Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2018.22529

117 (2018) 318–328
June

Hazard from sediments contaminated with persistent organic pollutants (POPs)

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Received 28 February 2018; Accepted 22 May 2018

ABSTRACT

In the paper, attention is paid to the global problem of sediments contamination with selected persistent organic pollutants (POPs) in European and Non-European countries. The level of pollution of sediments with respect to origin, age of sediments, sources of contamination, estimation of the ecological risk and their presence in aquatic organisms is highlighted. The total concentration of PAHs was in the range 20 ng/g dry matter (dm)–90 mg/kg dm. The sum of polychlorinated biphenyls varied from not detected values up to 2.3 mg/kg dm. Among selected dioxins and furans, the measured concentrations ranged up to 1,800 ng/kg dm. The highest concentration of selected pesticides was equal to 10 µg/kg dm-aldrin, 7.4 µg/kg dm-dieldrin and 269 µg/kg dm–dichlorodiphenyltrichloroethane (DDT), respectively. Among phthalates, di(2-ethylhexyl)phthalates occurred in the range from 0.04 mg/kg dm to 115 mg/kg dm. Some data are given with respect to toxic equivalency factor indicating the value of the contamination background, whereas others refer to sediment quality guidelines. Recent trends confirm the necessity of improving the quantitative understanding of the sources and emission of POPs in the water environment as these hazardous pollutants affect aquatic life and pose a risk to human health. Although legislation plays a significant role in environmental protection, regular, smart monitoring and sustainable management is required with respect to POPs in sediments to reduce contamination in the future.

Keywords: Sediments; Contamination; Hazard; Persistent organic pollutants

1. Introduction

It is commonly known that sediments contaminated with persistent organic pollutants (POPs) create a serious problem not only in Europe but also in other countries [1–7]. Although the sources, behaviour, fate and effects of their presence in the environment have been regularly controlled, POPs can still be found in aquatic ecosystems. In Europe, investigations into sediments are carried out under the Water Framework Directive [8,9]. This directive forces EU members to establish national monitoring programs of surface waters in order to deliver compatible information on both the ecological state and chemical waters in each catchment area. Another important piece of legislation is the Stockholm Convention on Persistent Organic Pollutants [10]. One of the aims of the European Union's Water Framework Directive is to ensure that extensively modified water bodies are required to meet good ecological potential criteria.

Studies of POPs concentration in sediments have been carried out for many years. The improvement of analytical techniques over the decades now allows concentrations of pollutants occurring in the environment, including sediments, in trace amounts to be analysed. The presence of micropollutants in sediments is a function of anthropopression, however it is strongly related to the mineral composition of the sediments (mainly to silt and ash fraction) and the contents of organic matter [2,3]. It is important to take into consideration the reasons for this phenomenon.

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Presented at the 13th Conference on Microcontaminants in Human Environment, 4–6 December 2017, Czestochowa, Poland.

Recently, not only the problem of the level of contamination of POPs in sediments has became increasingly important but also their origin, age of sediments, sources of contamination, estimation of the ecological risk and the occurrence of POPs in living organisms [4,5,11–18]. This has created a necessity to develop efficient methods of limiting pollutants entering waters that are settled in the sediments and the migration and re-contamination of pollutants to the water. The data available in the literary sources are often non-uniformed which makes it difficult to compare the results. In this paper, some data have been obtained by calculating the toxic equivalent quantity (TEQ) in order to determine the background value, whereas other data are compared with the sediment quality guidelines (SQGs). In addition, there are examples of contamination with organic pollutants in sediments originating from various countries around the world.

This paper takes into consideration selected organic micropollutants such as: polycyclic aromatic hydrocarbons (PAHs), polychlorinated dibenzodioxins (PCDDs), polychlorinated dibenzofurans (PCDFs), polychlorinated biphenyls (PCBs), selected pesticides listed by the Stockholm Convention as well as phthalates (DEHP), nonylphenols (NP) and their ethoxylates (NPE).

2. PAHs in sediments

PAHs are regarded as POPs in the environment. The sources of their origin are mainly related to anthropogenic activities such as combustion engines, industrial activities and burning of biomass [19]. For humans, the fact that PAHs have carcinogenic, mutagenic and teratogenic properties resulting in their accumulation in organisms is of prime importance [2]. PAHs move into aquatic system by wastewater discharge, runoff from land and atmospheric fall out. Due to the processes of sorption and sedimentation, PAHs become bound to solid particles such as sediments. High molecular weight PAHs are able to accumulate in sediments, especially under anaerobic conditions, playing a role of trap and secondary source of PAHs contamination [20]. PAHs have toxic effect on aquatic organisms, and so may cause negative long-term changes to the water environment. Recently, the attention has been placed on monitoring the sediment quality in order to assess the potential negative effects of PAHs on aquatic environment and public health. A Directive of European Parliament and Council of Europe (No. 2008/105/EU) with respect to environmental quality standards in water management identifies 33 priority compounds in the field of water management [9]. PAHs and biologically active substances being the remains of pesticides are included among them. The fact that contamination of sediments of flowing waters, lakes and oceans has been the subject of scientific interest all over the world confirms the ubiquitous scale of the problem.

Popenda and Włodarczyk-Makuła [3] analysed Polish monitoring data from 2014 and found that PAHs were present in sediments originating from 25 measurement points located on two main Polish rivers – the Vistula and the Oder; 6 measurement points were located on the river Vistula, while 7 points were located on the river Oder. The total content of 19 PAHs ranged in value up to 65 mg/kg dm (the Oder river in Chałupki and Widuchowa). It was concluded that the Vistula river was much less polluted with PAHs as values ranged from 0.3 to 6 mg/kg dm. In 2014, the monitoring of PAHs also included lake sediments [3]. It was observed that the total PAHs concentration ranged from 3.3 to 20.7 mg/kg dm. The highest values were found in lake Bysławskie (kujawsko-pomorskie voivodeship) and lake Garbicz (lubuskie voivodeship), respectively.

According to data given by Witt and Trost [21], the sum of PAHs in sediments from the Baltic Sea and from the German coastal waters varied between 11 ng/g dm for sandy sediments and 1,900 ng/g dm for muddy sediments in the Belt Sea and Arkona Sea [21]. Elevated concentrations up to 17,000 ng/g were observed in the surface sediments of internal coastal waters with the highest concentrations in the Warnow estuary and in the Oder Haff. This indicates the significant contribution of river discharge to the contamination of sediments with PAHs.

In surface sediments originating from stations on Santander Bay, Cantabric Sea, Northern Spain, the level of 16 PAHs varied from 0.02 to 344.6 μg/g dm [22]. The results show a very different environmental quality from heavily polluted/medium effects to non-polluted/below threshold effects depending on the geographical location. These results indicate that local sources of PAHs, especially estuary discharges, lead to very different qualities of sediments in coastal zones, where traffic and industrial activity is present.

Hussain et al. [12] found a concentration up to 90 mg/kg dm for sum of 13 PAHs in sediments originating from the Seine River basin, France. An aim of this study, among others, was to reconstruct a historical profile of sediments over the last 70 years in order to assess the historical trends in a long-term urbanised and industrialised area. Using sediment from various sources, the authors concluded that the source of PAHs was the transition from coal to natural gas.

Lorgeoux et al. [11] investigated the sources of PAHs in sediments in the Bharalu tributary of the river Brahmaputra flowing through the city of Guwahati in India. They studied 16 PAHs listed by the Environmental Protection Agency (EPA) and sum of PAHs concentrations were in the range 338–23,100 ng/g dm during post-monsoon and from 609 to 8,620 ng/g dm during the pre-monsoon, respectively. The concentration of benzo(a)pyrene (BaP) varied from 17.8712 ng/g dm during post monsoon to 21.9727 ng/g dm pre-monsoon seasons, respectively. It was found that the sediments were mainly polluted with three-, four- and six-ring PAHs. The authors also calculated the hazard risk of PAHs to organisms and ecosystems using a risk quotient. They concluded that Acy, Phen and Pyr indicated severe hazards during the post-monsoon and pre-monsoon season. The origin of PAHs was mainly related to the emission from anthropogenic activities connected with gasoline, diesel and wood combustion.

Li et al. [13] assessed the contamination of PAHs in surface sediments in Chinese sediments coming from the Bohai Sea (BS) and northern part of the Yellow Sea (NPYS). The total concentrations of PAHs varied from 149.24 to 1,211.81 ng/g dm for BS and from 148.28 to 907.47 ng/g dm for NPYS, respectively. By comparing the results to the SQGs, it was found that acenaphthylene, fluorene, phenanthrene, fluoranthene, pyrene and dibenzo(a,h)anthracene may occasionally have adverse biological effects at some stations. The authors also paid attention to the ecological risk of PAHs by calculating the toxic equivalent concentrations of carcinogenic BaP. These were in the range

12.87–64.6 ng/g dm-BaP in BS and 5.95–68.80 ng/g dm-BaP in NPYS leading to the conclusion that there was rather low ecological risk. With respect to the sources of PAHs in sediments, authors found that coal combustion (56.6%), vehicular emission (29.2%) and coke oven (14.2%) were mainly responsible for the PAHs presence in the sediments.

Ruiz-Fernández [14] investigated the presence of PAH in sediments located in the lower reach of the Coatzacoalcos River, coastal area of Mexico. The researchers found that the total concentration of the 16 PAHs listed by the EPA was rather low below 100 ng/g dm, except for a peak value of 1,000 ng/g dm. The authors concluded that sediments had been chronically contaminated with oil since 1950s. The origin of PAHs in sediments was mainly related to the contamination released from waste from the oil industry. Although, according to the US EPA benchmarks, the PAHs concentration does not pose a risk for the benthic biota, monitoring for environmental risk is required.

Belin et al. [15] analysed the concentration of PAHs in sediments coming from the West Port, the west coastal waters of Malaysia. They found ranges of PAHs from 100.3 to 3,446.9 μg/kg dm. The highest pollution, mainly with 4–6 rings PAH, occurred in the area having high shipping activities. The authors concluded that in the studied area there were no and if present only slight adverse biological effects were with PAH exposure in surface sediment.

Sapota et al. [23] examined the level of pollution of, among others, PAHs in surface sediments from selected fjords, tidal plains and lakes of the North Spitsbergen. They found that the sum of the 16 PAHs in fiords and tidal plains was 10 times lower (16 PAHs to 36 ng/g dm) than those in surface sediments from lakes (16 PAHs to 429 ng/g dm). The most dominant were PAHs of high molecular weight (fluorine, chrysene, benzo(a)fluoranthene, benzo(a)pyrene, benzo(g,h.i)perylene and indeno(1, 2, 3-cd)pyrene).

Bathi et al. [24] determined the content of some PAHs in urban creek sediments. They examined out relationships between sediment PAH concentration and sediment particle size, volatile organic content (VOC), and sediment chemical oxygen demand. The highest PAH concentrations and the highest VOC correlated with the distribution by particle size-large diameter sediments, which was statistically significant.

Kafilzadeh et al. [25] investigated the concentration of PAHs in sediments taken from four stations on the Kor River, Iran. They found that four-ring PAH (fluoranthene) was the most significant pollutant in the sediments with the concentration varying from 26.33 to 64.14 ng/g dm, whereas in water samples three-ring PAH (acenaphthene) was in the range 15.11–65.72 ng/L. The lowest mean content of PAHs 5.02 ng/L for water and 13.43 ng/g dm for sediment samples was found in summer, whereas the highest mean content of PAHs equal to 11.88 ng/L for water and 24.63 ng/g dm for sediment occur in autumn. The effect of photo-oxidation, volatilization and high degradation during the hot season, are the factors responsible for the obtained results [13]. The authors evaluated the origin of PAHs taking into their calculation the ratios of the phenanthrene/anthracene and fluoranthene/pyrene, concluding that there was a pyrolytic source of PAHs in both sediments and water samples.

Jiao et al. [6] determined concentrations of PAHs sediments, varying from 52.3 to 1,870.6 ng/g dm (average 472.0 ng/g dm) coming from Chinese coastal and estuarine areas of the northern Bohai and Yellow Seas. The highest concentrations of PAHs were related to anthropogenic origin, mainly in the area where wastewater from small factories is discharged into the river without treatment. The content of organic carbon was of prime importance in the concentration of hydrocarbons in sediments. High-temperature pyrolytic processes such as combustion of fossil fuel are the factors most responsible for the presence of PAHs in sediments. At most location points, the obtained values were below the SQGs leading to the conclusion that ecological risk was rather low.

Adeleyea et al. [26] determined the concentration of PAHs in Chinese sediments (Yangtze Estuary and Hangzhou Bay) in amounts varying from 32.10 to 171.70 ng/g dm. They also studied the ecological risk by analysing the contents of PAHs in benthic organisms in amounts from 43.20 to 291.20 μg/kg dm concluding that bioaccumulation in marine organisms presented a low health risk with respect to PAHs.

According to studies carried out by Dong et al. [27], the total concentrations of the 16 PAHs in sediments originating from an industrial port in the southern Kaohsiung Harbor of Taiwan was in the range 4.425–51.261 ng/g dm (average 13.196 ng/g dm). The authors found that PAHs content was quite high in the river mouth region, and gradually decreased towards the harbour region. Coal combustion was regarded to be a possible source of hydrocarbons in the industrial port area. According to SQGs, the determined PAH levels could cause acute biological damage.

In the studies conducted by Andersson et al. [28] with respect to PAH contamination in lake Lille Lungegårdsvannet (Norway), it was found that urban fires, gasworks and traffic are responsible for the PAH sources in the sediments.

Recent studies carried out by Guang Gua et al. [29] proved the presence of PAHs in surface sediments from Fuxian Lake, China. The total PAH concentrations varied from 83.2 to 261.7 ng/g (mean concentration of 143.3 ng/g). The most prominent in sediments were four-ring PAHs (39.1%–73.9%). Biomass, coal and petroleum combustion were the most dominant sources of PAHs. It was found that lake sediments had a 9% incidence of adverse biological effects according to the mean effects range-median quotient [29].

An et al. [30] investigated PAHs contamination in the sediments of Manwan Reservoir in the middle of the Lancang River, China, with respect to potential ecological risk assessment. They found that the total concentrations of 16 PAHs were in the range 14.4–137.7 ng/g (70.68 ng/g dm mean concentration). Higher PAHs concentration was observed in the areas with residential settlement on large tributaries and near a dam. The authors classified the sediments as low to moderately polluted with PAHs. The concentration of PAHs in sediments is included in Table 1. The results indicate the high pollution of PAHs especially in river sediments.

In the latest studies, Lawal [31] confirmed the global presence of PAHs in both river and lake sediments including: the Mediterranean coastal environment of Egypt, Lake Michigan, Gorgan Bay, Caspian Sea, Gulf of Mexico, Taiwan, Brighton, UK, Akaki River, Lake Awasa and Lake Ziway, Ethiopia Mekonnen, Chandravanshi, Imam Khomeini Port, Persian Gulf, Iran, Mar Piccolo in Taranto (Ionian Sea), Southern Italy, China, San Francisco Bay, CA, USA, multi-industrial

Table 1
Concentration of PAHs in sediments

Place of sampling	Range of PAHs content	Sediment origin	Reference
Seine river, France	Up to 90 mg/kg dm	River	[12]
River Oder, Poland	65 mg/kg dm	River	[3]
Vistula river, Poland	0.3–6 mg/kg dm	River	[3]
Brahmaputra River, India	338–23,100 ng/g dm	River	[11]
Oder Haff	17,000 ng/g dm	River	[21]
Coatzacoalcos River, Mexico	100–1,000 ng/g	River	[14]
Lancang River, China	14.4–137.7 ng/g dm	River	[30]
Kor River, Iran	24.63 ng/g	River	[25]
Lake Byslawskie, Poland	3.3–20.7 mg/kg dm	Lake	[3]
North Spitsbergen	Up to 429 ng/g dm	Lake	[23]
Fuxian Lake, China	83.2–261.7 ng/g dm	Lake	[29]
Bohai Sea	149.24–1,211.81 ng/g dm	Sea	[13]
Baltic sea	11 ng/g–1,900 ng/g dm	Sea	[21]
Cantabric Sea, Spain	0.02–344.6 μg/g dm	Estuary	[31]
Yangtze Estuary	32.10–171.70 ng/g dm	Estuary	[26]
Costal sediments of Malaysia	100.3–3,446.9 μg/g dm	Costal	[15]
Kaohsiung Harbor, Taiwan	4.425–51.261 ng/g dm	Harbour	[27]

city, South Korea, Sydney Harbour, Nova Scotia, Canada, Indian Himalayan Region, Malaysia and Japan.

3. PCDDs, PCDFs and PCBs in sediments

The origin of dioxin in the environment has a mainly anthropogenic background related to the level of industry and urbanization. TEQ can be helpful in determining the value of background. In urbanised areas, the factor varied from 7 to 20 TEQ pg/g. In rural areas, the TEQ values were in the range of 1–6 TEQ pg/g, whereas in sediments ranged it up to 60 TEQ pg/g, respectively [32].

Dioxins occur in the water environment mainly due to air deposition, run off from agricultural areas, which were exposed to plant protection agents, run off of non-controlled landfills of wastes containing polyvinyl chloride and municipal wastes, industrial, and municipal wastewater [33].

It was found that during treatment of municipal wastewater, the accumulation of dioxins and furans in sewage sludge may range up to 90% compared with their content in wastewater [34]. However, the remaining amount entering to surface water can significantly get uploaded to surface waters and sediments resulting in the presence in water environment for many years. In surface waters, dioxins are able to bind to organic and inorganic particles [34]. In the water environment, dioxins are up taken and cumulated by algae, insect larvae, snails and fishes. It has been determined that concentrations in the aforementioned organisms can be over 9,000 times higher than in the surrounding waters [35]. It should also be pointed out that fish can be the first link in the chain in the migration of dioxins through fodder and food.

PCBs are regarded as xenobiotics resistant to decomposition under physical, chemical and biological conditions. The physical and chemical properties of PCBs have an influence on the migration of these compounds [3]. Therefore, the most volatilised and low chlorinated (one or two chlorine atoms per particle) are identified in the air on distances non-polluted with PCBs.

On the other hand, congeners, including more than two chlorine atoms, are present in surface waters and sediments. It has also been confirmed the presence of PCBs in aquatic organisms in fat tissue, which is a result of their high solubility in lipids. In such a way, they enter the trophic chain and thereby tissues in higher organisms as well as human body [2]. PCBs enter into an organism by inhalation, and those most exposed to these compounds are workers in industrial waste incineration plants as well as workers in transformer stations. The uptake of PCBs by organisms may occur through the skin as well as through food. In surface water, PCBs cumulate in sediments and then, move up the food chain to include tissues of aquatic organisms and then higher organisms [33]. The presence of PCBs in sediments is not persistent as the compounds may be released into the waters under changing environmental conditions. An example of biomagnifications of PCBs is described by Rapp [36], highlighting the problem of the accumulation of PCBs in ducks from eating fish and other aquatic organisms coming from polluted rivers.

The level of water contamination with PCDD as well as PCDF is not exactly known. Investigations into PCDD and PCDF contamination in sediments are carried out irregularly. Dioxins due to their low solubility in water, often occur in water in trace concentrations or below the detection limit. However, the concentrations of dioxins and furans have been determined in sediments. Fragmental investigations carried out in 2002 did not indicate the presence of dioxins and furans in waters of two main Polish rivers – Vistula and Oder river, whereas the total concentration of the aforementioned compounds in sediments amounted to 0.83 μg/kg dm. The highest concentration of octachlorodibenzodioxins and octachlorodibenzofurans up to 0.79 and 0.20 μg/kg dm, respectively, were found in sediments [37]. In this study, it was also proved that dioxins and furans cumulate in the fat cell of fishes. The total concentration of dioxins and furans was up to 0.645 μg per kg of fat [37].

Studies on the concentration of dioxins and furans in sediments originating from reservoirs in Central Poland were carried out by Urbaniak et al [38]. 7 compounds of dioxins and 10 of furans were determined. In sediments taken from Barycz reservoir on the Grabi River, the total concentration reached the value of 172 ng/kg dm for dioxins, and 31 ng/kg dm for furans. In sediments from Reservoir of Dolny Staw on the Sokółka River (right tributary of Vistula river, Poland), the total concentration of dioxins and furans reached the values of 446 ng/kg dm and 89 ng/kg dm, respectively. The total concentration of dioxins and furans with respect to World Health Organization (WHO)-TEQ had the values of 2.3 ng/kg dm (Barycz Reservoir, Poland) and 7.9 ng/kg dm (Dolny Staw, Poland), respectively [38].

Niemirycz and Jankowska [39] determined the concentration of PCDD and PCDFs in sediments of Polish rivers and the Gdansk Basin – Baltic Sea. The concentration of dioxins in sediments examined in the Polish costal area which amounts to 5 pg TEQ/g dm is treated as 'ecologically clean' zone. Higher dioxin concentration (51.23 pg TEQ/g dm) has been found in Wloclawek Dam Reservoir. The highest concentrations of tetra- and penta-congeners are still present in the surface layer of Gdansk Deep as a result of atmospheric deposition.

In Japan, studies determining the level of contamination with dioxins, furans, and PCBs in sediments in the area of Tokyo Bay were conducted. 11 points were chosen and sediments sample were taken from six layers from the following depths: (0 to 50 cm) 8–10, 18–20, 28–30, 38–40 and 48–50 cm, respectively. In sediment samples taken from the surface layer (0–2 cm), the total concentration of PCDD and PCDF varied from 3.1 to 49 pg TEQ/g. the PCBs concentration was in the range 0.18–3.0 pg TEQ/g. It was proved that dioxins and furans are mainly adsorbed onto the lowest fraction of the sediments. Over 90% of the aforementioned compounds were accumulated on particles lower than 75 μm. 40% of the total dioxins content was accumulated on the fraction lower than 5 μm, whereas 52% were accumulated on the particles from 5 to 75 μm. It was concluded that the main sources of these compounds were herbicides and deposition from polluted air [40].

Studies carried out in the United States proved that the concentrations of dioxins and furans in sediments originating from lakes gradually increased, and it was concluded that the highest amounts were found in the 70's (1,550 ng/kg dm). In the second half of 80's, these compounds were observed to decrease by half. Studies conducted in Europe revealed a similar trend. The sediments coming from the Baltic Sea in the beginning of 80's contained up to 1,800 ng/kg dm of dioxins and furans [41]. This study of sediments, which determined the concentration of seven PCB congeners, proved that the highest concentrations of these compounds were found in the sediments of Oder River in Chałupki (Poland) with value of 84.6 μg/kg dm. The criteria of the sediment quality found in the foreign sources declare information that sediments containing the above 20 μg/kg dm of PCBs should be treated as contaminated. Considering the aforementioned criteria, it appears that sediments coming from Polish rivers have low level of PCBs contamination, due to fact that among 36 tested samples only in 2 samples the concentration of PCBs exceed the mentioned value [42]. Besides the sediments in the Oder river in Chałupki, sediments taken from the Ner river contained a concentration of PCBs of 24.5 μg/kg dm. In sediments originating from the Baltic Sea that were investigated in the 90's, the concentration of PCBs varied from 10 up to 1,380 μg/kg dm [43]. Studies into the level of contamination with PCBs in sediments were carried out by Rosińska and Dąbrowska [44]. The sediment samples were taken from the reservoir of Poraj located in the upper water flow of the Warta river, Poland (Table 2). The occurrence of PCBs depended on the location of the sediment, and the highest concentrations were found in the place where industrial wastewater flows into the reservoir [44].

Urbaniak et al. [45] determined the selected PCB congeners in sediments in water from Reservoir Sulejówski (Poland) as well as the sediments coming from the Pilica river (Poland) [45]. They found that the total concentration of coplanar biphenyls varied from 0.58 to 6.30 ng/kg dm, whereas the remaining studied PCBs did not exceed a value of 20 ng/kg dm. The total concentration of PCBs was in the range 0.11–0.96 ng TEQ/kg with respect to the toxicity coefficient according to WHO.

In another study carried out on the German Coast in the neighbourhood of Peene river an average amount of PCBs equal to 122 μg/kg dm was found, whereas in Seine River the

Table 2
Range of PCBs concentration in sediment from Poraj reservoir (Warta river) and in the estuary of the Vistula/Oder rivers [37,44]

PCB	Range of concentrations			Fishes μg/kg dm
	Poraj Reservoir, μg/kg dm	Water in the estuary of the Vistula/Oder rivers, ng/L	Sediments of Vistula and Oder rivers, μg/kg dm	
2,4,4′-trichlorobiphenyl PCB 28	22.2–61.1	n.d.	n.s.	11.4
2,5,2′,5′-tetrachlorobiphenyl PCB 52	25.7–41.2	1.5	2.0	20.3
2,4,5,2′,5′-pentachlorobiphenyl PCB 101	31.4–51.6	n.d.	2.4	37.4
2,4,5,3′,4′-pentachlorobiphenyl PCB 118	635.1–726.7	n.d.	0.9	68.1
2,3,4,2,4′,5′-hexachlorobiphenyl PCB 138	48.5–58.1	n.d.	4.9	15.3
2,4,5,2′,4′,5′-hexachlorobiphenyl PCB 153	76.9–91.7	4.9	6.9	1.7
2,3,4,5,2,4′,5′-heptachlorobiphenyl PCB 180	Below 0.1	2.9	4.7	29.1
Coplanar (PCB 77, PCB 126, PCB 169)	n.s.	n.d.	0.9	69.9

n.d. – not detected; n.s. – not studied.

average amount found was 300 μg/kg dm [41]. In the last few years, a decrease in PCBs concentrations in sediments has been observed as a consequence of causing their production and limiting their application. PCBs were analysed in marine sediment samples collected from the coastal area of Barcelona (Spain) and near a underwater outlet coming from a waste water treatment plant located at the mouth of the Besòs River [22]. Concentration levels ranging from 2.33 to 44.00 ng/g dm for PCBs were found in the coastal sediments, while for samples collected near to submarine emissary higher levels varying from 22.34 to 37.74 ng/g dm of PCBs were obtained.

Lorgeoux et al. [11] stated that the total concentration of 15 PCB ranged the value 2.3 mg/kg is sediments of the Seine River basin, France. The data collected over 50 years allows to a long-term picture of the long-term environmental behaviour of POP in areas of anthropogenic pressures to be formed. Kim et al. [46] investigated the concentration of PCBs in surface waters and sediments at 20 measuring points in Japan. PCB concentration was in the range 0.18–3.0 μg TEQ/kg in sediments of the surface layer (0–2 cm) [40]. Much higher amounts of PCB were found in the rivers and sediments of Chinese bays. The highest concentration reached a value of 150 mg/kg dm and appeared in the most contaminated part of the studied areas [47].

Zhao et al. [7] determined the concentrations of 32 PCBs in sediments coming from the Haihe River and Haihe Estuary Area, Tianjin, China. They found that the concentrations were in the range of n.d. (not detected) to 253 ng/g dm; average value: 66.8 ng/g dm and from 0.997 to 2,447 ng/g dm; average value: 738 ng/g dm, respectively. The researchers concluded that the concentrations of all POPs from the Haihe River were higher than those from the Haihe Estuary Area. Tri-PCBs were dominant in sediment from the Haihe River. The authors compared the results with data originating from other areas around the world concluding that while concentrations of POPs in sediments from the Haihe River were higher, the level of contamination was similar to other areas in the Haihe Estuary Area. It was pointed out that POPs in the studied area pose a potential ecological risk in comparison to established SQGs,

Adeleyea et al. [26] determined the concentration of PCBs in sediments from the Yangtze Estuary and Hangzhou Bay (East China) in amounts varying from 0.60 to 63.00 ng/g dm. They also studied the ecological risk by analysing the content of PCBs in benthic organisms in amounts from 2.60 to 96.20 μg/kg dm concluding that bioaccumulation in marine organisms pose a significant health risk with respect to PCBs.

The determination of PCBs and hexachlorobenzene (HCB) in sediments coming from the Lenga estuary (central Chile) was also done by Pozo et al. [4]. The total PCB concentrations were in the range 20–10,000 ng/g dm. The PCB profile was dominated by middle to highly chlorinated congeners (6-Cl = 40% and 7-Cl = 30%). HCB concentrations were lower than the PCBs and varied from 1 to 870 ng/g dm. The authors also highlighted that the obtained values are much higher than those found in other coastal sediments of the world.

In the studied carried out by Sapota et al. [23] a total sum of seven PCB congeners varied from <0.9 to 5.6 ng/g dm in sediments from fjords and tidal plains and from <0.1 to 14.6 ng/g dm in sediments from lakes of the North Spitsbergen, respectively.

4. Chlorinated pesticides in sediments

The pathways of pesticide fates in the environment are often assigned by physical, chemical and biological conditions. In waters, the composition of microflora differs from those of soil microorganisms, and radiation access may cause photochemical degradation [2]. At the same time, the bioaccumulation of pesticides in aquatic organisms as well as adsorption onto solid particles can be observed. Aquatic organisms are cells in the food chain, whereas solid particles settle forming sediments. In sediments, pesticides undergo anaerobic fates. It is stated in the literature that a lack of oxygen favours decomposition of chlorine hydrocarbons. Susceptibility of pesticides to the decomposition varies under aerobic conditions. Based on data, the removal of pesticides during biological wastewater treatment in the maximum concentration of selected compounds allowing for their decomposition was determined. For example, the permissible aldrin concentration was equal to 1 mg/L, whereas the permissible concentration of DDT was estimated at a level below 50 mg/L [48]. This indicates various possibilities of decomposition of these xenobiotics under environmental conditions.

Investigations into the determination of pesticide concentration listed by the Stockholm Convention were carried out at the beginning of 90's. A quite high concentration of pesticides was identified in the sediments of the studied rivers. In sediments of the Oder river (Poland), the DDT and metabolites ranged around the value of 51 μg/kg dm, whereas concentrations of α-HCH and δ-HCH ranged around the value of 42 μg/kg dm. The concentration of γ-HCH (lindane) was four times higher ranging in value up to 177 μg/kg dm. In contrast to the mentioned compounds, the lowest concentration, up to 14 μg/kg dm was determined for methoxychlor (DMDT) [37].

Confirmation of the sorption of pesticides onto river sediments was obtained in the studies carried out in sediments originating from the following Polish rivers: Warta, Oder and Oława [49]. Seven chlorinated active compounds such as: α-, β-, γ-HCH, metabolites of dichlorodiphenyldichloroethane (DDD), DDT, dichlorodiphenyldichloroethylene (DDE) and DMDT were investigated. The partition coefficients for sediment–water were in the range 1.3–4.1 L/kg dm. The highest concentrations were determined for sediments coming from the Oder river. These sediments contained the highest amount of organic matter. The partition coefficients for TOC (total organic carbon) were in the range 3.7–5.1 L/kg of TOC [49].

In 2004, monitoring studies were carried out in sediments originating from the whole of Poland with respect to pesticides. In 80 samples investigated, the presence of the following pesticides was found: aldrin up to 2.5 μg/kg dm, dieldrin up to 1.2 μg/kg dm, endrin up to 2.9 μg/kg dm, respectively. The highest content of DDT was equal to 269 μg/kg dm. Furthermore, four HCH derivatives endosulfan, heptachlor and DDE and DDD were identified. The authors pointed out that the pesticides were present even though that pesticides containing aldrin, dieldrin and endrin were only applied up to the end of 1978 (Table 3) [43].

Other sources state that concentration of DDT in contaminated sediments of upper tributary of the Vistula river ranged in value up to 28,200 mg/kg dm [48]. A much higher concentration of 96,000 mg/kg dm was found for malathion. Waters contain a concentration of the mentioned compounds in the range 0.1–6 mg/L. The results of the investigations confirm the thesis of the possible accumulation of pesticides (also which are banned) and their retention in sediments for many years. Sadecka [48] mentioned that, according to the results of the studies carried out by other authors, the following dependence between the concentration of the pesticides in surface water and sediment could be determined: if the concentration in waters was in the range of 0.1–0.2 μg/L, the concentrations of pesticide in sediment were in the range 20–100 μg/L, respectively.

Studies of pesticide concentration listed by the Stockholm Convention were carried out in the sediments from Wrocław reservoir. The lowest concentration not exceeding a value of 0.04 μg/kg dm, was found for dieldrin, whereas the contents of aldrin and endrin were equal to 0.32 and 0.54 μg/kg dm, respectively. The concentration of DDT 1.69 μg/kg dm and heptachlor 2.79 μg/kg dm were of one order of magnitude higher. In Table 4, the concentration of pesticides listed in the Stockholm Convention in waters, sediments and fishes are included [50].

On the German Coast in the neighbourhood of the river Peene, an average amount DDT of 70 μg/kg dm was found [50], whereas in Lake Orta (Italy) sediments contained from 0.7 to 119.7 μg/kg dm of DDT [51].

Wang et al. [17] studied the contamination of Chinese sediments. They analysed 21 organochlorine pesticides (OCPs) in the Daling River area. They found that surface sediments were the most polluted with hexachlorocyclohexanes (HCHs) from 1.1 to 30 ng/g dm and DDTs from 9.1 to 53 ng/g dm, respectively. The aim of the researchers was to identify the sources, distribution and contamination history and ecological risk of the aforementioned compounds in sediments. It was concluded that the temporal trends of HCHs and DDTs concentrations in the sediments core occurred due to emission and soil residues. As a result of agricultural activities, new sources of lindane and dicofol appeared. Furthermore, the authors identified a slight potential health risk with respect to benthic organisms in Daling River and Bohai Sea according to available SQGs.

In the study conducted by Sapota et al. [23], selected chloroorganic pesticides (HCH isomers, DDT and its metabolites, aldrin, dieldrin, endrin, heptachlor, endosulfan I and II, and methoxychlor) in surface sediments from selected fjords, tidal plains, and lakes originating from North Spitsbergen were studied. Endosulfan I and methoxychlor were detected in concentrations near the detection limit (LoD) in 14 sediment samples.

Adeleyea et al. [26] determined the concentration of OCPs in sediments coming from the Yangtze Estuary and Hangzhou Bay (East China); the amount of HCHs varied from not detected (n.d.) to 22.40 ng/g dm, whereas DDTs was in the range from n.d. to 5.10 ng/g dm. The sediment ecotoxicological risk showed the toxic effect of DDTs on benthic organisms.

Recent studies carried out by Kumar et al. [52] found that the concentration of DDT varied from <0.01 to 128,600 ng/g whereas, ΣHCH/BHC was in the range <001–234,000 ng/g in various regions of India, respectively.

5. DEHP, NP and NPEs in sediments

Phthalates undergo a process of sorption and photodegradation, biodegradation and anaerobic degradation, which means they are easily released into the environment [53].

Table 3
Concentration of pesticides in sediments originating from Polish rivers (μg/kg dm) [43]

Place of sampling	Aldrin	Place of sampling	Dieldrin	Place of sampling	DDT
Czarna Hańcza (Frącki)	2.5	Brynica (Sosnowiec)	1.2	Brda (Bydgoszcz)	269
Liwa (Biała Góra)	1.6	Jeziorka (Konstancin)	1.2	Wisła (Oświęcim)	117
Wierzyca (Gniew)	1.2	Bug (Kryłów)	1.0	Wisłą (Tyniec)	45
Gwda (Ujście)	1.2	Oder (Chałupki)	1.5	Przemsza (Chełmek)	34

Table 4
Concentration of pesticides listed in the Stockholm Convention in waters, sediments and fishes [37]

Active substance	Water in the estuary Vistula and Oder rivers, ng/L	Sediments (estuary of Vistula and Oder rivers), μg/kg dm	Sediments from Włocławek Reservoir, μg/kg dm	Fishes (estuary of Vistula and Oder rivers) μg/kg of fat
Aldrin	15.3	10.3	0.322	16.9
Dieldrin	2.5	7.4	0.042	81.3
Endrin	n.d.	1,5	0.543	1.1
Heptachlor	27.9	94.6	2.795	0.7
DDT	5.2	2.4	1.692	25.3
HCB	4.3	3.3	n.data	23.2
Cis-chlordane/trans-chlordane	n.d.	3.6/13.4	n.data	52.4/ n.d.
Mirex/toxaphene	n.d.	n.d.	n.data	n.d./8.9

n.d. – not detected; n.data – no data.

The sources of NP and their ethoxylates (NPE) in sediments are mainly related to run off and leachates from landfills [18]. In wastewater treatment plants, nonylphenolethoxylate degrades into nonylphenol. Then, the remaining nonylphenols in the treated wastewater are drained to surface waters and can be found in river, sediments, soil and groundwater. In water environment, NPE occur in surface waters, soils, and sediments for months before being degraded. According to Soares et al. [5], the half-life of nonylphenol in sediment is estimated to be more than 60 years. On the one hand, a decrease in the concentration of nonylphenol in the environment has been found but on the other hand it is still found at concentrations of 4.1 μg/L in river waters and 1 mg/kg in sediments [5].

NPE are moderately easily cumulated in organisms [18,54]. In general, they are resistant to the process of decomposition, however under certain conditions they undergo fates forming metabolites of ethoxylates which are much more toxic than the substrates. Due to their properties of being able to bioaccumulate and to be persistent they can be transported over long distances and found far from the site of pollution [10].

There is limited information in the literary sources on the results of phthalates in the waters. The available data concern American sources of potable water. The concentrations of these compounds were in the range 0.01–65 μg/L, the average concentration calculated based on 181 samples was equal to 0.83 μg/L. In surface waters, the concentrations of DEHP were in the range 0.11–36 μg/L [55]. Lin et al. [56] compared the results of DEPH in sediments originating from various rivers from all over the world (Table 5).

A determination of the levels of nonylphenols and their ethoxylates was carried out by Kannan et al. [18]. The researchers determined the levels of the mentioned compounds in surface waters, sediments and fishes. In waters the concentrations of nonylphenols did not exceed 18 μg/L. NPE were also determined in organisms such as fish and their concentrations did not exceed 20.6 μg/L [18].

6. POPs in sediments – recent trends and challenges

POPs are ubiquitous pollutants able to be transported via air and water so they can be deposited far from the place of their release, where they remain in terrestrial and aquatic environment. Although POPs have been extensively used since the early 40's, before being banned, they still occur in the water ecosystems.

Table 5
Concentration of DEHP in sediments [56]

Place of sampling	Range of DEHP concentration mg/kg dm
Great Britain	0.8–115.0
Italy	0.058–0.487
Germany	0.21–8.44
USA	0.04–16.0
Japan	1.0–2.0
Taiwan	0.1–23.9
Malaysia	0.49–15.0

The hazard posed by contaminated sediments should take into consideration several aspects. It is necessary to highlight the sources of pollutants in the sediments. Increasingly, it is not only the estimation of the POPs concentration in sediments which is of interest but also their origin, the age of the sediments, sources of contamination, estimation of the ecological risk and their presence in aquatic organisms. Studies on POPs in sediments indicate that long-range atmospheric transport and industrial activities are responsible for local sources of POPs. Taking into account the age of sediment with respect to POPs pollution, the researchers pointed out that the transition from coal to natural gas was the main source of POPs, especially PAHs in sediments. In the recent literary sources, there is some evidence confirming that leisure boats can be treated as the source of PAHs in sediments and in the neighbourhood of both marinas and natural harbours [57]. The concentration of selected POPs in sediments was investigated by Combi et al. [58] along the Adriatic mudwedge and in deep-sea regions in the southern Adriatic basin. The researchers found that the Po river can be regarded as a major source of pollutants in sediments of the Adriatic Sea. The authors concluded that the flow of pollutants from coastal waters to the open sea was possible due to the cascading of the North Adriatic dense water in deep sea areas in the southern Adriatic, and enabling the quick transport of suspended sediments and particle binding of pollutants. Heise and Förstner [59] examined historical polluted sediments in the Rhine basin. They confirmed the various impacts from the aquatic environment both the natural background levels and the anthropogenic accumulation of contaminants. The authors proved that harbour basins, depressions, lakes, lowlands, flood plain soils and sediments were responsible for the level of pollution. The literature data referring to the persistent organic micropollutants is extensive. It confirms both the presence of the aforementioned compounds in all environmental elements including sediments as well as their toxic effect on organisms. The harmful impact increases if the accumulation of these compounds in tissues of organisms occurs and the formation of much more toxic by-products of metabolic fates they undergo takes place. Some results point out that the ecological risk connected with POPs is rather low, whereas others show that the determined POP levels in sediments could cause acute biological damages.

Dredging is another important aspect related to sediment contamination. Dredging is required mainly for navigation purposes for national and international transport. During this unavoidable process, sediments are disturbed and the pollutants bound to the particle surfaces can be released into the water environment. Some researchers indicate that the problem of dredging presents various issues depending to the country. For example, in developing countries (Panama Canal) the main limitations such as poor facilities and limited environmental expertise are responsible for the risk of environmental negligence [60]. Despite the fact that the maritime industry in Malaysia is treated as a priority by the government, effective monitoring of the influence of dredging together with an integrated environmental management framework is required. In the European countries (e.g., Ireland) the polluted sediments are excavated and transported for treatment and disposal [61]. In most cases, the dredged, contaminated sediments originate from historical industrial sources or former practices in the dredging area. The treatment of sediments is cheaper in the other European countries such as: Belgium €21/t to €28/t [62] and Germany €39/t for landfill [63]. The main problem with the treatment is that if sediments contain more than one pollutant no specific method can be applied, making it uneconomic. Sheehan et al. [61] recommend a disposal at sea charge as an alternative method. In Ireland, the application fee for a Disposal at Sea license is equal to €63.49 [64] whereas in the United Kingdom dredge disposal varies from £3,650 to £43,500 depending on the quantity of material to be disposed of and the type of project involved [65]. In the United States in the State of Washington, fees range from €0.07/t to €0.32/t ($\$0.10/yd^3$ and $\$0.45/yd^3$) for open-water dredge sediment disposal including costs of management and environmental monitoring [66]. In Ireland, a ton of waste for disposal in landfill was charged €142 [67].

With respect to dredging in Poland, in the coming 15 years the Polish government has already planned to restore the Polish rivers in order to re-establish navigability within the frame of the Programme of development of inland waterways. 15 mld€ is planned to be spent for this purpose. The following three international aquatic trails are planned to be restored: E30 (the Danube-Bratislava-Baltic Sea-Szczecin), E40 (the Baltic Sea, Gdansk-the Black Sea, Odessa) and E70 (Atlantic Ocean, Antwerp-the Baltic Sea, Klaipeda) [68].

In some European countries such as: Ireland, the United Kingdom and Belgium as well as in non-European countries as United States and Canada, contaminated sediments are classified using SQGs before being dredged [69–71]. The aim of the activities is to prevent both environmental and human exposure to the pollutant [72]. Together with SQGs, Water Guideline Values (WGVs) are also applied during monitoring water chemicals during dredging. WGVs takes into consideration water quality in aquatic water systems as well as quality of water intended for drinking purpose.

Taking into consideration the policy, it should be noted that the Stockholm Convention is a regulation with respect to the international significance of the problem of POPs. It forces members to fulfill the obligations included in the Convention to eliminate and restrict POPs that have toxicity, resistance to degradation, and bio-accumulative properties. Some data on POPs contamination indicate that the organic contamination tends to decrease over time reflecting the impact of environmental policies. The aim of the Water Frame Directive, Priority Substances Daughter Directive and the Marine Strategy Framework Directive is to protect the water environment by preventing the resource base upon which marine-related economic and social activities depend, and by implementing environmental quality standards for priority substances and other contaminants [9,73,74]. The EPA encourages the monitoring of the water quality during dredging in order to collect data for determining the influence of dredging as well as sediment disposal. The Regional Sea Conventions in Europe [75,76]: Convention for the Protection of the Marine Environment of the North-East Atlantic (OSPAR) and Helsinki Commission (the governing body of the Convention on the Protection of the Marine Environment of the Baltic Sea Area, also called Helsinki Convention have strategic objectives of preventing pollution of each convention area by continuously reducing discharges, emissions and losses of hazardous substances, with the ultimate aim of achieving concentrations in the marine environment that approximate background values for naturally occurring substances. It should be noticed that although legislation tools continue to play a significant role in environment, protection challenges still exist.

Sustainable management of contaminated sediments is another challenge. Organic contaminants create a serious problem posing risk to the environment in the Baltic Sea Region. For example in Sweden about 700 sites, containing approximately 10,000 m^3 of contaminated sediments each have been identified. Treatment of contaminated sediments is expensive, and on landfill costs approximately 100–200 Euro per cubic meter [77]. Thus, an international project of sustainable development of contaminated sediments was conducted within the years 2007–2013. Sweden, Finland, Lithuania, Poland and Germany participated in it. The goal of the project was to achieve a more sustainable management of dredged contaminated sediments in ports and fairways. In the final report, it was concluded that there is a need for national strategies or policies developed for the management of sediments. It was highlighted that at present, sediments are not seen as a resource. It was concluded that contaminants should be controlled ideally at the source.

Due to fact that the contamination with POPs in sediments is regarded as a global problem, it is necessary to find solutions which allow cleaning and, if this is not possible, at least to limit the risk of spreading pollutants from the sediments. As a wide range of available techniques is possible, it is important to identify the most reasonable one. In-situ technologies have economic advantages but they present a risk of the presence the contaminated sediments from their location. On the other hand, while ex-situ technologies by removing sediments eliminate contaminants, they increase the cost of removing sediments. Nowadays, it seems to be reasonable to apply a combination of more than one technology with special attention paid to not only economic factors but also non-toxic, easily available and eco-friendly for sediments remediation. Recently, Agarwal and Liu [78] propose the innovative method using ionic liquids to enhance oil recovery from sediments.

It should be pointed out that it is extremely difficult to compare data of sediment contamination carried out by different authors due to the various locations of sampling points, origin of the sediments, sources of their contamination and applied analytical methods.

The results of this brief overview indicate that sediment contamination with POPs requires regular, smart monitoring and that management should target the implementation of regulation to reduce pollution in the future.

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