

# Removal of Polycyclic Aromatic Hydrocarbons from Polluted Water Using Constructed Wetlands: A Review

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## Author Details

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## Article History

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**Abstract:** During the last decades, Polycyclic Aromatic Hydrocarbons (PAHs) are detected in agricultural soils and water bodies at concentrations higher than permitted limits. The main sources of PAHs are fossil fuels, biomass burning, waste incineration, and pesticides. They harm aquatic ecosystems and organisms and pose a high risk to human health. For these reasons, several laws have been enacted to regulate the permitted concentration levels of PAHs in the environment and specifically, the wastewater discharged into aquatic ecosystems. Several methods for the removal of PAHs from wastewater exist which, although efficient, are not used extensively due to their high installation and operation cost. Constructed Wetlands (CWs) constitute an effective, practical, and low-cost option for PAHs removal from polluted waters. This paper focuses on the various treatment processes for the removal of PAHs. Research findings indicated that vertical flow CWs and hybrid systems are more effective in removing PAHs.

**Keywords:** PAHs; Constructed wetlands; Wastewater; Naphthalene; Anthracene; Phenanthrene; Benzo[a]anthracene; Tetracene; Chrysenium, Benzo[a]pyrene

## Introduction

During the last decades, environmental pollution and especially the atmosphere's contamination had a serious impact on increasing common environmental problems such as water and soil pollution. Analytically, we define water pollution as any form of unwanted changes in the physical, chemical, and biological characteristics of water. The main sources of this type of pollution are the point sources and non-point sources (or diffuse pollution).

Irrespective of the lakes' and rivers' size, we mark that pollutants could exist, caused by various human activities which are constantly increasing. One of the most dangerous pollutants for all living beings is Polycyclic Aromatic Hydrocarbons (PAHs).

PAHs have been studied extensively in recent bibliography where many researchers focus their works on water systems and current installations. Specifically, a property of great concern is the PAHs concentration values

that tend to exceed the limits set by the European Union (EU) legislation and the individual countries alike [1]. PAHs have been detected at concentrations of up to 1.2 µg/L and 2.16 µg/L in Raba river, Hungary [2] and in Svratka river, Czech Republic [3], respectively. The PAHs anthracene and fluoranthene have also been measured at concentrations 0.17 µg/L and 0.93 µg/L, respectively, in Nestos river N. Greece [4]. A two-year study performed by Witt [5] at the Baltic Sea, found out that the concentration values of PAHs in water were abnormal. Specifically, the results of this paper indicate that the levels of PAHs in the spring were significantly lower than those of the autumn. As shown by this study, the reason for this observation was attributed to the fact that in the autumn and winter, the temperature of the water is low, thus, the number of microorganisms is lower meaning that PAHs are not capable to oxidize by them. Moreover, it was observed that UV radiation during the autumn period was not sufficient for photooxidation. As a result, the percentage levels of PAHs in the fall ranged from 0.001 ng/L to 4.8 ng/L. Furthermore, the PAHs of the two and three benzene rings



were at higher concentrations than the PAHs of the six benzene rings, the average concentration of which was 0.1 ng/L. Lastly, it was noticed that the concentration levels of PAHs with six benzene rings remained unchanged throughout the year as the natural degradation processes of PAHs were insufficient.

Schulz-Bull et al. [6] studied the PAHs detection levels in the North Atlantic Ocean around Iceland. PAHs of three to five benzene rings with concentrations above the permissible limit of 5 pg/L were measured. In particular, the concentration of PAHs ranged between 5 and 30 pg/L with phenanthrene and fluoranthene having the highest concentrations. In Colombia, the lack of environmental controls and legislation regarding water pollution has resulted in significant quantities of PAHs being released annually in water systems. To put things into perspective, in 2007, the local newspaper El Timepo published that about 500 tons of waste per day were discharged into the country's water systems [7]. Moreover, Sarria-Villa et al. [7] measured the concentration of PAHs in water and sediments at eight stations along the Cauca River. The PAHs measured in the most water samples were: fluorene, acenaphthylene, and anthracene, while in the sediments were: benzo[b]fluoranthene, benzo[k]fluoranthene, and pyrene. The PAHs concentrations levels in water ranged from 1.60 to 101.8 ng/L and in sediments from 0.4 to 26.5 µg/kg. Due to the widespread presence of PAHs, it is important to further develop technologies for the treatment of water contaminated by them. For this purpose, several treatment technologies have been recommended such as chemical degradation, use of radioactivity and ultrasonic, advanced oxidation, biological degradation, and Constructed Wetlands (CWs) [8-10]. Among them, CWs are an efficient method with low operational and maintenance costs, and environmental impact.

This review aims at investigating the PAHs removal from wastewater and urban runoff, using CWs. For this, scientific literature was searched on Scopus, Science direct, Scholar Google, etc. Based on our findings, we have developed a database that includes the PAHs, the CW type as well as the design parameters of the CW such as dimensions, plant species, wastewater type, and the PAHs removal rate.

## PAHs- Definition

PAHs are a class of pollutants derived from the incomplete combustion of petroleum products. Other sources of PAHs infection are the following [11]:

- Natural gas
- Burning of wood/forests
- Volcanic eruptions
- Waste incineration
- Pesticides

Furthermore, PAHs pollution derives from our everyday activities and our recent lifestyle. As such, some of the most common sources creating PAHs are the following [12]:

- Carbon-containing fuels: used for optimizing engines' operation (petrol engines etc.).
- Construction materials: regarding both traditional and contemporary industry processes.
- Household products: such as paints, finishes, coatings of crude oils, etc.
- Grilling food: as most grill machines run on coal.

## Constructed Wetlands

CWs are natural systems that mimic the functions of natural wetlands (NW). Modern CWs have been designed and constructed for improved

treatment capacity. The size and type of CWs, in addition to economic factors and budget restrictions, depend on various factors such as: the type of pollutants, the volume of wastewater, the meteorological conditions and the climate of the area, and the degree of wastewater treatment [13].

The basic types of CWs, in terms of the hydrological mode of operation, are the following [14,15]:

- Free water surface (FWS) CW has areas of open water (Figure 1) while the water flows at the surface of the bed.
- Horizontal Subsurface Flow (HSF) CW, planted with wetland vegetation, and the water moves horizontally below the surface of the porous media from the inlet to outlet (Figure 2).
- Vertical Flow (VF) CW, designed to have vertical water flow. Similar to HSF they are planted and the water is treated as it percolates through the plant root zone (Figure 3).

CWs are the proposed solution for the treatment of various types of wastewaters such as: municipal, urban runoff, rainwater, agricultural wastewater, and industrial wastewater [16-20].

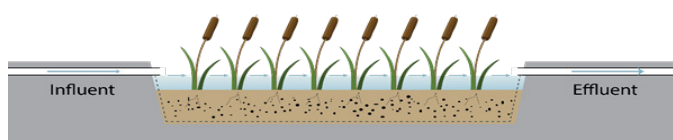


Figure 1: Free water surface CW.

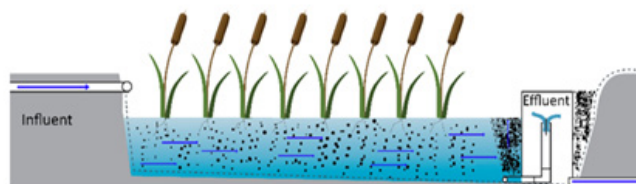


Figure 2: Horizontal subsurface flow CW.

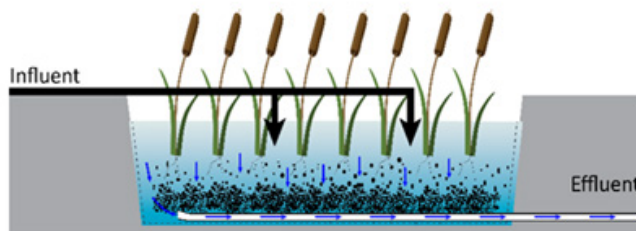


Figure 3: Vertical subsurface flow CW.

## Results

Based on our study, after examining several articles on the use of CWs for the removal of PAHs from the wastewaters, a Database (DB) containing information on design parameters and performance of CWs was developed. Specifically, we indicatively present the use of constructed wetlands (Table 1) from our constructed DB, as well as data (Figure 5) regarding interesting publications in the field. As evident, our analysis gathered information on various properties, such as CW type, size, plant species, PAHs, wastewater type and removal rates.

(Figure 4) shows the removal rates of PAHs for different processes where the highest percentage of PAHs removal rates existed in the process regarding lab scale CW-MP. This type of CW is filled with freshwater mussels. The removal efficiency for five PAHs (i.e.,  $C_{12}H_{10}$ ,  $C_{12}H_8$ ,  $C_{13}H_{10}$ ,  $C_{16}H_{10}$ ,  $C_{16}H_{10}$ ) was equal to 97%. The second highest percentage of PAHs removal occurred in processes of biological treatment in combination with (a) Ozone (Alto Seveso) as well as with (b) Peracetic acid (Nosedo)

and the removal efficiency was equal to 96,5% and 89,5% respectively. Furthermore, the third highest percentage of PAHs removal exists in the process of the Hybrid (VF+HSF) system of CW. The removal efficiency was equal to 94%. Also, the rest of the processes have a moderate efficiency of approximately 60-80 % in the removal of PAHs. Lastly, as presented in (Figure 4), when CW is used in individual systems, the mean removal efficiency is about 70%.

(Figure 5) presents box-whisker plots of the PAHs removal in FWS, HSF, VF, and Hybrid (VF+HSF) CWs. The line inside the box denotes median values, the upper box frame denotes the 75th percentile, the lower box frame denotes the 25th percentile, and the two whiskers denote the

**Table 1:** The use of constructed wetlands for PAHs removal.

CW type	Size	Plant species	PAHs <sup>a</sup>	Wastewater	Removal (%)	References
HSF	20 pilot scale systems 90x30x50 (cm) (L x W x H)	Phragmites - Vetiver	(5), (11), (16)	Artificial	66-83	[21]
HSF	45 m <sup>2</sup>	Phragmites australis and Arundo donax	(1) - (16)	Municipal	79	[22]
FWS	42 m <sup>2</sup>	Phragmites australis & Arundo donax	(1) - (16)	Municipal	68	[22]
FWS	5 m <sup>2</sup>	Unplanted	(1) - (16)	Municipal	73	[22]
FWS	2 systems: 11x3x0.4 (m) (L x W x H)	Arundo donax, Phragmites australis	(1) - (16)	Highway runoff	49-51 58-57	[23]
HSF	2 pilot scale systems: 4x8x0.5 (m) (L x W x H)	Arundo donax, Phragmites australis	(1) - (16)	Highway runoff	59-47 71-63	[23]
VF	14 pilot scale systems: high 70 cm, diameter 25cm	Acorus calamus	(8)	Artificial	83 - 99	[24]
HSF	4 pilot scale systems 60x40x50 (cm) (L x W x H)	Oenanthe javanica (Blume)	(1), (2), (13), (16)	Municipal	88 - 99	[25]
FWS	-		(3), (5) - (8), (12), (14), (15)	Municipal	5-31	[26]
VF	4 pilot scale systems: high 70 cm, diameter 25cm	Iris pseudacorus	(11)	Municipal	90 - 94	[27]
VF	3 pilot scale systems: 90 m <sup>2</sup>	Phragmites australis	(1), (3) - (6), (9), (11) - (13), (16)	Municipal	50 - 90	[28]
VF	-		(1), (4) - (7), (9), (11) - (16)	Municipal	> 80	[29]
VF	-	Dianella revoluta	(11)	Municipal	84	[30]
VF	4 pilot scale systems: 66x25x37 (cm) (L x W x H)	Acorus calamus, Canna indica, Phragmites australis	(5), (11), (16)	Municipal	> 99	[31]
HSF	Pilot scale: 30x30x30 (cm) (L x W x H)	L. articulate	(1) - (16)	Municipal	86-90	[32]
HSF VF	10 pilot scale systems, total 37.5 m <sup>2</sup>	Phragmites australis	(3) - (7), (9) - (14), (16)	Landfill leachate	94	[33]
HSF	10 pilot scale systems: 50x50x0,6 (m) (L x W x H)	Phragmites australis	(3) - (7), (9) - (14), (16)	Landfill leachate	63	[33]
FWS	2 pilot scale systems: 160x75x70 (cm) (LxWxH)	Phragmites australis	(3), (12)	Municipal	> 80	[34]

The correspondence of the numbers with the PAHs is as follows: (a) (1) Acenaphthene, (2) Acenaphthylene, (3) Anthracene, (4) Benzo(a)anthracene, (5) Benzo(a)pyrene, (6) Benzo(b)fluoranthene, (7) Benzo(g,h,i)perylene, (8) Benzo(k)fluoranthene, (9) Chrysene, (10) Dibenzo(a,h)anthracene, (11) Phenanthrene, (12) Fluoranthene, (13) Fluorene, (14) Indeno(1,2,3-c,d)pyrene, (15) Naphthalene, (16) Pyrene.

FWS: Free Water Surface Flow; HSF: Horizontal Subsurface Flow; VF: Vertical Flow

max and min values. Comparing the three CW types (i.e., FWS, HSF, and VF), higher PAHs mean removal was observed in VF CW. More specifically, the VF-CW showed better performance than HSF-CW and FWS-CW, and HSF-CW showed better performance than FWS-CW. The combination of VF and HSF-CWs (hybrid system) showed the highest PAHs removal.

(Figure 6) presents the performance of the technologies used to remove PAHs besides the CW. Analytically, the highest percentage rate of PAHs removal occurs in the photolysis and catalytic wet peroxide oxidation processes, as well as in all processes of photolysis (UV-C) whereas, the lowest effectiveness occurs in the methods of biological degradation.



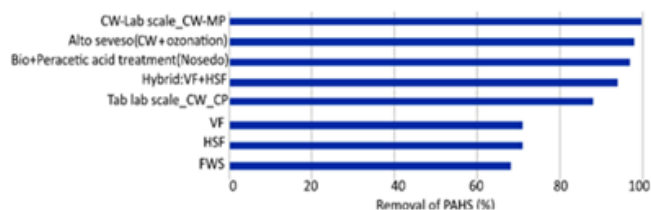


Figure 4: Comparative analysis of removal rates of PAHs with different CW systems.

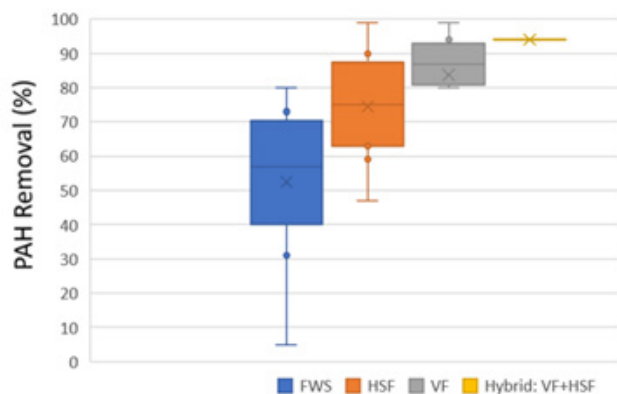


Figure 5: Removal efficiency of CW.

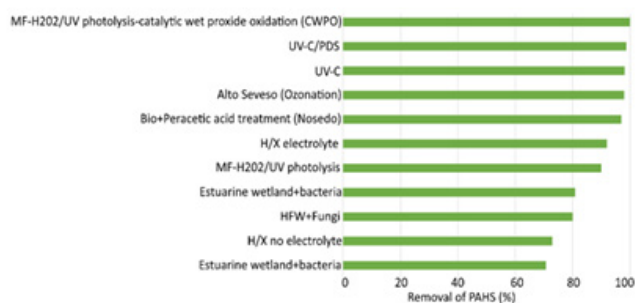


Figure 6: Comparative analysis of removal PAHs technologies.

## Conclusion

In this article, the effectiveness of CW in the removal of PAHs from wastewater and various other removal processes were studied. The combination of the biological method with ozonation showcased promising results whereas, the oxidation-based methods (e.g., electrochemical oxidation) released ions into treated effluents that were harmful to animals and human health. Moreover, in various articles, the photodegradation method was one of the most efficient methods in the removal of PAHs but the overall construction and operating cost make it difficult to produce and maintain. Lastly, the efficiency of the microbiological treatment method was low and perhaps negligible compared to other PAHs removal methods.

In conclusion, as evident from above, the CW technology offers the optimal trade-off between cost and efficiency as: (a) the PAHs removal rate is high and, in most cases, greater than 80%, (b) it uses natural materials for its construction, and (c) construction and operating cost is at reasonable levels, thus making it an affordable environmentally friendly solution.

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