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Determination of polycyclic aromatic hydrocarbons in algae : Giresun coastline-Eastern Black Sea Region

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Abstract: Polycyclic aromatic hydrocarbons (PAHs) are often measured in the atmosphere for air grade valuation, and sea creatures for environmental monitoring, in residue and for safety cases in food materials, in biological tissues for health-effects monitoring. In this study, Polycyclic aromatic hydrocarbon (PAH) concentrations in algae of *Ulva rigida* species, as an environmental indicator, in the spring of 2019 were collected from eigh station in Giresun costline in May. In general, algae samples are extracted by methanol solutions with Soxhlet apparatus. By applying the necessary purification processes to the obtained solution, the concentrations of 16 PAH components were determined with the GC/MS device. The amount of most of the 16 PAH compounds in the Keşap coast algae samples was the highest. The highest amount compounds are found to phenanthrene, fluorene and anthracene, and it is high in Bulancak, Giresun-Central and Keşap sites. It belongs to seaweed samples collected from Piraziz site, which has the lowest values among eight stations. The PAH compound concentration is determined 0.08-340.96 ppb values. In the spring sampling, the lowest value is Espiye and Tirebolu stations with Benzo[a]pyrene compound and the highest value is Keşap station with Phenanthrene compound. Since the benzo [g,h,i]perylene is lower than the detection levels in many stations. The main origins of PAHs were found as both pyrolytic and petrogenic at most stations.

Keywords: Algae, Polycyclic aromatic hydrocarbon, GC/MS, Eastern Black Sea

1. Introduction

PAH compounds are a family of many toxic compounds are mutagenic and carcinogenic potential. They are known that environmental pollution with two or more fused aromatic rings. PAH compounds are organic pollutants that are mixed into water, air and soil with industrial waste discharge, atmospheric particles and fossil fuels pouring into the water environment. (Kafouris et al. 2020; Lampa et al. 2018; Tongo et al. 2017).

PAH compounds consist of two sources: first of all, sustained sources such as domestic heating, aluminum and iron-steel industry, oil refinery, power and heat generation and mobile sources such as exhaust gases or aromatic hydrocarbons named pyrogenic-pyrolytic occur during forest fires origin. The other arises from fixed sources refinery facilities during petrogenic sources oil transportation. This is caused by ship accidents and oil transportation in areas with much ship traffic (Clark 1997; UNEP/IOC/IAEA 1982).

There is a significant accumulation of Polycyclic Aromatic Hydrocarbon (PAH) in the Black Sea because of the spillage or leakage of waste fuel burned by vehicles such as ships and tankers passing through the Black Sea, rivers exposure to discharge waters and flowing into the Black Sea. The entry of PAH compounds into seawater is linked to coastal sea waters, rivers, streams, and by waters (Güven et al. 2009).

Because of the low solubility in water, PAH compounds accumulates in the sediment at the bottom of the water. Due to the fact that sea creatures such as crab, lobster, shrimp, mussel, many fish species and algae that are buried in the sea bottom, are fed by filtering into their bodies, they accumulate PAH compounds in their tissues over time (Nwaichi and Ntorgbo 2016; Ololade et al. 2017; Yang et al. 2013; Zhang et al. 2015). Among these sea creatures, seaweed enters the body of people with food chain because its consumption as a foodstuff by many cultures. Since PAH compounds have toxic, carcinogenic and mutagenic properties. They cause many diseases such as lung, skin, bladder cancer in humans (Racovita et al. 2020). One of the best defined founded on their genotoxic and carcinogenic properties and most toxic of 16 species PAH is benzo(a) pyrene, which is generally used as the indicator of PAHs (Albuquerque et al. 2016). The International Agency for Research on Cancer has categorized benzo[a] pyrene into group 1, showing that it is “carcinogenic to humans”; dibenz [a,h] anthracene is in group 2A, displaying it is “probably carcinogenic to humans”(Honda et al. 2021).

The interest in macroalgae is increasing because they are generally used as food approximately 160 species in island countries and the Far East countires especially China, Japan and Korea. Since macroalgae are considered to be an excellent source of protein, it is expected that macroalgae will replace animal and vegetable protein sources in time to meet protein needs. They contains of high quality proteins as riboflavin and niacin, lipids and water-soluble fiber and are abundant in minerals and vitamin needed for human nutrition (Alçay et al. 2017; Øverland et al. 2019).

Green macroalgae species, especially *Ulva* or sea lettuce species such as *Ulva rigida* and *Ulva lactuca*, compose of major primary producers of marine ecosystems (Maghraby and Hassan 2021; Neckev et al. 2002; Rosioru et al. 2018). They are f ound in the rocky parts of shallow waters and this type of algae has leaves that look like thin plates. It grows naturally on the Black Sea coasts and the Bosphorus (Akyurt et al. 2011, Turna and Uzunköprü 2015). These *Ulva* species are dried into powder and added to fast food foods in Scandinavia, Great Britain, Ireland, China, Japan and Korea. Antioxidant polysaccharides called “ulvans” are obtained from sea lettuce species.

Due to it grows very much between 0-3 meters deep, prevalence in the marine ecosystem, high absorption capacity to the environment and accessible species and easy to collect algae material, it is widely preferred in enviromental studies as biomonitors and bioindicators of potential hazards in marine environments (Cheney et al. 2014; Pavoni et al. 2003; Zeroual et al. 2020).

Also, algae are highly preferred bioindicators for the detection of different types of pollution in aquatic environments. In many countries, various seaweed species is chosen as a bioindicator for the detection of soil, water and other environment pollutions because it accumulates PAH compounds, which are organic pollutants in the water, and are widely available (Kaur et al. 2020; Li et al. 2021; Namiesnik et al. 2012; Wang and Zhao 2007).

There are studies in which many samples generally air, water, sediment, sea creatures and algae are used for PAH measurements in Turkey and world. In previous works, while air, water, sediment, marine organism were preferred more in PAH studies, on seaweeds were more limited in Turkey(Ayyıldız and Esen 2020; Balcıoğlu et al. 2017; Eleren and Yücel 2021; Koçak 2021).

In this paper we report results on the ability of *Ulva* species seaweed to accumulate polycyclic aromatic hydrocarbons (PAHs). In fact, *Ulva* species especially *Ulva rigida* species are ubiquitous in the Black Sea region as well as other seas and brackish coastal ecosystems. *Ulva rigida* is an one of dominant algae species. To the best of our knowledge, there are no previous reports on determination of PAH in *Ulva* species in East Black Sea Region.

2. Materials and Method

The samples were collected from 8 different stations around Giresun central and district in 2019 at 1-2m depth by diving during the spring seasons. Stations 1 and 8 are far to the port of Giresun. Station are 1. Piraziz, 2. Bulancak, 3. Giresun Central, 4. Keşap , 5. Espiye-Opet, 6. Tirebolu, 7. Görele and 8. Eynesil station. The 1st station is near the airport Ordu site, and the 8th station is near Trabzon, and the stations are oriented in the east-west direction of Giresun. All coordinations are given Table 1. All samples were brought to the laboratory under cold chain and in polyethylene bottle. The collected samples were washed in seawater and kept in 0-4 ℃ until analysis. However, the physicochemical parameters of seawater at the time the sample was collected are shown in Table 2.

The algaes were authenticated immediately after collection. The green seaweed and image microscope are shown in Figure 1 and Figure 2.

Figure 1. Green seaweed (*Ulva rigida*) collected from Giresun, Turkey.

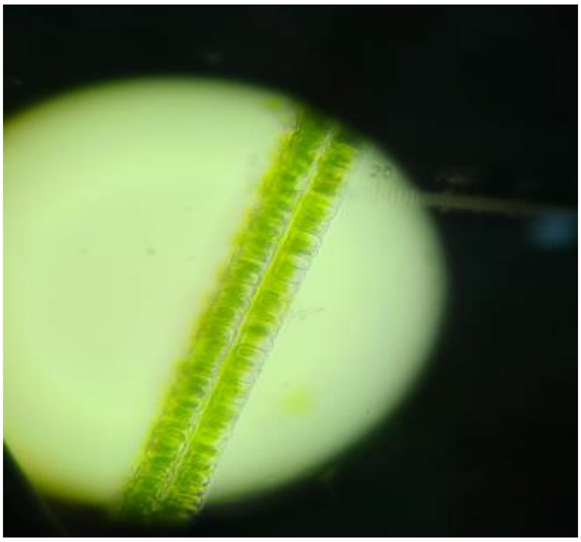

Figure 2. Screen microscopy of cross sections of cell layers

In this study, 16 PAH components which are in the priority contaminants list of the United States Environmental Protection Agency (US-EPA 610) were designated. Investigated PAHs are listed as naphthalene (NAP), acenaphthylene (ACL), fluorene (FL), acenaphthene (AC), phenanthrene (PHE), anthracene (AN), fluoranthene (FA), pyrene (PY), chrysene (CHR), benzo(a) anthracene (BaA), benzo(b) fluoranthene (B[b]FA), benzo[k] fluoranthene (B[k]FA), benzo(a) pyrene (BaP), dibenz-(a,h) anthracene (DBahA), indeno (1,2,3-cd) pyrene (IP), benzo(g,h,i) perylene (BghiP) in Table 2 (EPA 2009).

In general, the amount of seaweed samples to be tested was weighed around 15-20 g, placed in the Soxhlet cartridge, the cartridge was placed in the Soxhlet apparatus and 250 mL of HPLC grade methanol was added into the flask and extracted with Soxhlet for 12 hours. At the end of the period, 0.7 M KOH 20 mL and 30 mL distilled water were added to the solution, and the mixture was refluxed for 2 hours. The resulting solution was combined with n-hexane extracts 3 times (90 mL in 1st, 2nd and 3rd 50 mL hexane) and its volume was reduced to 5 mL in the evaporator. Purification was done through silica column. The first components from the hexane-conditioned column were aliphatic hydrocarbons, the later fractions were combined to consist of PAHs. PAH solutions in hexane were stored in a refrigerator to be given to the GC/MS device.

2.1. Chemicals

For the preparation of seaweed samples, potassium hydroxide, dichloromethane, methanol, anhydrous sodium sulfate, HPLC grade n-hexane were purchased from Sigma-Aldrich. PAH standards: naphthalene, fluoranthene, and phenanthrene, fluorene, pyrene from Supelco Sigma – Aldrich as PAH Calibration Mix was obtained from TraceCERT. Florisil 60 with 60–100 mesh size was obtained from Sigma-Aldirch.

2.2 PAH analysis-GC/MS

GC analysis is the Agilent 7890A-(5975C inert MSD) model. Gas chromatography conditions are; HP-5MS 5% Phenyl Methyl Silox column, GC furnace was equipped with an RTX5M 30 m capillary column (0.25 mm ID, 0.25 µm film thickness) with carrier gas helium at 1.5 mL min-1 flow rate. The analysis start temperature is 55 °C(1 min), the mold will reach 320 °C (16.6 min) at 5 °C/min. The final temperature was 320 °C and the analysis was terminated by staying at the same temperature for 1 minute. As the carrier gas (1.5 mL / min), the temperature of the injector and detector are 290 °C and 320 °C, respectively. Scan mode (70-300 m/z) is used for mass intake (Bates et al. 2008).

0.5, 1, 2 and 4 ppb (µg/kg) of the standard solution used in the PAH analysis were prepared by dissolving the stock solutions in hexane. Solutions were enjected in the GC-MS instrument and a chromatogram of the PAH components was generated by extracting the calibration curve.

3. Results

The lowest detected value, 0.08 ppb, was measured as benzo[a] pyrene compound in seaweed samples at Espiye and Tirebolu stations in the spring sampling. The highest value of 340.96 ppb is the concentration of phenanthrene (PHE) compound at the Keşap station in the spring sampling. The high values of all 16 PAH compounds were determined in Keşap station. It can be said that the amounts of especially phenanthrene, fluorene, benzoanthracene, anthracene, dibenzoanthracene, chrysene compounds are quite high. These concentraitons of PAH compounds are given in Table 3.

The amount of fluorene, phenanthrene and antresen are the highest compounds, and they are high concentrations in Bulancak, Giresun Central and Keşap. It belongs to the seaweed samples collected from the Piraziz site, which has the lowest values among 8 stations. Benzo [g,h,i] perylene could not be found since the detection values were lower than the level in many stations.

Some evaluations are made to understand whether the source of PAH is pyrolytic or petrogenic.
- Fluoranthene(FA) and pyrene(PY) compounds are PAH compounds of pyrogenic origin, and if their ratio (FA/PY>1) is greater than 1, they are thought to be PAH compounds from pyrolytic sources (Soclo et al. 2000).
- If the ratio of phenanthrene(PHE) and anthracene(AN) is lower than 10, it can be said to be from pyrolytic deposits, if it is greater than 15(PHE/AN>15), it can be said to be from petrogenic originated deposits (Soclo 1986).
- In some cases, it can be said that there is petrogenic pollution if the ratio of fluoranthene/pyrene (FA/PY), to the phenanthrene/anthracene(PHE/AN) value is less than 2(<2), if the pyrolytic is greater than 2(>2) (Sicre et al. 1987).
- Another PAH pollution source determination ratio is Benzo(a)antrecene and chrycene (BaA/CHR) ,formed as a result of low-temperature combustion, component ratios. If these components are higher than 0.9, it showed that petrogenic pollution (Gschwend and Hites 1981).

According to the results of algae samples on the concentrations of 16 PAH components in this study;
- When evaluated according to the ratios of floranthene/pyrene (PHE/AN) and benzo(a)anthracene/chrycene (BaA/CHR) in spring sampling; Among the 8 stations, the PHE/AN ratio was found to be 17 only in the Piraziz station.

According to this result, it was determined that the type of pollution was caused by petrogenic sources and the pollution in other stations was PAH pollution originating from pyrolytic.

- Sampling stations coordinates as shown in Table 1.
- Spring sampling physicochemical measurements as shown in Table 2.
- Seaweed samples PAH values on spring season (ppb) was shown in Table 3.

Table 1 Sampling stations coordinates

	Stations	Coordinates		Stations	Coordinates
1	Piraziz	40^{o} 70’ 56’’ N 38^{o} 33’ 50’’ E	5	Espiye-Opet	40^{o} 71’ 00’’ N 38^{o} 55’ 07’’ E
2	Bulancak	40^{o} 14’ 64’’ N 38^{o} 35’ 11’’ E	6	Tirebolu	41^{o} 84’ 05’’ N 38^{o} 81’ 12’’ E
3	Giresun Central	40^{o} 51’ 98’’ N 38^{o} 85’ 99’’ E	7	Görele	41^{o} 48’ 40’’ N 39^{o} 36’ 60’’ E
4	Keşap	40^{o} 46’ 70’’ N 38^{o} 66’ 28’’ E	8	Eynesil	41^{o} 02’ 10’’ N 39^{o} 53’ 60’’ E

Table 2. Spring sampling physicochemical measurements

	1	2	3	4	5	6	7	8
Temperature	16,6	19,1	17,8	19,3	15,7	16,7	16,4	16,8
pH	7,81	8,05	7,66	7,7	8,52	7,85	7,7	7,52
% O2	113,7	107,4	107	105,3	103,6	110,3	103,5	93,4
mg/L O2	10,83	9,73	9,93	9,53	10,5	10,46	9,85	8,78
Conductivity	22272	11702	21043	18723	2921	15931	18983	14476
Superconductivity	26523	13168	24680	21038	3519	19065	22742	17179
Salinity	16,2	7,6	15,1	12,6	11,9	11,3	13,8	10,2
ORP	-49	-59	-33	-31	-79	-47	-44	-23
TDS(mg/L)	17223	8578	16077	13367	2245	12374	14866	11150

Naphthalene, anthracene and phenanthrene is fossil fuel sourced. These compounds are formed as a result of coal combustion, pyrolysis of petroleum and incomplete combustion of organic compounds (Yalaki 2005). Naphthalene is one of the dangerous atmospheric pollutants. In our study, naphthalene levels were found to be higher at Bulancak, Keşap, Görele and Eynesil stations, where traffic density and industry are more intense, compared to other stations.

4. Discussion

In the literature, there are studies about the concentration determination of PAH compounds in seaweed samples in different seas of the world. However, the use of algae in the seas in PAH study in Turkey has remained fairly limited. In this study, EPA 16 PAH compound concentrations were determined in seaweed samples collected from a total of 8 stations along the Black Sea Giresun-Ordu coast and the pollutant types were determined with some parameters.

Algaes are widely used specimens in environmental studies in many countries around of the world. In addition, some types of algae are used as foodstuffs in many eastern countries. The human body obtains higher levels of PAH compounds from food than from atmospheric air or drinking water (Barranco et al. 2003).

Table 3. Seaweed samples PAH values on spring season(ppb)

	1	2	3	4	5	6	7	8
Naphthalene (NAP)	7.58	35.56	6.00	32.23	10.36	9.83	34.95	49.80
Acenaphthylene ACL)	0.24	2.91	5.88	14.42	0.98	0.84	0.85	1.17
Acenaphthene (AC)	4.97	12.34	3.09	44.23	1.21	1.10	7.54	18.29
Fluorene (FL)	0.82	9.16	7.13	187.67	2.78	2.00	1.77	2.63
Phenanthrene (PHE)	6.09	164.79	100.20	340.96	33.07	20.78	11.37	21.77
Anthracene (AN)	0.35	66.52	40.45	137.64	13.35	8.39	4.59	8.79
Fluoranthene (FA)	0.34	12.16	11.69	39.54	2.70	3.35	2.45	4.58
Pyrene (PY)	0.23	3.67	8.02	9.72	1.25	1.47	1.67	1.81
Benzo[a]anthrecene (BaA)	1.28	0.71	5.31	142.31	0.99	1.77	1.81	1.09
Chrysene (CHR)	0.49	0.27	2.04	54.57	0.38	0.68	0.70	0.42
Benzo[b]fluoranthene (B[b]FA)	0.95	1.41	5.19	12.99	0.88	2.32	0.94	0.95
Benzo[k]fluoranthene (B8k)FA)	0.48	0.71	2.62	6.55	0.44	1.17	0.48	0.48
Benzo[a]pyrene (BaP)	0.32	0.11	2.10	16.04	0.08	0.08	0.60	0.13
Dibenz[a,h]anthrecene (DBahA)	0.46	2.70	7.83	87.50	1.82	1.18	0.64	0.70
Indeno[1,2,3-cd]pyrene(IP)	0.18	0.23	1.72	14.39	0.46	0.80	3.41	0.63
Benzo[g,h,i]perylene (BghiP)	0.76	2.38	N.D.	N.D.	N.D.	N.D.	0.63	N.D.

The most important of these PAH compounds is benzopyrene (BaP). It is formed as a result of the combustion of automobile and other vehicle fuel types and reaches the atmosphere. This compound, which has a high carcinogenic effect, then reaches the land, drinking waters, and the sea, along with the precipitation, and enters with the organism. Considering the amounts of this compound, it was found as 0.08 ppb at most stations, while it was measured as 16.04 ppb at the Keşap station. The station with the highest PAH compounds analyzed in this study was found to be the Kesap station with a concentration of Phenanthrene compound of 340.96 ppb. The lowest concentration was analyzed as benzo [a] pyrene compound with 0.08 ppb at Espiye and Tirebolu stations.

High molecular weight compounds such as pyrene, fluorantene, phenanthrene, chrysene are more stable and are formed as a result of burning fossil fuels under high temperatures. The ratio of fluorantene and pyrene is used to determine the type of pollution. In most stations, along with other ratings; The ratio of these two compounds is greater than 1 (FA/PY>1), that is, it can be said that they are pyrolytic pollution. Also, determination of phenantren (PHE), pyrene (PY) and benzo(a)anthracene (BaA), acenaphthylene (ACL), acenaphthene (AC) and chrysene (CHR) components indicates that Keşap station has been exposed to continual pollution.

Güven et al. (2009), found higher PAH amounts in the samples from the eastern black sea region than the samples collected from the western and central Black Sea regions in their study to determine the level of oil pollution on mussels, snails and algae collected from the Black Sea coast. It can be said that especially PAH measure in port samples are more polluted.

In other study by Okay et al. (2000), they were studided that the accumulation in mussels algae-fed exposed to the common PAH benzopyrene pollutant. They found that the benzopyrene ratio in tissue of mussel samples fedding with algae was quite high. BaP can be say to be a contaminant whose presence and toxicity be passed through the food chain. İn this study; benzopyrene ratio is found to generally low level except for Keşap station. The Keşap station is also important for the food chain of sea creatures that feed on algae and for the health of people.

Karacık et al. (2009), were determined PAH amount and PAH's effects on marine organism in İstanbul strait. They identified 16 PAHs and calculated to assess the origins of PAHs (PHE/AN, FA/PYR and CRY/BaA) in samples., the Amounts of PHE, FA, PY, BaA, AN compounds were found to be high in the polluted stations similar to our study.

Another study Balcıoğlu et al. (2017) were found to similar PAH value in mussel samples in Mediterranean. The important contributors to PAH concentrations were chrysene, fluoranthene and benzo(a)pyrene (BaP) were determined in mussels. In this respect, it is similar to this study, chyresene and fluoranthene in Keşap station higher than other stations. Although some stations exposed to shipping activity was found to petrogenic origins related to instant traffic contamination.

Fogaça et al. (2018) were studied PAH determination in *Ulva* sp. species algae and shrimp. The highest value acenaphthene (49.11 ± 5.19), fluorene (1.78 ± 0.92) and naphthalene (0.96± 0.29) was identified by high-performance liquid chromatography. According to results of this study; acenaphthene (44.23), fluorene (187.67) and naphthalene (32.23) were found highest PAH concentrations in algae.

5. Conclusion

In recent years, interest in determination of PAHs in environmental samples stems from possible adverse health effects of PAHs. The present study revealed the EPA 16 PAH compound in seaweed samples collected from Giresun coastline for the first time. This study will provide a resource for future studies on PAH pollution in the Eastern Black Sea region.

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