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Assessment And Spatial Distribution Of Benzo(A) Pyrene In Soil Environment Of Panvel Taluka

Namrata Kislay[1], Dr. Geeta Malbhage[2*], Dr. Nilesh D. Wagh[3], Harshala V. Kasalkar[4]

Abstract

This study focuses on soil analysis from different sites (urban, peri-urban, and rural) to quantify the concentration of Benzo (a) Pyrene (B(a)P) in soil during pre-monsoon and post-monsoon seasons. This includes establishing the co-relation with physical parameters and assess its impact in Panvel Taluka (Dist. Raigad), Maharashtra (India).

Samples were collected for two consecutive years (Yr. 2021 & 2022) and from five distinct sites characterized by various human activities, including an agricultural area, an industrial area, residential districts with traffic congestion, and others. Using a spatial statistical tool, to evaluate the spatial characteristics of B(a)P accumulation. Concentration of B(a)P in soil for the pre-monsoon and post-monsoon differed between the two years. Concentration of B(a)P relates inversely to urbanization, indicating a correlation between urban development and the accumulation of pollutant in the soil.

Study observed that the concentration of B(a)P in soils shows a descending gradient in the order of urban > peri-urban > rural areas. Urban and industrial sites have 2-6 times greater B(a)P concentration in soil. In future, this study will help identify the long-term effects, remediation techniques, and policy implications of soil contamination.

1. Introduction

A strong economic foundation for an area is provided by its city's functional economy, which has evolved from agriculture to industry, services, trade, and commerce. This resulted in growth of industrial zones and motor vehicles. Both the industries and traffic contribute to pollution escalations in urban areas. On the other hand, only few industries run with proper treatment system, while others still go with old technologies and releases various environmental pollutants. Like emission of Polyaromatic Hydrocarbons (PAHs) from burning of crude-oil, coal, gasoline, garbage & others. Centres of PAH emissions are primarily found in urban areas. [1]Thus, in recent studies the focus has been on pattern of distribution, source identification, and risk estimation of PAHs in urban soil. Their presence is mostly explained by industrial activities, vehicular emissions, and others (Wang et al., 2017).

A class of chemical molecules known as polyaromatic hydrocarbons (PAHs) is made up of two or more fused benzene rings. The teratogenic, mutagenic, and carcinogenic effects of PAHs and their toxicity to life forms are well known. (Wang et al., 2017). The International Agency for Research on Cancer has categorised several PAH compounds as probable 2A or 2B category human carcinogens. Five-ringed PAH benzo[a]pyrene (B(a)P), which has been extensively studied for its higher carcinogenicity than others, is commonly employed as a marker of the potential harm of PAHs to both the natural world and human health

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[1] Amity School of Applied Sciences, Amity University, Mumbai-410206, India
*[2]Assistant Professor, Amity School of Applied Sciences, Amity University, Mumbai-410206, India
[3]Associate Professor, SK Somaiya College, Somaiya Vidyavihar University Mumbai 400077, India
[4]Amity School of Applied Sciences, Amity University, Mumbai-410206, India
*-Corresponding Author

(Pufulete et al., 2004; Pardo et al., 2016). It is extremely hydrophobic and biodegradation-resistant, making it mostly persistent in soils (Gua et al., 2021). 16 PAHs have been designated as priority pollutants by the United States Environmental Protection Agency (US EPA), with soil serving as the primary environmental sink for these contaminants (Wang et al., 2015b; Mojiri et al., 2019).

Soil is the main reservoir of PAH compounds in the environment. Both agricultural and industrial soils contain them because they are mostly combustion by-products of organic molecules. (Jaruga et al., 2019). Most combustion-derived PAHs are anticipated to be localised to the top layer of the soil because atmospheric deposition is the most frequent source of soil pollution (Zhang et al.,2023). Hydrophobic pollutants (like PAHs) persist in the soil matrix for a considerable amount of time after adhering to the soil's organic matter. In addition to being a risk to humans (via food contamination and unintentional ingestion), its presence in soil may also be hazardous to other biological constituents of the soil environment, including plants and microbes (Jaruga et al., 2019). To reduce the danger of human exposure and control pollution, it is crucial to identify and allocate the PAHs source in soil.

Indian researchers have published various studies on the levels of PAHs in sediments and airborne particles. Evidence about the PAHs spatial distribution in soils is rare. Furthermore, the PAHs concentration in soils is not yet defined in India, and only a few guidelines exist - Mexican standards (6 mg/kg), Dutch standards (0.02–0.05 mg/kg), Polish standards (0.2 –10 mg/kg). There is still no publication of Korean criteria for soil-bound PAH concentrations. (Agarwal et al.,2009; Roy et al., 2022).

Over the last few years, spatial analyst methods have been successfully applied to examine the spatial distribution of environmental variables such as PAHs in urban, peri-urban, and agricultural soils (Peng at al., 2016; Zhang et al.,2023). Moreover, spatial cluster analysis is crucial for quantifying spatial patterns (Peng at al., 2016). Recent research, notably those on soil pollution (Zhang et al. 2008), appear to show that Moran's I is a particularly well-liked approach for spatial cluster analysis compared to other indexes. Some of the benefits of using Moran's I includes varying level of statistical computes, summarizes spatial autocorrelation, easily interpretable, ability to apply clustering at local & global levels, can apply various types of spatial data.

The objective of this work is to quantify the B(a)P presence in soil for pre-monsoon and post-monsoon weather condition in Panvel Taluka, Raigad district, Maharashtra (India) that can be theoretically estimated using the B(a)P equivalent concentration (BaPeq). This study can eventually be used to identify the potential health risks associated with all PAHs and thus, help in devising the potential mitigation options. The study of B(a)P was not carried out earlier in this area. Benzo(a)Pyrene (B(a)P) is one of the most virulent mutagens, according to the World Health Organisation (WHO), and is commonly used as a generic indicator of PAHs and a benchmark for the carcinogenicity of all PAHs.

2. Study area and sampling description

The Mumbai Metropolitan Region, which includes the Panvel, is the largest urban agglomeration in the nation. It has a geographic area of 110.6 sq. km, with a population of 5.09 lakh (Census, 2011). None the less, the population must attain a good growth till current year 2023. It is situated at the latitude of 18°58′N and 19°70′N and the longitude of 73°2′E and 73°9′E. Sahyadri Ranges offshoot to the east, Navi Mumbai to the west, Thane and Dombivali to the north, and JNPT port and Uran to the south, surround it.

It is in the North Konkan Region and receives precipitation from monsoon winds. Maximum rainfall (95%) is received from south-west monsoon winds. High temperatures and evaporation are from March to May. The month of July recorded the most precipitation.

The mean maximum and minimum temperatures 38.5 ◦C and 25.0 ◦C in summer. The average annual temperature and annual rainfall is 27 ◦C and ~3267 mm respectively (Environmental status report,2020-21). Location of Panvel Taluka sampling sites is shown in Fig.1.

Bhatan is regarded as a rural and agricultural area. The main source of pollution in this area is the neighbouring highway, as well as the burning of biomass and coal in locales. Next two sites are Taloja and Panvel. Taloja is one of the highly developed industrial areas with businesses engaged in a range of commercial endeavours. Types of Industries in this area are varied and spread into Chemicals, Engineering, Dyes, Food processing, Steel, etc. whereas Panvel is one of the most populous cities in Maharashtra. It has experienced substantial urbanisation in the recent decade because of various development projects such as, the multi-corridor Project, the New Mumbai International Airport, etc. Numerous redevelopment projects are now underway and some of the major industries includes L&T, Reliance, ONGC, and others. A rising residential neighbourhood next to Panvel town is Karanjade. The future Navi Mumbai International Airport will be in Ulwe (Environmental status report,2020-21). It's a Navi Mumbai node that is still developing, and there are many structures and constructions being built there. Based on urban, suburban, and rural areas, five different locations were picked. Figure.1 depicts the study area.

Table 1 Sampling sites

Area	**Latitude**	**Longitude**	**Categorization**
Taloja	19.058538	73.112741	Industrial - Urban
Panvel	18.99340	73.116775	Residential - Urban
Ulwe	18.982386	73.078349	Agriculture Peri-Urban
Karanjade	18.983045	73.091544	Residential Peri-Urban
Bhatan	18.931531	73.163786	Agricultural - Rural

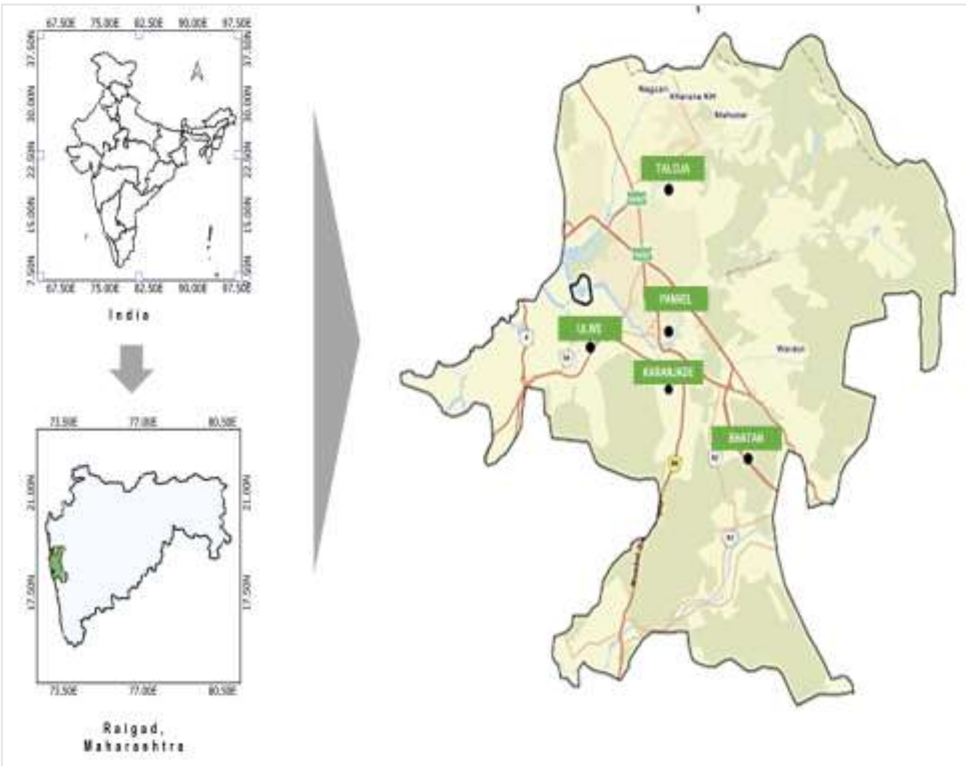


Fig.1 Sampling Sites

3. Materials and methods

3.1. Sample collection

Samples were collected from five different sites in Panvel Taluka during the pre-monsoon and post-monsoon for the year 2021-2022. The possible effects of rainfall on soil properties and pollutant behaviour are taken into consideration while analysing pre- and post-monsoon soil samples. Pre-monsoon helps evaluate the baseline pollution levels. Post-monsoon season can cause contaminants to build up in the soil. Rainfall may have an impact on how pollutants travel through the soil and how they degrade, which may alter their concentrations. Purpose to have pre-monsoon and post-monsoon sample is to study seasonal variation, hydrological effects for soil moisture contents, temperature influence and land use impacts.

Pre-monsoon sampling seasons was carried out in the month of March to May and post-monsoon sampling was in the month of October to December. In total, 120 soil samples from all the sites were collected in the years 2021 and 2022. Soil samples (collected at 6–8-inch depth) scooped with a soil probe and collected in labelled plastic bag. These samples were cleaned by removing stones, bricks, and other stray vegetation. The soil was first sun-dried and then oven-dried at temperature of 90°C for 1 Hr. duration. At the end with the use of mechanical sieve shaker, which is equipped with 75 and 150-mesh sieve, soil particles were obtained with less than 75 and 150 µm respectively.

3.2. Chemicals

To study the B(a)P level in the soil, a standard (1000 µ/mL in Acetone) containing the same were procured from Supelco (Bellefonte, USA). All solvents (methylene dichloride, acetone etc.) used for sample processing and analysis, were of analytical grade. Water used for the analysis was high purity deionized water taken from the Mili-Q system, to ensure no contaminants, enable precise measurements that eventually help maintain the reaction integrity and results outcome.

3.3 Sample extraction and analysis

The extraction of B(a)P from dry soil samples were performed using a 30-mL mixture of solvents (methylene dichloride/ acetone, 1:1, v/v) for overnight through Rotary shaker. These extracts were evaporated at 50°C using a rotary evaporator. The extraction efficiency was improved by combining polar and non-polar solvents. The satisfactory results were obtained with combinations of acetone and methylene dichloride. The B(a)P were analysed by gas chromatography mass spectrometry (GC-MS, Agilent7890B/5975C, Agilent Technologies) with a DB-5MS capillary column (15 m, 250 µm inner diameter × 0.25 µm film thickness). The carrier gas was helium (high purity, 99.99%) at a constant flow rate of 1 ml/min. A sample (2 µL) was injected using an automatic sampling system. Oven temperature was initially set at 100 °C held for 1 min, and then increased to 230 °C at 15 °C/min held for 0 min. Finally, the oven temperature was increased to 300 °C at 10 °C/min held for 5 min.

The internal calibration method was used for the quantitative analysis, and the B(a)P were identified by comparing their retention times to those of real standards. By injecting a standard reference solution of the B(a)P spiked with the internal standards, response factors for B(a)P were determined. Response factor and peak area responses were used to determine the concentration of B(a)P.

4. Data analysis and statistical method

Spatial autocorrelation analysis: In this study, the spatial autocorrelation of total B(a)P concentrations in soils were investigated using Moran's I index. The first assessment of spatial autocorrelation was Global Moran's I (Moran 1950). It has a value range of 1 to -1. "1" denotes perfect positive spatial autocorrelation (high or low values cluster together), "-1" denotes perfect negative spatial autocorrelation, and "0" denotes perfect spatial randomness (Tu and Xia 2008).

IBM SPSS Statistics' SPSS 24 was used to conduct the statistical analysis. Spatial analysis was performed with ArcGIS Pro 2.7. Inverse Distance Weighted (IDW) was used to carry out the spatial interpolation and to produce the spatial distribution maps for the subsequent spatial correlation analysis for the year 2021-22.

While global Moran's I evaluates spatial autocorrelation, Local Indicators of Spatial Association (LISA) uses local Moran's I to evaluate the level of spatial autocorrelation at each unique place (Anselin 1995). It is particularly useful for detecting Spatial outliers and local spatial cluster patterns (Harries 2006). The local Moran's I index (Levine 2004) can be written as:

$$I_i = \frac{z_i - \bar{z}}{\sigma^2} \sum_{j=1, j \neq i}^{n} \left[W_{ij} \left(z_j - \bar{z} \right) \right]$$

Where,

z_i= the value of the parameter z at the position i
z = the average value of z with n samples
z_j= the variable's value, z in all other locations (where $j \neq i$)
σ^2= the variance of variable z
W_{ij}= weight as the inverse of the distance d_{ij} between points i and j

A distance band can also be used to calculate the weight W_{ij}; samples inside the band are given with the exact weight, while those beyond it are given zero weight. The results of the local Moran's I index can be standardised, and its significance level can be verified using an anormal distribution assumption (Levine 2004).

When applying the local Moran’s, I index to analyse the spatial pattern, the results were influenced by the weight function definition, data modification, and the presence of extreme values. As a result, these parameters were considered to get accurate and steady findings. In soil contamination study, high-high clusters are considered "regional hotspots," while low-low clusters are considered "cool spots" (Zhang et al. 2008). A high positive local Moran's I value means that the target value is comparable to its neighbourhood, and the locations are then spatial clusters and are classified as high-high clusters and low-low clusters. On the other hand, a high negative local Moran’s I value indicates a spatial outlier, which means that the target value is obviously different from the values of its neighbourhood, and it comprises high-low and low-high outliers. High-low spatial outliers are known as "individual hotspots."

5. Results and discussion

5.1 Physio-chemical properties of soil

Physio-chemical characteristics of soils reflects that the accretion and retention of B(a)P are strongly influenced by soil properties; and can be affected by factors such organic matter content, pH, clay, silt content, and moisture levels. It was discovered that the examined soil samples were slightly acidic to slightly basic in composition. Majority of the soils being studied have a loamy texture. According to Lu et al., clay had the greatest PAH concentration, which dropped in the following order: clay > silt > coarse sand > fine sand. The sodium adsorption ranged from 3.9% (in the urban area), 3.7% (in the industrial area) and 4.0% (in the rural area).

5.2 Pre-monsoon and post-monsoon concentration of B(a)P and its spatial distribution

The concentrations of the overall B(a)P observed in pre-monsoon soils are:
- Year 2021: 0.566 to 1.628 mg/kg with a mean of 1.045 mg/kg
- Year 2022: 0.154 to 8.54 mg/kg with a mean of 4.34 mg/kg

While the concentrations of the overall B(a)P observed in post-monsoon soils are:
- Year 2021: ranged from 1.253 to 3.762 mg/kg with a mean of 2.1348 mg/kg.
- Year 2022: ranged from 1.166 to 9.411 mg/kg with a mean of 5.513 mg/kg.

Table 3 and 4 summarizes the statistical results of the B(a)P concentrations measured in pre-monsoon and post-monsoon soils of Panvel Taluka. The seasonal variation of B(a)P shows that the highest levels of B(a)P were discovered in the post-monsoon, followed by pre-monsoon seasons. This is because monsoon rains deposit the B(a)P contaminants in the soil. Different weather conditions experienced over these seasons can be used to explain variations in B(a)P content in soil. While during the monsoon season this area often receives frequent downpours and this results in the washout effects of contaminants. A wet deposition of B(a)P at the soil surface is also feasible during this season in addition to dry deposition, and this should have increased the amount of B(a)P in the soil, and variations in human activity may also contribute to this pattern at various Panvel Taluka locations.

Pre-monsoon pH values for year 2021 and 2022 fall between 6.7 to 7.5, indicating that the soil is either slightly acidic or slightly basic in nature. Post-monsoon pH values for the same year fall between 6.1 to 7.5, indicating that the soil is more acidic in nature than pre-monsoon. There is slight difference in the concentration of electrical conductivity in pre and post monsoon. Also, water holding capacity is slightly more in the post monsoon season as compared to pre monsoon season. Mean B(a)P concentrations were higher in the post-monsoon than in the pre-monsoon. Details are given in Table 2 and 3.

Table-2 Mean, SD, CV% of B(a)P in soil during pre-monsoon.

	Pre-monsoon Soil (n=30), Year 2021					**Pre-monsoon Soil (n=30), Year 2022**				
	B(a)P, mg/kg	**pH**	**EC (µS/cm)**	**SAR (%)**	**WHC (%)**	**B(a)P, mg/kg**	**pH**	**EC (µS/cm)**	**SAR (%)**	**WHC (%)**
Min	0.56	6.9	229	3	31	0.15	6.7	222	3.4	38
Max	1.62	7.4	402	4.2	44	8.54	7.5	382	4.52	52
Mean	1.04	7.12	282	3.68	37.93	4.34	7.08	277.06	3.97	43.93
SD	0.36	0.16	63.82	0.37	4.74	2.77	0.21	59.83	0.38	4.57
CV %	34.98	2.32	22.63	10.14	12.5	63.87	3.05	21.59	9.62	10.41

Table-3 Mean, SD, CV% of B(a)P in soil during post-monsoon.

	Post monsoon soil (n=30), Year 2021					**Post monsoon soil (n=30), Year 2022**				
	B(a)P, mg/kg	**pH**	**EC (µS/cm)**	**SAR (%)**	**WHC (%)**	**B(a)P, mg/kg**	**pH**	**EC (µS/cm)**	**SAR (%)**	**WHC (%)**
Min	1.25	6.48	210	3.4	40.5	1.16	6.11	235	3.7	40.7

Max	3.76	7.4	322	4.59	49.8	9.41	7.5	391	5.24	55.1
Mean	2.13	7.05	272.26	3.85	44.53	5.51	6.89	287.66	4.37	47.56
SD	0.91	0.3	42.35	0.4	3.28	2.91	0.42	55.14	0.59	5.02
CV%	43	4.27	15.55	10.57	7.37	52.93	6.2	19.17	13.66	10.57

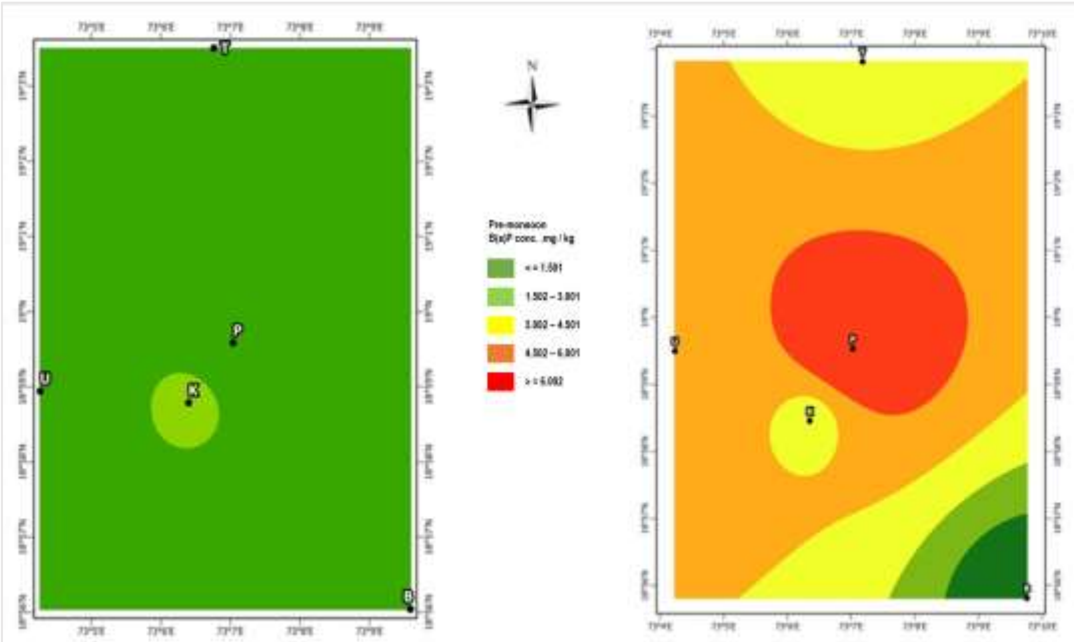
*Min minimum, Max maximum, SD standard deviation, CV coefficient of variation, EC electrical conductivity.

5.3 Spatial distribution map for pre-monsoon & post-monsoon soil

The spatial distribution of B(a)P in soils is generated using ArcGIS for all the five locations:

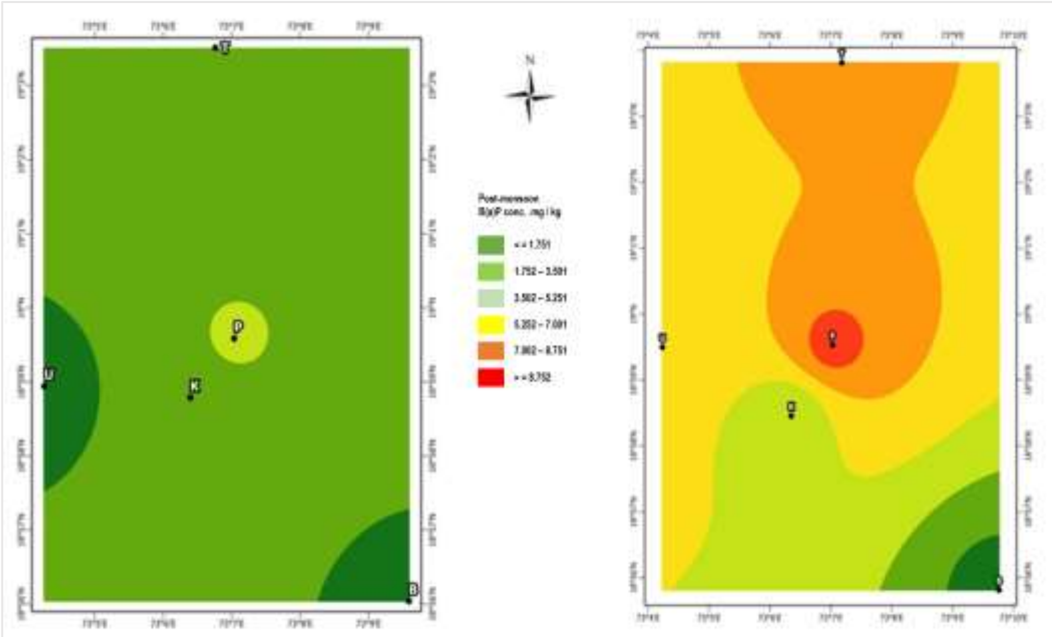
- K - Karanjade
- T - Taloja
- P - Panvel
- U - Ulwe
- B – Bhatan

These results show large concentrations of B(a)P in the Panvel and Karanjade (Figure 2 and 3)



Year 2021 Year 2022

Fig.2 Spatial distribution map of pre-monsoon soil for the year 2021-22



Year 2021 Year 2022

Fig.3 Spatial distribution map of post-monsoon soil for the year 2021-22

The result above reflects that B(a)P concentration was low for the year 2021, as compared to year 2022. The primary reason behind the decreased B(a)P in year 2021 is contributed primarily COVID lockdown impacts that includes less vehicular movements, low industrial and reduced construction activities. Similar studies were conducted in the Delhi (India) where there was significant drop in the pollution level during COVID 19 lockdown (Tyagi et al, 2022).

5.4 Spatial cluster and outlier analyses

Figure-4 depicts the findings of LISA analysis and global Moran's I statistics. Significantly positive autocorrelations evidenced by global Moran's I value were discovered for concentrations of B(a)P in soil, indicating the presence of spatial patterns in their spatial distributions (Fu et al. 2011).

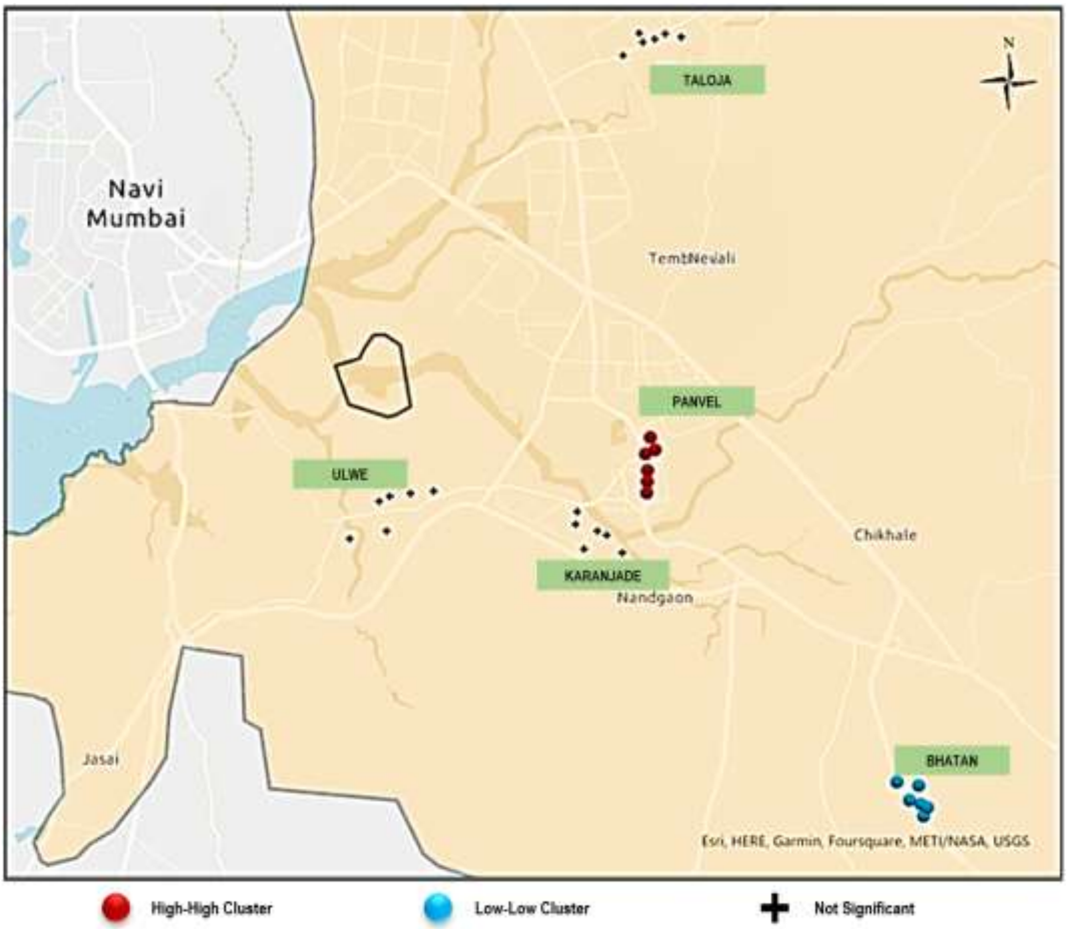


Fig.4 Map showing spatial cluster analysis.

There were clear spatial clusters for total B(a)P in soils for both pre-monsoon and post-monsoon, including a large 'High-High Cluster' at Panvel and a 'Low-Low Cluster' at Bhatan. The presence of high-high clusters of total B(a)P in soils was closely linked to industrial and construction activities as well as due to redevelopment projects (Fig. 2). Another reason for the overall elevated B(a)P concentrations in soils were also due to vehicular movement. The low-low spatial cluster in Bhatan revealed that overall concentrations of B(a)P in soils were generally low since this area was far from industry and still practised traditional agricultural management. While Ulwe, Taloja and Karanjade reflects 'Not Significant' levels.

5.5 B(a)P distributions in soils under diverse land-uses

B(a)P distributions under diverse land-use helps in assessing the impact of human activities. This brings an opportunity to study the varying levels of B(a)P into the soils. Figure 5(a) and 5(b) summarizes the pre-monsoon and post-monsoon averaged concentrations of B(a)P in soil at four different sites composition at Panvel Taluka, i.e., industrial, residential, roadside, and agricultural. In this area, the soil B(a)P concentrations varied according to land use classifications. The residential area had significantly higher soil B(a)P concentrations than the other land use categories. The average B(a)P concentration in soils for the residential areas was in the range of 1.12-8.49 mg/kg and 3.76-9.41 mg/kg in pre-monsoon and post-monsoon respectively, which was significantly higher than that of the industrial, roadside, and agricultural areas.

The soil B(a)P concentrations in the agricultural areas was in the range of 0.157-0.568 mg/kg and 1.167-1.443 mg/kg in pre-monsoon and post-monsoon respectively.

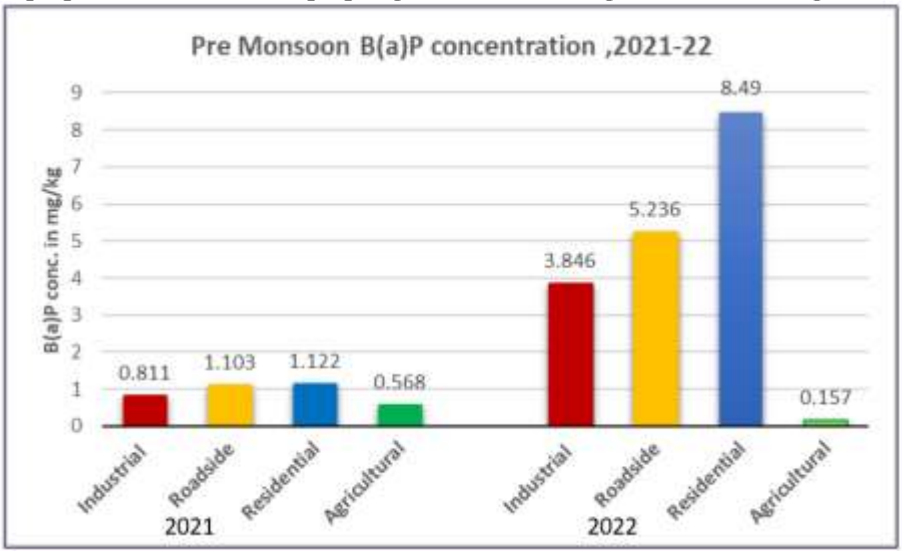


Fig.5(a) Pre-monsoon averaged concentrations of B(a)P

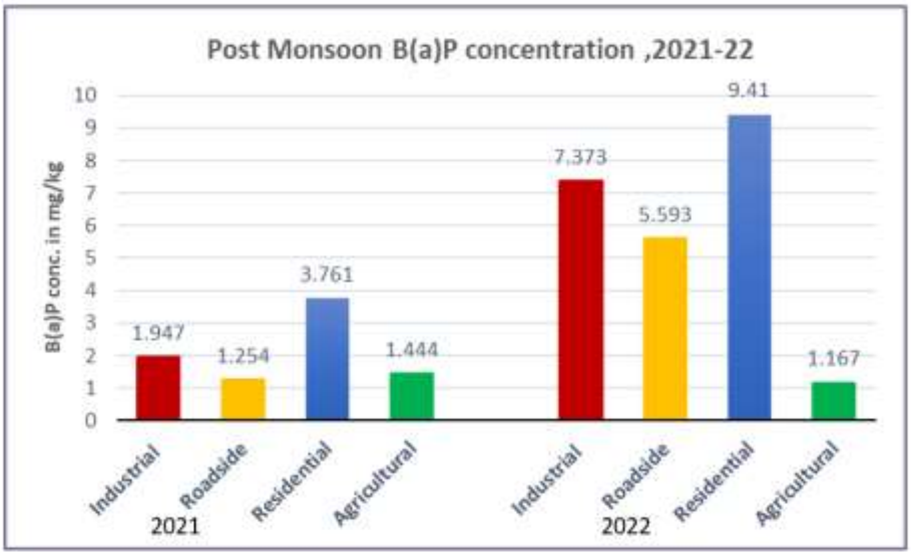


Fig.5(b) Post-monsoon averaged concentrations of B(a)P

Similar patterns of soil PAH distribution among various land uses have also been observed in cities of Europe, the United States, and China, including London, Miami, and Shanxi Province. (Banger et al., 2010; Vane et al., 2014; Ji et al., 2022). Soil B(a)P concentrations were significantly influenced by the type of city (including its economics, stage of development, and so forth), annual temperature, environmental quality, and land use. These variables were the reason for the variations in the spatial distribution of soil B(a)P in Panvel Taluka cities.

The low B(a)P concentrations in agricultural soils revealed that agricultural practises were not the primary contributor to the B(a)P build-up in soils. The deposition of airborne B(a)P brought from the industrial area and from roadways could land in the agricultural area. It is critical to understand the different levels of B(a)P in soil. This will enable the development of necessary mitigation and remediation approaches that can include land management practices.

5.6 Correlation of soil parameters with B(a)P

The link between B(a)P and soil pH is complex and can be influenced by a variety of circumstances. The PAH-contaminated soils may retain the persistent pollutants for long period of time depending on their physicochemical properties (pH, organic carbon, grain size, and porosity) and its microbial diversity. The pH of the soil can influence the behaviour of many chemicals, including B(a)P. Higher accumulation of PAH can attribute to lower pH values with less biological activity in soil environment (Lasota et.al 2021). This can potentially lead to ill health of the soil, which may affect the crop pattern and increases the chances of groundwater pollution if the situation persists for longer time.

The sorption of B(a)P to soil particles is influenced by pH; and in general, B(a)P tends to sorb more strongly to soils with lower pH values. The degradation and transformation of B(a)P in soil can be pH dependent. Certain microorganisms and enzymes involved in B(a)P degradation have optimal pH ranges for activity. Consequently, pH levels outside of these ranges can affect the microbial degradation capacity, potentially impacting B(a)P persistence in soil. The accumulation or concentration of benzo (a) pyrene in soil is not correlated with electrical conductivity.

Several research have concluded that PAH binding occurs primarily on finer grain size, i.e., clay and silt (Magi et al. 2002). The clay particles have a larger surface area and, as a result, have more bonding sites, which lead to the PAHs adsorption from the finer soil fraction. Fine particles also have less porosity and thus less mobility of adsorbed pollutants over time, resulting in long-term toxicity and effects. This type of pollutants which rupture the texture of the soil, which leads to adverse impacts on soil characteristics like water holding capacity, microbial population etc. (Sakshi et.al. 2019)

Conclusion

Study has provided a significant and valuable insights into the complex interplay of factors contributing to B(a)P compound in the soil. These data can serve in assessing environmental risk and developing appropriate mitigation strategies by lowering exposure to B(a)P in urban and peri-urban sites. It also reflects the importance of implementing soil management policies curated for the larger requirements that includes stricter soil contamination controls, green infrastructure development, and public awareness campaigns. In future studies, it is also equally important to start using the Artificial Intelligence (AI) in relation to B(a)P in soil for faster research outcomes. AI can be used to develop models that predicts the pattern of B(a)P in soil basis historical data and distribution patterns and build the necessary control measures. This includes identifying B(a)P concentrations in various soil samples and comparing them to relevant criteria such as land use patterns, proximity to pollution sources, and soil characteristics. AI combined with GIS will help create faster and detailed maps for PM and B(a)P in areas and can be a potential next step of this research. By harnessing all the available approaches and investing in new ways, institutions can work toward effective control mechanisms and public health protection strategies in Panvel Taluka (Maharashtra).

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