

Emerging Pollutants in the Marine Coastal Environment of the World's Largest Ship Breaking Yard- Alang, India

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ABSTRACT

Since the last decade, organic contaminants in marine environment have been a major source of worry across the country. The aim of this study was to investigate the occurrence of emerging pollutants at Alang coast in Gujarat, India from February 2021 to December 2021, as well as to examine physico-chemical variables in marine waters and sediments. Gas chromatography-mass spectrometry (GC-MS) was used to detect the organic chemical contaminants from water and sediment. Results indicate that there were total 208 organic compound including 168 aliphatic and 40 aromatic hydrocarbon were present in marine water and sediment sample. Hundreds of ships including cargo vessels, oil tankers, passenger liners and warships have been dismantled and based on the results obtained we can concluded that the ship breaking activities carried out at Alang contrib-

ute a source of petrogenic hydrocarbon, plasticizer, alkane and alkanolic acid as well as it affect marine water quality.

Keywords GC-MS analysis, Emerging pollutant, Physico-chemical properties of marine water and sediment, Polycyclic aromatic hydrocarbon.

INTRODUCTION

Marine ecosystem faces a number of threats including climate change, sewage disposal, shipping, industrial chemical discharge, oil spills, industrial overgrowth, excessive modernization of agriculture and the development of maritime transport, antifouling coatings, mariculture and off-shore operations such as offshore oil and gas exploration and production and seabed mining as well as runoff from surface water, soils and groundwater (Chatterjee 2017), (Hanke 2016) which release a broad variety of synthetic or naturally occurring organic compounds that are not commonly monitored in the environment but have potential to enter into the environment and cause different adverse ecological and health effects are known as emerging contaminants (ECs) (Egbuna *et al.* 2021). Organic compounds are also known as contaminants of emerging concern (CECs), which are chemicals released into the environment for which no regulations are currently established (Shah *et al.* 2022). Most of these compounds were classified as persistent organic pollutants (POPs) because they persist in the environment, possess the ability to be transported over long ranges, biomagnify across the food chain, bioaccumulate in human and animal systems (Olisah *et al.* 2021) with a worldwide occurrence

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Fig. 1. Map showing study area.

in the marine environment because of their resistance to environmental degradation.

The high lipophilicity makes them capable to bioaccumulate in large quantities in tissues of marine organisms (Agostino *et al.* 2020). Since the latter half of the twentieth century, the use of synthetic chemicals, such as cosmetics, insecticides, personal care products, medicines, and steroid hormones, has increased exponentially (Pintado-Herrera *et al.* 2017) as they are essential for contemporary societies around the world. Each year, the global output of these pollutants is expected to rise from 1 million to 500 million tonnes (Khan *et al.* 2022). On May 17, 2001, a convention on POPs was sanctioned and entered into force by 152 countries in Stockholm, Sweden (<https://www.unido.org>). The main goal of this convention is to protect humans and the environment from POP contamination, with the intent of eventually eliminating them. The toxic effects of some of these POPs on

humans and the environment became apparent soon after they were widely released into the environment in the 1960s and 1970s, prompting legal action against their production and use (Pintado-Herrera *et al.* 2017). These chemicals include organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polybrominated diphenyls (PBDEs) (Olisah *et al.* 2021) which are also ubiquitous in estuarine waters, sediments and biota (Anim *et al.* 2017). Thakur and Patharia (2020) found that the highest levels of POPs are found in marine mammals, which lead to vitamin and thyroid deficiencies and cause microbial infections and reproductive disorders. In 2020 Qiu *et al.* (2020). examine the concentrations of persistent organic pollutants (POPs) in sediments sample of the Eastern Indian Ocean by the help of GC-MS to learn more about contamination, distribution, and potential sources and risks. Lalwani *et al.* (2020) investigate the presence of Bisphenol A (BPA) in surface waters worldwide and its estrogenic effects in humans are

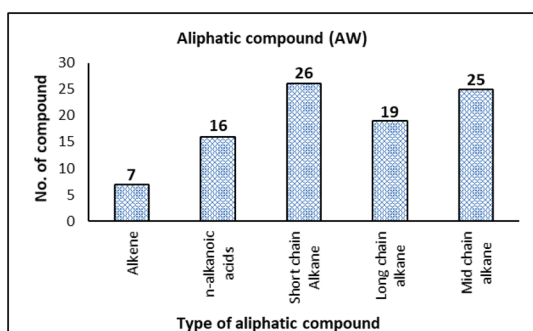


Fig. 2. Different type of aliphatic compound present in water sample.

well documented. Ruan *et al.* (2019) carried out first nationwide biomonitoring of PFAS in Indians. In 2018 Gosai *et al.* (2018) investigated the levels of PAHs, their input prediction and potential risks to bacterial abundance and human health along Gujarat coastline. Therefore, the present work was carried out to investigate the status of emerging pollutants with marine water and sediment quality at world's largest shipbreaking yard Alang as well as to identify the possible source of all detected organic compound. Same work was also carried out by Shah *et al.* (2022) to investigate the occurrence and distribution of organic compounds in nine cities of Gujarat, India.

Experimental methodology

Site description

Alang-Sosiya ship-scraping yard is one of the largest ship-scraping yards in the world. Geographically it is situated 21°5' 21°29' towards the north and 72°5' 72°15' towards the east on the western coast of the Gulf of Khambhat (Fig. 1). Alang has 153 plots or ship-breaking yards developed on a 10-kilometer long coast in Bhavnagar district. The yard has a gentle slope of around 10 degrees with a firm and hard rocky bottom, which is suitable for bringing ships right up to the scrapping yard afloat with minimum investment and risk factors. Nearly half of the world's ocean-going ships are being dismantled and recycled in India, of which around 95% are scrapped in Alang-Sosiya yards. Annually, hundreds of ships mainly including cargo vessels, oil tankers, passenger liners, and warships have been dismantled, which adds up to millions of Light Displacement Tonnage. Mithi

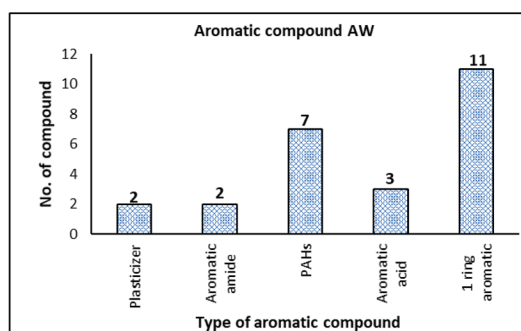


Fig. 3. Different type of aromatic compound present in water sample.

Virdi is a proposed site consisting of six reactors with a total capacity of 6,600 MW about 3 km (2 mi) north of the ship breaking beach.

Sample collection

The samples of coastal surface sediments were collected from the depth of 40 to 50 cm in plastic bags while coastal surface seawater samples were collected in clean bottles during February 2021 to December 2021 on bimonthly basis. The sediment and seawater samples were collected at low tides from the intertidal zone. Each collected sample was used for organic compound analysis as well as for heavy metal analysis.

Extraction of organic pollutants from water and sediments

Marine water and sediment sample extraction

Liquid-Liquid extraction (LLE) is a method to separate compounds based on their relative solubilities in different immiscible liquids. In the LLE procedure, the 800 mL of water sample poured into a separatory funnel and the mixture of 100 mL n-hexane and dichloromethane (1:1 v/v) was added and shaken for 2 min. The water phase was drained and then the organic phase was poured into a glass funnel containing 20 g of anhydrous sodium sulfate and re-extracted with 50 ml of the same solvent mixture. The extract was concentrated prior for organic compound analysis (Mahgoub 2016). Marine sediment sample was air dried followed by oven dried to absorb excessive moisture after drying of sediment the sample was

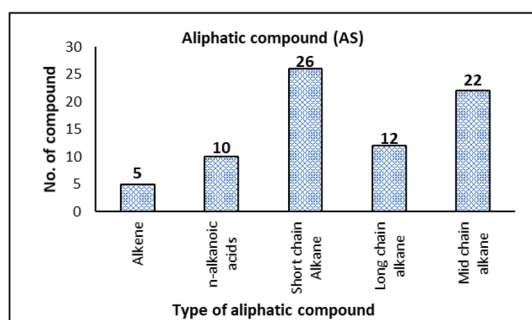


Fig. 4. Different type of aliphatic compound present in sediment sample.

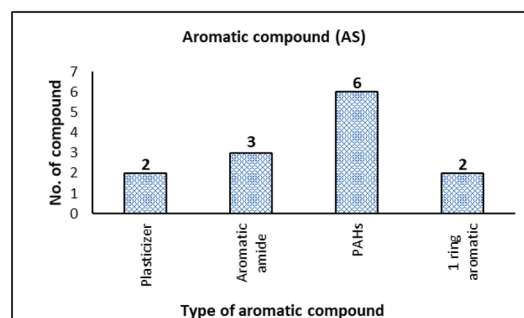


Fig. 5. Different type of aromatic compound present in sediment sample.

grounded in mortar and pestle then sieved through 5 mm sieve. 2 g of finely grounded sample was taken in PP tub then 2 mL of extraction solvent mixture of Methanol and Dichloromethane (DCM) (1:1) was added to same tube followed by ultrasonication (Sonicator USA) and vortex mixing then the extract was concentrated in a hot water bath for up to 2 mL and kept in the refrigerator until the analysis (Giri *et al.* 2013).

GC-MS analysis of sample

The extracted marine water and sediment samples were analyzed by using GC 2010 plus coupled with a Shimadzu Mass Spectrometer at the Central Salt and Marine Chemicals Research Institute, Bhavnagar, Gujarat with a flow rate of 1 mL/min, a cone volt-

age of 0.3 kV and an ion source temperature of 300°C, good quality helium gas was employed as the carrier. It's worth noting that all of the identified aliphatic and aromatic chemical values were calculated using the chromatogram's peak height percent. For each chemical, chromatogram from Library : NIST17. lib m/z and pub Chm were used to identify it. The interference was removed by performing a blank during the organic chemistry analysis.

Physico-chemical parameter

Marine water samples and sediments from the selected site were collected simultaneously in clean TARSON bottles and analyses were carried out as per the standard methodology outlined in Strickland and Parsons (1972) and American Public Health

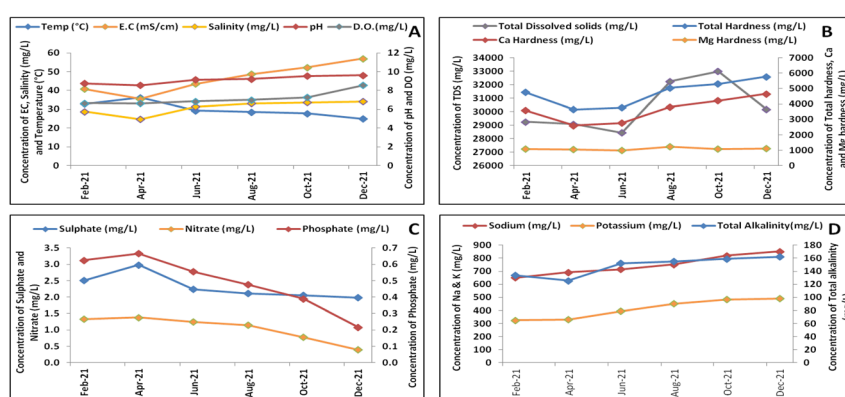


Fig. 6. A) C Temp., EC, salinity, pH and DO B) Total dissolved solids, total hardness, calcium and magnesium hardness C) Sulfate, phosphate and nitrate sample D) Sodium, potassium and alkalinity of water sample.

Table 1. Organic compounds present in marine water of Alang.

Peak area (%)	No. of compound	Dominant compound name	Structure	Functionality	Structure
0.05 to 0.19	34	Tridecane, 4-methyl-		Alkane	$C_{14}H_{30}$
		1,8-Dioxa-5-thiaoctane, 8-(9-borabicyclo[3.3.1]non-9-yl)-3-(9-borabicyclo[3.3.1]non-9-yl)-1-phenyl-		Aromatic	$C_{27}H_{42}B_2OS$
		Carbonic acid, eicosyl vinyl ester		n-Alkanoic acid	$C_{23}H_{44}O_3$
		Methoxyacetic acid, 3-tridecyl ester		n-Alkanoic acid	$C_{16}H_{32}O_3$
		1-(4-Chlorophenoxy)-1-(1H-imidazol-1-yl)-3,3-dimethylbutan-2-one		Aromatic ketone	$C_{15}H_{17}ClN_2O_2$
0.2 to 0.35	47	Triacontane, 1-bromo-		Alkane	$C_{30}H_{61}Br$
		Cetene		Alkene	$C_{16}H_{32}$
		4-Methylnonanoic acid		n-Alkanoic acid	$C_{10}H_{20}O_2$
		4-[4-Chlorophenyl]-1-methyl-.alpha.-[3-[2-methyl-2-butoxy]propyl]-4-piperidine methanol		PAHs	$C_{21}H_{34}ClNO_2$
		Silane, trichlorooctadecyl-		Alkane	$C_{18}H_{37}C_3Si$
		Disulfide, di-tert-dodecyl		Alkane	$C_{24}H_{50}S_2$
		Sulfurous acid, 2-propyl tridecyl ester		n-Alkanoic acid	$C_{16}H_{34}O_3S$
		Ethane, 1,2-Bis(1-phenylcyclopropyl)-		PAHs	$C_{20}H_{22}$
		Geranyl isovalerate		Alkene	$C_{15}H_{26}O_2$
0.36 to 0.55	26	Benzenesulfonic acid p-fluoro, 3,5-dichloro-2,6-dimethyl-4-pyridyl ester		Aromatic acid	$C_{13}H_{10}Cl_2FNO_3S$

Table 1. Continued.

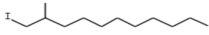

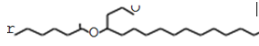
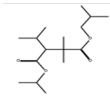
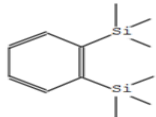
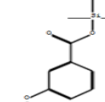
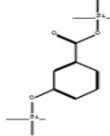
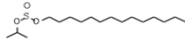
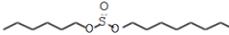
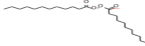



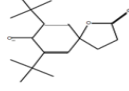
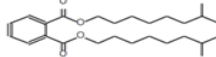


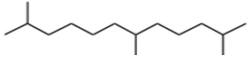

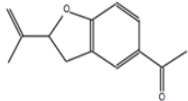
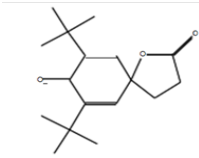
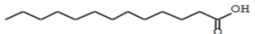
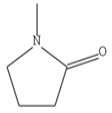
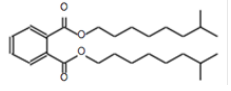


Peak area (%)	No. of compound	Dominant compound name	Structure	Functionality	Structure
		1-Iodo-2-methylundecane		Alkane	$C_{12}H_{25}I$
		9-Eicosene, (E)-		Alkene	$C_{20}H_{40}$
		6-Bromohexanoic acid, 4-hexadecyl ester		n-alkanoic acid	$C_{22}H_{43}BrO_2$
		Pentanoic acid, 2,2,4-trimethyl-3-carboxyisopropyl, isobutyl ester		n-alkanoic acid	$C_{16}H_{30}O_4$
		1, 2-Bis (trimethylsilyl) benzene		Aromatic silyl	$C_{12}H_{22}Si_2$
		3-Hydroxybenzoic acid, 2 TMS derivative		Aromatic acid	$C_{13}H_{22}O_3Si_2$
0.56 to 1	05	Sulfurous acid, 2-propyl tetradecyl ester		n-alkanoic acid	$C_{17}H_{36}O_3S$
		Sulfurous acid, hexyl octyl ester		n-alkanoic acid	$C_{14}H_{30}O_3S$
		Dotriacontane, 1-iodo-		Alkane	$C_{32}H_{65}I$
		Lauroyl peroxide		Alkane	$C_{24}H_{46}O_4$
		Hexacosane, 1-iodo-		Alkane	$C_{26}H_{53}I$
Above 1	07	Octacosane		Alkane	$C_{28}H_{58}$
		Tetratetracontane		Alkane	$C_{44}H_{90}$
		7,9-Di-tert-butyl-1-oxaspiro (4,5)deca-6,9-diene -2,8-dione		PAHs	$C_{17}H_{24}O_3$
		Phthalic acid, bis (7-methyloctyl)ester		Plasticizer	$C_{26}H_{42}O_4$
		Heneicosane		Alkane	$C_{21}H_{44}$
		2-methyloctacosane		Alkane	$C_{29}H_{60}$
		Dodecane, 2,6,11-trimethyl-		Alkane	$C_{15}H_{32}$

Table 2. Organic compounds present in marine sediment of Alang.

Peak area (%)	No. of compound	Dominant compound name	Structure	Functionality	Structure
0.05 to 0.19	29	Tetracosane		Alkane	$C_{24}H_{50}$
		Nonahexacontanoic acid		n-alkanoic acid	$C_{69}H_{138}O_2$
		4-Hydroxy-4-methylhex-5-enoic acid, tert.-butyl ester		n-alkanoic acid	$C_{11}H_{20}O_3$
		5, 5-Diethylheptadecane		Alkane	$C_{21}H_{44}$
		Benzenesulfonyl fluoride, 4-(hexadecyloxy)-3- nitro-		Aromatic	$C_{22}H_{36}FNO_5S$
0.2 to 0.35	29	Heptacosane, 1-chloro-		Alkane	$C_{27}H_{55}Cl$
		Sulfurous acid, octadecyl pentyl ester		n-alkanoic acid	$C_{23}H_{48}O_3S$
		Oxalic acid, 6-ethyloct-3-ylheptyl ester		n-alkanoic acid	$C_{19}H_{36}O_4$
		Carbonic acid, decyl undecyl ester		n-alkanoic acid	$C_{22}H_{44}O_3$
		1-(2-Methylbutoxy)-7-isoheptyl-2,2,4,4,6,6-hexamethyl-1,3,5,7-tetraoxa-2,4,6-trisilaheptane		Alkane	$C_{17}H_{42}O_4Si_3$
0.36 to 0.55	20	Bis (tridecyl) phthalate		Plasticizer	$C_{34}H_{58}O_4$
		Octadecanoic acid		n-alkanoic acid	$C_{18}H_{36}O_2$
		2-Isopropyl-5-methyl-1-heptanol		Alkane	$C_{11}H_{24}O$
		Sulfurous acid, hexy octyl ester		n-alkanoic acid	$C_{14}H_{30}O_3S$
		Heptacosane		Alkane	$C_{27}H_{56}$
0.56 to 1	04	Octacosane, 1-iodo-		Alkane	$C_{28}H_{57}I$
		Heptadecane		Alkane	$C_{17}H_{36}$

Table 2. Continued.

Peak area (%)	No. of compound	Dominant compound name	Structure	Functionality	Structure
		Octyl tetradecyl ether		Alkane	$C_{22}H_{46}O$
		(+/-)-Tremetone		PAHs	$C_{13}H_{14}O_2$
		7, 9-Di-tert-butyl-oxaspiro (4, 5) deca-6, 9-dien-2, 8-dione		PAHs	$C_{17}H_{24}O_3$
		Tridecanoic acid		n-alkanoic acid	$C_{13}H_{26}O_2$
Above 1	06	2-Pyrrolidinone, 1-methyl-		Aromatic	C_5H_9NO
		Phthalic acid, bis (7-methyloctyl) ester		Plasticizer	$C_{26}H_{42}O_4$
		Eicosanoic acid		n-alkanoic acid	$C_{20}H_{40}O_2$
		Undecane, 2-methyl-		Alkane	$C_{12}H_{26}$

Association 23rd edition (2017) (APHA).

RESULTS AND DISCUSSION

Occurrence of organic compounds

GC-MS analysis of water and sediment sample shows the presence of aliphatic and aromatic hydrocarbon which includes alkane, alkene, ketones, ester, alcohol, n-alkanoic acid, PAHs, halogenated hydrocarbon, plasticizer. A total of 119 organic chemical compounds were detected in water sample of Alang among which 95 aliphatic compounds (Fig. 2) and 24 aromatic compounds (Fig. 3) has been detected whereas 88 organic chemical compounds were detected in sediment sample of Alang out of which 76

aliphatic compounds (Fig. 4) and 12 aromatic (Fig. 5) compounds has been detected.

Total aliphatic hydrocarbon (TAHs) present in study area

The petrogenic hydrocarbon in marine sediments and water may have originated from a variety of hydrocarbon sources, both naturally occurring and anthropogenically produced. Oil can enter marine systems naturally through petroleum extraction, crude oil transportation, sewage discharge, and roadside runoff, as well as through natural seeps or erosion of carbon-rich formation. Out of 95 aliphatic compounds 72 n-alkane compounds including short chain, mid chain and long chain (C₅₄ to C₅), 16 alkanolic

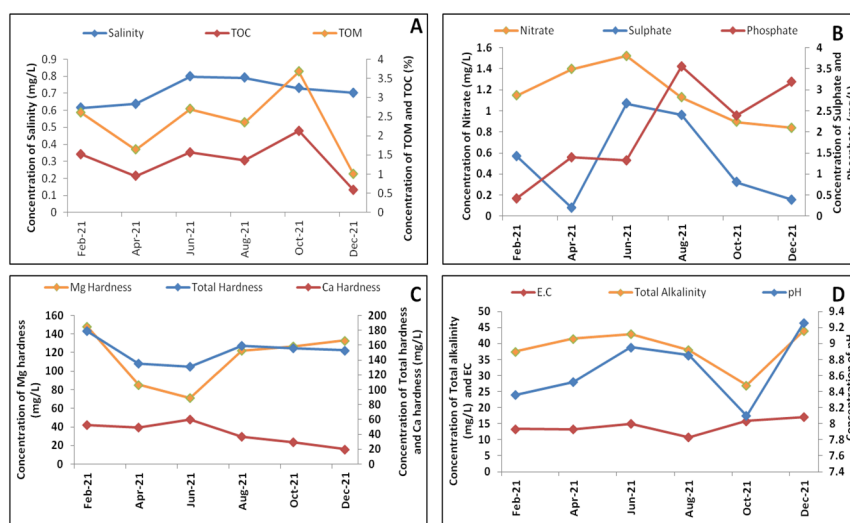


Fig. 7. A) Salinity, TOC and TOM B) Sulfate, phosphate and nitrate sample C) Total hardness, calcium and magnesium hardness D) EC, pH and total alkalinity of sediment sample.

acid compounds and 7 alkene (Fig. 2), compound were detected in marine water and out of 76 aliphatic compounds 60 n-alkane compounds including (C40 to C5), 10 alkanic acid and 5 alkane compound (Fig. 4) were detected in marine sediment sample. Short chain alkane were predominant in both the sample as marine phytoplankton produces mostly short-chain n-alkanes, mostly n-C15, n-C17 and n-C19 as well as fossil fuel input contribute to the production of short-chain n-alkanes (Gao *et al.* 2021). Based on GC-MS result organic compound are separated in 5 different group based on their peak height (%). Among alkane compounds Octacosane, 2-methyl-octacosane, Tridecanol, 2-ethyl-2-methyl-, Hexacosane, Heneicosane, Dotriacontane, 1-iodo-, Lauroyl peroxide, Octadecane, 1,1'-[1,3-propanediylbis(oxy)] bis were dominant in marine water sample of Alang (Table 1) whereas 7,9-Di-tert-butyl-1-oxaspiro(4,5) deca-6,9-dien, Tridecanoic acid, 2-Pyrrolidinone, 1-methyl-, Phthalic acid, bis (7-methyloctyl) ester, Eicosanoic acid, Undecane, 2-methyl-, Octacosane, 1-iodo-, Heptadecane were predominant in sediment sample of Alang (Table 2). According to a recent study, incomplete combustion of fossil fuels and petroleum residues, wood combustion and ceramic dust might be the major source for n-alkanes in the cold season (Boreddy *et al.* 2018). Short chain al-

kanoic acid like Sulfurous acid, 2-propyl tetradecyl ester, Sulfurous acid, hexyl octyl ester, 6-Bromohexanoic acid, 4-hexadecyl ester, Pentanoic acid, 2,2,4-trimethyl-3-carboxyisopropyl, isobutyl ester, Sulfurous acid, dodecyl 2-propyl ester were predominant in marine water (Table 1) whereas Tridecanoic acid, Eicosanoic acid, Octadecanoic acid, Sulfurous acid, hexyl octyl ester, Sulfurous acid, octadecyl pentyl ester, Oxalic acid, 6-ethyloct-3-yl heptyl ester were dominant in sediment sample (Table 2).

Aromatic compound present in study area

Aquatic contaminants like pesticides, polycyclic aromatic hydrocarbons, polychlorinated biphenyls, organophosphorus esters, and phthalate esters have been reported to be released and dispersed by microplastic (Gugliandolo *et al.* 2020). The group of xenobiotic chemicals known as polycyclic aromatic hydrocarbons (PAHs) is composed of carbon and hydrogen. They are a class of contaminants that are frequently found in coastal marine environments that are impacted by industry and shipping traffic. They have high melting and boiling points, low vapour pressure, and very low water solubility. 4-[4-Chlorophenyl]-1-methyl-.alpha.-[3-[2-methyl-2-butoxy]propyl]-4-piperidinemethanol, Pyridate, Ethane,

1,2-Bis(1-phenylcyclopropyl)- were dominant in marine water sample (Table 1) and (+/-)-Tremetone, Menthyl salicylate, 4-[2-(4-Fluorophenyl)ethyl] piperidine were detected in sediment sample (Table 2), whereas 7,9-Di-tert-butyl-1-oxaspiro(4,5)deca-6,9-diene -2,8-dione was found in water and sediment sample with maximum peak height. Aromatic amine including trans-2,4,5-Trimethoxy-.beta.-methyl-.beta.-nitrostyrene and 2-Piperidinone, N-[4-bromo-n-butyl]- were detected in marine water sample and 2,5-Dimethoxy-4-propoxy-.beta.-methyl-.beta. and Azacycloheptane, 3-amino-1-t-butoxycarbonyl were detected in sediment sample which represent a category of chemical agents of considerable importance as witnessed by their widespread use as intermediates in the manufacture of drugs, pesticides and plastics, as antioxidants in the preparation of rubber for the manufacture of tires and cables and as curing agents in the preparation of various plastics. They are also widely used as precursors in the preparation of dyes and pigments, which are used to color a variety of products including textiles, leather, rubber, printing inks, paints, lacquers, metal finishes, plastic, and paper as well as semi-permanent coloring materials (Fishbein 1984). In marine the most abundant and frequently detected plasticizer were dibutyl phthalate, diisobutyl phthalate, di (2-ethylhexyl)phthalate, di(2-ethylhexyl) terephthalate confirming that such compounds bioaccumulate through the food chain (Gugliandolo *et al.* 2020).

Physico-chemical parameter of water and sediment

During the study period marine water temperature found highest during April (36.2°C) and lowest in December (24.9°C). Salinity ranged between to 24.7 to 34.1%, maximum recorded in the month of December whereas minimum encountered in April. The salinity is greatly influenced by the runoff of surface water, so that the lowest salinity was observed during monsoon season (Kumar *et al.* 2009). The pH recorded maximum during December and less in April. The Maximum Electrical conductivity was recorded in December (57 mS/cm). Ions and dissolved compounds had the biggest impact on EC. The increase in conductivity may be caused by the water's high ion concentration whereas dissolve oxygen is

varying from 6.6 to 8.5 mg/L (Fig. 6A). Calcium hardness was ranged from 2600 to 4658 mg/L, maximum calcium hardness was recorded in December. Maximum total dissolved solid was found in October 32985 mg/L (Fig. 6B). Phosphate, Nitrate and Sulfate content were encountered greater in the month of April i.e., 0.666 mg/L, 1.381 mg/L and 2.981 mg/L respectively (Fig. 6C) same range of phosphate and nitrate was also observed by Barot (2017). Alkalinity was found maximum in December 162 mg/L. Kumar *et al.* (2009) also reported same range of alkalinity in surface water of estuary area. Na and K content was range between 651 to 852 mg/L and 325 to 492 mg/L respectively (Fig. 6D).

Salinity of sediment ranged between to 0.61 to 0.79 mg/L, minimum recorded in the month of February. Total organic carbon content ranged between 0.58 to 2.13 % (Fig.7A). Maximum sulfate and nitrate content found in June i.e., 2.66 and 1.52 mg/L respectively. Phosphate was encountered greater in the month of August (3.55 mg/L) (Fig. 7B) Barot (2017) also reported elevated concentration of phosphate during monsoon season. Calcium and total hardness ranged from 19.9 to 59.8 mg/L and 131 to 179 mg/L respectively (Fig. 7C). Alkalinity was found maximum 44 mg/L in December. The pH recorded maximum during December (9.26) and minimum in October. The Maximum Electrical conductivity was recorded in December (17.1 mS/cm) (Fig.7D).

CONCLUSION

The organic chemical analysis was done using GC-MS instrument and were grouped by indices scale based on the peak height (%) and total of 208 compounds were detected and different distribution patterns were observed in marine water and sediment sample. This study provides vital information about occurrence and distribution of the organic pollutants in world's largest ship breaking yard-Alang where Octacosane, Tetratetracontane, Heneicosane and 2-methyloctacosane were found to be dominant in water sample and Tridecanoic acid, 2-Pyrrolidinone, 1-methyl-, were found in sediment sample whereas 7,9-Di-tert-butyl-1-oxaspiro(4,5)deca-6,9-dien and phthalic acid, bis (7-methyloctyl) ester detected in both the sample with maximum peak height which

demands further detail study on spatial, temporal variations and its impact on health issues. Based on result we can conclude that the marine water quality was greatly influenced by seasonal variation as well as it affected by various anthropogenic activities carried out at shipbreaking yard.

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