

INVESTIGATION OF POLYCYCLIC AROMATIC HYDROCARBONS (PAHs) CONTAMINATION IN THE AIROLI MANGROVE SEDIMENTS USING ENVIRONMENTAL MAGNETIC METHODS

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ABSTRACT

In present article polycyclic aromatic hydrocarbons (PAHs) are analyzed due to their persistent as pollutants in marine ecosystem. These hydrocarbons may be responsible for long-term adverse effect to the mangrove ecosystem. Polycyclic aromatic hydrocarbons in mangrove sediments are analyzed using chemical and environmental magnetic methodology. Concentration, depth of contamination and sources of 16 PAHs observed in Airoli, Navi Mumbai mangrove sediments are investigated. Ten sediment samples from different depths were analyzed and presence of 16 polynuclear aromatic hydrocarbons (PAHs) is detected via gas chromatography/mass spectrometry. Sixteen PAHs i.e. Naphthalene (NAP), Acenaphthalene (ACY), Acenaphthene (ACN), Fluorine (FL), Phenanthrene (PHT), Anthracene (ANT), Fluoranthene (FLA), Pyrene (PYR), Benz (a) Anthracene (BaA), Chrysene (CHR), Benzo (k) Fluoranthene (BkF), Benz (e) Acephenanthrylene (BeA), Ideno (1, 2, 3, -cd) Pyrene (INP), Dibenz (a, h) Anthracene (DBA), Benzo (a) Pyrene (BAP) and Benzo (ghi) Perylene are identified in most of the samples. Environmental magnetic parameters magnetic susceptibility (χ), anhysteretic remanent magnetization, Saturation Isothermal Remanent Magnetization and useful ratios of some required parameters are used to ascertain grain size of the magnetic mineral assemblage. Present article is an attempt to discuss importance of integrated study that the mineral magnetic measurements along with chemical analyses may be of great value to understand environmental conditions that prevailed in the mangrove sediments.

Keywords: Polycyclic Aromatic Hydrocarbons (PAHs), Mangrove, Sediments, Susceptibility.

1. INTRODUCTION:

Polycyclic Aromatic Hydrocarbons have been found in marine, lacustrine, and fluvial sediments around the globe and are well documented class of chemicals (Neff, 1980). These hydrocarbons enter the environment in spills and seepages of crude oil and refinery products. PAHs may be produced *in situ* through diagenetic transformations of plant materials. For results of pyrolytic processes and anthropogenic (Lipiqtou, 1991) unsubstantiated (non alkylated) compounds are produced by high temperature combustion of fossils fuels. Also, these compounds are detected in refined lead free petroleum (Westerholm, et al., 1988). Typically, unsubstantiated (non alkylated) compounds are the most abundant PAHs found in surficial sediments. For analysis of changes in pollution inputs over time in marine sediments several researchers have used variations in the distribution of Pyrolytic PAHs compounds (Gschwend, P.M. and Hites, R.A., 1981; Baumard, P., et al., 1999).

The structure of PAHs contains two or more fused six member carbon atom aromatic rings e.g. naphthalene, benzopyrine they have low water solubility. The molecular weight of PAHs is directly proportional to the number of benzene rings. Most of them are solid with increase in melting point with molecular weight (Jones and Leber, 1979). They have low vapor pressure and are formed in environment which is oxygen deficient. Incomplete pyrolysis or high temperature pyrolytic process during combustions of fossil fuels may result formation of PAHs. Natural process like pyrosynthesis may also form PAHs. Thus it can be estimated that PAHs are the constituents of the products of incomplete combustion (Kulkarni and Venkataraman, 2000; Daisey, J.M., et al., 1979).

On the other hand, mangroves provide habitat for various marine and terrestrial flora and fauna (Kumaran, K.P.N. et al., 2004). In environmental magnetism sediments are considered to retain records of past environmental and climatic changes in natural archives (Thompson and Oldfield, 1986). Iron is one among the most common available elements in the Earth's crust. Hence, magnetic minerals, mainly iron oxides occur more or less in the

world. These oxides can be detected rapidly, easily and non-destructively by magnetic mineralogy (Thompson, R., et al., 1980).

Because all matter consists of atoms with circulating charged particles. Therefore, everything in this universe is magnetic. So, we can describe objects by their magnetic properties. For this study we have considered magnetic properties of environmentally important minerals, like those of iron-ferromagnetism. Although, there is no completely reliable methodology to differentiate between biogenic and abiogenic magnetic materials (Stolz, et al., 1990; Chang and Kirschvink, 1989). The aim of present study is to investigate present and past environmental and climatic conditions at Airoli, Navi Mumbai that prevailed in the past along with chemical analyses.

2. METHODOLOGY:

2.1 Sampling: The study was carried out on soil sedimentary cores from mangrove sedimentary area in Airoli, Navi Mumbai, Maharashtra. Two sedimentary profiles were selected adjacent localities, The site was excavated for construction work about 200 cm depth below it was hard bed rock. Surface soil was removed because it was land filled. Core portion cleaned and muddy sites mangrove sediments samples were taken at different depths using Livingstone type piston corer. Ten samples were analyzed spanning depth from 5 cm to 180 cm by GC/MS technique adopted by (APHA-6440). All ten samples were dried at 40°C. To remove coarse materials these samples were grinded and sieved.

Samples used in environmental magnetism measurements were prepared by drying the soil samples in porcelain containers in an electric oven for about 72 hr. The samples were heated at temperature range of 40°C and not more than that. The dried samples were later on powdered and then weighed and filled in the 10 mm plastic sample bottles. Powdered soil was wrapped in a thin polyethylene film, which was practically nonmagnetic. This was to prevent any movement of the sample particles while measurements.

2.2 Instrumentation: Analyses of PAHs Sample was done by GC/MS (VARIAN SATURAN 2200) with split/split less injection system and capillary column. Condition is as below-

GC/MS	:	VARIAN GC/MS SATURAN 2200
Column	:	Capillary column DB-5
Carrier gas	:	Helium 30ml/min
Injector temperature	:	250°C
Detector mass range	:	50-450 amu Detector
FID Oven	:	125°C hold for 2 min at rate of 10°C/min.

For mineral magnetic studies the AGICO KLY4S Kappabridge was used for magnetic susceptibility measurements. ARM measurements for intensity of 0.05mT were also carried out on the unoriented samples using the ASC Scientific D2000 AF demagnetizer. IRM measurements were carried out using a combination of the Molspin pulse magnetizer and Molspin spinner.

2.3 Extraction: A measured quantity of sample (approx. 10 gm) was extracted. For extraction solid liquid partitioning method was used using suitable solvent such as Methylene Dichloride. Concentrated extract to small volume was cleaned up by using rotary evaporator. Residues of polycyclic aromatic hydrocarbons is reconstituted to 2 ml, again by dissolving into suitable solvent hexane and injected on GC-MS- APHA (AMERICAN PUBLIC HEALTH ASSOCIATION 6440) method.

2.4 Chemicals: All chemicals used were of pure quality and were of branded manufactures like sigma Aldrich and Merck. A working stock solution was prepared from PAHs compound mix procured from ACCU standards containing 10 µg of each in 1ml methanol.

3. SITE LOCATION:

The geographic location of the sampling site is 19°10'41" N, 72°59'48" E at Airoli a suburban of Navi Mumbai, Maharashtra state in India. The site location is situated adjacent to the mangrove forests which cover the banks of the Thane Creek (fig.1).

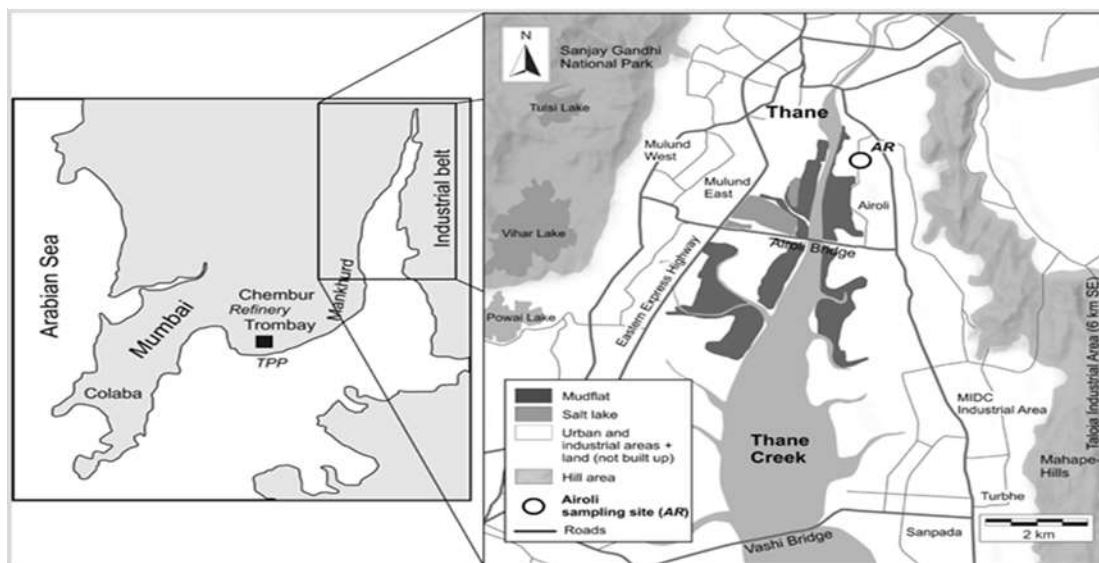


Fig.1-Maps of Mumbai city with the Trombay power plant (TPP) and Thane Creek (THC) with the Airoli sampling site.

The industrial area, eastern express highway, steel plant, industries are adjacent to the eastern side of Thane creek. A steel plant of Mukand Iron & Steel is also situated to the North-east of the site. These are the sources of sediment pollution (Kulkarni and Venkataraman, 2000). The selected site was previously under the mangrove cover and had been cleaned for the construction of the MIDC, IT PARK. The total length of the excavated core is ~2 m. It is interesting to observe that the area is situated on the banks of the Thane Creek which is connected to the sea, and hence is subjected to the transgressions of the sea. The Parsik Hills, Kharegon, Kalwa is situated near by to the East of the creek, these hills are almost barren and hence is a probable source of input into the creek. All these sediments seem to be entrapped in the rigid root framework of the mangroves.

4. ENVIRONMENTAL MAGNETIC PARAMETERS:

In the late 1970s Oldfield and his coworkers proposed concept of using magnetic properties as proxy parameters and correlation tool in environmental context. An article in journal 'Science' (Thompson et al., 1980) generally reflects formal definition of environmental or mineral magnetism. Determination of regional relationship between low field susceptibility and percentage carbonate is another approach to detect reduction of PAHs in marine sediments. Comparison of the ratio ARM/SIRM with the susceptibility (Maher and Taylor, 1988) resolves the problem of poor discrimination. Simultaneous matching of magnetic data is a non-trivial exercise (Martinson et al., 1987). Variations in the particle size of the magnetic mineral in mangrove sediments are primarily reflected in interparametric ratios ARM/ χ and SIRM/ χ . Ratio ARM/ χ and SIRM/ χ vary inversely with magnetic particle size (Bolemendal et al., 1988; Maher and Taylor, 1988). S-ratio is used to determine proportion of higher coercive magnetic minerals to lower coercive magnetic minerals in sediments material. High proportion of magnetite in material is indicated by high values of S-ratio ~1.0 and increasing proportion of hematite and goethite is indicated by lower values of S-ratio (Robinson, 1990; Bolemendal et al., 1988; Thompson and Oldfield, 1986). In 1982, Kent first demonstrated strong correlation between variations in magnetic parameters from marine sediments.

5. DISCUSSION:

5.1 Chemical Analyses: Compounds Naphthalene, Acenaphthalene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz (a) Anthracene, Chrysene, Benzo (k) Fluoranthene, Benz (e) Acephenanthrylene, Ideno (1, 2, 3, -cd) Pyrene, Dibenz (a, h) Anthracene, Benzo (a) Pyrene and Benzo (ghi) Perylene are identified and quantified in Airoli mangrove sediments. Concentrations of these compounds at different core depths are given in Table 1. Total PAHs concentrations at 5, 25, 45, 65, 85, 110, 120, 130, 150, 170 cm depths are 0.320, 0.266, 0.215, 0.1238, 0.036, 0.01348, 0.00314, 0.00433, 0.00404, 0.00406 $\mu\text{g g}^{-1}$.

Table 1: PAHs variation of concentration ($\mu\text{g g}^{-1}$) with depth (cm)

Name	Depth 5 cm	Depth 25 cm	Depth 45 cm	Depth 65 cm	Depth 85 cm	Depth 110 cm	Depth 120 cm	Depth 130 cm	Depth 150 cm	Depth 170 cm
NAP	0.020	0.017	0.013	0.0087	0.0021	0.00084	0.00014	0.00021	0.00026	0.00029
ACY	0.019	0.017	0.013	0.0072	0.0027	0.00084	0.00012	0.00022	0.00025	0.00022

ACN	0.019	0.017	0.014	0.0073	0.0021	0.00096	0.00012	0.00027	0.00028	0.00031
FL	0.021	0.017	0.013	0.0081	0.0020	0.00085	0.00010	0.00030	0.00032	0.00030
PHT	0.022	0.016	0.013	0.0087	0.0020	0.00081	0.00016	0.00025	0.00024	0.00032
ANT	0.020	0.016	0.015	0.0082	0.0022	0.00078	0.00012	0.00031	0.00028	0.00025
FLA	0.019	0.015	0.014	0.0082	0.0021	0.00083	0.00013	0.00025	0.00023	0.00026
PYR	0.020	0.016	0.014	0.0076	0.0021	0.00097	0.00013	0.00026	0.00022	0.00023
BaA	0.020	0.016	0.013	0.0073	0.0023	0.00078	0.00013	0.00028	0.00022	0.00026
CHR	0.020	0.018	0.014	0.0075	0.0023	0.00080	0.00013	0.00032	0.00027	0.00032
BkF	0.020	0.017	0.014	0.0073	0.0025	0.00086	0.00011	0.00023	0.00020	0.00021
BeA	0.019	0.017	0.014	0.0072	0.0022	0.00077	0.00011	0.00028	0.00027	0.00028
INP	0.021	0.018	0.014	0.0076	0.0021	0.00085	0.00010	0.00024	0.00024	0.00025
DBA	0.020	0.015	0.014	0.0075	0.0024	0.00090	0.00012	0.00031	0.00026	0.00026
BAP	0.020	0.016	0.013	0.0075	0.0024	0.00078	0.0013	0.00031	0.00029	0.00030
BGP	0.020	0.018	0.010	0.0079	0.0025	0.00086	0.00012	0.00029	0.00021	0.00023.
Total PAH	0.320	0.266	0.215	0.1238	0.036	0.01348	0.00314	0.00433	0.00404	0.00406

A graph of variation of concentration with depth is shown in fig. 2. PAHs input in sediments are in different trend. Curves plotted show three different Zones. From 120 to 85 cm, there is an increase in PAHs content, zone 85 to 45 cm exhibits high PAHs content, zone from 45 to 5 cm has variable PAHs levels associated. A depth of 120 cm therefore, approximately, corresponds to background. The primarily increase in PAHs content above 120 represents a record of the initial urbanization, second rise in PAHs values is observed at around 85 cm to 45 cm. Increase in industrialization in this region may be explained by this zone. The value of PAHs at a depth of 120 cm was taken as representing the pre-industrial background level (Daisey, J.M., et al., 1986; Miguel and Pereira, 1989; Li and Kamens, 1993). Enhancement in degree to this background is represented by all other values of PAHs.

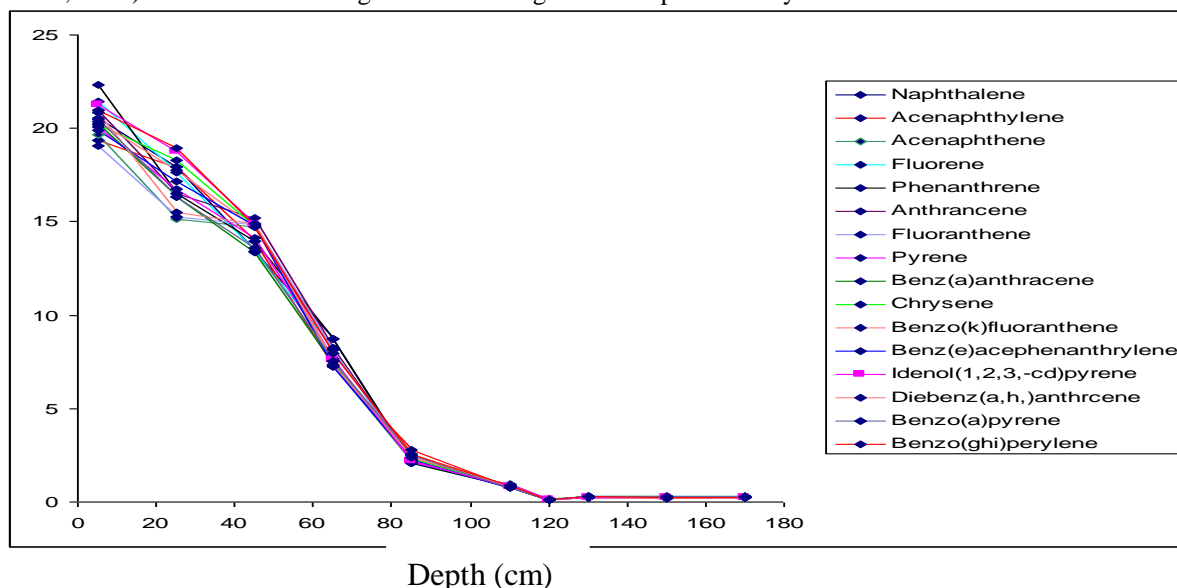


Fig.2-Graph showing variation of concentrations with depth.

5.2 PAHs markers for various sources in urban environment: The statistical data for individual PAH and total PAHs for outside of Airoli mangrove sediments are tabulated in table-2. The PAHs concentrations ranged from 0.0813-0.152 $\mu\text{g g}^{-1}$. Mean concentration of total PAHs for the entire samples collected was 0.1132 $\mu\text{g g}^{-1}$. The concentration for individual sixteen PAHs are shown in fig.3. From fig.3, it appears that the concentration of Benzo(k)fluoranthene, Diebenz(a,h,) anthracene, Benzo(ghi) perylene is high, sources of these PAHs are from diesel powered vehicles. Coal and kerosene burning, Fluorene, Phenanthrene, Anthracene, shows similar input concentration major sources (Table-3) of these are coal combustion, and coke production (Daisey et al., 1986; Miguel and Pereira, 1989; Li and Kamens, 1993; Venkataraman and Friedlander, 1994a; Khalili et al., 1995). The

dominant presence of unsubstituted compounds 4-ring Pyrene, Fluorene, Chrysene, 3-ring Phenanthrene, Anthracene shows similar input trends sources of these PAHs are coal combustion and coke production, diesel emission.

Table 2: Total concentration of PAHs in mangrove sediments ($\mu\text{g g}^{-1}$)

PAHs	Mean Concentration	Range of Concentration
Naphthalene	0.00709	0.0046-0.011
Acenaphthalene	0.00698	0.0046-0.0088
Acenaphthene	0.00675	0.0052-0.0091
Fluorene	0.00717	0.0048-0.010
Phenanthrene	0.00723	0.0049-0.010
Anthracene	0.00719	0.0053-0.0098
Fluoranthene	0.00678	0.0052-0.0091
Pyrene	0.00693	0.0050-0.0092
Benz(a)anthracene	0.00677	0.0047-0.0092
Chrysene	0.0072	0.0052-0.0092
Benzo(k)fluoranthene	0.00749	0.0052-0.0092
Benzo(e)acephenanthrylene	0.00699	0.0052-0.0092
Idenol(1,2,3,-cd)pyrene	0.00691	0.0048-0.0094
Diebenz(a,h,)anthracene	0.00735	0.0053-0.0097
Benzo(a)pyrene	0.00697	0.0053-0.0094
Benzo(ghi)perylene	0.0074	0.0052-0.0097
Total PAHs	0.1132	0.0813-0.152

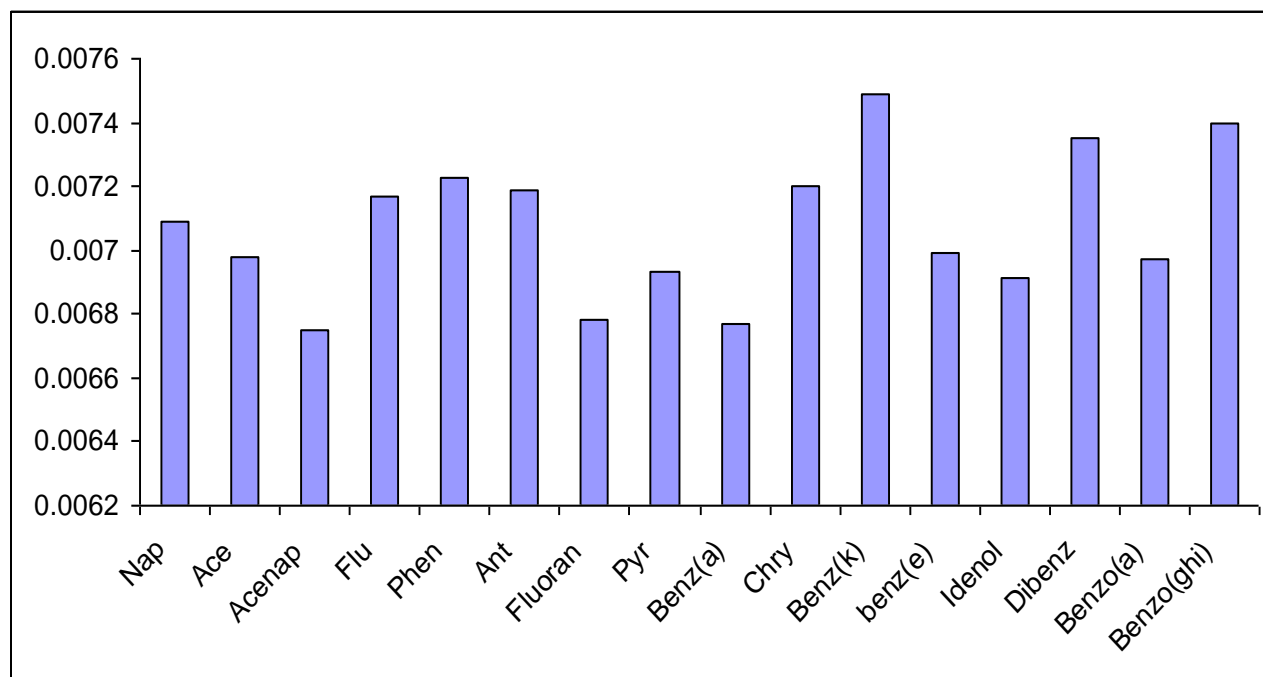


Fig.3-Individual concentration of PAHs in $\mu\text{g g}^{-1}$.

Table-3: Sources of concentration of PAHs in mangrove sediments

Name of PAHs	Sources
Phenanthrene , Fluoranthene ,Pyrene. Anthracene ,Phenanthrene , benzo(a) pyrene. Pyrene ,phenanthrene , Fluoranthene. benzo(a) pyrene , Fluoranthene. Fluoranthene Pyrene Chrysene. Benzo(a) pyrene, Indeno (123 cd) pyrene, Coronene. Fluoranthene - Pyrene , benzo(b)fluoranthene, Benzo(k) fluoranthene.	Coal combustion. Coke production Incineration, Wood combustion , Industrial oil burning Petrol –Powered vehicles Diesel powered vehicles

5.3 Environmental Magnetic Analysis: Environmental magnetic parameters, various ratios made and their inference are shown in fig. 4. From the analyses of the graphs, high values of χ , χ_{ARM} , and SIRM are observed throughout the upper half of the profiles upto about 100 cm. This can be explained by the presence of magnetite in this section of the profile. The super paramagnetic substances can be the pollutant input into the sedimentary section from the steel plant and the adjoining anthropogenic activity. The lower part of the profile section that is from 100 cm downwards consists mainly of multidomain particles. This may consist mainly of primary hematite, as well as magnetite which are both of natural origin.

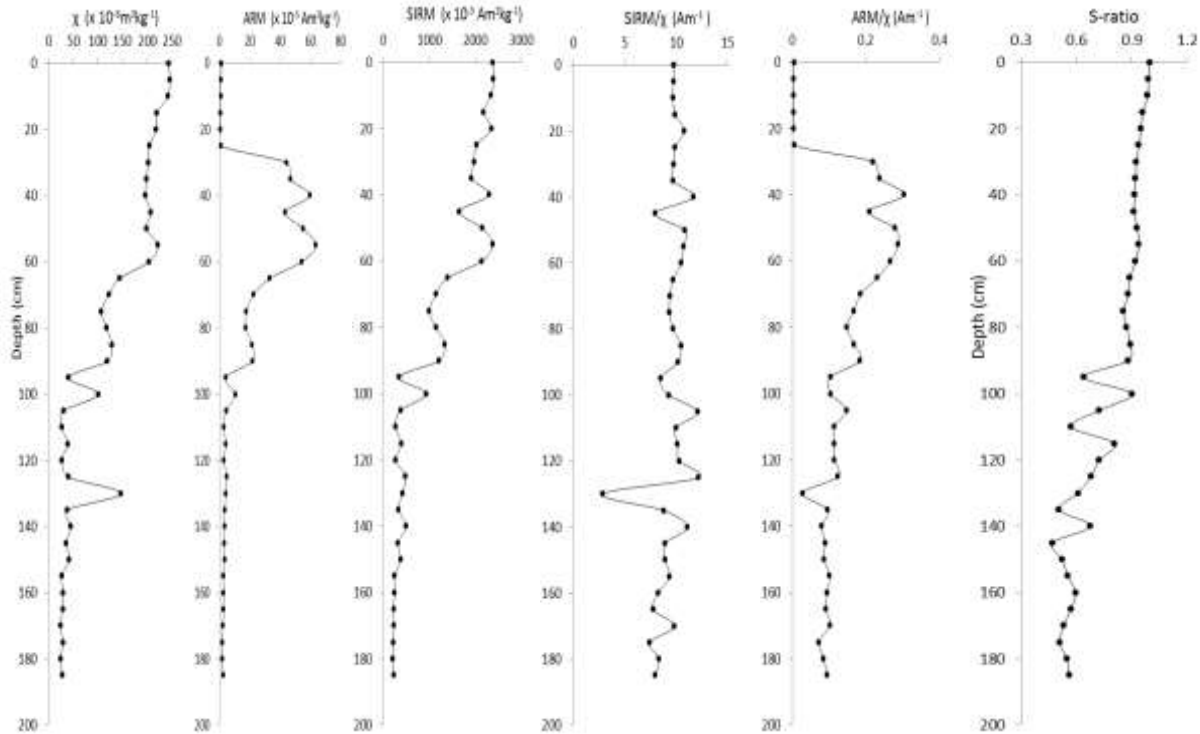


Fig.4- Environmental magnetic parameters, various ratios and their inference.

The S-ratio is the ratio of the Soft IRM to the hard IRM, in the graph plotted we find that the proportion of the hard and the soft components are almost same upto 100 cm depth, while below this, its value fluctuates this is indicative of the dominance of hematite in the lower part of the profile (Mudgal, T.R., 2012). It is interesting that hematite is present throughout the section while the magnetite is varied. The main inference is that contrary to a general understanding of multidomain magnetite of industrial origin, the occurrence of super paramagnetic and single domain magnetite is observed in the segment interpreted as the accumulation of pollutants in and around sampled area (Mudgal, T.R., et al., 2022).

6. RESULTS AND CONCLUSION:

With the help of the environmental magnetism parameters it is inferred that the upper portion of the soil section is dominated by the magnetite is of anthropogenic origin. This is inputted into the mangrove environment from the nearby developmental activity and also from the steel plant which came up at the 1980s and that to a depth of 100 cm, while the hematite or the hard component is produced by the oxidation of the Basalt rocks which are prominent in the area, the Deccan traps. This is a resultant of natural oxidation of iron to form its hydrated oxide. Hematite is present all throughout the system whereas the magnetite dominates only on the top part. Thus with the magnetic technique we can clearly mark out the boundary between the polluted and the unpolluted sediments. This boundary is placed at about 100 cm, corresponding to the year 1946 according to estimated sedimentation rate of 1.6cm/year. It is inferred that the area got exposed to pollution since this magnetic take-off point at 100 cm (Mudgal, T.R., et al., 2022).

From present work it is evident that pre-industrial background level value may be taken at a depth of 120 cm (fig.3). Degree of enhancement relative to this background value is represented by all other values. Thus assessment of impacts of organic contamination on ecosystems and sustainability of local aquaculture in the area after the industrialization can be understood. This result enhances the understanding of current contamination levels.

This investigation was carried out adjacent to the Thane - Belapur road highway which is passed near to MIDC (TBIA) is the major sources of PAHs in sediment. The total mean concentration of PAHs in sediments is observed $0.1132 \mu\text{g g}^{-1}$ (Table-2). A set of sixteen PAHs (Table-1) is found in sediments including Naphthalene, Acenaphthalene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz(a)anthracene, Chrysene, Benzo(k)fluoranthene, Benz(e)acephenanthrylene, Idenol(1,2,3,-cd)pyrene, Diebenz(a,h)anthracene, Benzo(a)pyrene, Benzo(ghi)perylene. Sources of these PAHs are from diesel powered vehicles, coal and kerosene burning, Fluorene, Phenanthrene, Anthracene shows similar input concentration major sources of these are coal combustion and coke production (Baumard, P., et al., 1999; Kulkarni and Venkataraman, 2000; Daisey, J.M., et al., 1979). Hence, from this study it is inferred that mineral magnetic techniques along with chemical analyses may be a fast, rapid and reliable methodology for study of pollution in marine mangrove ecosystems.

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