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Status of polychlorinated biphenyl (PCBs) contamination in several marine and freshwater sediments in the world during the last three decades

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Status of polychlorinated biphenyl (PCBs) contamination in several marine and freshwater sediments in the world during the last three decades

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Abstract. Despite the research on PCBs occurrence in the various matrices in the world, no review has been carried out on the status of these substance contaminations in marine and freshwater sediments during the current periods throughout the globe. The objectives of this paper are to briefly discuss the occurrence of PCBs concentrations within marine and freshwater sediments in several places in the world and to determine which part of marine and freshwater sediments are the most contaminated in the world. The emphasis is on the last three decades namely from 1985 to 2020. The results suggested that during 1985 to 2010, the PCBs concentrations in the sediments ranged between $2 \mu\text{g.kg}^{-1}$ and $1648 \mu\text{g.kg}^{-1}$. The most elevated was found in Ionian Sea, Italia as the site where connected with high activities of industries, urban and harbor. The lowest one was reported in Bering sea and the Gulf of Alaska as the uncontaminated site during this era. Subsequently, a decade later which was recorded from 2011 to 2020 found the PCBs concentrations in the sediments ranged from 0.036 to $3730 \mu\text{g.kg}^{-1}$. The highest concentration was reported at coastal area at the center of Vietnam where linked with the urban and industries sites. Conversely, the lowest one was found at Chao River sediments where connected with the agriculture field. The evaluation of sediments guide lines quality through ERL (Effects Ranged Low) and ERM (Effects Ranged Medium) showed that the most contaminated marine and freshwater sediment in the world during 1985 to 2010 was at Ionian Sea, Italy, whereas in the recent period (2011 to 2020) was found in the Coastal area of Vietnam. According to this evaluation, these locations are high risk to the toxic effect particularly to the benthic organisms. Therefore, these places are high concerned in terms of protection and conservation of some marine biotas.

1. Introduction

Marine environment is considered as an important component that playing the role in determining the contaminants transport, fate and sinks in the environment such as persistent organic pollutants (POPs) including PCBs at local and worldwide scales [1], [2]. Prior to the regulation of PCBs under the Toxic Substances Control Act (TSCA) in 1976, PCBs were released (both accidentally and intentionally) into the atmosphere, water and land through sewers, smokestacks, storm-water runoff, spills and direct application to the environment. Large volumes of PCBs have been introduced into the environment through the burning of PCB-containing products, vaporization from PCBs-containing coatings and



materials, releases into sewers and streams, improper disposal facilities and by other routes (such as ocean dumping) [3]. Based on the current regulation of PCBs, the current primary (new) sources of PCBs contamination are limited to out dated or illegal landfills and scrap yards and leaks or explosions of electrical equipment and others (such as locomotive transformers) that may still contain PCBs [3]. Other sources are facilities or sites that were previously contaminated with PCBs are emitted and re-deposited to the environment via volatilization from water and soil, wet and dry depositions, and re-volatilization [4].

PCBs could be released into the environment from the sources as mentioned above through various mediums. Therefore, these substances have been detected in the different environmental matrices from various area throughout the globe. In the marine sediments [5], [6], [7], soils [8], [9], [10], [11] aerosols [8], [12], [13], [10] and biota [6], [13], [7].

Model study revealed the interconnection among several components such as soil, sediments, water, fish and air as the mediums where PCBs are transported in the river. A model has been developed and indicated that fish and sediments represent environmental compartments with the highest PCBs concentrations [15]. Nevertheless, the greatest mass of PCBs (over 70%) is likely to remain in the soil. As emissions decline, soil could then act as a significant secondary source of PCBs with the river bed-sediment functioning as a long-term reservoir of PCBs. They also mentioned that the most significant result was a tendency for climate change to enhance the evaporation of PCBs from soil to air. Therefore, PCBs as one of the persistent organic pollutants (POPs) are potential to be long-range transport [15].

However, sediments are not always being guarantee as a secure repository for contaminants. The alteration in external conditions (e.g., climate) in the sediments and the internal physical and biogeochemical conditions in the sediments and dredging spoil (e.g., pH, salinity and redox potential) could result in particles and dissolved contaminants being mobilized and entering the overlying water [16], [17], [18].

The existence of these hydrophobic and semi-volatile substances is of a particular interest because of their ubiquity, toxicity and accumulation in the components of food webs [2]. It was well-explained that water column dynamics have a significant impact on the air-sea exchanged of these compounds, as their efficient removal from the mixed surface layer decreases the volatilization rates and captures atmospheric pollutants [1], [19], [7].

On the other hand, in the aquatic ecosystems, the main indicators of pollution are contaminated sediments, which are the primary repository of chemical elements in the marine and freshwater environment. Thus, it is interested in studying on the distribution and the extended of these compounds either in marine sediments or freshwater sediments as their final repository. This paper is aimed at identifying and determining at least 30 years research on the occurrence of these substances in the environment, particularly in the marine and freshwater sediments. It is also evaluating the contamination level of PCBs during the last three decades and determining which part are the most polluted in the world. This information is expected to allow the future coastal guideline management for regional development in the several marine and estuary areas particularly a wise undertaking in the light of the ecological fragility of some areas, monitoring and surveillance of xenobiotic remediation that would be undertaken as well as the appropriate mitigation measures.

2. Materials and methods

The whole information from 1985 to 2020 were obtained through the original articles tracking from www.science.com as described within Table 1 in results and discussions below.

3. Results and discussions

3.1. *The distribution of the PCBs concentrations in the sediments around the world*

PCBs analysis from the sediment samples collected from marine and freshwater environment throughout the world have been long time exposed to the nature according to this review. Even though they have undergone variation degradation processes including UV-induced photodegradation, thermal

degradation, mechanical action, and biological interactions (e.g biodegradation). The properties such as hydrophobicity and semi-volatile substances would be greatly altered from the original sources and production. However, once these substances enter into the aquatic environments such as ocean, lake and river, they would be adsorbed onto the surface of sediments and other organic matter. After undergoing a path through the soil and atmospheric route, these PCBs will remain in the environment for long duration of time. The matrix where they would be accumulated in high proportion is sediments therefore this is the most interesting repository to study for the long time. This is because granulometric factors which promote their adsorption such as clay and organic matter [20]. Moreover, PCBs evenly transferred into water which would be evaporate into the atmosphere. Finally, they would be deposited through the rain and snow falls and some of them would be transferred by insect when they come into contact with them.

Table 1. The distribution of PCBs concentrations in the marine and freshwater sediments in the world

No	Sources	Concentrations	Locations	References
1	Deep Sea sediment	\sum PCBs ($130 \mu\text{g.kg}^{-1}$)	Gulf of Maine	[21]
2	Depth sediments	\sum PCBs ($6 - 65 \mu\text{g.kg}^{-1}$)	North Green Land	[22]
3	Harbor area	\sum PCBs ($7.2 - 81 \mu\text{g.kg}^{-1}$)	Waukagen Harbor, US	[23]
4	Local industry, agricultural, and domestic sewage	\sum PCBs ($0.006 - 2.2 \mu\text{g.kg}^{-1}$)	Aquatic ecosystem, Hongkong	[24]
5	Urban and industrial site	\sum PCBs ($2 - 116 \mu\text{g.kg}^{-1}$)	Essex Coast,	[25]
6	Uncontaminated area	\sum PCBs ($0.13 - 2 \mu\text{g.kg}^{-1}$)	Bering Sea and Gulf of Alaska	[26]
7	Agriculture and industrial area	\sum PCBs ($0.47 - 28.1 \mu\text{g.kg}^{-1}$)	Coast of North Vietnam	[27]
8	A very densely populated and highly industrialised	\sum PCBs ($105 - 400 \mu\text{g.kg}^{-1}$)	Western Scheldt river (Belgium)	[28]
9	Sewage sludge input from the river	\sum PCBs ($6.5 - 32.9 \mu\text{g.kg}^{-1}$)	Coastal Sea of Qing Dao, China	[29]
10	Industrial, Urban and Harbor activities	\sum PCBs ($2 - 1648 \mu\text{g.kg}^{-1}$)	Ionian Sea, Southern Italy	[30]
11	Industrial effluents, petroleum refineries, petrochemical plants	\sum PCBs ($184.16 \mu\text{g.kg}^{-1}$)	Guanaba Bay, Rio de Janeiro, Brazil	[31]
12	Sewage discharged without treatment	\sum PCBs ($1.3 - 384 \mu\text{g.kg}^{-1}$)		[32]
13	Entrance river, aquaculture area, slightly human activities	\sum PCBs ($0.91 - 6.48 \mu\text{g.kg}^{-1}$)	Bay Yangdian Lake, North China	[33]
14	High industrialization and urbanization	\sum PCBs ($< \text{DL} - 420 \mu\text{g.kg}^{-1}$)	Surabaya Coastal Area, Indonesia	[34]
15	Industrialized bays	\sum PCBs ($294 \mu\text{g.kg}^{-1}$)	Benthic ecosystem in Gwangyang Bay, South Korea	[35]
16	Harbor, urban runoff, effluent from sewages	\sum PCBs ($62 - 601 \mu\text{g.kg}^{-1}$)	Port of Spain, Trinidad and Tobago	[36]
17	Industrial, agricultural, urban effluents	\sum PCBs ($0.20 - 1.92 \mu\text{g.kg}^{-1}$)	Mediterranean Coast, Egypt	[37]

18	River Sediments	$\sum \text{PCBs}$ ($\sum \text{PCBs}$ ($15.1 \mu\text{g.kg}^{-1}$) ¹⁾)	Rio Negro River, Argentinean Patagonian, Argentine	[38]
19	Agricultural area	$\sum \text{PCBs}$ ($0.0039 - 0.0365 \mu\text{g.kg}^{-1}$)	Chao River, China	[39]
20	Industrial area, chemical industry, food and agriculture	$\sum \text{PCBs}$ ($2.8 - 435 \mu\text{g.kg}^{-1}$)	Huveaune River, France	[40]
21	Industrial/municipal wastewater discharges	$\sum \text{PCBs}$ ($0.02 - 15 \mu\text{g.kg}^{-1}$)	Pialassa Baiona, NW Adriatic, Italy	[41]
22	Agricultural area	$\sum \text{PCBs}$ ($0.6 - 35.6 \mu\text{g.kg}^{-1}$)	The Canang Island, Spain	[42]
23	Industrialization and oil refined	$\sum \text{PCBs}$ ($1.25 - 78.4 \mu\text{g.kg}^{-1}$)	State of Kuwait	[43]
24	Urban and industrial area	$\sum \text{PCBs}$ ($0.42 - 1232 \mu\text{g.kg}^{-1}$)	Jobos Bay, Puerto Rico	[44]
25	Industrialization, urbanization, agriculture	$\sum \text{PCBs}$ ($17.68 - 169.26 \mu\text{g.kg}^{-1}$) ¹⁾	Pearl River Estuary, China	[45]
26	Urban and industrial port, associated with anthropogenic activities	$\sum \text{PCBs}$ ($0.61 - 17.6 \mu\text{g.kg}^{-1}$)	Buenos Aires, Argentine	[46]
27	Industrial and urban area	$\sum \text{PCBs}$ ($<\text{DL} - 5.87 \mu\text{g.kg}^{-1}$)	Estuary of Babytonga Bay, Brazil	[47]
28	Urban Area	$\sum \text{PCBs}$ ($9.72 - 3730 \text{ ng.g}^{-1}$)	Coastal Area of Central Vietnam	[48]
29	Commercial Port, Port of Power Plant, Touristic beach	$\sum \text{PCBs}$ ($143 - 303 \text{ ng.g}^{-1}$)	Lebanese coastal zone	[49]
30	Agriculture and Industries	$\sum \text{PCBs}$ ($4.6 - 187.4 \text{ ng.g}^{-1}$)	Pearl river estuarien	[50]

As described by [20] in Figure 1, Bioaccumulation of PCBs refers to how a pollutant enters the food chain starting from the absorption process by phytoplankton and other microorganisms. Whereas, biomagnification refers to the tendency of pollutants to be concentrated and move from one trophic level to the next. As explained in Figure 1 [20] that when the accumulation that occurs at the level of phytoplankton, small fish, medium-sized fish, marine mammals to humans as the highest consumer level is the recipient of risk where the largest accumulation of these pollutants.

Like other organic micro pollutants, PCBs are able to induce mobile biomagnification [51], [52] with a residence time of 10 -15 years, [53], they are fat soluble and biologically active. Therefore, these chemical substances are persistent in the environment.

When the bioaccumulation process takes place, it will proceed to the biomagnification which will involve the food chain as a link. In biomagnification, there is an increase in the concentration of chemicals at each trophic level, so the higher the trophic level will be followed by increasing levels of these chemicals [51], [52]. There is the tendency of increasing levels of chemicals as trophic levels increase in the food chains. This process begins when producers take nutrients from the surrounding environment to be synthesized into complex molecules that are useful for biological processes. Because the availability of nutrients is limited in the environment, plants generally use their energy to pump nutrients actively enter the cell, they sometimes take more than needed and store it in the tissue, and finally the concentration of nutrients in plant tissue will be higher than the concentration in the

surrounding environment. These substances are chemically the same as some inorganic nutrients, they will also be absorbed and stored in plant body tissues.

The mechanism of contamination in fish [54], [55], which starts from the gills and goes to the skin while the contamination of food or particles that settles in water is absorbed and into the digestive system in fish. The effects of these poisons change according to the characteristics of each material that is contaminated and accumulates in the tissue system.

Humans as the highest level of consumers are inevitable from the effects of PCBs contamination. Several research results have proven that PCBs contamination can trigger several dangerous diseases such as gastrointestinal, breast and lung cancer [56].

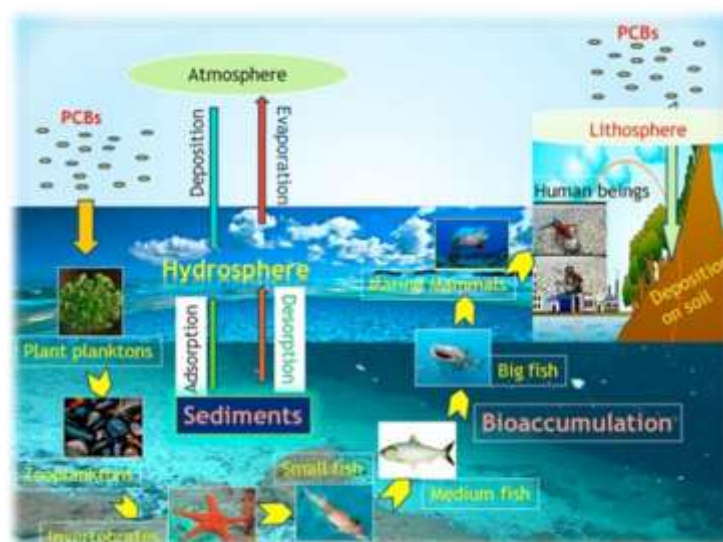


Figure 1. The PCBs transport in the environment from the sources to the different levels of food chain in the environment [20].

There were 44 research on PCBs occurrence within sediments from Asia to Africa and from Europe to America through the different methods. They are categorized in two cluster, the first one is from 1985 to 2010 and the second one is from 2011 to 2020. These works attributed to the new insight and gaining our understanding on how much these substances have been existing in the aquatic sediments such as marine and freshwater area according to the research which have been carried out. It is described the various concentrations among those publications. from not-detected to the highest concentrations which posed high risk to the benthic organisms. During these three decades, www.science.direct has published the investigation of PCBs concentrations isolated from the sediments from Asia as the highest frequently works of 43.18% from the total of 44 publications, followed by Europe of 22.72%. The third part was research in the America Continent of 18.18% and in the Middle East of 6.81%. Whereas in Africa people are less concerned to assess these chemicals in the environment particularly in the sediments. Asia continent is not only regarded as the center of industrial countries such as China and Hongkong but also as the most populated on earth. Additionally, Indonesia, Thailand, the Philippines and Vietnam as the developing countries extensively used the PCBs as an important group of industrial chemicals that were widely used as dielectric and coolant fluids in electrical equipment and as additives in many industrial and consumer [48]. Therefore, scientists are probably more concerned to investigate the occurrence of these compounds in the area. Since these contaminants have high accumulation potential in marine and freshwater environments where close to urban and industrial areas such in several developing countries and can be adsorbed in marine and freshwater sediment. This is probably bio-accumulated in

fatty tissues of living organisms through biomagnification processes [48]. The data also provided that Europe and America as the second and the third part of interesting places to be studied with the percentage of 22.72% and 18.18%. Whereas, Middle East and Africa were the last places with 6.81% and 4.54% of the investigations.

Since it has been divided into two large groups of the data in this work namely data from 1985 to 2010 and from 2011 to 2020 thus, it will be discussed first in the era of the 80s to the beginning of the 2000s. In this period there were 15 data in which the concentrations of PCBs ranged from 2 to 1648 $\mu\text{g.kg}^{-1}$. It was found that the highest concentration of PCBs in sediments was in the Ionian Sea, Southern Italy which is closely related to urban, industrial, and touristic activities (Table 1 and Figure 2). It is probably that the high concentration of this chemical compounds is also induced by the lack of wastewater treatment plant (WWTPs) in certain areas. During this period, advanced industrial countries such as China and the United States of America found less concentrations which only ranged between 2.2 - 384 $\mu\text{g.kg}^{-1}$. This is likely that better treatment of WWTPs and probably the application of capping or bioremediation that have been well implemented in some places.

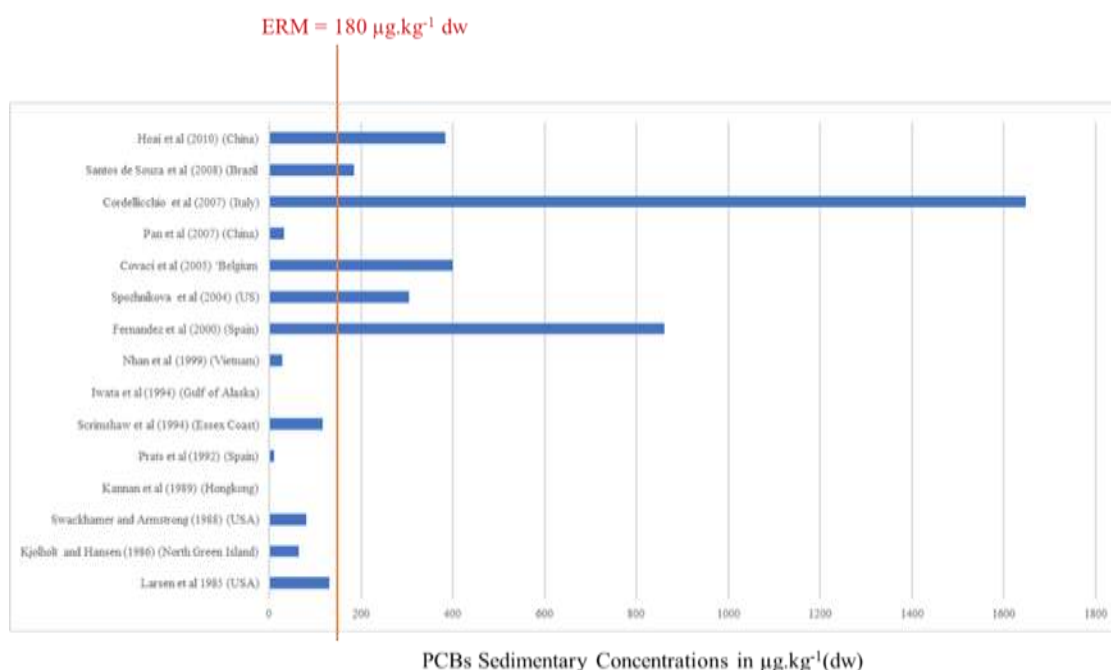


Figure 2. The environmental risk evaluation of PCBs contamination in the sediments during 1985 to 2010 throughout the world.

In the last decade, in the period of 2011 to 2020 there were 29 results of the research on the presence of PCBs concentrations in marine and freshwater sediments (Figure 3). It showed a range between 0.036 and 3410 $\mu\text{g.kg}^{-1}$. The highest concentration was found in Vietnam Sea sediment which was related to urban and industrial areas. As mentioned above, the high concentration of PCBs in the environment is most likely related to high industrial activity and high population density in the local area. This is very closely related to the needs and use of PCBs in various purposes. When it is compared to the two time periods from 44 data reviewed, it shows that over time the concentration of PCBs in the environment, especially aquatic sediment, is getting higher. The average concentration of PCBs in the 1985 - 2010 period was only 238 $\mu\text{g.kg}^{-1}$ while the last decade showed an average concentration of 403 $\mu\text{g.kg}^{-1}$.

In order to determine the environmental significance of those PCBs concentrations, we implemented the ERL (effect range low) and ERM (effect range median) sediment guide values.

These PCBs ERL-ERM type guide values have been used by the USEPA (United States Environmental Protection Agency) and are widely used around the world [30], [57]. According to figure 2. The distribution of PCBs concentrations during the era of 1985 to 2010 shows that 40% of the data far passed the ERM. This means that almost a half of the research suggested that PCBs are at high risk and toxic effect to the benthic organisms. Meanwhile the rest 40% probably posed the less risks. It was found 20% of the data which was uncontaminated. As discussed earlier, the most contaminated area during this period was discovered in Ionian Sea, Italy [30].

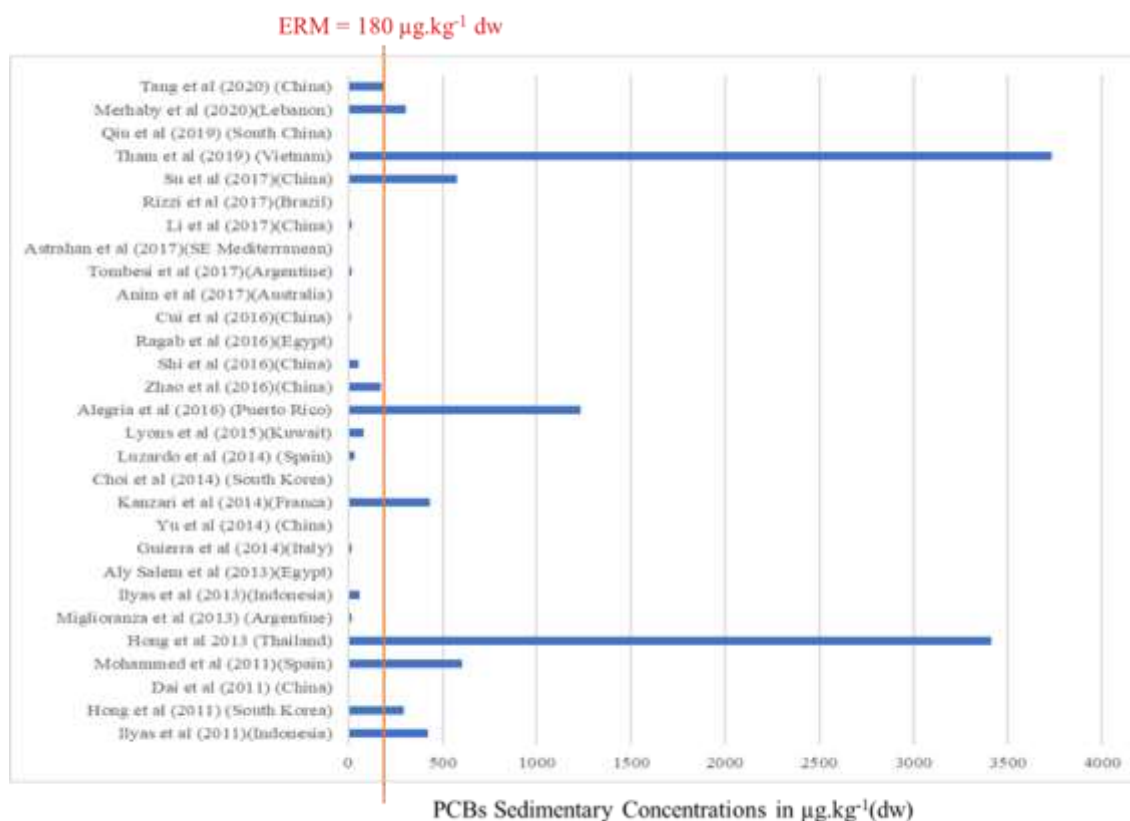


Figure 3. The environmental risk evaluation of PCBs contamination in the sediments during 2011 to 2020 throughout the world.

Subsequently, recent decade shows that 34.4% of the data exceeded the ERM. It is not significantly different compared to the past period which was 40%. It could be due to more research data during current works. However, the concentrations between ERL and ERM was only 17.24% much lower than in the past decades. Whereas the uncontaminated area was 48.36% of the data which is larger than in the era of 1985 to 2010. The highest polluted area by PCBs was found at Vietnam Sea, in the Southeast Asia which influenced from the industrial, urban and agricultural activities which can be the source of such pollutants.

4. Conclusion and perspective

Almost half (40 %) of the benthic aquatic ecosystem were at high risk of the PCBs contamination according to the data during at least the last three decades. Particularly in Asia and Europe, indicated that the biologically diverse of marine and freshwater organisms are threatened to be toxic by these substances.

Since the large number of benthic ecosystems in the world, both marine and freshwater, are at high risk of PCB toxicity, it is time to all stakeholders of taking into account to the right steps of the mitigation such as the improvement of WWTPs, capping or bioremediation.

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