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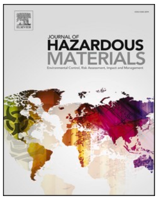

Enhanced photoelectrocatalysis for oxidation of organic pollutants and metal recovery from polluted water and sediments

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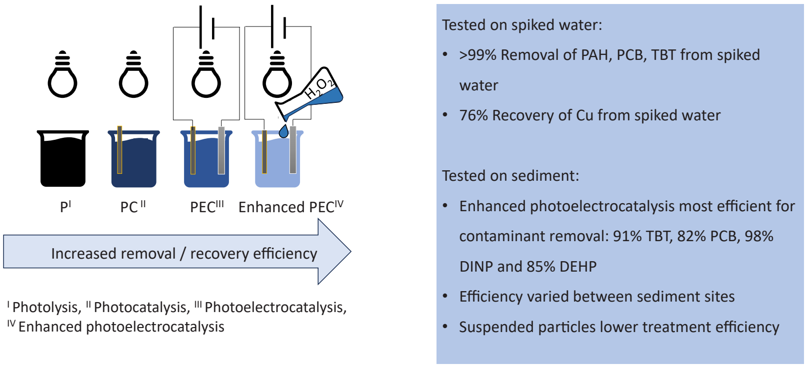
HIGHLIGHTS

- Effective photocatalytic TiO_2 electrodes for PEC made by suspension thermal spraying.
- Effective PEC oxidation of TBT, PCBs & PAHs (>99 %) in clear water solutions.
- PEC achieves 76 % Cu recovery from clear water solutions at the cathode.
- PEC combined with H_2O_2 reagent treated the polluted sediments most effectively.
- Removal efficiencies from sediment: 91 % TBT, 82 % PCB, 98 % DINP, and 85 % DEHP.

GRAPHICAL ABSTRACT



Tested on spiked water:
- >99% Removal of PAH, PCB, TBT from spiked water
- 76% Recovery of Cu from spiked water

Tested on sediment:
- Enhanced photoelectrocatalysis most efficient for contaminant removal: 91% TBT, 82% PCB, 98% DINP and 85% DEHP
- Efficiency varied between sediment sites
- Suspended particles lower treatment efficiency

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ABSTRACT

Sediments often contain a complex mixture of organic and inorganic pollutants, including polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), aliphatic and aromatic hydrocarbons, phthalates, tributyltin (TBT), and metals. Since large quantities of contaminated sediments are regularly dredged, it is necessary to develop methods to simultaneously treat the different contaminants. These techniques should also handle complex sediment matrices where pollutants may be strongly bound, e.g., TBT in paint flakes and PAHs in tire particles. This work has focused on the development of photoelectrocatalytic (PEC) processes to degrade organic pollutants (OPs) and simultaneously recover metals from marine, brackish and stormwater sediments. The remediation efficiencies were studied in real contaminated sediments and clear water solutions spiked with PAHs, PCBs, TBT, and metals. In spiked water, PEC yielded a > 99 % reduction of PAHs, PCBs, and TBT, and recovered 76 % of copper. The pollutant removal from the sediments by PEC was less efficient, especially for

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PAHs in stormwater sediment. However, the combination of PEC and H_2O_2 reduced other groups of OPs: TBT by 91 %, PCB by 82 %, and phthalates DINP and DEHP by 98 % and 85 %, respectively. The release of pollutants from sediments into the water phase is the key to successful PEC application.

1. Introduction

Today, the global economy depends on shipping, with more than 80 % of global trade volume transported by sea [1]. Dredging of sediment is vital for maintaining water depths and allowing cargo ships access to ports and other critical marine structures. In Europe, the annual volume of dredged sediment is estimated at over 200 million m3 [2], and large volumes are often handled at major ports. As an example, the largest port in Scandinavia, the Port of Gothenburg (Sweden), needs to remove around 200,000 m^3 of dredged material every five years, of which 75 % is polluted with tributyltin (TBT) and metals, mainly due to previous activities [3]. In addition to severe contamination in large ports, many urban waterways and coastlines are contaminated with metals and organic pollutants, such as polychlorinated biphenyls (PCBs), due to ongoing and past activities [4–6]. To reduce the risk posed by contaminated sediment, dredging is sometimes needed. Urban stormwater and road runoff from highly trafficked areas are also highly polluted with a cocktail of pollutants, including metals, tire wear particles and organic pollutants such as polycyclic aromatic hydrocarbons (PAHs) [7–9].

While it is possible to use dredged material in construction projects, fine-grained dredged sediment is often limited due to contamination, and it is therefore often deposited in landfills. This, however, is not compatible with sustainable development ambitions, and finding sites for landfilling is becoming increasingly complex owing to environmental goals and regulations. It is therefore urgent to develop alternative solutions, including sediment treatment methods [10]. Pollutants in sediments often occur in a complex mixture of organic pollutants and metals, and organic pollutants such as PAHs may be strongly bound in oils and tire and road wear particles [11]. In marine sediments, organotin compounds may also be strongly bound in old paint flakes [12]. Methods used to remove or degrade organic pollutants in sediment include leaching, biological degradation, phytoremediation, chemical oxidation, photocatalysis, electrolysis, and thermal treatment [2,13–17]. Among these, a combination of photocatalytic and electrolytic processes, known as photoelectrocatalysis (PEC), has demonstrated promising results for the oxidation of organic pollutants in water [18,19]. Photoelectrocatalysis involves a series of chemical reactions. In the photocatalytic phase, photons are absorbed by a semiconductor photocatalyst (e.g., TiO_2) [20,21], which converts light energy into chemical energy. The photon energy (hv) excites electrons from the valence band to the conduction band, generating electron-hole pairs (e^--h^+). These can lead to the formation of reactive radicals, such as the most commonly formed hydroxide radicals (•OH), and superoxide radicals ($\bullet O_2^-$), through either photogenerated electron-driven oxygen reduction reactions (Eqs. 1, 2 and 3) or hole-dominated water oxidation reactions (Eqs. 4 and 5) [22,23]. Also, the highly reactive singlet oxygen ($^1O_2$) is an essential species in advanced oxidation processes, including photocatalysis and electrocatalysis [24]. The radicals then degrade organic molecules (RH) (Eqs. 6 and 7) [25,26]. Applying an electric current reduces electron-hole pair recombination, increasing the efficiency of photocatalytic reactions. When a cathode is added, metal recovery becomes possible (Eq. 8), as the dissolved metal ions are attracted, reduced, and deposited on the cathode surface via electron transfer [26]. At the anode, water is oxidized and •OH is generated proportional to the electrical current applied (Eq. 9) [13].

Electrolytical methods alone have also proven suitable for fine-grained sediments, as the particles can adsorb most metals, resulting in high electrical conductivity and a strong electric field [2,27]. A study using boron-doped diamond (BDD) electrodes showed promising results in oxidizing organic pollutants, such as a 58 % reduction in TBT [13]. To further improve the removal of organic pollutants, enhanced photooxidation could be achieved by adding strong oxidant hydrogen peroxide (H_2O_2) [13,15,28,29]. The addition of H_2O_2 increases the formation of radicals and prevents the recombination of electron-hole pairs (Eq. 10) [25]·H_2O_2 can also react with ferrous iron (Fe^{2+}) in the sediment, leading to the Fenton reaction (Eq. 11) [13]. However, there seems to be a lack of literature on PEC studies that aim to degrade organic pollutants while simultaneously recovering metals from sediment.

$$O_2 + 2e^- + 2H^+ \rightarrow H_2O_2 \tag{1}$$

$$O_2 + e^- \rightarrow \bullet O_2^- \tag{2}$$

$$\bullet O_2^- + e^- + 2H^+ \rightarrow H_2O_2 \tag{3}$$

$$2H_2O + 2\,h^+ \rightarrow H_2O_2 + 2H^+ \tag{4}$$

$$H_2O + h^+ \rightarrow \bullet OH + H^+ \tag{5}$$

$$RH + \bullet OH \rightarrow H2O + R. \rightarrow \text{further oxidation} \tag{6}$$

$$C_xH_yO_z + h^+/\bullet OH \rightarrow CO_2 + H_2O_2 \tag{7}$$

$$\text{Cathode: } M^{n+} + ne^- \rightarrow M(s) \tag{8}$$

$$\text{Anode: } H_2O \rightarrow \bullet OH + H^+ + e^- \tag{9}$$

$$H_2O_2 + hv \rightarrow 2\bullet OH \tag{10}$$

$$Fe^{2+} + H_2O_2 \rightarrow \bullet OH + OH^- + Fe^{3\,+} \tag{11}$$

This study aimed to develop an effective laboratory-scale method for treating contaminated water and sediment by combining photocatalysis and electrolysis to degrade organic pollutants and recover metals simultaneously. To the authors' knowledge, this is the first time a PEC method has been used on contaminated sediments. This study used different electrode materials and electrode potentials to assess the oxidation of TBT, PAHs, and PCBs in water solutions and TBT, DBT, MBT, PAHs, PCBs, aliphatic hydrocarbons, and phthalates in sediment-water-slurries from different sites with varying sediment properties, such as morphology, chemical composition, and pollutant concentration and sources. Additionally, the metal recovery potentials for Cu, Zn, Pb, Cd, Ni, and As were studied. The PEC experiments were used to develop and evaluate the method's potential, rather than to give statistically reliable results. The studied sites are located in Gothenburg, Sweden, and the samples included TBT polluted sediment from the port area, PCB polluted sediment from a bay, and PAH polluted sediment from a sedimentation pond in an urban area highly impacted by traffic.

2. Experimental

An overview of the steps performed is shown in Fig. 1. To begin with, electrodes were developed, and the degradation of methylene blue in aqueous solution was investigated to assess the treatment efficiency under controlled conditions. The methods were then tested on spiked water solutions. Finally, the methods were applied to four different sediment samples.

2.1. Photocatalytic electrodes

The photocatalytic electrode consisted of a 50 mm × 30 mm × 3 mm titanium alloy (Ti-6Al-4V) plate coated with a photocatalytic (TiO_2) layer. The TiO_2 layer was deposited by Suspension Plasma Spraying using an Axial III thermal spray gun (Northwest Mettech Corp., Canada)

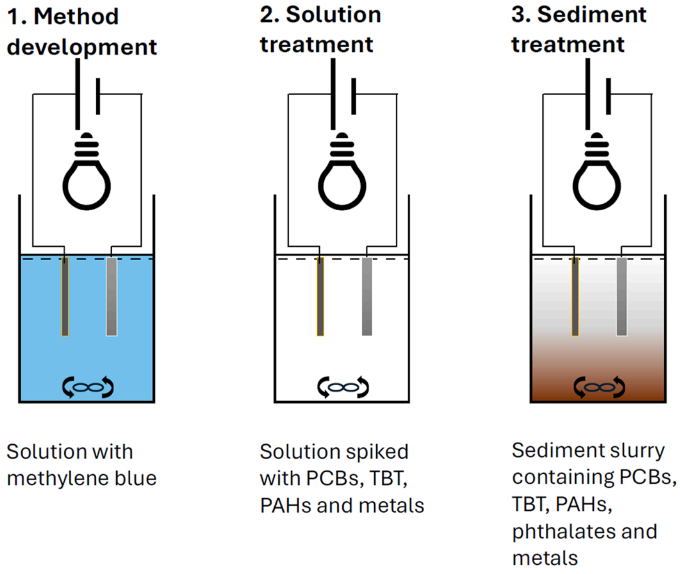


Fig. 1. Schematic illustration of the method's procedure.

and, as feedstock material, a suspension of 25 wt% solid load (TiO_2 powder) in water. A commercially available nanosized TiO_2 powder (Nanoshel-UK Ltd.) with a nominal particle size range of 10–25 nm and 99.9 % purity (Anatase phase) was used to prepare the suspension. Before coating, the Ti-6Al-4V plates were grit-blasted with Al_2O_3 powder to an average surface roughness (Ra) of 3.2 µm. The grit-blasted samples were then ultrasonically cleaned with acetone, and five preheating strokes were performed to eliminate any volatile impurities. The thermal spray parameters used to deposit the TiO_2 layers are listed in Table 1. The TiO_2 layers were sprayed to a thickness of approximately 50 µm with a total gas flow of 180 PSI; only one side of the Ti-6Al-4V substrate was coated.

2.2. Photocatalytic and photoelectrocatalytic experimental setup

The bench-scale photocatalytic experimental setup consisted of two UV lamps (UVP, AnalytikJena) placed 13–18 cm above glass beakers containing a liquid solution or sediment suspension. The irradiance levels were approximately 2.0, 1.2, and 0.9 mW/cm^2 for UVA, UVB, and UVC lamps, respectively. The solutions and suspensions in the beakers were mixed using a magnetic stirrer. A photocatalytic anode was placed in each beaker, with the photocatalyst facing up towards the light source. In photoelectrocatalysis experiments, a titanium cathode was also placed in each beaker. The anodes and cathodes were connected to a galvanostat (KP07, Bank IC), which controlled the electrical current.

2.3. Photoelectrochemical tests

To assess the oxidation efficiency of PEC and to optimize the operational parameters, an experimental setup was employed using methylene blue (MB) solutions (Sigma-Aldrich) diluted to 5 mg/L in water solutions containing 5800 mg/L NaCl. Glass beakers with a diameter of 14 cm were filled with 150 mL MB solution and placed under the UV lamps as previously described. Lamps (15 W) with UVA (365 nm), UVB (302 nm), and UVC (254 nm) were tested. Methylene blue is a commonly used model compound in photoelectrocatalytic experiments [30,31]. Approximately 4 mL of solution was extracted from the beakers at regular intervals, and the methylene blue concentration was measured by spectrophotometry.

Table 1
Spray parameters for coating deposition of the TiO_2 on the Ti-6Al-4V plates.

Parameters	
Powder injection	Axial
Current (A)	180
Power (kW)	86
Stand-off distance (mm)	70
$Ar\%/N_2\%/H_2\%$	50/40/10
Powder feed rate (g/min)	45
Total gas flow (psi/MPa)	180/1.24

2.4. Treatment of organics and metals in solutions

Three 2-L water solutions containing organic pollutants and metals commonly found in contaminated sediment were prepared. The spiked solutions contained 10 % methanol to keep the organics in solution. Organotin compounds were added as TBT 47 ng/L (containing impurities of DBT and MBT). The concentrations for each specific PAH were 1.5 µg/L, and PAH-16 = 24 µg/L which is the sum of sixteen common PAHs; naphthalene, acenaphthylene, acenaphthene, fluorene, phenanthrene, anthracene, fluoranthene, pyrene, benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthene, benzo(a)pyrene, dibenzo(ah)anthracene, benzo(ghi)perylene, and indeno(123 cd)pyrene. There are 209 congeners of PCBs, arranged according to current nomenclature and numbered from 1 to 209 in order of increasing chlorine content. In the spiked solution, the concentration of each specific PCB was 0.031 µg/L, and PCB-7 = 0.22 µg/L, which is the sum of seven common PCBs; PCB 28 (2,4,4′-trichlorobiphenyl, 2,4,4′-PCB), PCB 52 (2,2′,5,5′-tetrachlorobiphenyl). PCB 101 (2,2′,4,5,5′-pentachlorobiphenyl), PCB 118 2,2′,3,4,4′,5,5′-heptachlorobiphenyl), PCB 138 (2,2′,3,4,4′,5′-hexachlorobiphenyl), PCB 153 (2,2′,4,4′,5,5′-hexachlorobiphenyl, PCB 180 (2,2′,3,4,4′,5,5′-heptachlorobiphenyl). The concentrations of metals were 70 µg/L for each (Al, Ba, Pb, Fe, Cd, Co, Cr, Cu, Mn, Ni, Zn). One of the solutions was used as a control, kept in the dark without stirring next to the experimental setup, and the concentrations in this solution were used to calculate removal efficiencies. The other two solutions were placed in beakers for PEC treatment. A TiO_2-coated photocatalytic electrode was submerged in each solution. The solutions were mixed using magnetic stirrers and exposed to UVC. Solution S1 was only treated photocatalytically with UVC and a TiO_2-coated Ti alloy plate. Solution S2 also contained a titanium alloy cathode, and a current of 4 mA was applied between the photocatalytic anode and the cathode. The changes in concentrations of PAHs, PCBs, and metals in S1 and S2 were measured after 72 h of the experimental period and compared with the control.

The removal efficiency was calculated using the Eq. 12.

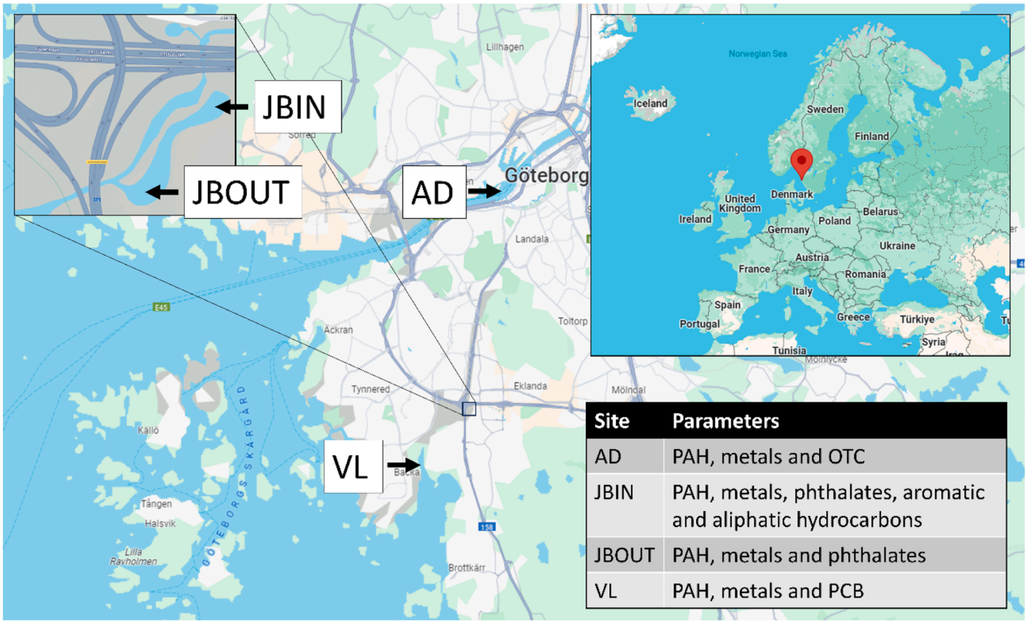
$$R = 100 \times \frac{c_0 - c_1}{c_0} \tag{12}$$

R=removal [%], c_0 = initial concentration, c_1=concentration after treatment

2.5. Sediment samples

Sediments were collected from three sites in Gothenburg, Sweden (Fig. 2). The Arendal sediments (AD) were collected during the dredging of the river Göta Älv. Samples were also collected from the Välen Bay (VL), and from the Järnbrott stormwater pond inlet (JBIN) and outlet (JBOUT), using an Ekman grab sampler. The samples were homogenized and stored in opaque plastic containers and kept at 4°C until analysis and experiments were conducted. The sediment samples were analyzed for metals and PAHs. Based on prior knowledge of contamination at the sampling sites, the AD sediment was additionally analyzed for TBT, DBT, and MBT; the VL sediment for PCBs; and the JBIN and JBOUT sediments for phthalates and aliphatic and aromatic hydrocarbons.

The initial sediment concentrations can be found in Table 2. The results have been compared to guidelines and reference values used to classify Swedish sediments. All four sediments had a high Cu content,



Site	Parameters
AD	PAH, metals and OTC
JBIN	PAH, metals, phthalates, aromatic and aliphatic hydrocarbons
JBOUT	PAH, metals and phthalates
VL	PAH, metals and PCB

Fig. 2. Site locations in Gothenburg, Sweden, and parameters analyzed in the sediment.

showing a distinct deviation from preindustrial levels [32,33]. The concentrations of PAH-M, PAH-H, and PAH-11 in the stormwater sediments JBIN and JBOUT have been classified as very high compared with other sites in Sweden [34]. The marine sediment VL showed high concentrations of PAH-M, PAH-H, and PAH-11. The AD sediment had high PAH-M and PAH-11 concentrations and PAH-H at moderate levels. The TBT concentration in AD and the PCB-7 concentration in VL sediments were also classified as very high.

2.6. Treatment of organics and metals in sediments

Different setups were used to test the efficiency of the developed method on the sediments, as described below. The tests are named according to the abbreviations and explanations in Table 3. The names include sediment, liquid-to-solid (LS) ratio, and experimental conditions. A summary of the tested setups is presented in Supplementary Material A.

After each experiment, the sediment slurry was placed in glass beakers in a fume hood. Fans were used to accelerate the water evaporation until nearly all the liquid had disappeared. This was done to show the actual oxidation and removal of the pollutants from the sediment, rather than dilution or transition to the water phase. Thereafter, samples were sent to an accredited external laboratory for analysis. Duplicate samples were not prepared, as the primary objective was to evaluate the applicability of the methods across a diverse range of sediment types, rather than to statistically validate their efficiency or determine precise post-treatment pollutant concentrations in each sample.

2.7. Analytical methods

The concentration of methylene blue was measured as absorbance at 680 nm using a spectrophotometer (UV-1800, Shimadzu). Irradiance was measured using an SDL470 light meter for UVA and UVC (Extech), and an IM-213 UV AB meter (RS Pro).

An accredited external laboratory carried out all analyses of pollutants in sediment and water. The limit of reporting for the analyzed water and sediment is presented in Supplementary Material B. Sediment samples from all sampling sites were analyzed for dry weight (DW) (SS-EN 028113), total organic carbon (TOC) (CSN ISO 10694:1995), metals, and PAHs. Metal concentrations were determined after digestion and analyzed using Inductively Coupled Plasma – Atomic Emission Spectroscopy (ICP-AES) (US EPA 200.7, CSN EN ISO 11885, US EPA 6010, SM 3120). The metals As, Ba, Cd, Co, Cr, Cu, Fe, Hg, Ni, Pb, V, Zn were digested using $HNO_{3,}$ while Ag, Mo, Sb, and Sn were digested using aqua regia (US EPA 3050, CSN EN 13657, ISO 11466). The PAH-16 concentration was analyzed based on US EPA 429, US EPA 1668, US EPA 3550 using Gas Chromatography-Mass Spectrometry (GC-MS). The analyzed PAHs included low molecular weight PAH (PAH-L) (naphthalene, acenaphtylene, acenaphtene), medium molecular weight PAH (PAH-M) (fluorene, phenanthrene, anthracene, fluoranthene and pyrene), and high molecular weight PAH (PAH-H) (benzo(a)anthracene, chrysene, benzo(b)fluoranthene, benzo(k)fluoranthrene, benzo(a)pyrene, indeno (1,2,3,*c*,d)pyrene, dibenzo(a,h)anthracene, benzo(g,*h*,i)perylene). The concentration of organotin compounds (TBT, DBT and MBT) was analyzed in AD samples (SS-EN ISO 23161:2011). The PCB concentration was analyzed in JBIN, JBOUT, and VL samples (DIN ISO 10382). Additionally, JBIN and JBOUT were analyzed for the concentration of phthalates (DIN 19742: 2014–08) and aliphatic and aromatic hydrocarbons (using GC-MS according to SPIMFAB quality manual).

The water samples were analyzed for concentrations of metals (SS-EN ISO 11885:2009 and US EPA Method 200.7:1994), organotin compounds (ISO 17353:2005), PCBs and PAHs (method based on US EPA 8270D, US EPA 8082 A, CSN EN ISO 6468 and US EPA 8000D).

2.8. Cost estimation

A simplified cost estimation was conducted based on local conditions in Gothenburg, Sweden. The estimation includes costs for materials (titanium plates, UV lamps, water, electricity, H_2O_2. The prices for materials and electricity, with references, are presented in Supplementary Material C. For the titanium plates, only the cost of raw materials was considered, as the plates used in this study were custom-made at University West specifically for this work. For water, municipal tap water costs were used, assuming ultrapure laboratory-grade water would not be used in a larger-scale project. Electricity costs were based on the average annual energy price in the Gothenburg area for 2024. The cost of H_2O_2 was obtained from online suppliers of laboratory-grade chemicals. In a full-scale dredging operation, both the price and the required purity of H_2O_2 may differ significantly due to bulk purchasing and varying quality requirements. The cost for UVC lamps refers to the

Table 2
Average initial concentrations of metals, PAHs, PCBs, phthalates, organotin compounds, aliphatic and aromatic hydrocarbon in the original samples and identified as concentrations higher than the level of quantification in at least one of the samples from Arendal (AD), Välen (VL), Järnbrott inlet (JBIN), and Järnbrott outlet (JBOUT).

Sediment	Unit	AD	VL	JBIN	JBOUT
Dry weight (DW)	%	53	35	28	25
Loss on ignition (LOI)	% DW	6.6	8.8	17	17
Cd	mg/kg DW	0.28	0.58	0.87	1.6
Cu	mg/kg DW	32	120	220	320
Ni	mg/kg DW	14	20	29	39
Pb	mg/kg DW	24	66	60	82
Zn	mg/kg DW	130	290	720	990
PAH−16	mg/kg DW	1.3	1.9	6.6	9.0
PAH-L	mg/kg DW	0.063	0.14	0.18	0.32
PAH-M	mg/kg DW	0.58	0.73	2.8	3.5
PAH-H	mg/kg DW	0.69	1.0	3.6	5.2
PCB−7	mg/kg DW	n.a.[a]	0.34	n.a.	n.a.
Di(2-etylhexyl) phthalate (DEHP)	mg/kg DW	n.a.	n.a.	15	24
Diisodecyl phthalate (DIDP)	mg/kg DW	n.a.	n.a.	< 100	< 50
Diisononyl phthalate (DINP)	mg/kg DW	n.a.	n.a.	160	85
Aliphatics C_5-C_8	mg/kg DW	n.a.	n.a.	< 10	n.a.
Aliphatics C_8-C_{10}	mg/kg DW	n.a.	n.a.	13	n.a.
Aliphatics C_{10}-C_{12}	mg/kg DW	n.a.	n.a.	103	n.a.
Aliphatics C_{12}-C_{16}	mg/kg DW	n.a.	n.a.	107	n.a.
Aliphatics C_{16}-C_{35}	mg/kg DW	n.a.	n.a.	972	n.a.
Aromatics C_8-C_{10}	mg/kg DW	n.a.	n.a.	1.2	n.a.
Aromatics C_{10}-C_{16}	mg/kg DW	n.a.	n.a.	< 1.0	n.a.
Methylpyrene/ methylfluoranthene	mg/kg DW	n.a.	n.a.	1.25	n.a.
Methylchrysene/ methylbenso(a) anthracene	mg/kg DW	n.a.	n.a.	1.25	n.a.
Aromatics C_{16}-C_{35}	mg/kg DW	n.a.	n.a.	2.6	n.a.
Sum TEX[b]	mg/kg DW	0.44	n.a.	n.a.	n.a.
Monobutyltin (MBT)	µg/kg DW	14.8	n.a.	n.a.	n.a.
Dibutyltin (DBT)	µg/kg DW	32.5	n.a.	n.a.	n.a.
Tributyltin (TBT)	µg/kg DW	104	n.a.	n.a.	n.a.

[a] n.a. = not analyzed [b] Sum TEX = Sum of toluene, ethylbenzene and xylenes (incl. ortho-, meta-, and para-xylene)

price paid for the UV-lamps used in this study. The cost was calculated for treating one tonne (1000 kg) of wet dredged sediment. Costs related to potential sediment dewatering, as well as the construction and operation of a treatment facility, were not included in this estimate.

Table 3
Overview of sample names and experimental setup.

Sediment from **Arendal (AD)**, **Välen (VL)**, **Järnbrott inlet (JBIN)** or **outlet (JBOUT)**	The sediments JBIN, JBOUT, and VL were diluted with a saline (5.8 g/L NaCl) water solution. In experiments with AD marine sediments, only ultrapure water was used as the sediments naturally contained salts. The suspension was stirred in all experiments.
Liquid-to-solid ratio (LS) 10, 40, 80, or 133	Most experiments were carried out at an LS of 40. For comparison, additional experiments were done with LS of 10, 80, or 133.
Photolysis (P)	Experiments without the presence of photocatalytic TiO_2 were performed with **UVB** or **UVC** using two setups. Setup 1: UVB fluorescent tubes (wavelength 302 nm) or UVC fluorescent tubes (wavelength 254 nm) were placed above beakers with water-sediment-slurry. The experimental setup was identical to the setup described for water samples. Setup 2: The water-sediment-slurry was recirculated through a chamber with an 18 W UVC lamp (wavelength 254 nm) using a peristaltic pump (**Chamber**).
Photocatalysis (PC)	In all PC experiments, TiO_2-coated titanium plates measuring 3×5 cm^2 were used.
Photoelectrocatalysis (PEC)	In PEC experiments, the TiO_2-coated titanium plate (3×5 cm^2) served as an anode, and a Ti alloy substrate (3×5 cm^2 plate) served as a cathode in an electrochemical system with an applied current of 4 mA.
Hydrogen peroxide (H_2O_2)	In some experiments, hydrogen peroxide (H_2O_2) was added to the beakers. To optimize the reaction, the pH of the water-sediment-slurry was lowered to pH 3 by adding H_2SO_4 [28, 35]. No iron was added as in a typical Fenton's reaction, as a previous study on the AD sediment had shown that it already contained iron at sufficient levels to form radicals [13]. To optimize the pollutant oxidation, H_2O_2 (100 mL) was added in batches at 0, 3, 9, 24, 32, and 48 h.
Experiment duration 3, 7, 10, or 12 days	Most experimental runs lasted 3 days, and a few lasted 7, 10 or 12 days.

3. Results and discussion

3.1. Optimization of photoelectrocatalysis with methylene blue

Illuminating a methylene blue (MB) solution with UVB in the absence of a photocatalyst resulted in minimal oxidation of MB. However, with a TiO_2-coated electrode immersed in the solution, the MB oxidation rate was 0.097 $g{\times}m^{-2}{\times}h^{-1}$. The oxidation rate increased to 0.11 $g{\times}m^{-2}{\times}h^{-1}$ when a current of 1 mA (0.67 A m^{-2}) was applied between the TiO_2-coated electrode and a titanium plate counter electrode. At a current of 4 mA (2.67 A m^{-2}), the MB concentration rapidly dropped to zero and the MB oxidation rate could not be accurately quantified but was at least 0.33 $g{\times}m^{-2}{\times}h^{-1}$ (Fig. 3 A). An electrical current is known to increase the efficiency of photocatalysis by minimizing the recombination of electrons and holes generated when the photocatalyst is illuminated [36]. The photoelectrocatalysis rate with a current of 4 mA was tested with three UV lamps. The MB oxidation rate increased from 0.64 $g{\times}m^{-2}{\times}h^{-1}$ for UVA to 0.93 $g{\times}m^{-2}{\times}h^{-1}$ for UVB and 1.09 $g{\times}m^{-2}{\times}h^{-1}$ for UVC (Fig. 3B). A comparison of TiO_2 and ZnO photocatalysts was also carried out, in which TiO_2 gave a 3-fold higher

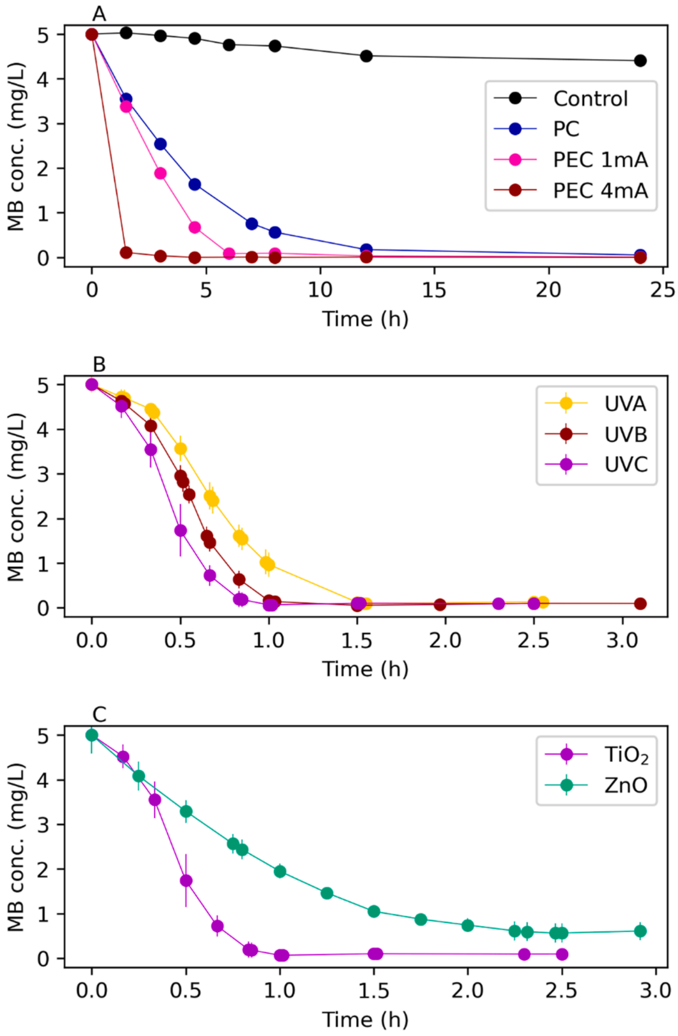


Fig. 3. (A) Effect of photoelectrocatalysis (PEC) on methylene blue (MB) oxidation during exposure to UVB illumination. The control did not contain a photocatalytic electrode; PC is TiO_2 photocatalysis without an applied electrical current, PEC 1 mA and 4 mA are TiO_2 photoelectrocatalysis with 1 mA and 4 mA applied current, respectively. (B) Effect of light wavelength on TiO_2 photoelectrochemical (4 mA) oxidation of MB. (C) Effect of photocatalyst on photoelectrochemical (4 mA) oxidation of MB with UVC illumination. The error bars in panels B and C are standard deviations based on 4–5 replicate tests. The tests in panel A were carried out once.

oxidation rate (Fig. 3 C). TiO2has previously been shown to achieve higher photocatalytic oxidation rates than ZnO, for example, in the degradation of formaldehyde. This could be explained by a higher degree of recombination of photoinduced electron-hole pairs in ZnO [37].

3.2. Treatment of organic compounds and metals in water solutions

Photocatalysis (PC) and photoelectrocatalysis (PEC) were found to efficiently oxidize organic pollutants in spiked water solutions (Fig. 4). Both methods removed > 98 % of PCB-7 and > 99 % of PAH-16 after 3 days of treatment, showing that PC and especially PEC efficiently degrade highly persistent organic pollutants. The combined concentration of organotin compounds (TBT, DBT, and MBT) was efficiently reduced after three days of PEC and PC treatments, with a total decrease of 88 % and 84 %, respectively. The concentrations of TBT decreased by > 99 % (Fig. 5), which is similar to the results from a previous study by Brosillon, Bancon-Montigny [38]. During the treatment, TBT was degraded into DBT and subsequently into MBT and finally Sn through debutylization (Eq. 13) [13]. This explains the increase in MBT concentrations in the spiked water solution after treatment. The PEC treatment was somewhat more effective than PC, resulting in lower concentrations of both DBT and MBT in the water solution after treatment. The applied electric current produced hydroxyl radicals at the anode and reduced the recombination of electron-hole pairs, thereby enhancing the efficiency of the photocatalytic reactions as well.

$$C_{12}H_{28}Sn + 80\bullet OH + 4H^{+} \rightarrow 12CO_2 + Sn^{4+} + 56H_2O \tag{13}$$

The concentrations of all metals except Zn were reduced by PEC treatment of the spiked solutions (Fig. 5). The removal efficiency of PEC was 73 % for Cu, 41 % for Pb, 30 % for Cd, 28 % for Ni, 22 % for Co, and 10 % for Cr. The higher removal efficiency of Cu is assumed to be due to its higher reduction potential compared to other metals [39]. Without an applied current (i.e. photocatalysis), metal concentrations were similar to or somewhat higher than those in the control sample. As metal ion removal from the spiked solution is expected to occur via electrochemical reduction at the cathode, it is logical that PEC showed metal removal, but PC did not. The results show that PEC can effectively treat water contaminated with both organic pollutants and metals while simultaneously recovering metals. The removal efficiencies found in this study may be further optimized by adjusting the pH or by combining PC with sorption to a solid substrate [40,41].

3.3. Treatment of organic compounds and metals in sediments

3.3.1. Polycyclic aromatic hydrocarbons

Large variations in PAH oxidation were observed across the different sediments and treatments (Fig. 6). In samples from the marine sediments AD and VL (LS40_PEC_UVC_3d, Fig. 6C, and LS40_3d, Fig. 6B), the PAH

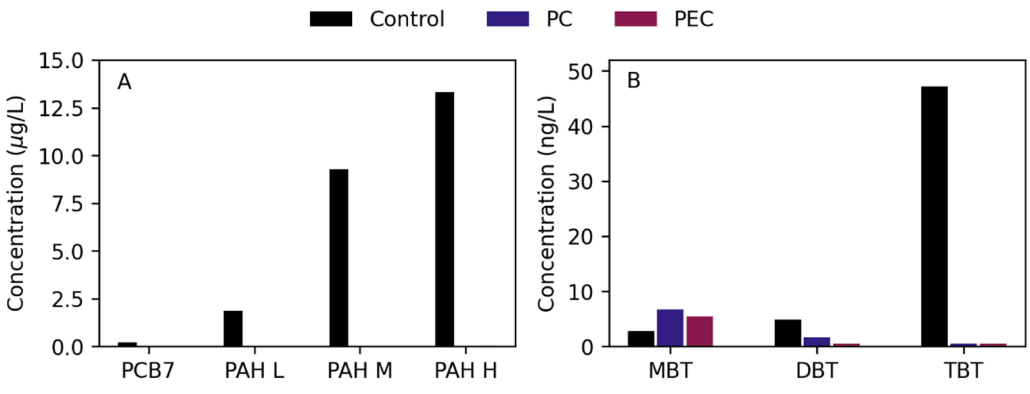


Fig. 4. Concentrations of PCBs, PAHs, (A) and organotin compounds (B) in spiked water before and after 3 days of treatment with photocatalysis (PC) and photoelectrocatalysis (PEC), based on a single measurement.

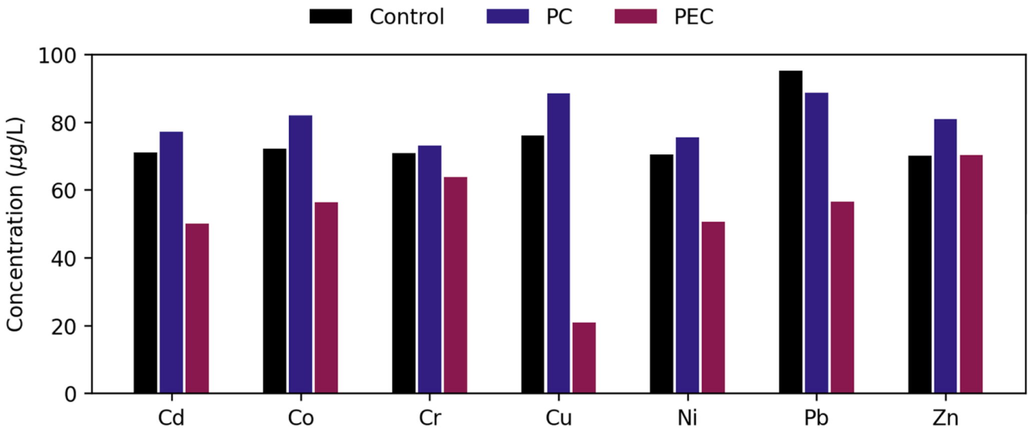


Fig. 5. Concentrations of metals in spiked water samples before and after 3 days of treatment with photocatalysis (PC) and photoelectrocatalysis (PEC), based on a single measurement.

concentrations increased after treatment, by 25 % and 330 % respectively, indicating more heterogeneous PAH concentrations than in the stormwater sediments JBIN and JBOUT. Such an increase in concentration is sometimes observed for various parameters in sediment and soil samples after treatment. It could be due to heterogeneity, but also because the treatment enhances the availability of the compounds during laboratory analysis [12]. A possible explanation is that the treatment caused a release of PAHs strongly bound in oils and tire and road wear particles, which, after this treatment, were more easily extracted from the sediments during the chemical analysis procedure [42]. In Lu, Su [43], it was seen that UV light degrades tire particles and increased the PAH concentration 1.2 times in the produced leachate, compared to leachate not exposed to UV light. Sediment characteristics and the binding of PAHs in matrices have been shown to affect their availability and treatment performance [12,44]. Differences in sediment characteristics, such as organic content (Table 2), may also affect the effectiveness of PAH oxidation, as the stormwater sediments' organic content was almost twice as high as that of the marine sediments, indicating that the PAHs were more strongly bound in the asphalt material and tire wear particles in the stormwater sediments. The hydroxyl radicals formed during the treatments may primarily target other, more reactive substances in organically rich material before degrading more persistent compounds such as PAHs [44]. Tailored catalysts could be used to selectively degrade specific organic pollutants by controlling the formation of reactive oxygen species [45]. The relative composition of the specific PAHs in the stormwater sediment samples after the different treatments seemed to be reasonably stable, indicating that the PAHs may have been trapped in matrices, e.g., tire rubber particles [46], and not easily accessible to UV light, PC, or PEC. In the VL and AD samples, the PAH composition was more varied after treatment, but no clear trend could be identified (Fig. 6).

The most effective PAH removal was 78 % in JBIN sediments using H_2O_2 as the only oxidation method. This result is similar to results from previous studies where PAHs were removed from soil and water using Fenton and photo-Fenton, with a reduction rate of 76–99 % for PC, a reduction rate of 70–100 % in soil and water, and a reduction rate of 82–93 % for electrolysis [47,48]. In general, methods that included H_2O_2 achieved higher removal efficiencies than treatments without H_2O_2 for JBIN, VL and AD. For VL, the highest PAH reduction of 37 % was achieved by using H_2O_2 alone, and for AD, the highest PAH removal of 42 % was achieved with H_2O_2 combined with PC. The use of electricity had a low impact on degradation, but various parameters could be optimized to achieve higher oxidation, such as current intensity, plate size and distance, exposure time, and the effect of the UVC lamp [49].

Tests with a liquid-to-solid (LS) ratio of 40 showed more effective oxidation than those with an LS ratio of 10. These results align with the understanding that a higher dilution of suspended particles that absorb UV photons results in more efficient photooxidation. These results have been confirmed by other studies using TiO_2 photocatalysis along with advanced oxidation techniques to degrade PAHs in water and soil [15]. However, it appears that more strongly bound PAHs are transformed into a form more accessible for chemical analysis if the solution is highly diluted (e.g., LS 133); the treatment had a limited effect on removal efficiency. The results demonstrate the complexity of degrading strongly bound PAHS in sediments containing oils, asphalt, and tire wear particles. On the one hand, more strongly bound PAHS may be more readily treated, but on the other hand, they may occur in forms more accessible to living organisms in sediments.

3.3.2. Organotin compounds

The organotin compound TBT degrades into DBT, then MBT, and finally inorganic Sn through debutylization. The highest efficiency in treating organotin-polluted sediments was achieved with H2O2 and PEC treatment. Here, high oxidation rates of 91 %, 72 %, and 87 % were achieved for TBT, DBT, and MBT, respectively (Fig. 7). These results surpassed previous treatments of the same sediment using Fenton's reagent or only H_2O_2, which achieved a maximum reduction in TBT concentration of 64 % [13]. As a result, the classification of the sediments improved from "very high concentration" to "moderate concentration" [34]. Using H_2O_2 with PC, the degradation was lower, at 83 % for TBT, 58 % for DBT, and 45 % for MBT, resulting in the sediments having a "high" TBT concentration. The oxidation of organotin compounds through a combination of H_2O_2 and PC was also observed by Rodríguez-Chueca, Ferreira [29]. In contrast, no degradation was observed in the experiments without H_2O_2, indicating that H_2O_2 is needed for effective TBT oxidation. This may be explained by the ability of H_2O_2 to degrade paint flakes containing TBT, thereby releasing more TBT than treatments without H_2O_2 [12]. Photoelectrocatalysis can further enhance the oxidation, as PEC and H_2O_2 work synergistically by trapping the conduction band e^-, which forms more OH radicals [35]. Previous experiments have suggested that current intensity affects oxidation, and that a stronger electrical current could therefore further enhance it [13,44].

3.3.3. Polychlorinated biphenyls

As for organotin compounds, the method that performed best for PCB oxidation was PEC, enhanced by the addition of H_2O_2, for which a reduction of 82 % was observed for VL sediments (Fig. 8A, LS_40_PEC_UVC_H2O2_3d), and 49 % for JBIN sediments (Fig. 8B, LS40_PEC_UVC_H2O2_3d). These results are in line with findings from the literature, where PCBs were degraded at rates from 13 % to up to 100 % after treatment with PC, PEC, and H_2O_2 of polluted soil [50,51]. The

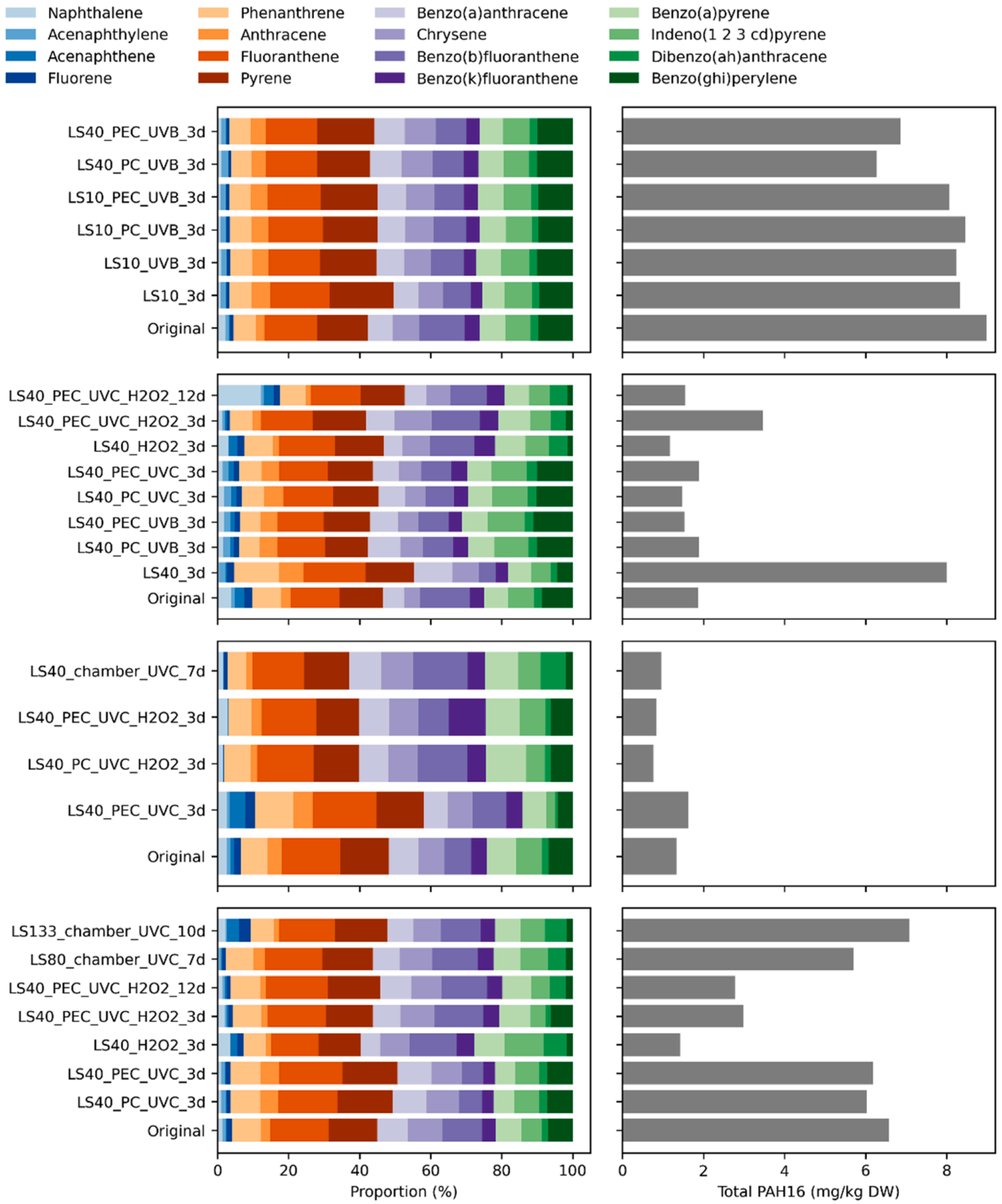


Fig. 6. Proportions and total concentrations of PAH-16 for sediments from JBOUT (A), VL (B), AD (C), and JBIN (D) after treatment with photolysis (chamber), photocatalysis (PC) and photoelectrocatalysis (PEC) with and without addition of H_2O_2 at different liquid-to-solid ratios and treatment times, based on a single measurement.

reduction of the PCBs was lower without the addition of H_2O_2 (Fig. 8), which may imply that other organic compounds took precedence in the oxidation in the sediments, similar to the discussion above for the oxidation of PAHs. The PCB removal may be enhanced if the electrolysis treatment is coupled with surfactants such as saponin and citric acid to increase PCB solubility and desorption from sediment [52]. In the VL

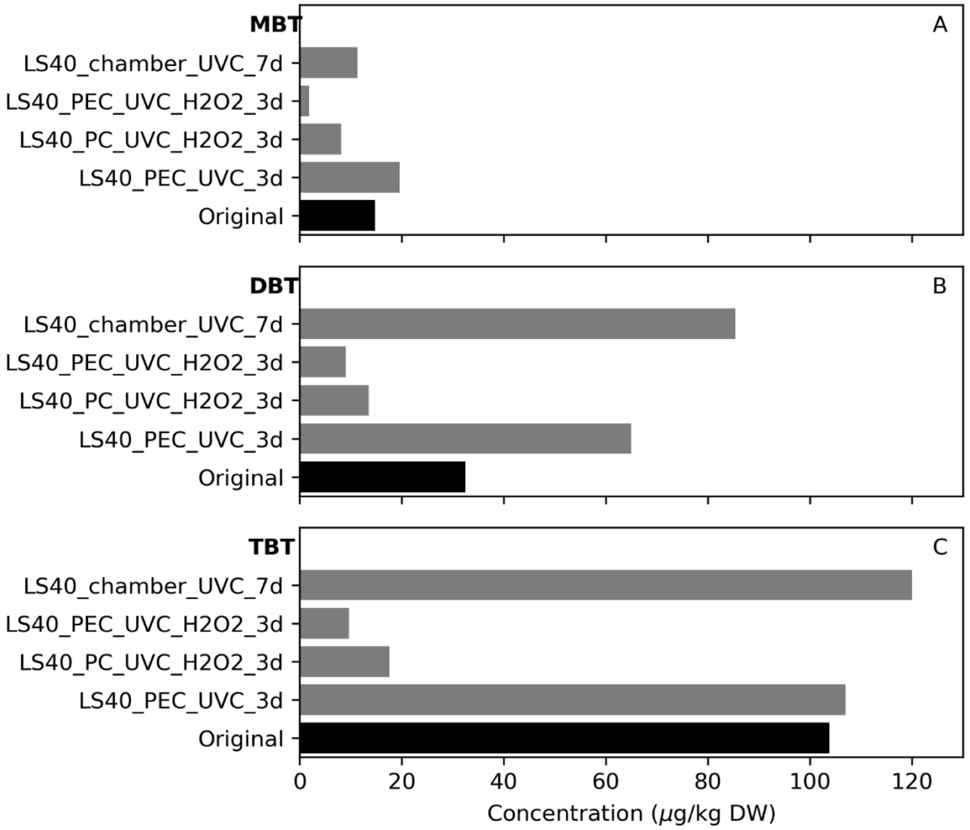


Fig. 7. Concentrations of tributyltin (TBT) (A), dibutyltin (DBT) (B), and monobutyltin (MBT) (C) in AD sediment after treatment with photolysis (chamber), photocatalysis (PC), or photoelectrocatalysis (PEC), with and without addition of H_2O_2 and at different treatment times, based on a single measurement.

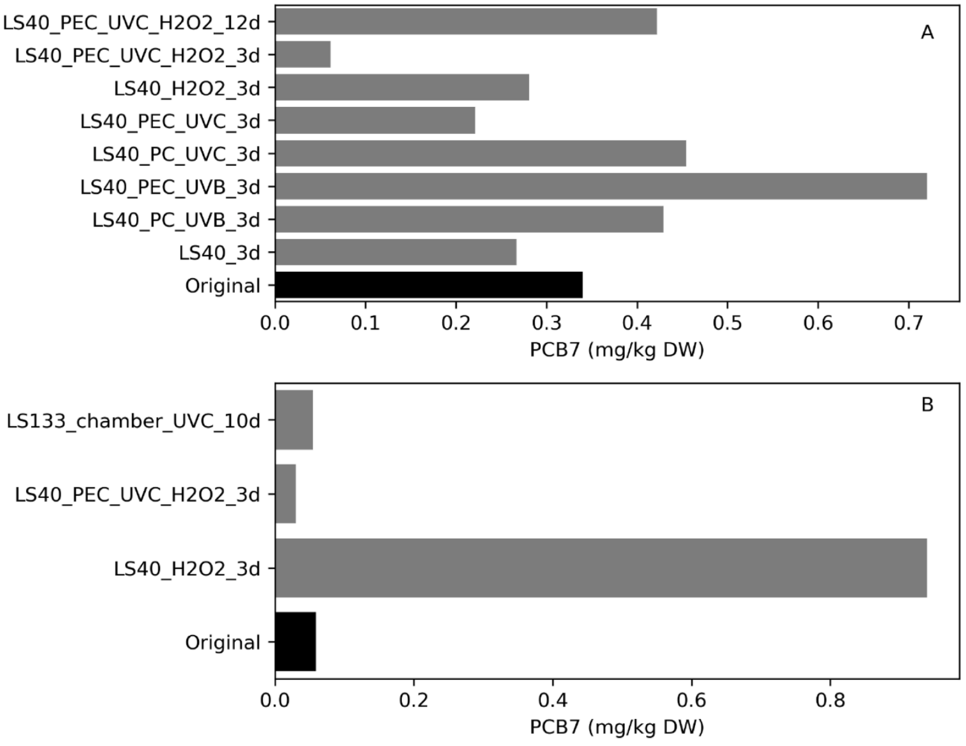


Fig. 8. Concentrations of PCB-7 in VL (A) and JBIN (B) sediment before and after treatment with UVB or UVC, photolysis (chamber), photocatalysis (PC), or photoelectrocatalysis (PEC), with and without addition of H_2O_2 at different liquid-to-solid ratios and treatment times, based on a single measurement.

sediments, PCB concentrations varied significantly, suggesting that the sediment had a highly heterogeneous distribution of PCBs (Fig. 8). The VL samples also had higher uncertainty in the chemical analysis protocols (approximately 30 % compared to 10 % at other sites), which may have resulted in more uncertain analysis results. PCBs are degraded through oxidation by hydroxyl radicals in complex reactions via the addition of the hydroxyl group to one of the halogenated sites of the molecule [53]. The higher congeners degrade more slowly due to unreactive halogenated sites, and within a homologue group, chlorine on the meta and para positions results in a lower reaction rate, depending on steric hindrance from hydroxyl radical attack. Fig. 8 illustrates that UVC is more efficient than UVB for the oxidation of PCB molecules.

3.3.4. *Phthalates*

The reduction in phthalate concentration in sediment after treatment varied between JBIN (Fig. 9) and JBOUT (Fig. 10). Phthalates were found to be more easily oxidized than PAHs in corresponding sediments. This may be explained by the higher water solubility of phthalates compared to PAHs, which makes them more readily susceptible to oxidation during the treatment experiments.

For JBIN samples, degradation was found to be in the order DINP>DIDP>DEHP (Fig. 9), regardless of the method used. 97 % of the DINP and 85 % of DEHP were removed from JBIN (Fig. 9) using H_2O_2, which also led to high oxidation of PAHs in JBIN, AD, and VL, and of organotin compounds in AD samples. Other studies have shown that photolysis enhanced by the addition of H_2O_2 yielded higher phthalate removal than PC [54,55]. Even without H2O2, PEC achieved high phthalate removal (DINP 83 %, DIDP 74 %, and DEHP 50 %) and PC (DINP 76 %, DIDP 58 %, and DEHP 34 %).

Photocatalytic electrolysis yielded the highest removal of DINP (44 %), DIDP (24 %), and DEHP (29 %) for JBOUT (Fig. 10, LS_PEC_UVB_3d). The results were similar, but slightly lower when electrolysis was excluded from the treatment, indicating that adding electrolysis was less critical for the phthalate removal from JBOUT sediment. The results also showed a higher degradation for phthalates with a higher LS ratio, which can be explained by decreased turbidity [55].

3.3.5. *Aromatic and aliphatic hydrocarbons*

Fig. 11 shows the concentrations of aromatics in sediment from JBIN after treatment with different PC and PEC methods, with and without the addition of H_2O_2. It can be observed that all aromatics were effectively degraded by photooxidation alone in the circulating system, as well as in the system with PEC and added H_2O_2. The highest reduction was observed for aromatic hydrocarbons C10–16, with a removal efficiency of 54 %, which is less than the average reported removal for photolysis in water [56]. In Fig. 11, corresponding results for aliphatics are presented, clearly indicating that aliphatics with higher molecular weight, i.e. less water-soluble, are more challenging to degrade. The aliphatics may be more difficult to treat, as they are more prone to bind to particles and fatty acids in the sediment [57].

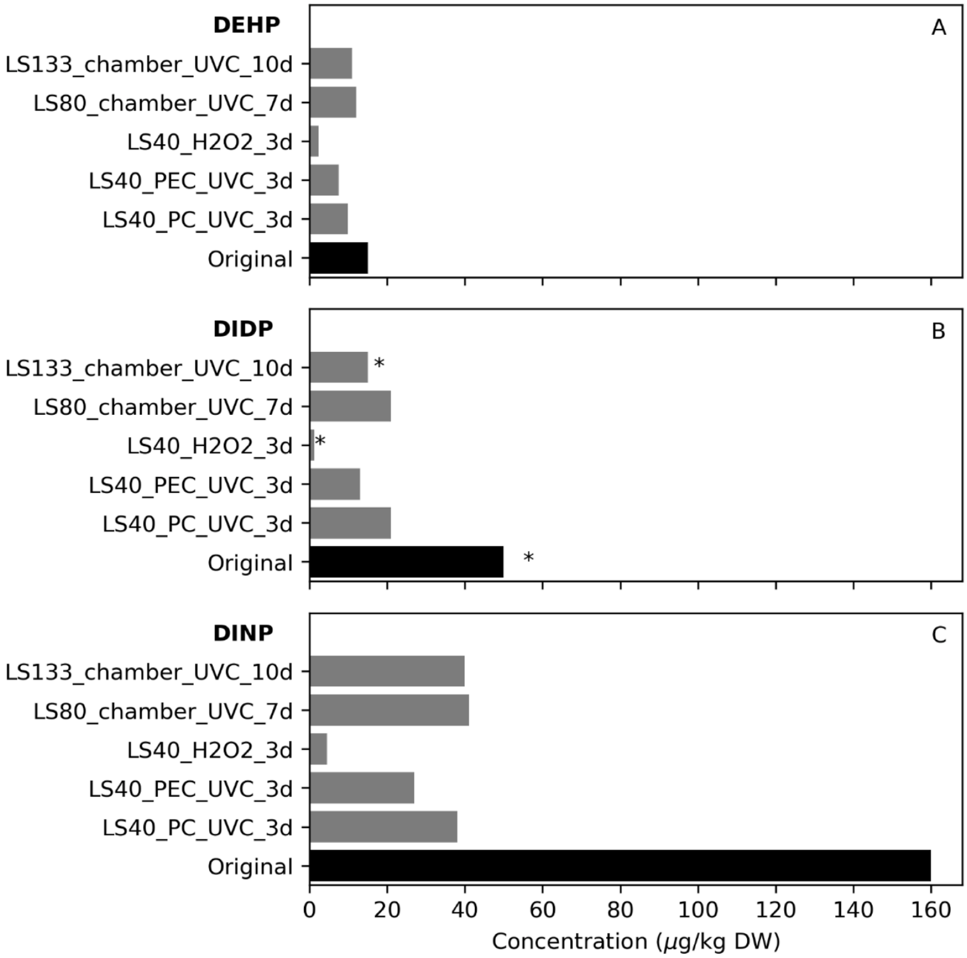


Fig. 9. Concentrations of the phthalates DEHP (A), DIDP (B), and DINP (C) in JBIN treated with UVC photolysis (chamber), photocatalysis (PC), or photoelectrocatalysis (PEC), with and without addition of H_2O_2 at different liquid-to-solid ratios and treatment times, based on a single measurement. *Indicates that half of the level of quantification has been used.

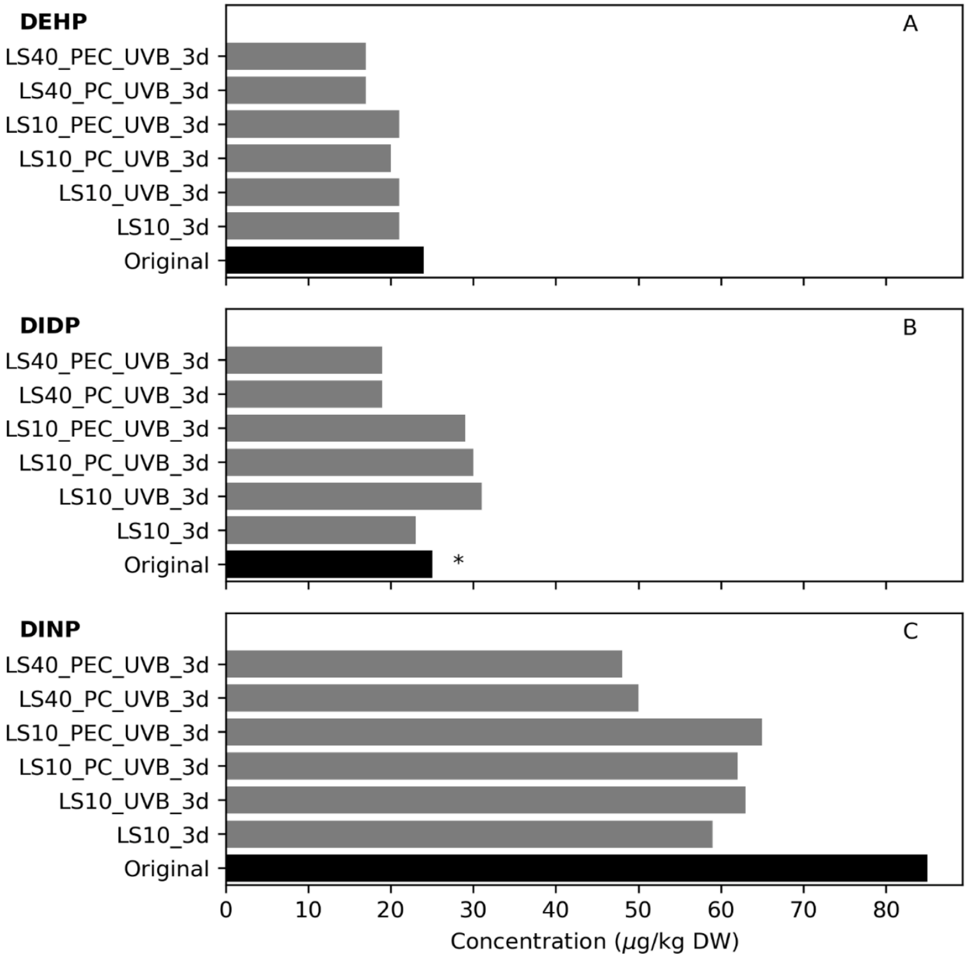


Fig. 10. Concentrations of the phthalates DEHP (A), DIDP (B), and DINP (C) in JBOUT treated with UV, photocatalysis (PC), or photoelectrocatalysis (PEC), with and without addition of H_2O_2 at different liquid-to-solid ratios and treatment times, based on a single measurement. *Indicates that half of the report level has been used.

3.3.6. *Metals*

Metal concentrations were measured to assess the feasibility of extracting metals from the contaminated sediments. However, the extraction was low for all metals at LS10 regardless of the treatment used (i.e., Cu, Zn, Pb, Cd, Ni <15 %) (see JBOUT samples in Figs. D1–5 in Supplementary Material D). This is in line with another study treating sediment with electrolysis [52]. At LS40, the amounts of removed metals were higher, ranging from 10 % to 28 %. There was no clear difference between samples with or without current. The most effective method for metal removal was stirring with ultrapure water at LS40 (i.e., Cu, Zn, Pb, Cd, Ni at 55–65 % for VL sediment (see VL samples in Figs. D1–5 in Supplementary Material D)). This highlights the importance of the LS ratio for metal removal. The results indicate that the sediment sorbed the metals and that their release is linked to the LS ratio and mechanical removal, rather than the electrolysis. Additionally, hydroxides formed during electrolysis, along with those already present in the alkaline water-sediment slurry, may precipitate with the metals, preventing their sorption onto the cathode [58–60]. Another possible reason for the low recovery of metals is that the treatment techniques used may increase the presence of organic colloids, as the radicals generated degrade organic substances [61]. These organics could form complexes with metal ions, thereby hindering metal recovery [62].

Based on the results for the PC treatment of water, little to no metal extraction was expected by using this method. The low reduction indicates a heterogeneous metal distribution within the sediment. As metals were recovered from the clear water solutions by PEC, this suggests that the metals remained bound to the sediment and were governed by different water-sediment interactions, e.g., pH, redox conditions, sorption/desorption, dissolution/precipitation, as well as sediment type and organic content [63]. More investigations to better understand the mechanisms of metal binding to different fractions within a sediment (e.g., sulfites, carbonates) and alterations to the electrolysis procedures should be carried out to improve removal efficiency. For instance, sequential extraction schemes could provide valuable information regarding metal mobility, e.g., [64] or modifications thereof. To increase the metal recovery, the pH of the water-sediment-slurry could be reduced [65], solubilizers used (e.g., chelating agents or surfactants [12,66]), or electrodes placed closer together [49,58].

The results show that electrolytic techniques cannot recover a large quantity of metals from sediment in a one-step treatment. A water sample was taken after the experiment JBIN LS40_PEC_UVC_H2O2_12 but before the water-sediment-slurry was dried, and had a Cu concentration of ~1 mg/L. This corresponds to a sediment concentration reduction of 40 mg/kg DW. As no removal was observed for the corresponding sediment sample, it can be assumed that the metal was attached to colloids suspended in the water and reintroduced to the sediment sample during the drying process. Other techniques could potentially be used to first remove metals from the sediment, after which electrolysis could be used on any extraction liquid to recover the metals.

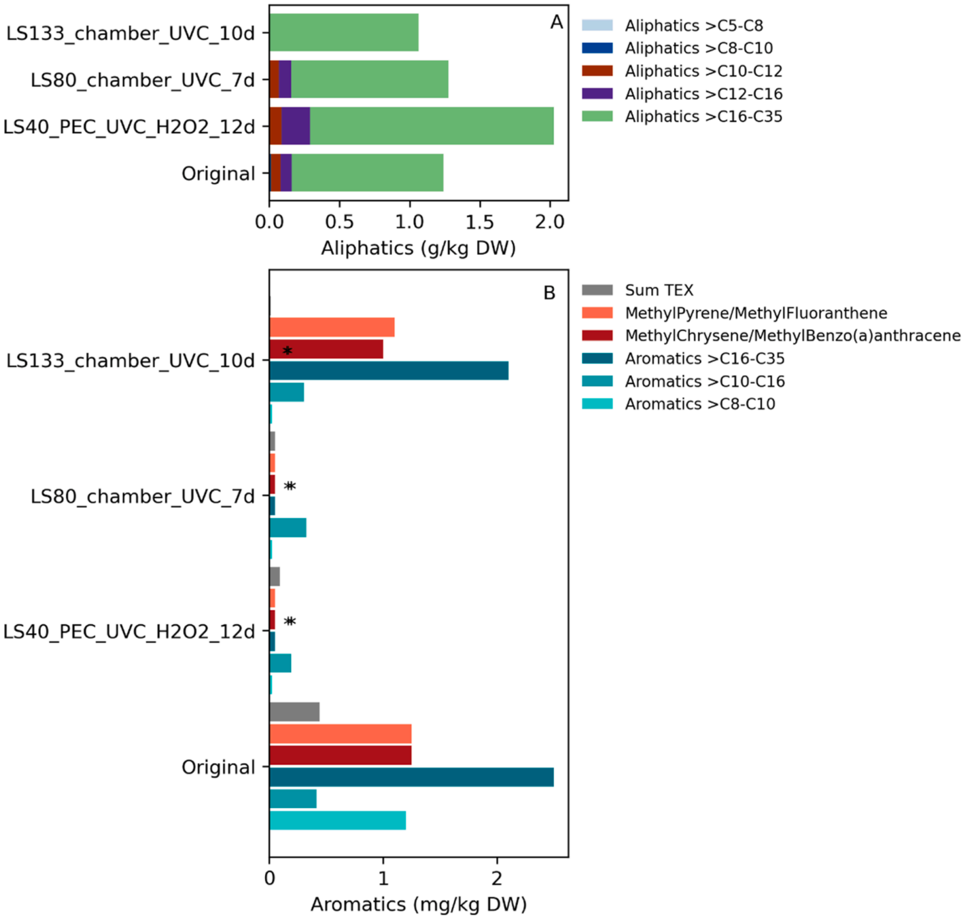


Fig. 11. Concentrations of aliphatic (A) and aromatic (B) hydrocarbons in JBIN treated with UVC photolysis (chamber) or photoelectrocatalysis (PEC), with and without addition of H_2O_2 at different liquid-to-solid ratios and treatment times, based on a single measurement. *Indicates that half of the report level has been used.

3.4. Cost estimation

The cost of treating sediment using PEC and PC was estimated to be $1466 USD/tonne of treated wet sediment. The low cost difference is due to the low difference in energy used by electrolysis. The PEC method used 4 % more energy than the PC method (0.004 kWh/kg), due to the low energy use by the electrolysis. For water treatment, PEC was estimated to cost 527 USD/L and PC 526 USD/L. For the water treatment the energy consumption while using PEC and PC was negligible (1.09 and 1.08 kWh/L, respectively). The energy use is highly dependent on the treatment time but also the amount of liquid and sediment to be treated. In this estimate, the cost was for materials (titanium powder and plates, lamps), which were for small scale experiments. In a large-scale project, these costs are likely to be lower since large quantities are purchased. Taxes and quality of the products may be different, and after modifications and optimization for scaling up the treatment, the cost may differ greatly and would likely be lower than in this estimate.

3.5. Future research for PEC treatment of sediment

In future research, the mechanisms of generation and quenching of reactive oxygen species should be analyzed to better understand the degradation processes and outcomes, along with other reactive species such as reactive sulfur species and active chlorine [67]. This includes investigating the presence of reactive intermediates that may influence the degradation of pollutants. Specific degradation pathways could then be examined. Since the results from this study indicate that sediment characteristics significantly affect pollutant removal efficiency, it would be useful to explore how removal mechanisms are influenced by factors such as sediment pH, organic content, and chemical composition. Future studies are encouraged to replicate these experiments to ensure statistical robustness and reproducibility. Using superhydrophilic TiO_2 could benefit the management of high organic loads in the sediments, improve photocatalytic performance, and reduce fouling on electrodes, which would be advantageous for long-term applications such as a remediation project [68].

3.6. Strategic outlook for PEC-based sediment remediation

Based on the results of this study, the following is a proposed strategy for future research. It is not a method validated within the current work. Therefore, the development of a PEC-based method in two stages is suggested: (1) the extraction of metals and organic pollutants into the liquid phase and (2) subsequent use of PEC on the liquid phase to degrade organic contaminants and simultaneously extract metals.

In step 1, the sediments should be washed to leach out metals and OPs. Since the studied marine and stormwater sediments are already fine-particle materials, no separation of coarser material is needed before purification with the soil washing method. In soil washing, fine soil particles, which often have the most pollutants sorbed onto them, are separated from bulk soil in a water-based system and extracted/leached based on particle size. Washing with ultra-pure water is reasonably effective for metals as previously shown [12]. However, the effectiveness of the washwater can be enhanced by using leaching agents, surfactants, pH adjustment, or chelating agent to help remove organic pollutants and metals [12,16,69]. Depending on the pollutant composition in the sediments, different pH levels can be used to extract the pollutants. For most metals, a low pH favors the release of metals from the sediment [70]. Tributyltin exhibits the strongest sorption to sediment at around pH 8; therefore, the pH of the sediment could be either increased or decreased to enhance TBT release [12]. Lowering the pH could potentially be beneficial in promoting the release of TBT and, in particular, metals from the sediment. However, the pH may need to be readjusted after treatment for further management. The sorption and release of other OPs (PAHs, PCBs, phthalates, aliphatic and aromatic hydrocarbons), are generally not significantly affected by changes in pH. Instead, different solvents could be tested to promote release [71,72]. If possible, strong chemicals should be avoided so that the treated sediment is safe to handle, either for disposal or, preferably, reuse [12]. For metals, chelators such as EDTA could be used [12]. A promising technique for extracting hydrophobic organic pollutants is supercritical fluid extraction (SFE), which utilize CO_2 [73]. In the method, CO_2 is pressurized and heated until it reaches a supercritical state behaving either as a gas or a liquid. Vegetable oil is another option that has been utilized to extract OPs from soil, with the benefit that residual oil after treatment acts as a nutrient for plant growth [73,74]. In addition, because a large proportion of organic pollutants are assumed to be tightly bound within complex matrices in the sediments themselves, as well as in color flakes, rubber tire particles, asphalt, and thick oils, density separation using salt solutions can be employed to isolate microplastics, color flakes, and rubber particles, which are prevalent in both marine and stormwater sediments [43,75–77].

In step 2, PEC can be applied to degrade OPs and extract metals from the liquid from step 1. Photoelectrocatalytic oxidation of various organic pollutants in aqueous solutions is feasible; however, its efficiency is significantly influenced by the presence of suspended particles. These particles likely scatter UV light, reducing its penetration and thereby limiting the oxidation of pollutants [78]. By altering the pH or the ionic strength, unwanted interactions of competing ions (e.g., Cl^-, SO_4^{2-}) can interfere with the PEC reactions [79]. The method also needs to be tested at a larger scale in a pilot study to determine its feasibility in sediment remediation projects.

et al. [10] investigated the sustainability of various sediment treatments and pollution levels, and the electrolysis used in their study is similar to the PEC used in this study. It was found that the CO_2 and other emissions into the air were lower with electrochemical treatment of the sediment than with no treatment. This was the case even when the production of electrodes and the electricity used for treatment were taken into account. This can be explained by the fact that the reduced pollutant concentration in the sediment offers more deposition or usage alternatives, reducing the need for transportation and associated air pollution emissions. Environmental effects of sediment management options include impacts on land and sea organisms, human health, land use, and air quality, as well as CO_2 emissions. Enhanced PEC is beneficial for degrading organic compounds, and the H2O2 dose can be adjusted after the organic load. If H_2O_2 is used, the environmental impact of H_2O_2 production must be considered, along with the method of hydrogen production used [80]. To reduce the environmental impact, another option may be to use of UV LED lights instead of fluorescent tubes [81]. Treatment of water with photo-Fenton has shown no toxic effects [82], but the effects on biota need to be investigated if the treated sediment is to be returned to the sea (e.g., deep-sea disposal). Additionally, the impact of the method on sediment from a health, environmental, and market perspective must be further studied. The impact on the quality of the residual sediment after treatment should also be further investigated to ensure safe handling and optimal utilization, including analyzing the generation of active chlorine species. In summary, further optimization of the method, such as dosage adjustments, and exploration of other treatment methods should be carried out before it is tested at a larger scale in a pilot study to determine the feasibility of the method in sediment remediation projects, and to assess the health, environmental and market aspects of the method in sediment remediation projects.

4. Conclusions

This study explored the application of photoelectrocatalysis (PEC) as a treatment method for water and sediment contaminated with organic pollutants and metals. The method was initially optimized using methylene blue in water. The PEC method demonstrated high efficiency in degrading a range of organic pollutants (>99 % removal of PAHs and PCBs and >84 % removal of TBT/DBT/MBT) and for recovering metals, particularly copper (76 %), in clear water solutions.

When applied to real sediment samples, the method was less effective, possibly due to limited light penetration caused by the presence of particles, as well as the more complex nature of sediment matrices. However, the addition of H_2O_2 significantly enhanced the degradation of several pollutant groups. The highest removal efficiencies were observed for TBT (91 %), PCB (82 %), and for the phthalates DINP (98 %) and DEHP (85 %), while other pollutants showed lower degradation rates.

Despite these challenges, the study marks the first documented application of PEC for sediment remediation. It revealed that treatment efficiency is highly dependent on site-specific sediment properties and the composition of pollutants. The findings underscore the complexity of treating contaminated sediments and the challenges of developing a treatment method that can effectively remove pollutant groups.

These results suggest that the treatment should be carried out in two steps. The first step is to release pollutants into the solution phase. The second approach is to treat the resulting solution using PEC, which leads to the oxidation of organic pollutants and the recovery of metals at the cathode. Future research should focus on optimizing system parameters, such as exposure time, pH, H_2O_2 dosage, the liquid-to-solid (LS) ratio, as well as exploring the use of circulating systems, stronger UVC light, varied flow rates, and additives like solubilizers or biosurfactants to further enhance treatment performance.

Environmental implication

Contaminated sediments from ports and urban waterways contain hazardous levels of metals and persistent organic pollutants (POPs) such as TBT, PAHs, and PCBs. This study presents a novel photoelectrocatalytic (PEC) method with the potential to simultaneously degrade organic pollutants and recover metals from real sediment samples with diverse contamination profiles. The method, especially when combined with H_2O_2, shows promise for treating various sediment types. By reducing pollutant loads, PEC enhances reuse potential and lowers environmental risks linked to sediment disposal, thereby supporting sustainable sediment management.

CRediT authorship contribution statement

Oskar Modin: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Conceptualization. **Anna Norén:** Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Funding acquisition, Formal analysis. **Ann-Margret Strömvall:** Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Yvonne Andersson-Sköld:** Writing – review & editing, Funding acquisition. **Nicolaie Markocsan:** Writing – review & editing, Resources. **Aravind Kumar Thoutam:** Writing – review & editing, Resources. **Stefan Björklund:** Resources. **Karin Karlfeldt Fedje:** Writing – review & editing, Writing – original draft, Funding acquisition. **Sebastien Rauch:** Writing – review & editing, Writing – original draft, Methodology, Investigation, Funding acquisition, Conceptualization.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2025.140375.

Data availability

Data will be made available on request.

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