



REVIEW ARTICLE

The contamination of bottom sediments in the Southern Baltic with polycyclic aromatic hydrocarbons (PAHs)

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Abstract – The study involved a comparative analysis of PAH (polycyclic aromatic hydrocarbons) concentrations in the bottom sediments of the southern part of the Baltic Sea. It was determined that the contamination of sediments with PAHs is considerable (>500 ng/g), with the predominance of pyrogenic PAHs. Polycyclic aromatic hydrocarbons become deposited mainly in anaerobic or extremely anaerobic areas, with a large proportion of the <0.063 mm fraction and a sizeable content of organic matter. In the studied regions, the contamination of sediments was the consequence of anthropogenic activity.

Keywords – biogenic, diagenetic, petrogenic, pyrogenic PAHs, sediments

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Characterisation of PAHs and their influence on living organisms

The *National Institute of Standards and Technology (NIST)* distinguishes between more than 600 structural formulas of *polycyclic aromatic hydrocarbons (PAHs)* (Kubiak 2013). These form a class of nonpolar organic compounds, which are characterised by high lipophilicity and hydrophobicity. They are composed of a large number of conjugated aromatic rings, the most commonly found hydrocarbons in the environment being composed of two (naphthalene) to seven (coronene) conjugations of benzene rings. The boiling point and solubility of PAHs depend on their molecular mass (the higher it is, the higher the boiling point is, while the compound's solubility in water decreases). Lighter hydrocarbons (e.g. naphthalene) are more soluble in water than heavier compounds, meaning that they are less durable in the environment as it is easier for them to become diluted, evaporate, or undergo bacterial decomposition (Page et al. 1999).

Polycyclic aromatic hydrocarbons are substances that pose a threat to living organisms. While in their pure form, they do not show any carcinogenic activity. However, their derivatives, which are formed as a result of the activity of metabolic enzymes, are carcinogens (Neff 2002). The most carcinogenic PAHs include benzo(a)pyrene, dibenz(a,h)anthracene and benzo(b)fluoranthene (Collins et al. 1991). Nevertheless, even the hydrocarbons which are less harmful as such can augment the synergic effect of other PAHs.

PAHs are introduced into living organisms with food, through the skin or via inhalation. Hydrocarbons are highly soluble in fats, a quality which aids their accumulation in living organisms. Moreover, they are characterised by chronic toxicity. This means that a prolonged and regular intake of PAHs even in small doses can lead to illness. The accumulation of hydrocarbons in the tissues of marine animals is conditioned by their metabolic ability. Organisms which accumulate smaller amounts of PAHs in a short time are, for example, fish, whereas shellfish and molluscs accumulate more hydrocarbons. This, however, requires a longer period of time as the metabolism of invertebrates protects them from carcinogenic hydrocarbons. As a consequence, the accumulation of PAHs at higher trophic levels is low (low biomagnification). In the marine environment PAHs negatively affect mainly the species diversity and changes in the percentage share of particular taxons (Balcioglu et al. 2014). However, any contact of PAHs with cell membranes or other cell organelle interferes with their structure and functioning. Membrane permeability changes are leading to dysfunctions in the transportation of substances within the cell. Next the energetic balance weakens. In clams this is manifested by a decrease in body size. In oysters the result is a drop in tissue volume. Cells which are heavily burdened with aromatic hydrocarbons undergo autolysis as a consequence. PAHs interact with lipids, nucleic acids (DNA, RNA) and proteins (e.g. enzymes). The binding of hydrocarbons to DNA can lead to mutations, and if repair mechanisms are overloaded, cancer develops. Polycyclic aromatic hydrocarbons also negatively affect the immunological system. Fish caught in badly

contaminated areas have been found to have a reduced number of lymph nodes and a considerably lower number of lymphocytes. Other diseases caused by PAHs are skin conditions, liver problems or cataract in fish (Knutzen 1995). The most dangerous are petrogenic PAHs.

In humans PAHs exhibit photosensitizing properties. This is an ability to damage cells in the presence of light and oxygen. It has been proven that during energy transformation living organisms can produce free oxygen, which is reactive and together with PAHs leads to cell damage. Yet the greatest danger to humans is posed by atmospheric pollution, contaminated food and tobacco smoke for smokers (Kubiak 2013).

Processes which lead to the formation of PAHs in marine bottom sediments

Hydrocarbons can be found globally in all sectors of the environment and are formed as a result of both natural and anthropogenic processes. They occur as mixtures of homocyclic and heterocyclic compounds (Kubiak 2013) and, in the marine environment, are found in the form of mixtures of compounds containing three or more condensed aromatic rings. Some PAHs are typically found in conjunction with alkyl chains. Heavier hydrocarbons are the most likely to penetrate into the sea bed. This is related to the sorption of these contaminants onto organic particles, which then become sedimented or decomposed by bacteria in sediments (Neff 2002).

In marine bottom sediments aromatic hydrocarbons can be formed in various conditions. The first of the processes leading to their formation is diagenesis – the slow transformation of sediments, initiated at the stage of sediment deposition. However, diagenesis proper starts only after that sediment has been covered by younger sediments. This is a relatively speedy process, lasting between a few days and a year, occurring at a low temperature (about 70°C). The presence of organic matter in sediment is necessary for the occurrence of diagenesis that enables PAHs to form. In anaerobic conditions, at a low temperature and pressure, sediments become cemented before biological transformations take place. The PAH that this process typically leads to the formation of is perylene. The main source of perylene in marine sediments is considered to be the diagenesis of organic matter originating from diatoms.

Other hydrocarbons can be formed in the process of biosynthesis as a result of biological activity of organisms (e.g. bacteria) in the marine environment. Biogenic PAHs are characterised by the simplest structure, a result of fewer formative biosynthesizing reactions (Savinov et al. 2000). An example of such a hydrocarbon is rethen, which originates in some algae and/or bacteria. It is very common in coastal sediments all over the world (Morrison and Murphy 2006).

More complex PAH mixtures should be linked to fossil fuel deposits in geological structures. These are called petrogenic hydrocarbons and their formation takes millions of years, occurring at increased pressure and high temperatures of between 100°C and 300°C. The structural complexity of hydrocarbons found in fossil fuels is an effect of biodegradation and complicated long-term diagenetic processes. The composition of petrogenic hydrocarbons includes a large number of highly alkylated, light hydrocarbons (Page et al. 1996). In marine sediments, petrogenic aromatic hydrocarbons are formed when organic matter transforms into petroleum (which can contain from 0.2% to over 7% PAHs).

The last group are pyrogenic PAHs. These are formed at a high temperature, reaching over 500°C, at which organic compounds do not undergo total combustion, but form an aromatic ring. Typical sources of pyrogenic hydrocarbons are wood burning and fossil fuel combustion in engines. In a marine environment, pyrogenic hydrocarbons most often originate from the emission of exhaust fumes from sea transportation and harbour activity. The concentration level of PAHs produced in sediments and their composition depend on the type of fuel burnt. The type of engine and age of vehicle, as well as burning conditions (oxygen content, humidity and temperature), all have great significance. The concentration of pyrogenic PAH in sediments is mostly determined by oxygen concentration, while temperature is responsible for its composition. Burning at a lower temperature leads to the generation of alkyl hydrocarbons, while a higher temperature is conducive to the production of matrix PAH compounds (Dalsøren et al. 2007).

The origin of PAHs can be determined based on their structure and composition. This is done using molecular indicators based on thermodynamic stability. For example, among three-ring isomers, phenanthrene and anthracene are compared against one another, of which the former is more stable thermodynamically. A phenanthrene/anthracene ratio below 10 indicates PAHs of pyrogenic origin. When the sample contains petrogenic hydrocarbons the ratio increases over 25. Another indicator is the fluoranthene/pyrene ratio, where fluoranthene is less stable thermodynamically than pyrene. For this reason, when pyrene is predominant in sediments, PAHs are treated as a transformation product of petrogenic hydrocarbons. If the opposite is true and fluoranthene prevails, the PAHs in sediments are of pyrogenic origin (Budzinski et al. 1997).

High values of PAH concentrations in sediments and the tissues of marine organisms, mainly those inhabiting the seabed, are also an indicator of anthropogenic contamination. On their basis it is possible to determine the ecological condition of the sea and its response to human activity.

Legal regulations concerning the protection of the Baltic Sea environment against pollution

The contamination of the marine environment is the act of introducing to it (and river outlets), directly or indirectly, substances or energy which cause or can cause harmful outcomes such as: damage to living resources and life in the sea, a decrease in the quality of utilised sea water and the worsening of recreational conditions (Convention on the Law of the Sea, 1982).

Given that elevated concentrations of PAHs can be found in most urbanised coastal areas in the world (Neff 2002), polycyclic aromatic hydrocarbons are placed on many international priority lists of environmental pollutants. Among them are the list of threats from the Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR), and the list of the United States Environmental Protection Agency (US EPA). The latter specifies the main 16 PAHs which should be monitored. The European Commission (Directive 2008/105/WE), on the other hand, named eight priority PAHs: anthracene, benzo(a)pyrene, benzo(b)fluoranthene, benzo(g,h,i)perylene, benzo(k)fluoranthene, indeno(1,2,3-cd)pyrene, fluoranthene and naphthalene.

The Baltic is an intercontinental sea where the inflow of water from rivers and precipitation is greater than the inflow of sea water. The basin is characterised by low water dynamics, particularly in deeper areas, which become seasonally stagnant. Lack of water mixing leads to the formation of extremely anaerobic areas emitting hydrogen sulfide. The low water dynamics is also a consequence of the limited connection with the global ocean (only through the Danish Straits). The large number of seaside agglomerations located along the Baltic coastline generate a higher load of pollutants per individual inhabitant than is the case with open seas. Loads of harmful substances are delivered by rivers, via surface run-off and through atmospheric deposition. In the case of the latter, hydrocarbons adsorb onto aerosols

and the organic matter bound to them. The contamination of the Baltic Sea environment is also caused by industrial usage of the water. This includes harbour activity, such as cargo transshipment, washing cargo containers with water and detergents, emergency discharge of oils and oil mixtures, removal of waste and other materials from ships or platforms, or fuel combustion. The legislation implemented to protect the environment of the Baltic is mainly intended to support activities that prevent the disruption of environmental balance and promote a way of utilising the marine environment which protects it from losing its ability to clean itself. With this purpose in mind, in 1974 the Baltic Marine Environment Protection Commission (HELCOM) was created. It is an international organisation proclaimed by the Helsinki convention as its executive body and its aim is to monitor and protect the natural environment of the Baltic Sea. It also sets out the priority groups of harmful substances and these include 15 polycyclic aromatic hydrocarbon congeners found in Baltic sediments (Tab. 1) (HELCOM 2010).

In 1983 an international convention on the prevention of the contamination of the sea by ships came into force (MARPOL 73/78). It had been drawn up in 1973 at an International Conference on the Pollution of the Seas. The convention contains recommendations for using fuel of suitable quality and regulations concerning the removal of solid waste from ships. It prohibits dumping plastic debris into the sea and also contains rules for loading operations, aimed at protecting sea water from petrogenic pollutants. The convention is particularly restrictive with regards to the protection of special defined regions (i.e. basins that are in particular danger owing to their ecological sensitivity or levels of shipping traffic), of which the Baltic Sea is one. In 1997 an appendix was added to the MARPOL convention, regarding the limitation of air pollution by ships (MARPOL 2006).

Table 1. The official limits of PAH concentrations [$\mu\text{g}/\text{kg}$] in sediments (HELCOM 2010)

Name of PAH	Concentration limit	Name of PAH	Concentration limit
Pyrene ³	665	Benzo(g,h,i)perylene ⁵	85
Fluoranthene ³	600	Anthracene ²	85
Benzo(a)pyrene ⁴	430	Dibenzo(a,h)anthracene ⁴	63
Chrysene ³	348	Acenaphthylene ²	44
Benzo(a)anthracene ³	261	Fluorene ²	19
Phenanthrene ²	240	Acenaphthene ²	16
Indeno(1,2,3-c,d)pyrene ⁵	240	Benzo(b)fluoranthene ⁴	-
Naphthalene ¹	160	Benzo(k)fluoranthene ⁴	-

Number of rings: ¹two rings, ²three rings, ³four rings, ⁴five rings, ⁵six rings

A direct source of contamination in Baltic sediments is also the dredging of the seabed in harbour areas. This is a common method used in order to maintain the water tracks so that they can perform their industrial and

economic functions. Part of the dredged material is deposited in the sea, at specially designated dump sites, and this leads to the formation of a natural geosorbent

which accumulates pollutants introduced into the marine environment.

The remaining part is deposited in silt dump sites on land. The dredged sediment undergoes procedures described by the *Ordinance of the Minister of Transportation and Construction on the Procedures of Granting Permission for Removing Dredged Material into the Sea and Dumping Waste and Other Substances in the Sea* (2006). The sediment is considered to be contaminated when at least one of the chemical indicators exceeds the limit. The maximum acceptable values are set out in the *Ordinance of the Minister of the Environment on the Types of Substances that Cause Dredged Material to Be Contaminated* (2002). If the dredging spoil is contaminated, it is treated as dangerous waste and deposited on land. The above-mentioned ordinance includes polycyclic aromatic hydrocarbons on the list of substances causing dump material to be contaminated, along with metals and polychlorinated biphenyls (PCBs). The ordinance also assumes that marine sediments should not contain benzo(ghi)perylene, dibenzo(a,h)anthracene, benzo(a)pyrene or indeno(1,2,3-c,d)pyrene in concentrations higher than 1.0 mg/kg dw (dry weight) Furthermore, according to the current norm, the concentrations of benzo(a)anthracene, benzo(b)fluoranthene and benzo(k)fluoranthene should not exceed 1.5 mg/kg dw.

Contamination of harbour bed sediments with PAHs

The Baltic Sea has some of the heaviest shipping traffic levels in the world, accounting for up to 15% of the world's cargo transportation. Both the number and the size of ships have grown in recent years, especially in

respect to oil tankers, and this trend is expected to continue. Forecasts indicate that due to long-term economic growth, especially in the eastern part of the Baltic region, the amounts of cargo shipped on the Baltic Sea will grow 64% by 2020. The number of large tankers is expected to grow, with more tankers carrying 100,000-150,000 tonnes of oil (HELCOM 2010). As shipping traffic increases, a consequent rise in PAH concentrations in the marine environment can be expected. At present there are 66 ports and harbours functioning on the Polish coast. One of them is the port in Gdynia, which has connections with 37 other ports, including ones in Belgium, Finland, Guatemala, Spain, Holland, the USA, Germany, Russia, Venezuela and the UK. The level of transshipment in ports is also increasing, with activity levels in Gdansk and Gdynia going up by 26.8% and 7.9% respectively in the years 2012/2013 (www.portgdansk.pl; www.port.gdynia.pl). Moreover, the port of Gdynia is preparing for the reception of oceanic containers, which entails a development of harbour infrastructure and increased activity in the ports. It is estimated that the general transshipment ability for containers in Gdynia will have increased by 143% by the year 2025 in relation to year 2014 (www.port.gdynia.pl).

Increased activity requires the harbour management to analyse the quality of sediments found at the bottom of harbour areas. Such analyses are currently based on the guidelines of the Helsinki Convention HELCOM (*Guidelines for the disposal of dredged material at sea*, adopted in June 2007). Samples are tested for the concentrations of contaminants, including eight PAHs.



Figure 1. The location of selected areas covered by PAH concentration tests (Gdynia Harbour is marked with an asterisk).

A study into the coastal area utilised as a port in Gdynia was carried out in 2003 (77 scoop samples, 0.5 m in thickness). In two studied areas the accepted concentration limits were found to have been exceeded by 80% for dibenzo(a)anthracene and by 20% for

benzo(a)pyrene. In 2005, 50 core samples were collected (at an average depth of 3 m), in which above-the-norm PAH concentrations were determined in 3 cores. A different situation was observed in 2006, when 26 samples taken from a depth of 2.5 m were analysed

and no exceedances were found. The following two years of tests (2008 and 2013) involved collecting a joint total of 151 harbour sediment samples, at respective depths of 1 m and 1.5 m. In both test campaigns the bottom sediments in the port area were found to be contaminated with PAHs whose concentrations exceeded the accepted norm (www.port.gdynia.pl). The question of harbour activity is directly linked to the dredging of port basins. Analyses of dredging spoil make it possible to assess whether it is characterised by elevated concentrations of harmful substances. In the Southern Baltic region, most silt dredged up from harbours has been deposited at nine sea dump sites (21 mln m³ on average) (Staniszewska 2014) (Fig. 1).

Studies into the contamination of sediments from the sea dump site area near Gdynia (6.4 km²) took place in 2012 and covered the actual area of the dump site (29 sediment samples), as well as the neighbouring area (21 sediment samples). The PAH concentration values in the collected samples did not show any exceedances of the accepted limits. Therefore that surface layer of dredging sediment was considered to be non-contaminated. However, the results were marginally higher than those obtained in 2009 (www.port.gdynia.pl). In the years 2003-2007 PAH tests were carried out in the sediments of the south-eastern part of the Baltic Sea, in the Gulf of Gdansk (Fig. 1A). Measurements covered the total of $\Sigma 12$ PAH concentrations. Their values ranged from 9 to 5100 ng/g dw. The mean concentration values in layers reaching 0-1 cm, 1-5 cm and 5-10 cm in depth were respectively: 3570 ng/g dw, 3400 ng/g dw and 3600 ng/g dw.

Owing to the fact that the concentrations exceeded 500 ng/g dw, it can be concluded that sediment in that area was highly contaminated. The sediment from the region of the Gdansk Deep was particularly notable, that station being characterised by an overwhelming proportion of grains measuring <0.063 mm (95%) and elevated values of total organic carbon concentrations (TOC).

Moreover, PAH concentration increased as oxygen concentration dropped in the near-bottom water. This proves that the durability of PAH compounds increases in extremely anaerobic and anaerobic conditions which occur in the Gdansk Deep. Furthermore it was estimated that the River Vistula is responsible for the introduction of 50% of the hydrocarbon load to the coastal zone of the Gulf of Gdansk (Lubecki and Kowalewska 2010). The lowest $\Sigma 12$ WWA concentrations were observed in sandy areas found closer to the shore. There were statistically significant differences between the mean values of $\Sigma 12$ WWA for coastal areas and those from the Gdansk Deep (the Kolmogorov-Smirnov test, $p < 0.05$). Tests did not however make it possible to determine any temporal trend of changes in the PAH load introduced to the sediments (Lubecki and Kowalewska 2010).

A wide range of PAH concentrations (from 3 to 30 000 ng/g dw) was noted in port sediments collected near industrialised towns and in sediments collected in the outlet of the River Oder (Fig. 1B) (Baumard et al. 1999). The study covered the total of 31 hydrocarbons. The areas of Kiel (Germany) and the Polish River Oder were found to be highly contaminated, as the mean hydrocarbon concentrations obtained for those regions were 8000 ng/g dw and 6600 ng/g dw, respectively. The most contaminated sediments though were those collected in Kiel's coastal zone, where maximum PAH concentration values were 14,900 ng/g dw and 18,700 ng/g dw. Such high concentration values can be attributed to anthropogenic activity, Kiel being a highly industrialised port city with shipyards, inhabited by over 300,000 people. The level of sediment contamination with PAHs decreased towards the open sea, where it amounted to 470 ng/g dw. The sediments of the River Oder were contaminated much more evenly, with the range of concentrations found ranging from 6600 ng/g dw to 9700 ng/g dw.

Sediment contamination with hydrocarbons was found to be lower in the German harbours of Peenemünde and Warnemünde, where the values were between 3 ng/g dw and 1600 ng/g dw. Mean PAH concentration values in harbour villages was 30 ng/g dw in Peenemünde, and 400 ng/g dw in Warnemünde. In both areas the level of PAH contamination reached its maximum in the harbour and decreased gradually away from the coastal zone. Here harbour activity was recognised as being the main reason for high hydrocarbon concentrations. In the areas covered by the study, most of the assayed PAHs originated from high-temperature burning (pyrogenic PAHs) while the proportion of PAHs of petrogenic origin was minor. The main sources of these latter contaminants were industry, fossil fuel burning and the heating of homes during the winter season (Baumard et al. 1999).

In the Bay of Mecklenburg studies were conducted in the area belonging to the Baltic Sea (Fig. 1C.). In the years 1950-1960 industrial waste from the city of Lübeck, highly rich in heavy metals and PAHs, was dumped there. After twenty years it was discovered that the contaminated material deposited had extended its range, reaching the surrounding area of the sea bed. Geochemical measurements of the surface layer of sediments showed elevated concentrations of heavy metals and PAHs in that region. The maximum concentration value was determined in the centre of anomaly, which is an unequivocal indication of anthropogenic influence. The accumulation of sediments was estimated at about 3 mm per year. The highest PAH concentration, amounting 80 μ g/g dw, was found at a depth of 16-18 cm into the sediment. Closer to the surface the concentration value amounted to 2 μ g/g dw (Leipe et al. 2005). Of the analysed PAHs, the compounds with the highest values, reaching up to 30 μ g/g dw, were of pyrogenic origin: phenanthrene,

fluoranthene and pyrene. The total amount of polycyclic aromatic hydrocarbons in the sediments dated at the time of dumping was 140 µg/g dw, which was 100 times higher than the value at the reference station (0.5 µg/g dw).

Conclusions

The enclosed basin of the Baltic Sea is exposed to anthropogenic activity and can be described as highly contaminated with polycyclic aromatic hydrocarbon compounds. PAHs are introduced mainly via the inflow of the Vistula and the Oder and through anthropogenic activity, mainly that related to the functioning of ports. The dynamics of marine currents results in PAHs being introduced into the Baltic deeps, the latter being a sort of sedimentation trap. Anaerobic conditions and a large proportion of the small sediment fraction are conducive to the accumulation of hydrocarbons and help increase their durability. Very polluted areas have a characteristic distribution of concentrations, with the maximum values in the anomaly centre, exceeding the values of background concentrations by as many as 1000 times, a fact which indicates the dumping of substances containing PAHs.

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