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Regional and Seasonal Variations of Polychlorinated Biphenyls (PCBs) in Water of Shatt AL-Arab River, Iraq

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ABSTRACT

The concentrations of polychlorinated biphenyls (PCBs) were determined from Autumn season 2019 to summer season 2020 at six sites they are: (Al-Qurna, Al-Deer, Al-Qarma, Al-Ashar, Abu-Al-khasib, Al-Fao) in Shatt Al-Arab river, Basrah city, Iraq. Water samples were taken at each site and were analyzed using gas chromatography-mass spectrometry (GC–MS) type Agilent 7820A for PCB levels. The results indicate that PCBs are detected in all water samples and its concentration range from 2.61ng /l to 6.65 ng /l, 14.01ng /l to 21.19 ng /l, 7.67ng /l to 20.90 ng /l and 8.36 ng /l to 15.12 ng /l in Autumn, Winter, Spring and Summer seasons, respectively. The Al-Qurna and Al-Ashar sites were found to have the highest and lowest mean levels of PCB concentrations 0.201ng /l and 24.83ng /l, respectively. Total congener PCB (PCB 18, 29, 31,28, 44, 52, 101, 141, 149, 138,153, 189, and 194) concentrations at the water samples for all sites ranged from 2.61 ng/l in Al-Qurna station during autumn season to 24.83 ng/l in Al-Ashar station during winter season. Óhexa-PCBs and Ótri-PCBs were dominating in comparison with others PCBs congeners. This study is a first of its kind in the region and could give available information and could be uses as a baseline study in the futures.

Key words: *PCBs, Shatt AL-Arab River, Iraq*

Introduction

The Shatt al Arab River is formed after the confluence of the Euphrates and the Tigris Rivers near the city of Al-Qurna in southern Iraq. Downstream of Al-Qurna, the area draining to the Shatt Al-Arab region is shared between Iran and Iraq. In addition to the Euphrates and Tigris Rivers, the Karkheh and the Karun sub-basins contribute water to the Shatt Al-Arab. Both the Karkheh and the Karun Rivers originate in the Zagros Mountains in Iran and discharge into the Shatt Al-Arab (Al-Saad *et al.*, 2015).

PCBs are synthetic, aromatic compounds formed in such a manner that the hydrogen atoms on the biphenyl molecule (two benzene rings bonded together by a single carbon-carbon bond) may be replaced by up to 10 chlorine atoms related to biphenyl, which is a molecule composed of two benzene rings with a chemical formula of $C_{12}H_{10-x}Cl_x$, where X = 1-10, (Apitz *et al*., 2006).

PCBs are a class of non-polar toxic chemical compounds out of which only about 130 (di- to deca-PCBs) are found in commercial mixtures(Adeyemi *et al.*, 2009). They are also persistent organic pollutants (POPs) whose effects remain long after their usage (Wang and Zhong, 2011). They were synthesized for the first time in the last century and produced at an industrial level in the 1930's (Jang and Townsend, 2003).

Polychlorinated biphenyls (PCBs) were among the first twelve compounds included in the list of persistent organic pollutants (POPs) in May 2001 by the Stockholm Convection on POPs. (Zhang *et al*., 2014). Their physicochemical properties cause them to be persistent in the environment, and to exhibit tendencies for bioaccumulation, biomagnifications, long-range transport and deposition in places far removed from emission sources. In addition, they cause chronic toxicity and are ubiquitous in ecosystems and humans(Li *et al.*, 2011).

All congeners of PCBs are lipophilic and their lipophilicity increases with increasing degree of chlorination. However, they have very low water solubilities. Congeners with a lower degree of chlorination are more volatile than those with a higher degree. Pure individual PCB congeners are colourless and often crystalline. Commercial PCB mixtures are clear to light yellow oils or resins and they do not crystallize, even at low temperatures (Apitz *et al.*, 2006).

PCBs have many applications including their use as additives in paints, and as coolants and insulating liquids in power capacitors and transformers (Duan *et al.*, 2013). In addition, PCBs have been used as pesticide extenders, hydraulic fluids, flame retardants, cutting oils, in carbonless copy paper, as stabilizing additives in PVC coatings, adhesives, sealants, wood floor finishes, and casting agents (Batterman *et al.*, 2009; Halfadji *et al*., 2013).

The adverse effects of PCBs on humans include impaired reproduction, cancer, neuro-developmental effects in infants, endocrine disruption, and Immunotoxicity (Barakat *et al.,* 2013). In addition, exposure to dioxin-like PCBs is known to cause adverse changes in hepatic micronutrient homeostasis. These changes exacerbate severe liver damage (Klaren *et al.,* 2015). PCBs stimulate changes in the DNA sequence (Ludewig and Robertson, 2013) and oxidative stress (Buha *et al*., 2015). For these reasons, the production and use of PCBs has been banned globally, but large quantities of these compounds are still found in old power capacitors and transformers (Iwegbue, 2016). PCBs from these sources can be released into the environment through runoff, volatilization, oil leakage, waste discharges, and dry and wet deposition, thus causing pervasive environmental effects (Li *et al*., 2013; Zhang *et al.*, 2007)

PCBs may be released into the environment from hazardous waste sites containing PCBs, illegal or inappropriate dumping of PCB waste, and leaks from electrical transformers containing PCBs. PCBs can be carried long distances in the air and remain in the air for about 10 days. In the water, a small amount of PCBs may remain dissolved, but most of them stick to organic molecules and sediments. PCBs accumulate in water in fish and marine mammals and can reach levels thousands of times higher than those found in water (Rahuman *et al.*, 2000).

Shatt Al-Arab River receives many pollutants when it is passing through Basrah City, due to many human activities and factories which discharge their wastewater into the river without any real treatments. It has been for that reason, Shatt Al-Arab River used as a sink for wastes from agricultural, industry and other human activities due to its flow and ecological nature.

The present work was aimed at determining the concentrations of PCBs for water samples in Shatt Al-Arab River in Basrah city, south of Iraq. We believe that this project is the first work that reports PCBs concentrations in Iraq country. So, the main objective of the present study in shatt Al-Arab was to:

- Determination of the concentrations and types of PCBs in six different stations along the Shatt al-Arab from the cross of the Tigris and Euphrates in the city of Qurna to its estuary in the Arabian Gulf in the Basra Governorate, southern Iraq.

Materials and Methods

Water samples were collected seasonally during the period from autumn season 2019 to summer season 2020 at six stations in Shatt Al-Arab river at Basrah city for analysis and estimation the concentration of some polychlorinated biphenyls (PCBs). GPS instru-

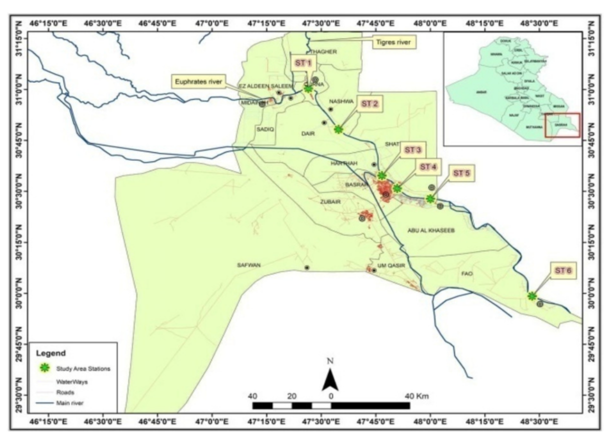

Fig. 1. The study stations

ment is used to fix the positions of these stations Figure (1).

The stations are : Al-Qurna (St.1), Al-Deer (St.2), Al-Qarma (St.3), Al-Ashar (St.4), Abu-Alkhasib (St.5) and Al-Fao (St.6) as shown in above map

Water samples were collected from six stations with dark glass bottle 5L at a depth about 25 cm below the water surface in the middle of the river and added 10 ml of chloroform to fix of samples. water samples were transferred into a Whatman Soxhlet cellulose thimble (previously extracted with Dichloromethan DCM). The sample-standard mixture was subjected to Soxhlet extraction for 18h with 50 ml of DCM/n-hexane (1:1 v/v). Elemental sulfur was removed from the extracts using activated elemental copper in order to avoid sulfur interferences when using gas chromatography.

The PCB compounds concentrated on a rotary evaporator, transferred to a vial, and the volume was adjusted to 1 ml . The concentrated extract was purified on a column packed from bottom to top with Glass Wool, 1 g each of anhydrous Na_2SO_4 and silica gel. Elution with 50 ml of DCM provided the PCB fraction then transformed to vial until measurement. The extract of water samples was determined according to the method of (USEPA, 2006).

Results and Discussion

The PCB concentrations in the water are displayed in Tables (1-4).The results show the concentrations of ©13PCBs compounds in water samples ranged from (2.62 ng/l) in the Qurna station during autumn season to (24.83 ng/l) found in the Al-Ashar station

Table 1. Concentrations of PCBs (ng/l) in water samples at the studied stations during Autumn season.

Compound Name	Al-Qurna	Al-Deer	Al-Qarma	Al-Ashar	Abu-Al Khasib	Al-Fao
PCB 18	0.05	0.12	0.17	0.21	0.03	1.19
PCB 29	0.00	0.02	0.09	0.33	0.05	0.36
PCB 31	0.00	0.25	0.51	0.63	0.47	0.18
PCB 28	0.13	0.17	0.16	0.12	0.09	0.15
PCB 44	0.00	0.00	0.1	0.03	0.01	0.00
PCB 52	0.59	0.64	0.76	0.41	0.14	0.14
PCB 101	0.21	0.32	0.13	0.26	0.18	0.22
PCB 141	0.67	0.21	0.29	1.14	0.76	1.77
PCB 149	0.05	0.09	0.13	0.12	0.19	0.27
PCB 138	0.00	0.45	1.23	1.31	1.81	1.91
PCB 153	0.22	0.27	0.14	0.12	0.00	0.00
PCB 189	0.00	0.45	0.00	0.00	0.00	0.00
PCB 194	0.69	0.32	0.18	0.6	0.33	0.46
TOTAL PCBs	2.61	3.31	3.89	5.28	4.06	6.65
MEAN	0.201	0.255	0.299	0.406	0.312	0.512

Table 2. Concentrations of PCBs (ng/l) in water samples at the studied stations during Winter season

Compound Name	Al-Qurna	Al-Deer	Al-Qarma	Al-Ashar	Abu-Al Khasib	Al-Fao
PCB 18	0.55	0.75	0.30	1.27	0.22	1.49
PCB 29	1.91	1.86	2.62	1.45	3.31	1.44
PCB 31	0.00	0.00	1.89	2.19	0.00	0.00
PCB 28	1.32	1.00	1.41	2.20	1.37	1.01
PCB 44	0.05	0.12	0.12	1.25	0.10	0.04
PCB 52	1.69	1.66	3.52	1.81	2.17	2.02
PCB 101	1.31	1.84	1.97	1.05	4.42	3.87
PCB 141	1.57	1.29	2.18	2.81	2.20	2.55
PCB 149	0.72	1.47	1.76	0.21	1.80	1.58
PCB 138	2.32	2.39	2.25	2.66	3.15	2.83
PCB 153	0.00	0.00	0.00	0.00	0.00	0.00
PCB 189	0.00	0.00	0.00	4.03	0.00	2.33
PCB 194	2.57	2.69	1.34	3.90	2.10	2.03
TOTAL PCBs	14.01	15.07	19.36	24.83	20.84	21.19
MEAN	1.078	1.159	1.489	1.91	1.603	1.63

Table 3. Concentrations of PCBs (ng/l) in water samples at the studied stations during Spring season.

Compound Name	Al-Qurna	Al-Deer	Al-Qarma	Al-Ashar	Abu-Al Khasib	Al-Fao
PCB 18	0.70	1.40	1.25	1.42	3.78	0.16
PCB 29	1.28	0.00	0.00	1.04	1.00	0.00
PCB 31	0.00	0.40	0.00	0.00	0.75	0.00
PCB 28	0.30	0.00	0.25	0.74	0.82	0.13
PCB 44	0.01	0.98	0.00	0.29	0.04	0.03
PCB 52	1.82	2.04	3.07	1.29	1.52	5.76
PCB 101	1.80	2.10	2.13	3.79	0.98	5.74
PCB 141	0.00	0.00	0.00	3.30	0.00	0.00
PCB 149	1.76	1.57	2.96	1.18	0.33	2.81
PCB 138	0.00	2.45	3.84	4.73	1.75	2.48
PCB 153	0.00	1.33	0.00	0.00	0.00	0.00
PCB 189	0.00	1.38	0.00	0.00	0.00	0.00
PCB 194	0.00	1.86	1.72	1.36	6.15	3.79
TOTAL PCBs	7.67	15.51	15.22	19.14	17.12	20.90
MEAN	0.590	1.193	1.171	1.472	1.317	1.608

Table 4. Concentrations of PCBs (ng/l) in water samples at the studied stations during Summer season.

Compound Name	Al-Qurna	Al-Deer	Al-Qarma	Al-Ashar	Abu-Al Khasib	Al-Fao
PCB 18	0.31	0.57	1.45	3.76	0.65	0.59
PCB 29	0.00	0.00	1.37	1.58	0.00	0.00
PCB 31	2.10	2.70	2.40	0.00	4.18	1.15
PCB 28	0.03	0.09	0.58	0.62	0.10	0.19
PCB 44	0.00	0.01	0.03	0.12	0.01	0.04
PCB 52	1.20	2.61	1.13	1.63	1.08	1.16
PCB 101	1.88	1.75	1.42	2.21	2.20	1.57
PCB 141	0.00	0.73	0.00	0.00	0.00	0.00
PCB 149	1.61	1.69	2.25	1.23	3.73	3.52
PCB 138	0.31	1.86	3.38	4.92	3.69	4.49
PCB 153	0.00	0.00	0.00	0.00	0.00	0.00
PCB 189	0.00	0.00	0.00	0.00	0.00	0.00
PCB 194	0.92	1.42	2.42	3.91	1.95	2.41
TOTAL PCBs	8.36	13.43	16.43	19.98	17.59	15.12
MEAN	0.643	1.033	1.264	1.537	1.353	1.163

during winter. The results of the study showed that PCBs compounds decreased during the Autumn season while increased during the winter season, as the concentrations in the Qurna station ranged between 2.61 ng/l to 14.01 ng/l, Al- Deer station concentrations ranged between 3.31 ng/l to 15.51ng/l , Al-Qarma station ranged between 3.89 ng/l to 19.36 ng/l, Al-Ashar station, the values ranged between 5.28 ng/l to 24.83 ng/l, Abu Al-khasibstation ranged between 4.06 ng/l to 20.84 ng/l, and the values in Al-Faw station ranged between 6.65 ng/l to 21.19 ng/l (Figure 2).

Significant differences (Pd≤0.05)were found among seasons for PCB29, PCB31, PCB28, PCB52, PCB101, PCB141, PCB149, PCB138 and 4PCB19.The highest mean value (2.098ng/l) was recorded in

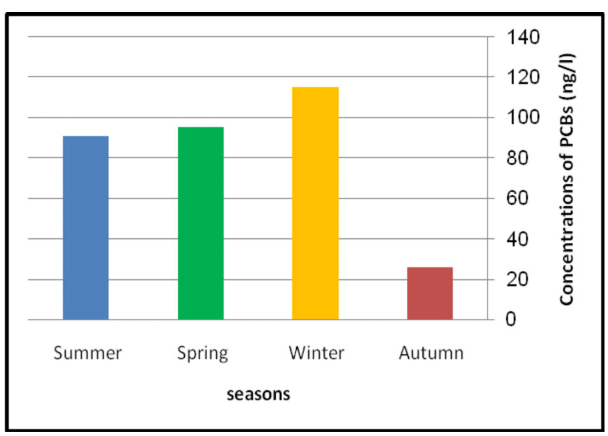

Fig. 2. Seasonal variations of total of PCBs concentrations Values at study stations

winter, while the lowest mean value (0.138ng/l) was recorded in autumn for PCB29, The highest mean value (2.088ng/l) was recorded in summer, while the lowest mean value (0.191ng/l) was recorded in spring for PCB31, The highest mean value (1.385ng/l) was recorded in winter, while the lowest mean value (0.268ng/l) was recorded in summer for PCB28, The highest mean value (2.583 ng/l) was recorded in spring, while the lowest mean value (0.446ng/l) was recorded in autumn for PCB52, The highest mean value (2.410ng/l) was recorded in winter, while the lowest mean value (0.220ng/l) was recorded in autumn for PCB101, The highest mean value (2.100ng/l) was recorded in winter, while the lowest mean value (0.550ng/l) was recorded in spring for PCB141, The highest mean value (2.338ng/l) was recorded in summer, while the lowest mean value (0.141ng/l) was recorded in autumn for PCB149, The highest mean value (3.108ng/l) was recorded in summer, while the lowest mean value (1.118ng/l) was recorded in autumn for PCB138 and the highest mean value (2.480ng/l) was recorded in spring, while the lowest mean value (0.430ng/l) was recorded in autumn for PCB138 Figure 3.

Significant differences were found among stations for PCB28 and PCB138, the highest mean value (1.047ng/l) was recorded in station 2, while the lowest mean value (0.335ng/l) was recorded in station 4 for PCB28, The highest mean value (3.405ng/l) was recorded in station 4, while the lowest mean value (0.657ng/l) was recorded in station 1 for PCB138.

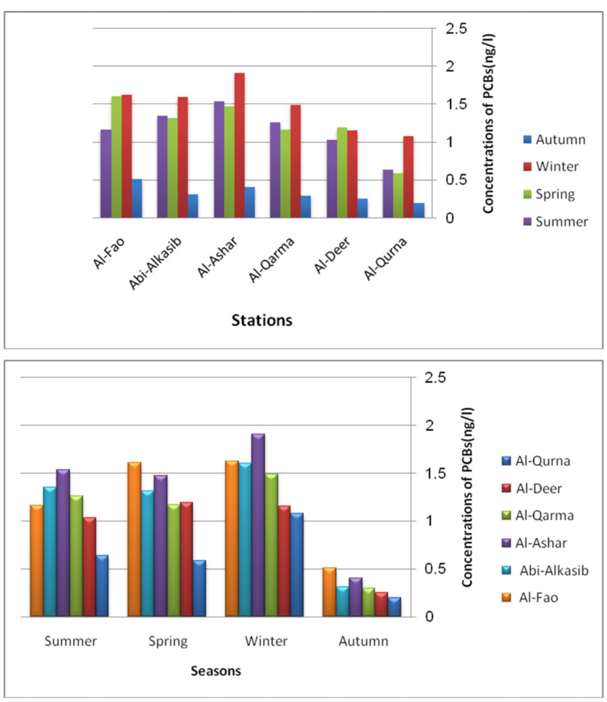

Fig. 3. Mean concentrations of PCBs Compounds at study stations

There was a positive significant correlation between PCB18 and between PCB194(r= 0.690, P<0.01)and PCB138 (r= 0.433, P<0.05), A positive significant correlation was recorded between PCB29 and between PCB141(r= 0.555, P<0.01) and PCB28 (r= 0.782, P<0.05), A positive significant correlation was recorded between PCB28 and between PCB189 (r= 0.529, P<0.01) and PCB141(r= 0.708, P<0.01) and PCB44 (r= 0.429, P<0.05), A positive significant correlation was recorded between PCB44 and between PCB189 (r= 0.784, P<0.01) and PCB153 (r= 0.524, P<0.01), A positive significant correlation was recorded between PCB52 and between PCB149 (r= 0.498, P<0.05) and PCB101 (r= 0.739, P<0.01), A positive significant correlation was recorded between PCB101 and between PCB138 (r= 0.458, P<0.05) and PCB149 (r= 0.577, P<0.01), A positive significant correlation was recorded between PCB141 and between PCB189 (r= 0.430, P<0.05), A positive significant correlation was recorded between PCB149 and between PCB138 (r= 0.529, P<0.01) and a positive significant correlation was recorded between PCB18 and between PCB194 (r= 0.477, P<0.05).

Pollution of surface water and groundwater through unstable sources has been considered an important environmental issue in the last decade. The increasing exploitation of natural resources, the development of oil industries, refineries, domestic wastewater and industrial wastewater lead to the entry of chemical pollutants into aquatic ecosystems (Mills and Chichester, 2005). Among the most important pollutants of rivers, are hydrocarbons in petroleum, which consist of thousands of chemicals in different physical forms (gas, liquid, solid), and pose a great danger to aquatic life and lead to compound formations such as biphenyl (Esmaili *et al.*, 2015).

PCB congeners reach water bodies by runoff and/or air transport. In aquatic ecosystems, PCBs can bioaccumulate in the food chain and accumulate in top predators through the consumption of contaminated water and organisms (Bjermo *et al.*, 2013).

Results of the present study showed the highest levels of PCB in water during winter season ,while the lowest levels observed during autumn season, which indicates the effect of temperature on the removal most of PCB from water (Thomas, 1982).

Temperatures affect in two main directions, the first one is to increase evaporation, in this process, carbon compounds with low molecular weights evaporate as well as the breakdown of carbon compounds which have high molecular weights(Lyons *et al.,* 2015). The second affect of temperature including increase the enzymatic activity of microorganism that use oil as an important source of organic carbon (Shamshoom *et al.,* 1990). Harnstrom *et al.* (2009) demonstrated that about 20 to 50 % of PCB compounds will be lost during evaporation processes, also the increase of enzyme activity of microorganisms increases when temperature in the range 30 CÚ to 40 CÚ. The increasing in the total concentration of PCB compounds in spring and winter is due to the fact that compounds that are input to the environment are higher in spring and winter due to the increasing in burning fuel. As well as the low rate of evaporation processes to the PCB compounds in the winter and decreasing the effectiveness of different microorganisms in degradation processes of these compounds with low temperatures. In addition high levels of PCB were shown during winter this may be attributed to the increase in fall of airborne materials from atmosphere during rain of these compounds which occur during winter (Leahy and Colwell, 1990).

Al-Ashar station showed increase in PCBs concentrations in the water due to human activities and various industrial wastes that are widely spread at the Basra Governorate center, Then the concentrations increase in the Abu Al-Khasib and Al-Faw stations, respectively, which discharge pollutants from the industrial facilities scattered near those stations. The increase of PCBs may be due to the presence of large numbers of fishing boats, and hydrocarbons transporting boats such as petroleum, These pollutants are released into the water during transportation, leakage and accidents, in addition to the presence of the Abu Flos port on the western side of the Shatt al-Arab in the Abu al-Khasib district and the Abadan refinery on the eastern side located in the Siba area which adds many industrial pollutants to the Shatt al-Arab estuary, as well as sewage and agricultural activities pollutants. Therefore, this study showed an increase in PCBs in the central and southern part, compared with the northern part of the Shatt al-Arab river.

The low concentrations in the autumn season are attributed to the warm climate of Iraq, as high temperatures cause evaporation of PCB compounds in water. The high temperatures also stimulate the microorganisms to break down these compounds, especially the low molecular weights. In addition, the oxidation processes due to the long period of brightness and intensity of solar radiation. so, the other factor is photo-oxidation, which results in the breakdown of compounds in the water column, so that this process is very important to change these compounds into simple ones (Ehrhardt and Petrick, 1993).

A negative significant correlation was recorded between PCB31and between dissolved oxygen ($r=-0.512$, $P<0.05$) and recorded same compound a positive significant correlation between each of water temperature ($r= 0.508$, $P<0.05$) and air temperature ($r= 0.601$, $P<0.01$), A positive significant correlation was recorded between PCB52 and between salinity ($r= 0.452$, $P<0.05$) and E.C ($r= 0.473$, $P<0.05$) and recorded same compound a negative significant correlation with water temperature ($r= - 0.424$, $P<0.05$), A positive significant correlation was recorded between PCB101 and between each of salinity ($r= 0.426$, $P<0.05$) and E.C ($r= 0.434$, $P<0.05$), A positive significant correlation was recorded between PCB141 and between dissolved oxygen($r= 0.571$, $P<0.01$) and a positive significant correlation was recorded between PCB149 and between each of salinity ($r= 0.504$, $P<0.05$) and E.C ($r= 0.511$, $P<0.05$)

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