	15.184933	73.962433	3.5
8-May-2018	SS4	15.175983	73.958833	2.5
8-May-2018	SS5	15.172700	73.949933	2.5
8-May-2018	SS6	15.163600	73.948333	2.5
8-May-2018	SS7	15.155583	73.952850	3.5
8-May-2018	SS8	15.211467	73.964933	2.0
8-May-2018	SS9	15.203817	73.959567	2.0
8-May-2018	SS10	15.149333	73.951083	3.5

Galgibaga

17-May-2018	GM1	14.962867	74.050933	0.5
17-May-2018	GM2	14.964083	74.053483	2.5
17-May-2018	GM3	14.965267	74.055817	1.5
17-May-2018	GM4	14.967617	74.058250	1.5
17-May-2018	GM5	14.969683	74.059800	4.0
17-May-2018	GM6	14.971367	74.062050	4.0
17-May-2018	GM7	14.972500	74.065000	2.0
17-May-2018	GM8	14.958017	74.052883	1.5
17-May-2018	GM9	14.958017	74.052883	2.0
17-May-2018	GM10	14.959367	74.049717	1.0

Divar Island mangrove

8-Jan-2019	D1	15.523322	73.913219	--
8-Jan-2019	D2	15.518112	73.910130	--
8-Jan-2019	D3	15.510669	73.900002	--
8-Jan-2019	D4	15.506368	73.884001	--
8-Jan-2019	D5	15.507526	73.875454	--
