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Biodegradation of polycyclic aromatic hydrocarbons: using microbial bioelectrochemical systems to overcome an impasse

Maria Kronenberg [a], Eric Trably [a], Nicolas Bernet [a], Dominique Patureau [a]

[a] LBE, INRA, 102 avenue des Etangs, 11100 Narbonne, France, izabel.kronenberg@supagro.inra.fr

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Abstract

Polycyclic aromatic hydrocarbons (PAHs) are hardly biodegradable carcinogenic organic compounds. Bioremediation is a commonly used method for treating PAH contaminated environments such as soils, sediment, water bodies and wastewater. However, bioremediation has various drawbacks including the low abundance, diversity and activity of indigenous hydrocarbon degrading bacteria, their slow growth rates and especially a limited bioavailability of PAHs in the aqueous phase. Addition of nutrients, electron acceptors or co-substrates to enhance indigenous microbial activity is costly and added chemicals often diffuse away from the target compound, thus pointing out an impasse for the bioremediation of PAHs. A promising solution is the adoption of bioelectrochemical systems. They guarantee a permanent electron supply and withdrawal for microorganisms, thereby circumventing the traditional shortcomings of bioremediation. These systems combine biological treatment with electrochemical oxidation/reduction by supplying an anode and a cathode that serve as an electron exchange facility for the biocatalyst. Here, recent achievements in polycyclic aromatic hydrocarbon

removal using bioelectrochemical systems have been reviewed. This also concerns PAH precursors: total petroleum hydrocarbons and diesel. Removal performances of PAH biodegradation in bioelectrochemical systems are discussed, focussing on configurational parameters such as anode and cathode designs as well as environmental parameters like porosity, salinity, adsorption and conductivity of soil and sediment that affect PAH biodegradation in BESs. The still scarcely available information on microbiological aspects of bioelectrochemical PAH removal is summarised here. This comprehensive review offers a better understanding of the parameters that affect the removal of PAHs within bioelectrochemical systems. In addition, future experimental setups are proposed in order to study syntrophic relationships between PAH degraders and exoelectrogens. This synopsis can help as guide for researchers in their choices for future experimental designs aiming at increasing the power densities and PAH biodegradation rates using microbial bioelectrochemistry.

Capsule:

This review can help as guide for researchers in their choices for future experimental designs aiming at increasing the power densities and polycyclic aromatic hydrocarbon biodegradation rates using microbial bioelectrochemistry.

Keywords:

Polycyclic aromatic hydrocarbons; microbial fuel cell; microbial electrolysis cell; exoelectrogen; bioremediation

Abbreviations:

BES: Bioelectrochemical system

MFC: Microbial fuel cell

MEC: Microbial electrolysis cell

PAH: Polycyclic aromatic hydrocarbon

HDB: Hydrocarbon degrading bacteria

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are part of a large group of compounds characterized by a structure composed of carbon and hydrogen atoms. PAHs derive from the incomplete combustion of organic matter, i.e. from transportation fuel (Lan et al., 2016), coal mining (Yakovleva et al., 2016), emissions from power plants and petroleum spills (Lübeck et al., 2016) as well as other anthropogenic sources. All PAHs are typically “water-hating” and “fat-loving” chemicals (Jones and de Voogt, 1999) and are very difficult to biodegrade (Barrie et al., 1992). The fate of PAHs in the environment is associated with multiple abiotic and biotic processes including volatilisation, microbial and chemical transformation and sorption (Reid et al., 2000). Once these water-hating molecules are released into water bodies or soils, they have a tendency to preferentially adsorb to organic particles, thus becoming less available for

biological degradation in aqueous phases (Palm et al., 2004). In consequence, they are inclined to accumulate in soils (Yang et al., 2014) and sediments (Feng et al., 2012). Inside organisms they accumulate in the fatty tissues of the cell rather than the aqueous compartments where most substrate bio-transformation takes place. Hence, apart from their thermodynamic stability, the degradation of PAHs by microorganisms is an issue since their likeliness to adsorb on non-polar molecules makes them less bio-available (Huang et al., 2013; She et al., 2013) while the metabolic pathways involved in their degradation are slow (Katsoyiannis, 2016). Exposure of humans to polycyclic hydrocarbons in the environment involves inhalation of vapours or air-suspended particles , dermal contact as well as dietary ingestion (Domingo, 2004). A massive environmental and societal challenge is at stake to limit PAH bioaccumulation in the food chain since they have been widely recognized as carcinogenic, teratogenic and mutagenic contaminants (Kim et al., 2013).

2. Limits of bioremediation of polycyclic aromatic hydrocarbons

Bacteria are known to play a fundamental role in the removal of contaminants from the environment (Hatamian-Zarmi et al., 2009; Pieper and Seeger, 2008). However, considering that most contaminated sediments and soils are oxygen depleted, *in situ* bioremediation of PAHs is often associated with anoxic or anaerobic conditions. Bioventing is a technique that supplies oxygen so as to create aerobic conditions. Numerous aerobic bacteria that utilise PAHs as a carbon and energy source have been isolated (Venkata Mohan et al., 2006), thus revealing that PAHs containing 2 to 4 rings can be easily biodegraded under oxic conditions (Cerniglia, 1993). However, the delivery of oxygen to sediments has been criticized as ineffective (Cruz Viggi et al., 2015) since oxygen rapidly diffuses away from where it was applied (Zhang et al., 2010). Previous studies mainly focused on anaerobic PAH degradation by mixed cultures under nitrate, sulfate or iron reducing conditions(C.-H. Li et al., 2010a; Winderl et al., 2010; M. Xu et al., 2015). Even though certain strains for PAH removal under anaerobic conditions have been identified, the microbial degradation of PAH is still limited by five factors: (i) low abundance, diversity and activity of indigenous PAH degrading bacteria (Fagervold et al., 2005), (ii) slow growth rates of the organisms with doubling times ranging from several weeks to months (Kazumi et al., 1997; Meckenstock et al., 2004), (iii) high partial pressure of hydrogen (Christensen et al., 2004), (iv) undershot threshold concentrations of pollutants and/or pollutant specific microorganisms (Rhee et al., 2001; Young-Cheol Cho et al., 2003) and (v) rare bioavailability of PAHs in the aqueous phase (Barret et al., 2010; Venkata Mohan et al., 2006).

How limitations of PAH bioremediation are addressed at present

Research efforts consist in accelerating the indigenous microbial turnover of PAHs (Chang et al., 2003; Coates et al., 1996). On the one hand, bio-stimulated growth of naturally occurring pollutant-degrading microorganisms can be achieved by the addition of electron acceptors, co-substrates and nutrients. Electron acceptors such as nitrate, ferric ion and sulfate are used by anaerobic microbes for the discharge of electrons that they obtain from organic matter utilisation. It has been observed that nitrate amendment enriches microbial functional genes allowing for *in situ* PAH bioremediation (Xu et al., 2014). Nutrients and co-substrates, additionally, promote microorganism cell growth and enhance their bioremediation capacity. Examples include methanol and acetate that have been employed as additional

carbon sources for microorganisms in order to increase the natural attenuation of PAHs in a marine sediment (Zhang and Lo, 2015). Biostimulation, on the other hand, is another common approach for removing organic pollutants. However, in the case of nitrates, biostimulation leads to secondary pollution issues (Pandey and Fulekar, 2012). A limited dispersion of chemicals through the soil matrix (Anneser et al., 2010), their consumption via abiotic reactions or their diffusion away from PAHs (Chun et al., 2013; Yu et al., 2016) can decrease the efficiency of biostimulation (Jobelius et al., 2011). This implies that a periodic replenishment of chemicals renders biostimulation economically unfavourable (Mittal and Rockne, 2010). Bioaugmentation represents a third option for PAH bioremediation. This technique is based on the application of indigenous or exogenous microorganisms such as fungi (Yanto et al., 2017) or bacteria (Song et al., 2017; Trably et al., 2003) to polluted and hazardous wastes in order to accelerate the removal of PAHs (Liu et al., 2014; Payne et al., 2011). Nevertheless, as the preparation of the inoculum and the loss in microbial activity during inoculation are energy and time consuming, bioaugmentation appears to be an even less economical solution (Laszlova et al., 2016).

3. Bioremediation of polycyclic aromatic hydrocarbons assisted by microbial bioelectrochemical systems

Basics of microbial bioelectrochemistry

Microbial bioelectrochemical systems (BESs) are one or two compartment electrochemical devices in which an anode and a cathode favour redox-reactions that are catalysed by microorganisms (Allen and Bennetto, 1993): this is the electroactive biocatalyst. Electrons produced by microorganisms from organic matter oxidation are transferred to the anode and flow via a conductive material to the cathode where they are consumed by biotic or abiotic reduction reactions (Logan et al., 2006). In this way, microbes convert organic matter into electric current (Kiely et al., 2011) which can be used to drive an electrical load. The principle of bioelectrochemistry is the ability of certain microorganisms to interchange electrons with insoluble conductive materials via extracellular electron transfer (Reimers et al., 2001; Tender et al., 2008). On the anode side, these are called anode-respiring bacteria or exoelectrogens.

MFC and MEC

The prototype of a BES is the microbial fuel cell (MFC) that generates electricity based on organic matter oxidation in the anode chamber (Venkata Mohan et al., 2014). Oxygen is traditionally used as electron-acceptor in the cathode compartment or at the air-cathode due to its unlimited availability and high standard redox potential (Bond and Lovley, 2003; Hou et al., 2012). Depending on the complexity of the substrate, typical maximum power densities of MFCs lie between 2 and 3 W/m^2 of projected electrode surface (mostly cathode) at optimum conditions (30°C, neutral pH, 20 mS/cm conductivity, buffered solution) (Logan and Rabaey, 2012). A rise in electrode area per volume of reactor running on wastewater achieved power densities up to 1.55 kW/m^3 (2.77 W/m^2) (Reimers et al., 2001). MFCs can be run on inexpensive organic wastes such as wastewater, sludge and biowastes (Venkata Mohan et al., 2014), in which PAHs can be found at low concentrations. The second type of BES is the microbial electrolysis cell (MEC) that uses a small amount of energy to fuel chosen reactions in the cathode

146 chamber. In a MEC, oxygen is eliminated from the cathode chamber so that electrons released from the
147 cathode can be exploited for other uses than oxygen reduction. Mostly, protons from aqueous solutions
148 are reduced to hydrogen gas (Villano et al., 2010) although other terminal electron acceptors have also
149 been suggested. These comprise chlorophenols (Kong et al., 2014), nitrobenzene (Mu et al., 2009; Wang
150 et al., 2011), polychlorinated ethenes (Aulenta et al., 2008; Strycharz et al., 2008) and cobalt(II) (Huang
151 et al., 2014). In most studies, it is assumed that electroactive microorganisms catalyse the oxidation and
152 reduction reactions in the anode and cathode chambers (Jafary et al., 2015; Rozendal et al., 2008).
153 Nevertheless, the thermodynamic reaction barrier of these reactions still needs to be circumvented with
154 a small external voltage delivered by a power source (Sun et al., 2008) or by setting an electrode
155 potential using a potentiostat (Logan and Rabaey, 2012).

156 ***The application of microbial bioelectrochemistry to polycyclic aromatic hydrocarbon removal***

157 The biocatalytic principle of a BES contributes to drive thermodynamically unfavourable reactions and
158 can thus be an alternative electron source for improving PAH removal. Recalcitrant PAHs may be more
159 easily transformed to less complex compounds if the metabolism of PAH degraders is directly or
160 indirectly boosted by a bioelectrochemical activity in the vicinity of the electrodes (Chandrasekhar and
161 Venkata Mohan, 2012). As an example, in an experiment, the number of hydrocarbon-degrading bacteria
162 (HDB) close to the anode of a MFC increased in comparison to an open circuit control when an electric
163 current was applied (Wang et al., 2012). When research on BES began 15 years ago, scientists focused on
164 the production of energy vectors (hydrogen or electricity) while evidence of aromatic hydrocarbon
165 removal by BESs only appeared in 2010 (Zhang et al., 2010). Using ^{14}C-labeling, Zhang et al.
166 demonstrated that toluene could be quantitatively oxidized to carbon dioxide in a MFC in the presence
167 of *Geobacter metallireducens*, a popular exoelectrogenic bacteria found in numerous MFC publications
168 (Zhang et al., 2010). Since the oxidation of toluene was achieved in the absence of an electron acceptor
169 other than the anode, this discovery allowed them to ascertain that electrodes can stimulate the
170 degradation of aromatic hydrocarbons in contaminated anaerobic sediments. In the same study,
171 naphtalene was completely degraded after only 9 days in comparison to zero removal in the heat treated
172 abiotic control and in the open circuit (OC) control. As depicted in Figure 1, research on biocatalyzed
173 remediation in MFCs only accelerated in 2014. Mohan & Chandrasekhar (2011) found further evidence
174 of PAH removal in BESs: they observed that a soil MFC was capable of easily attacking 5-6 ring
175 aromatics within petroleum sludge while anaerobic degradation was only restricted to 2-3 ring aromatics
176 (Venkata Mohan and Chandrasekhar, 2011). Since then, within six years, only about 23 publications
177 (Table 1) focus on the potentials of PAH remediation within bioelectrochemical systems, first and
178 foremost from TPH contaminated soil, diesel and engine oil which contain hydrocarbon mixtures of
179 alkanes, aromatic hydrocarbons and resin-asphaltenes as sole carbon sources. In these studies, co-
180 substrates were never used except for one study in which electricity generation and PAH degradation
181 were improved after glucose addition to a saline soil MFC (Li et al., 2016a).

182 ***Range of PAH concentrations, experiment duration, working volumes, voltages and current***
183 ***densities***

184 Initial hydrocarbon concentrations in soil, sediment, domestic wastewater and synthetic media range
185 from 8 mg to 12 g/kg of dry soil, 2-10 mg/kg of dry matter of sediment and 0.02 g to 30 g/L in aqueous
186 media (Table 1). The studies employ single aromatic hydrocarbons such as naphtalene, anthracene,

phenanthrene and pyrene for spiking but also contaminants from polluted sites such as total petroleum hydrocarbons, engine oil or diesel. All experiments were achieved under batch or fed-batch regime within MFCs. Experiment duration can vary between 17 and 376 days while reactor working volumes range between laboratory scale (100-500 ml) and pilot scale (10-50 L) experiments. The potentials that were measured across external resistors between 100 and 1000 Ω range from 16 to 366 mV. Highest current and power densities, respectively, among all MFCs that processed hydrocarbons attained 299 mA/m^2 (Li et al., 2016b) in rather dry conditions, reaching as high as 198 mA/m^2 (21 mW/m^2) in watery media (Chandrasekhar and Venkata Mohan, 2012). An exceptionally strong current density of 6 A/m^2 was reported in a soil MFC (Zhou et al., 2016) as described further in chapter 4, section "Anode surface & buildup".

Differences in the performances of PAH-degrading MFCs in solution, sediment and soil

The removal of PAHs is generally high in liquid media, often reaching 100 % when a minimal media (Adelaja et al., 2015, 2014) were used. It varied between 55 and 82 % for PAH-containing liquids such as real-field petrochemical wastewater (Yeruva et al., 2015), waste engine oil from saline water (Sabina et al., 2014) or diesel-contaminated media (Morris et al., 2009). In wastewater, the highest removal rate of an aromatic fraction in MFC in aqueous media reached 70 % within 17 days with an initial load of 3 g TPH/L. As for sediment, the first study that employed electrodes for increasing bioremediation in sediments achieved 24 % removal of a petroleum hydrocarbon mixture. This was a 12 fold improvement compared to the natural attenuation of the compounds (Morris and Jin, 2012). The highest removal for sediment microbial fuel cells (SMFCs) was 95 to 99 % for phenanthrene and pyrene, respectively, when BES treatment was combined with a ferrihydroxide addition (Yan et al., 2012). However it was suspected that biodegradation in SMFCs might depend on the polarity of the contaminant. In comparison to the control, more polar hydrocarbons such as alcohols and carboxylic acids were better removed in a SMFC than non-polar compounds like PAHs and nitrogen or sulfur containing cyclo- hexanes and -pentanes (Xia et al., 2015). In soil, degradation efficiencies of MFCs were generally lower (maximal 53 % at an initial load of 30 g/L). Supposedly this is because soil has a 90 % lower water content than sediment (soil water content is usually less than 60 %) (Wang et al., 2012).

4. Configuration of microbial bioelectrochemical systems for polycyclic aromatic hydrocarbon removal

Anode surface & buildup

Zhou et al. recently explored the electricity producing capacity of petroleum hydrocarbon degraders in soil that were isolated from the same soil (Zhou et al., 2016). They observed current and power densities reaching 6 A/m^2 (220 mW/m^2). These values are much higher than previously reported and are attributed to the larger electrode surface of the carbon cloth (Lu et al., 2014a, 2014b). The corresponding degradation results have not yet been reported. Other authors have determined that bioelectrochemical remediation of petroleum hydrocarbons in soil can be achieved beyond a certain distance using a multi-anode configuration which is designed to increase the anode surface. This resulted in higher degradation rates of 18 % (TPH), 36 % (PAH) and 29 % (n-alkanes) compared to natural attenuation (Li et al., 2014). As an example, net concentrations of phenanthrene in all soil layers (1-5 cm

227 from the air-cathode) decreased below 1000 ng/g dry soil while natural attenuation showed a lower
228 performance with a remaining phenanthrene concentration above 1500 ng/g dry soil (data from figure).
229 The TPH removal within a distance of 5 cm from the electrode was explained by an enhanced collection
230 of electrons in the vicinity of each anode layer (Li et al., 2014). At present, the use of multi-anodes has
231 resulted in a seven fold higher accumulation of charge compared to a corresponding single anode setup.
232 A possible up-scale for field applications was proposed by Lu et al. who used a pilot scale MFC of 50 L
233 with biochar and graphite granules as anode material (Lu et al., 2014b). TPH biodegradation was
234 improved by 241 % with respect to the control. In the column-type reactor, TPH removal extended to the
235 sides of the reactor, producing an 82 to 90 % efficiency within an operating radius of 34 cm. Another
236 bioelectrochemical build-up that accelerated the biodegradation of petroleum hydrocarbons in sediment
237 involved a simple conductive graphite rod that was used to connect the anoxic sediment with the
238 overlying oxic water in order to provide the bacteria with oxygen (Cruz Viggi et al., 2015). Oxygen, as a
239 high redox potential electron acceptor, should consequently accelerate oxidative biodegradation.
240 Indeed, after 200 days, biodegradation of petroleum hydrocarbons led to a 12 % and 21 % removal with
241 one or three graphite rods, respectively, whereas only a small removal rate was observed for the biotic
242 control without graphite..

243 ***Anaerobic versus aerobic cathode (MEC versus MFC)***

244 To our knowledge, only two publications concerning PAH removal in a BES with an anaerobic cathode
245 (MEC) are available. The first study used an anaerobic cathode for the bioremediation of naphthalene
246 (100µM) in a dual chamber BES where both electrode chambers were flushed with a mixture of nitrogen
247 and carbon dioxide (95/5) to maintain anaerobic conditions (Zhang et al., 2010). Naphtalene was
248 completely removed after only 9 days while the sterile and open circuit controls did not show any
249 removal. This outcome was the first evidence that an electrode as electron acceptor can stimulate the
250 anaerobic oxidation of aromatic hydrocarbons in contaminated sediments. In the second study,
251 Sherafatmand and Ng tested the action of a SMFC on naphthalene, acenaphtene and phenanthrene
252 removal with either an aerobic or an anaerobic cathode that they obtained by both diffusing air in the
253 overlying water column of the SMFC or by sealing the reactor (Sherafatmand and Ng, 2015).
254 Interestingly, results for the anaerobic cathode removal were much higher for two of the PAHs (76.9 %
255 naphthalene, 52.5 % acenaphtene, 36.8 % phenanthrene) than results for the degradation with an
256 aerobic cathode (41.7 % naphtalene, 31.4 % acenaphtene, 36.2 % phenanthrene). Nevertheless, both
257 treatments exceeded the natural attenuation of all PAHs, some by a factor of three (natural: 29 %
258 naphtalene, 29 % acenaphtene, 12 % phenanthrene). The authors stipulated that the potential
259 difference between anode and cathode led to a change in the physical and chemical properties of soil
260 that consequently stimulated the prevailing microorganisms. These pioneering studies exemplify that
261 there is a significant opportunity to biodegrade PAHs in BESs where oxygen is completely absent from
262 both electrode chambers.

263 ***Redox contrasting microenvironments***

264 As already discussed in this chapter, PAHs can be degraded under various redox conditions. Yan et al.
265 combined the addition of ferric hydroxide with the treatment of PAHs in a SMFC (Yan et al., 2012). The
266 addition of electrodes under ferric conditions was found to enhance phenanthrene and pyrene
267 degradation rates more than twofold. A 99.5 % phenanthrene removal was achieved after 240 days in

comparison to Fe(III) addition alone (87.6 %) and natural attenuation (80.3 %). The combined use, testified for accelerated humic acid consumption and final sequestration of PAHs to less bio-available fractions of the sediment with removal efficiencies of phenanthrene and pyrene reaching 99.5 % and 94.8 % respectively. Lastly, the combined treatment of macrophyte plantations and the use of a SMFC accelerated the biodegradation rates of benzo(a)pyrene and pyrene by about 70 % in comparison with separate macrophyte and SMFC treatments(Yan et al., 2015). The SMFC enabled the development of anoxic zones in the sediment, thereby increasing the number of aerobic as well as anaerobic PAH degraders. The creation of redox contrasting environments most probably enhanced the syntrophic metabolism between aerobic and anaerobic microbial communities for a successful removal of higher weight PAHs.

5. Impact of environmental parameters on polycyclic aromatic hydrocarbon removal in microbial bioelectrochemical systems

Conductivity

Hydrocarbon degrading MFC in soil was optimal both in terms of current density (299 mA/m^2) and remediation efficiency (484 % higher degradation rate than the control) when carbon fibre was added after having rinsed saline soil with distilled water (Li et al., 2016b). Accordingly, net PAH removal in the connected MFCs ranged between 44 and 88 %. While 1-3 ring PAHs were removed very efficiently the concentration of 4-6 ring PAHs also decreased. This bioremediation success resulted from electron transfer promotion and current collection by the conductive carbon material. The soil electrical conductivity of the control MFC without carbon fibre addition was 0.61 mS/cm whereas the carbon fibre supplemented soil MFC showed a 2.5 fold higher electrical conductivity of 1.5 mS/cm. The corresponding open circuit controls had 110 % and 419 % lower conductivities. In this study, the authors correlated strong degradation rates of hydrocarbons with stronger naphthalene dioxygenase and xylene monooxygenase activities in comparison relative to the non-carbon fibre control. Interestingly, even the high molecular weight fractions of hydrocarbons (C28-C36, 4-6 ring aromatics) could be degraded in this MFC. Moreover, the easy recovery of the carbon fibre could allow for its reuse. As a comparison, the soil conductivity of another saline soil MFC that processed petroleum hydrocarbons ranged between 7 and 14 mS/cm. The MFC with the highest water content (33 %) and highest conductivity achieved a 120 % higher TPH degradation rate than the open circuit control (Wang et al., 2012). A comparison between conductivities indicates that even though conductivity appears to enhance microbial electron transfer reactions, the measurement of conductivity alone cannot be a sufficient indicator of removal success. For example, salts such as sodium chloride can cause microbial inhibition in spite of enhanced soil conductivity. On the contrary, conductive materials such as carbon fibre act as electron transport promoters without inhibiting microbial metabolisms. Interspecies electron transfer (IET) via electric currents through carbon materials and other conductors exhibits faster electron transfer mechanisms compared to interspecies hydrogen transfer (Cruz Viggi et al., 2014) and could therefore explain enhanced bioremediation of PAHs. As another example, the addition of 1 % (w/w) carbon fibre to a soil MFC could further enhance PAH removal compared with the MFC without carbon fibre. In open circuit controls without carbon fibre, the net concentration of 16 PAHs decreased by 5% whereas the equivalent

closed circuits showed a 5 fold removal. However, the carbon fibre MFC outcompeted the classic MFC with a 7.5 times higher removal rate than in the open circuit (Li et al., 2016c). Interestingly, the open circuit control containing carbon fibre similarly outcompeted the open circuit control without carbon fibre. This confirms that the sole presence of carbon fibre can enhance PAH removal even if electrodes are not connected. Likewise, semi-conductive minerals (Kato et al., 2012) as well as granular activated carbon have boosted methane production through improved IET when added to anaerobic digestors (Liu et al., 2012). This explains why conductive carbon materials such as activated carbon of different sizes (S. Xu et al., 2015) , biochar (Chen et al., 2014a) and carbon cloth (Chen et al., 2014b) have been proposed as a tool for accelerated electron transfers between species.

Correlation between power density and polycyclic aromatic hydrocarbon removal

The relationship between the power density of a MFC and biodegradation efficiency has been poorly investigated. However, a correlation between current or power density and the degree of PAH biodegradation inside BESs could be an easy approach for monitoring *in situ* PAH removal issues. In certain electrokinetic studies, the degradation of diesel (Mena et al., 2014) and petroleum hydrocarbons (T. Li et al., 2010) was found to increase as the current intensity rose. Petroleum was mixed to soil at a level of 50 g/kg dry soil and an electric current was constantly supplied at a voltage of 1 V/cm. There are two reasons for the accelerated removal of petroleum hydrocarbons: in general the supplied current supports microbial activity; but more particularly it can boost the metabolism of HDB that use oil as a sole carbon substrate. MECs are also equipped with a small current that is applied to the existing microbial community. Until present, only a few studies on the above-mentioned anaerobic cathodes (MEC setups) are available in the literature. Nevertheless, the suggestion that the removal of hydrocarbons can be improved with the assistance of an anode in MFCs is supported by the close relationship between charge output, the presence of HDB and PAH removal. Various studies show an increase in the number of HDB in soil close to the anode (1cm). Wang et al. tested anodes that were precultured with exoelectrogenic bacteria for TPH removal in petroleum in contaminated soil (Wang et al., 2012). As a result, the number of HDB within 1 cm from the anode increased by almost two orders of magnitude, rising from $8*10^3$ CFU/g-soil to $373*10^3$ CFU/g-soil in comparison with the open circuit control. Consequently, the net biodegradation of 16 PAHs increased from 30 to 42 % within 25 days when the soil MFC had a water content of 33 %. The substantial increase in the amount of HDB highlights the biostimulation of their growth by the colonisation of exoelectrogenic bacteria in the same system. The authors stipulated that the limited biostimulation beyond a certain distance from the anode results from the need for exoelectrogenic bacteria to connect to the anode either through direct contact or through a conductive network which is limited in length (Wang et al., 2012). The study that testified the highest current density (299 mA/m^2) also reported, after 65 days, the strongest removal rate with a 153 to 484 % higher TPH degradation rate relative to the control. Nevertheless, other authors did not find any correlation between power densities and biodegradation rates; they have attributed this observation to oxygen intrusion (Adelaja et al., 2015), substrate to biomass conversion (Hu et al., 2011) and electron losses due to electron transfer from substrates to other electron acceptors such as sulfates in solution and/or assimilation of electrons for bacterial growth (Liu et al., 2005).

Porosity & mass transfer

The addition of sand was found to increase the soil porosity inside bioelectrochemical reactors; this probably enhanced PAH transfer simultaneously with a general improvement in mass transfer (Li et al., 2015). The authors pointed to a reduced ohmic resistance (by 46 %) in such a way that petroleum hydrocarbon removal was improved by 84 % within 135 days compared to the control MFC without any sand supplement. In particular, the degradation of n-alkanes and PAHs with high molecular weights was accelerated. However, the enhanced porosity of the soil also enabled more oxygen diffusion into the soil layers thus suggesting the possibility that some PAH degradation may be due to aerobic processes. The enrichment of the species *Alkanivorax* in the layer closest to the cathode supposedly accounts for the biodegradation of petroleum hydrocarbons. In addition, *Geobacteraceae* species were detected according to the supposed exoelectrogenic activity in this layer. Another approach to improve mass transfer for enhanced TPH degradation in soil was proposed by Zhang et al. The group compared hydrocarbon degrading MFCs (contaminated soil from real-field) with a horizontal (HA) and a vertical arrangement (VA) of anodes. A lower resistance of mass transport in the HA than that of the VA seemed to result in increased power generation and more TPH degradation (Zhang et al., 2015).

Salinity

Since 40 % of the worlds' soils are saline environments, Serrano and Gaxiola (1994) decided to investigate the effect of salinity on PAH removal in BES. They found that the performance of a hydrocarbon degrading saline soil MFC could be enhanced by rinsing the saline soil with distilled water. This rise in microbial activity was achieved following the reduction in salinity that was initially causing inhibition (Li et al., 2016b). Before rinsing, the soluble salt content was 8.6 times higher than after rinsing (2.8 % compared to 0.3 %). Optimum conditions for soil MFCs included a 1 % salt content (w/v) for which 91.6 % and 79.1 % of hydrocarbons and COD, respectively, were removed, with a maximum power density of 1.06 mW/m^2 (Adelaja et al., 2015). Furthermore, waste engine oil was used as an organic substrate in a desalination microbial fuel cell that achieved biodegradation of branched and cyclic aliphatic hydrocarbons (70.1 ± 0.5 %) along with desalination (68.3 %) and power production (3.1 mW/m^2) (Sabina et al., 2014). In consequence, 8 % more hydrocarbon was removed than when preliminary flask studies were carried out with the same *Bacillus subtilis* moh3 culture isolated from waste engine oil.

Sorption of polycyclic aromatic hydrocarbons on soil and on electrodes

Since PAHs are hydrophobic contaminants, they tend to adsorb equally to hydrophobic surfaces. An observed removal can be related either to the biodegradation of a contaminant, to its sequestration to organic matter (Yan et al., 2012) or to adsorption to the electrode of a MFC as demonstrated for benzoic (Yang et al., 2013) and trace organic compounds (Wang et al., 2015). Indeed, adsorption controls should be performed so that the actual remains of an environmental pollutant inside a matrix or its decomposition to a metabolite can be distinguished. For instance, adsorption controls have revealed that the carbon anode of a MFC exhibits a high affinity for phenanthrene with a maximum adsorption capacity of 0.45 mg/cm^2 (Adelaja et al., 2015). Yet, sterile adsorption controls in order to monitor the abiotic physical parameters have not been performed systematically throughout all studies.

An exemplary study determined the degradation efficiencies and rates of petroleum hydrocarbons based on their remains in solution and on hydrocarbons adsorbed to the electrode at the end of a MFC operation (Adelaja et al., 2015). Phenanthrene concentrations in the bulk solution rapidly decreased by 90 % within 24 hours, eventually reaching more than 95 % in all fed-batch cycles after 60 days. This fast decrease was attributed to the disappearance of phenanthrene from solution by adsorption to the electrode; microbial degradation of the compound then accounts for the subsequent gradual decrease. It has been previously reported that microorganisms can degrade aromatic hydrocarbons on an electrode and in solution; however, the hydrocarbons need to be initially adsorbed (Zhang et al., 2010). This point of view has already been described by Lovley et al. who stated that electrodes co-localise micropollutants and electrophilic microbes at the site where degradation finally takes place (Lovley, 2006). Therefore, PAHs may desorb from the electrode then follow a biodegradation process which may occur directly on the electrode. Both processes might also take place concomitantly. At present the underlying mechanisms are not at all clear. Chandrasekhar and Venkata Mohan (2012) hypothesized that the oxidation of lower aromatic compounds at the anode could result in OH·-radicals that might attack higher ring structures. In their view, aromatic hydrocarbons adsorbed to the anode surface can serve as a source of carbon for microorganisms

The above-mentioned hypotheses strongly depend on the electrode material. Two studies on the bioavailability of phenanthrene adsorbed to black carbon (BC) without application of a current both showed different results. In the first study, the addition of BC to soil led to a significant reduction in microbial mineralization of phenanthrene (Rhodes et al., 2008); in the second, microbes could utilize about 84 % of phenanthrene adsorbed to BC after aging for 21 days (Xia et al., 2010).

Desorption of polycyclic aromatic hydrocarbons by surfactant addition

Interestingly, surfactant (Triton x-100) addition to a soil MFC equipped with a carbon cloth anode led to a negative impact on the electricity production and TPH removal (Lu et al., 2014a). These authors assumed that accessibility of TPH for bioelectrochemical and microbial degradation in the aqueous phase was reduced when the surfactant sorption to soil at a surfactant concentration of 500 mg/L increased. They suggested that the soil-surfactant-sorption also resulted in increased TPH partitioning onto soil particles. On the contrary, Alcantara et al. and Gomez et al. demonstrated that PAH removal could be enhanced by surfactant addition (Tween 80) to soil. 80 to 85 % of fluoranthene, benzo(a)anthracene and pyrene were desorbed from a soil matrix into the aqueous phase and subsequently electrochemically degraded (Gómez et al., 2010). In a similar experimental setup 86 % phenanthrene was desorbed from soil after ten days and 96 % was removed with the application of a 5 V tension for 3 days (Alcántara et al., 2009). Given these contradictory results, the right combination between soil, surfactant and PAHs still needs to be determined so PAHs may be desorbed from soil or sediment and so biodegradation within BESs can be enhanced.

6. Microbial insights on polycyclic aromatic hydrocarbon removal in microbial bioelectrochemical systems

Syntrophy

Electroactive species can be found in diverse habitats and are affiliated to many different microorganisms such as *Clostridia*, *Bacilli* and different *Proteobacteria*, thus pointing to the absence of an ecological niche for exoelectrogens (Koch et al., 2014). In addition, the complex nature of organic waste requires a diverse microbial community for dissembling its chemical components (Logan and Rabaey, 2012), thus giving rise to syntrophic degraders. The positive effect of electricity in PAH bioremediation is probably related to bioelectrochemical catalysis in which HDB are favoured by the presence of electroactive microorganisms. A syntrophic relationship between exoelectrogens and various bacterial phyla has already been determined by various scientists (Kiely et al., 2011; Kimura and Okabe, 2013; Lu et al., 2012). Fermentative bacteria and acetate consuming anode respiring bacteria (Parameswaran et al., 2009) as well as methanogenic communities and exoelectrogens (Rotaru et al., 2014) have been observed to interact on the basis of cooperative metabolite production and consumption. This is no surprise since syntrophic bacteria drive the anaerobic degradation of certain fermentation products (e.g., butyrate, ethanol, propionate) into intermediary substrates (e.g., H_2, formate, acetate) (Schmidt et al., 2016) that can eventually be used by exoelectrogens and/or methanogens. Although a syntrophic relationship between HDB and an exoelectrogen has not yet been proven, a metabolic cooperation could be imagined between heterotrophs in soil from the *α*- or *β*-*Proteobacteria* phylum such as *Sphingomonas* or *Burkholderia* genus. These organisms use diverse substrates among other recalcitrant aromatic and phenolic compounds as carbon sources (Aislabie et al., 2013). Interestingly, *Burkholderia cenocepacia* was identified as the microorganism that produced the oxidant for exoelectrogens in a MFC (Hunter and Manter, 2011). *Burkholderia species* were also acknowledged as one of the dominant members of an anode chamber microbial community in a MFC using acetate as anodic substrate (Quan et al., 2012). The results described in chapter 5, section "Correlation between power density and polycyclic aromatic hydrocarbon removal" suggest that electricity generating bacteria enhance HDB activity (Zhang et al., 2012) through a syntrophic metabolism, namely by accepting additional electrons from the metabolites of HDB degradation. By shuttling them to the anode, they supposedly alleviate thermodynamic limitations of PAH removal similar to those described under methanogenic conditions (Christensen et al., 2004) when *Archaea* consume hydrogen to keep hydrogen partial pressure low enough for an exergonic reaction to occur (Gieg et al., 2014). Furthermore, microbial community analysis from an olive mill wastewater treating MFC has shown that both exoelectrogenic and phenol-degrading microorganisms from the *Sphingomonas* phylum were concentrated in the anode biofilm (Bermek et al., 2014).

Promising microbes for PAH removal in BESs

Several studies cite the *Proteobacteria*, *Firmicutes* and *Bacteriodetes* phyla comprising bacteria belonging to the *Enterobacteriaceae* and *Pseudomonadaceae* families that could be potential key players of PAH removal in BESs.. Likewise, *Escherichia* species (Li et al., 2014; Lu et al., 2014a) and the *Gamma*- and *Delta-Proteobacteria* as well as *Negativicutes* and *Bacilli* (*Firmicutes*) classes are possible candidates for successful PAH biodegradation in MFCs (Table 2) since their abundances have shown positive

correlations with hydrocarbon degradation (Li et al., 2016a; Venkidusamy et al., 2016). Differences in functional bacteria can be observed for crude oil and diesel degradation. Accordingly, 71 % of the predominant γ-Proteobacteria in a crude oil degrading MFC were *Escherichia sp.* (Li et al., 2016a) whereas the dominant phylum following bioremediation of diesel contaminated soil was *β-Proteobacteria* including mainly genera of *Achromobacter*, *Alcaligenes*, *Bordetella*, *Comamonas*, and *Pusillimonas* (Lu et al., 2014a). As an example for microbial selection during bioelectrochemical treatment, the abundance of *Firmicutes* increased by 346 % on average during the BES-treatment of a petroleum contaminated soil, at a much higher degree than for conventional bioremediation (Popp et al., 2006). Furthermore, a study by Lu et al., pointed out that all exoelectrogens belonged to *Proteobacteria*. More specifically, exoelectrogens that could also degrade hydrocarbons belong to the *Enterobacter* and *Pseudomonas* genera. The recognised hydrocarbon degraders and exoelectrogenic bacteria *Pseudomonas putida* and *Pseudomonas aeruginosa* were also part of the anode biofilm. *Pseudomonas* species respire anaerobically via the production of phenazines and pyocyanin. These electron shuttling compounds can in turn help microbial co-oxidation of contaminants such as phenanthrene via electron transfer to the anode (Qiao et al., 2015). In addition, Venkidusamy et al. reported sequence similarities with *Geobacillus* and *Megasphaera* species that both participate in current generation of mediator-less MFCs and diesel degradation (Liu et al., 2009; Venkidusamy et al., 2016). The anode biofilm of another diesel degrading MFC contained *Shewanella* and *Dechloromonas* species. Certain *Dechloromonas* species such as *Dechloromonas aromatica* are capable of degrading aromatics, (Salinero et al., 2009). They can use toluene, benzene and xylene as electron donors under anaerobic conditions coupled to nitrate reduction (Chakraborty et al., 2005). The discovery of the *Shewanella* species is promising since *Shewanella oneidensis* and *Shewanella putrefaciens* are part of many MFC anode biofilms (Gorby et al., 2009). Moreover, *Shewanella* species have been found to possess nanowires which are believed to improve the utilization of the anode as an electron acceptor (Gorby et al., 2009).

An excellent microbe for PAH removal in BESs: Geobacter?

Disregarding the fact that high power densities from complex sources of organic matter are typically associated with the presence of *Geobacteraceae* in the anodic community (Kiely et al., 2011), *Geobacter metallireducens* (Zhang et al., 2013) and *Geobacter sulfurreducens* (Zhou et al., 2016), in particular, can initiate the degradation of benzene and naphtalene respectively. Beside their capacity of mediated and unmediated electron exchange using self-produced flavins (Park et al., 2016; TerAvest and Angenent, 2014), they have also been found to facilitate substrate degradation and power production in MFCs through nanowires (Bond and Lovley, 2003; Shu et al., 2016). The pilin proteins potentially enable direct electron exchange with co-cultured species as seen for a co-culture of *G. metallireducens* and the methanogen *Methanosarcina barkeri* (Cheng and Call, 2016) as well as direct interaction with an electrode (Ding et al., 2016; Malvankar and Lovley, 2014). *Geobacteraceae* have been found in two petroleum removing MFCs (Li et al., 2014; Zhou et al., 2016) and one toluene removing MEC (Zhang et al., 2010), thus representing three out of nine publications that investigated the presence of specific microbial phyla in PAH degrading BESs (Table 2). Interestingly, the capacity of a soil-MFC to selectively enrich exoelectrogenic hydrocarbon degraders was convincingly demonstrated by the isolation of two

exoelectrogens from *Geobacter* and *Ochrobactrum* species that used petroleum hydrocarbons as a sole carbon source (Zhou et al., 2016).

7. Perspectives for polycyclic aromatic hydrocarbon removal in microbial bioelectrochemical systems

Enrichment cultures for PAH bioremediation in BESs

Correlation analysis testified that the biodiversity in soil beyond a certain distance from the cathode was mainly determined by the concentration of petroleum hydrocarbons, while the biodiversity in soil close to the cathode was dominated by the current (Li et al., 2016a). The change in microbial community structure in hydrocarbon-degrading MFCs in comparison to their open circuit controls clearly demonstrated that BESs significantly impact the metabolic pathways of biota present in soil or sediment. BESs assumedly favour syntrophic cooperation between different phyla of microorganisms such as *Geobacter* and *Ochrobactrum*. Studies with enrichment cultures that employ both known PAH degraders as well as exoelectrogens should be encouraged to advance our knowledge on the syntrophic relationships between them, to determine how PAH removal can be improved in these systems and to apply acquired knowledge to *in situ* experiments.

Conductive materials for PAH bioremediation in BESs

The more conductive carbon materials have proved to enhance PAH removal in MFCs (Li et al., 2016b, 2016c) and to improve IET in anaerobic digestion (Liu et al., 2012). Nevertheless, the detailed mechanisms explaining how these materials contribute to the observed performances still have to be determined (Kouzuma et al., 2015). To shed light on this matter, experiments investigating the relationship between soil conductivity, the addition of conductive materials and the biostimulation success of PAHs by BESs should be encouraged. A second unknown factor is the relationship between the supplied current and degradation efficiency of PAHs in MECs. However, a positive correlation between the biodegradation of PAHs and the amount of supplied current has been demonstrated in several electrokinetic studies (T. Li et al., 2010; Mena et al., 2014). Examples of positive correlations between current density and PAH biodegradation in MFCs have been put forward although, it is yet unclear whether the success of bioremediation through the application of current in MECs is due to a change in soil properties or whether the metabolism of HDBs is accelerated more directly. There is still a need for further research to be conducted in order to verify these hypotheses.

Anaerobic cathodes for PAH bioremediation

To our knowledge, few published studies have investigated the use of anaerobic cathodes to remove PAHs inside BESs, and none have yet attempted the use of biocathodes. Nonetheless, the few existing studies all point to opportunities for successfully removing PAHs, as discussed in chapter 4, section "Anaerobic versus aerobic cathode". Further studies are still required using anaerobic cathodes in order to answer the question whether MECs can be as efficient for PAH removal as MFC setups. Bioremediation in MECs could become an auspicious technology for treating contaminants in strictly anaerobic aqueous or solid media. For instance, Liang et al. degraded the chloramphenicol antibiotic (CAP) in a MEC in which denitrification and dechlorination of the compound took place in the biocathode

538 chamber. The MEC attained removal efficiencies of 96 %, a result which is 20 % higher than CAP removal
539 in the control experiment with an abiotic cathode (Liang et al., 2013). In aqueous media it could be
540 possible to use a single cell containing a bioanode and biocathode in the same compartment. This would
541 favour sequential redox-reactions of PAHs where ring fission could be followed by further decomposition
542 of alkylchains. However, knowledge concerning the microbial key players of anaerobic biocathodes in
543 MECs is still sparse (Jafary et al., 2015; Parkhey and Gupta, 2017). Biocathodes in MECs are most often
544 only employed for enhancing hydrogen production (Kadier et al., 2016; Lu and Ren, 2016), therefore it is
545 crucial that research on PAH bioremediation in MECs should be initiated.

546 ***Conventional versus bioelectrochemical PAH bioremediation***

547 Bioelectrochemical remediation of contaminated soil, sludge and sediment can be an inexpensive
548 alternative to conventional *in situ* bioremediation. Currently, certain authors rather consider the
549 performance of MFC technology for waste or wastewater treatment than for the generation of high
550 current densities (Li and Yu, 2015; Zhang et al., 2016). The low but stable electricity gain of SMFCs could
551 be used to power biosensors that monitor the on-site state of pollution during real time operations. For
552 instance, King et al. proved the utility of MFCs as biosensors for the detection of recalcitrant organics
553 such as bisphenol A in water (King et al., 2014). Not least, the use of SMFCs as long term electrical power
554 sources for sensors and buoys has been confirmed (Tender et al., 2008). Indeed, with the help of a SMFC,
555 Yang et al. charged batteries for electrical devices such as a camera and mobile phone (Yang et al., 2015).
556 Moreover, in cases where *in situ* bioremediation would require a small voltage, solar energy could be
557 collected by placing a biocathode at the subsurface of a polluted site, thus benefitting from the
558 advantage of low maintenance costs (Lovley, 2011).

559 ***Scaling of bioelectrochemical systems for PAH bioremediation***

560 Most MFC studies, for the removal of both organic pollutants and COD, are still limited to laboratory or
561 pilot-scale experiments. Problems arise at these scales with low power output owing to ohmic losses
562 (Cusick et al., 2011) and unfavourable biomass production in continuous flow operations (Logan, 2010).
563 For this reason, field experiments on hydrocarbon removal from contaminated sites should be
564 encouraged in order to validate *in situ* applications, especially when authors are confident towards large
565 field implementations without loss of removal efficiency (Lu et al., 2014b). When aiming at large scale
566 applications for recalcitrant pollutant treatments with BESs, it would be preferable to focus research
567 efforts on the study of PAH treatment with mixed cultures. These would be more robust when adapted
568 to changing microenvironments and less susceptible to contaminations. This demand for real-field up-
569 scaling goes hand in hand with the need for lab-scale studies during which experimental parameters can
570 be better controlled and treatment efficiencies are comparable between experiments. Particularly, the
571 question whether contaminants are adsorbed or degraded after sorption to the electrode material
572 and/or the polluted soil should be critically examined with the help of sorption controls. Simultaneously,
573 as there are knowledge gaps in the capacity to control the microbiology of such systems, there is a clear
574 need for fundamental research on pure or simplified culture media. This would contribute to the future
575 engineering of site-specific processes, and to a better understanding of the electron transfer
576 mechanisms between microbes and the surface of electrodes.

8. Conclusion

This review has identified the frontiers of the state of the art in PAH treatment by BESs in order to assist researchers in the field on the decisions they would have to take regarding the design of their next experimental plans in terms of configuration of the BES and of the environmental parameters that are employed. Pioneer studies conclusively demonstrate the success of the BES technology as an *in situ* approach for enhanced bioremediation of recalcitrant compounds such as PAHs, TPHs and diesel in contaminated soil, sediment and liquid media. Apparently, BESs can respond to two of the five stated limitations of PAH degradation: First and foremost, microbial bioelectrochemical systems can improve the activity of indigenous PAH degrading bacteria, thereby increasing their abundance; and secondly, they can partially circumvent the lack of PAHs in the aqueous phase by adsorbing contaminants to the surface of the electrode where they can be degraded before or after desorption. Additionally, several suggestions for further research have been put forward, taking into account the microbial aspects (e. g. culture studies with hydrocarbon degraders and exoelectrogenic bacteria) and portraying innovative methods to increase interspecies electron transfer (addition of conductive carbon materials) in BESs for an enhanced biodegradation of PAHs.

The main advantages of BESs for PAH bioremediation include the facts (a) that electrodes can serve as permanent terminal electron acceptors for electron discharge of biota, (b) that removal efficiencies of PAHs, TPHs and diesel can be improved without the need for a periodical replenishment of chemicals, (c) that a BES can be integrated into existing *ex situ* treatments with a potential for *in situ* treatment, (d) that the use of electrodes as electron donors and/or acceptors can generate a current for supporting electronic equipment, (e) that inexpensive and environmental friendly conductive carbon materials can be used inside BESs to further increase biodegradation and (f) that recalcitrant organic pollutants can be eliminated simultaneously with an enhanced organic matter conversion. In addition, the use of sediment or soil MFCs could alleviate the present drawbacks of *in situ* bioremediation such as expensive post-treatment for recycling or eliminating added chemicals. For this reason the use of BESs for PAH bioremediation is probably more environmentally friendly and less threatening to human health than conventional *in situ* bioremediation techniques. Albeit its challenges as a recent technology, the use of BESs for PAH biodegradation could be the way out of an impasse, since the removal of PAHs from anaerobic environments with BESs has proved to be satisfyingly diverse and efficient.

9. Acknowledgements

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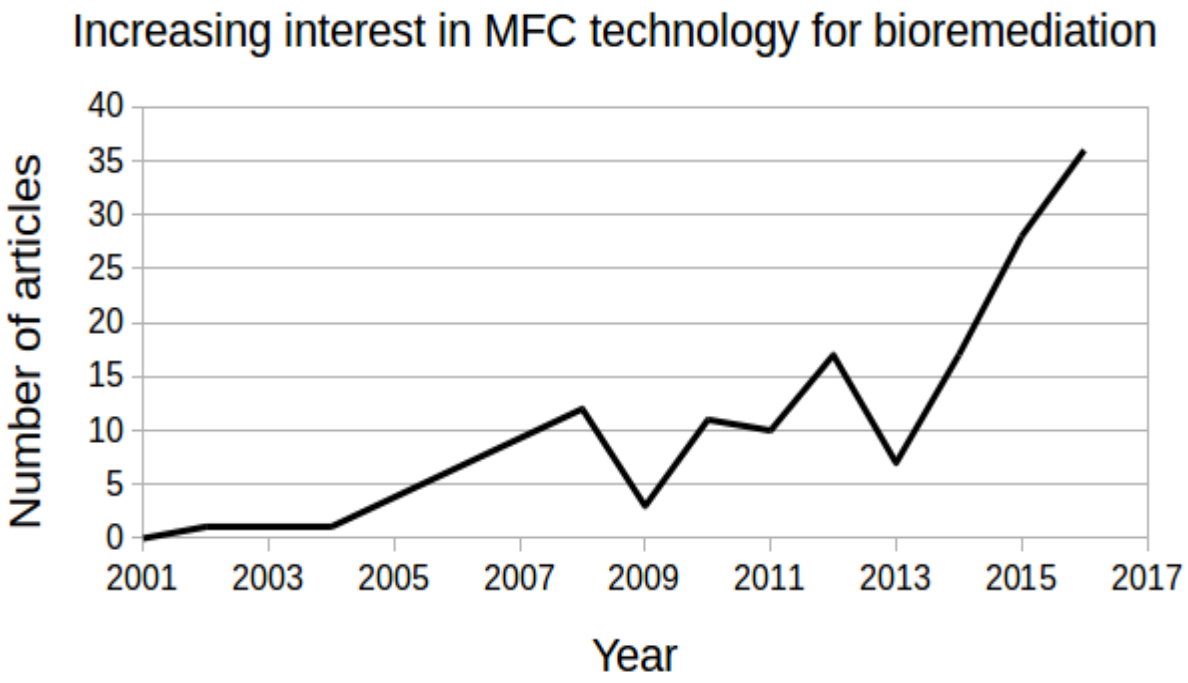

Figure 1: Number of hits of relevant articles per year (reviews excluded) on “Web of Science” in February 2017 using the key words "bioremediation" and "microbial&fuel&cell"

Author	Type of BES	Matrix	Pollutant	Start Concentration	Removal [%] (Degradation rate if indicated)	Controls (types and performances)	Experiment length (days)	Max. current density (CD)/ max power density (PD)* normalized by anodic or cathodic surface area
(Adelaja et al., 2014)	MFC	Synthetic medium	Benzene, phenanthrene	200 mg/L benzene, 30 mg/L phenanthrene	> 96 (for all inocula) best: mixed culture+ P. aeruginosa: 27.3 µM/d (phenanthrene), 65.6 (COD) Others: Co-culture and pure strains (P. aeruginosa, S. oneidensis), same strains mixed with digested sludge,	For each inoculum: abiotic MFC, disconnected MFC, anaerobic control using the same inocula	60	1.25 mW/m^2 (normalized to the projected total surface area of the anode (40 cm^2), 18 mA/m^2 (data from figure), max. voltage: 65 mV (data from figure)
(Adelaja et al., 2015)	MFC	Minimal medium	Benzene, phenanthrene	100 mg/L benzene 100 mg/L phenanthrene	91.6 (benzene +phenanthrene) 79.1 (COD)	Abiotic: 20 (benzene), 10 (phenanthrene)	60	4.89 mA/m^2, 0.72 mW/m^2
(Chandrasekhar and Venkata Mohan, 2012).	MFC	Domestic wastewater	Real field petroleum sludge	3000, 9000, 15000, 30000 mg TPH/L sewage OL1 (3 g) OL2 (9 g) OL3 (15 g) OL4 (30 g)	58 (aromatics at 30 g TPH/L) 70 (aromatics at 3 g TPH/L)	Four organic loading (OL) conditions: Low load better for power generation, high load better for substrate degradation	17	20.6 mW/m^2, 198.3 mA/m^2 at 3g TPH/L, 11.8 mA/m^2 at 30g TPH/L
(Li et al., 2016b)	SMFC	Saline Soil	TPHs, n-alkanes, PAHs	Max: 0.484 mg/kg (PAH), Max:20.181 mg/kg (n-alkanes)	153-484 % higher TPH degradation rate in all MFCs compared to CK, RM>MC>RS>CK RM: 60+-9 (TPH), OSOC: 3 (TPH), PAH removal: 59 -92, n-alkanes: 44 -88, 4-6 ring PAHs particularly removed in RM	CK: unmodified soil MFC RS: rinsed soil MFC MC: carbon fiber (2 %) + original soi MFC RM: carbon fiber (2 %) + rinsed soil MFC RMOC, RSOC, OSOC, MCOC: open circuit configurations of RM,RS, CK and MC	65	299 mA/m^2 (RM): 595 % higher than CK
(Li et al., 2016a)	SMFC	Saline soil	TPHs, n-alkanes, PAHs (phenanthrene,	Contaminated soil: 945 mg/kg PAHs 28.322 mg/kg	TPH: 21 (LG, Layer 1, 62 % higher removal than CK) TPH removal: LG > HG > CK > LGOC > HGOC > OC	CK: closed circuit OC: open circuit LG: soil + current + low glucose HG: soil + current + high	135	HG: 43 mW/m^2, ca. 108 mA/m^2 (with U= 0.4 V data from figure), LG: 35 mW/m^2, 79 % and 46 % higher

			fluoranthene, pyrene, Chrysene accounting for 70 % of all PAHs)	(n-alkanes), LG (low glucose 0.1 % w/v) HG (high glucose 0.5 % w/v)	PAH: 34 (LG) 44 (HG)	glucose LGOC, HGOC: open circuit controls		than that of CK, CK: 24 mW/ m^2
(Li et al., 2014)	SMFC	Saline Soil	C8-C40 n-alkanes, 16 PAHs	5.653 mg/kg (total PAHs) 609 mg/kg	Max. in soil layer four: 18 (TPH), 36 (PAH), 29 (n-alkane) compared to control with natural attenuation, degradation rates depending on soil layer [SL] in distance to anodes (SL 4 > SL 1 > SL 2 > SL 3), SL4 closest to air-cathode	open circuit	180	37 mW/m^2, 366 mV, 102 mA/m^2 on day 5
(Li et al., 2015)	MFC	Aged soil	TPHs, n-alkanes, 16 PAHs	Max: 945 mg/kg (PAH) Max: 27.891 mg/kg (n-alkane)	22 (TPHs) in high ratio sand/soil MFC, (+52 % versus LS), (+84 % HS versus CK), (+268 % HS versus OC), higher removal for PAH with sand addition	Soil only (OC); soil + current (CK); sand + soil +current: low sand/soil and high sand/soil ratio= (LS)=5:1 (HS)=2:1, 15 (less sand MFC) 12 (without sand but connected electrodes) 6 (OC)	135	0.28 mA/m^2 /g soil 2.76 mW/m^2/g soil
(Li et al., 2016c)	MFC	Soil	16 PAHs, 30 n-alkanes	7942 ± 7476 ng/g (PAHs), 701 ± 736 µ g/g (n-alkanes)	5 % (CKOC), Compared to CKOC: CK showed 5-fold removal, MC showed 7.4-fold removal, MCOC 4.4 times higher than CKOC	Original soil (OS), OS + carbon fiber (MCOS), Carbon fiber MFC (CK), OS-MFC (MC), Open circuit controls : CKOC, MCOC	144	203 mA/m^2, 0.8 mW/m^2, 100 Ω, Average over 144 days: 120.5 mA/m^2 for CK, 7.5 mA/m^2 for MC
(Lu et al., 2014b)	MFC	Soil	Hydrocarbons from raw diesel	12300 mg/kg dry soil	82-90 (1-34 cm radius)	68	120	70.4 mA/m^2

(Lu et al., 2014a)	MFC	Soil	TPHs	11460 mg/kg dry soil	78.7 (BCA) 73.1 (CCA) 62 (CCA-S; taken from figure)	41.6 (OC of BCA) 40.7(OC of CCA) CCA-S: CCA with surfactant addition	64	85.9 mA/m^2, 39.1 mW/m^2 (BCA) 73.0 mA/m^2, 17.7 W/m^2 (CCA), 21.2 mA/m^2, 2.9 mW/m^2 (CCA-S)
(Morris and Jin, 2012)	MFC	Sediment	TPHs	16000 mg/kg sediment	24 (closed circuit)	2 (open circuit)	66	190 mV, 2.2 mW/m^3
(Morris et al., 2009))	MFC	Ground water	Diesel range organics (DRO) C10-C28 aliphatics	176-241 mg/L	82 (DRO) 6.9 mg DRO/(L day) (or 34.5 µmol/day)	31 (cathode removed)	21	32 mW/m^2 Data from figure: ~175 mA/m^2 average 160 mV (CD and PD calculated per cathode surface)
(Venkata Mohan and Chandrasekhar, 2011)	MFC	Soil	TPHs	2-480 mg/L soil (calculated by us: 480000 mg/kg soil)	41 (Total TPH), 98 (benzo(g,h,i)perylene), 96 (dibenzo(a,h)anthracene)	Anaerobic controls: 21 (total) 68 (Benzo(g,h,i)perylene) 56 (Dibenzo(a,h)anthracene)	17	20.62 mW/m^2, 198.31 mA/m^2, 104 mV
(Sabina et al., 2014)	MFC	Saline water	Waste engine oil (branched, long-chain & cyclic hydrocarbons)	1 % (v/v) (100ml/L)	70.1 in desalination MFC (gravimetrical decrease in engine oil)	62.9 % (gravimetrical; shake flask study with Bacillus subtilis moh3)	14	3.1 mW/m^2 (based on the cross-sectional area of anode)
(Sherafatmand and Ng, 2015)	SMFC/ SMEC	Sediment	PAHs, naphtalene, acenaphtene, phenanthrene	50 ppm for all three PAHs (50 mg/kg)	Anaerobic cathode: 76.9 Naphthalene, 52.5 acenaphtene, 36.8 phenanthrene	Natural attenuation: 29 (Naphtalene), 29 (acenaphtene), 12 (phenanthrene), Aerobic cathode: 41.7 (Naphtalene), 31.4 (acenaphtene), 36.2 (phenanthrene)	45	2 mW/m^2 in anaerobic, 5.8 mW/m^2 in aerobic
(Venkidusamy et al., 2016)	MFC	Synthetic medium	Diesel	800 mg/L	100 (1 year selected biofilm anode)	83 (freshly inoculated anode)	30	90.8 mW/m^2, 115mA/m^2 normalized to cathode surface

(Cruz Viggi et al., 2015)	SMFC	sediment	Spiked crude oil (TPH)	11300-12300 mg/kg dry sediment	After 200 d: 12 (S) 21 (S3) 0 (C)	S: One graphite rod, S3: three graphite rods, C: biotic without graphite rod, B: autoclaved+ one graphite rod, B3: autoclaved+ three rods,	417	No external electrical charge input or output
(Wang et al., 2012)	MFC	Saline soil	TPHs, 16 PAHs (phenanthrene, chrysene, pyrene and phenanthrene representing 75 %)	0.5- 0.8 mg/kg soil	15.2 (TPH), 120 % higher degradation rate in CC (TPH) than OC in 1 cm distance of the anode	6.9 (open circuit), CC: closed circuit	25	0.85 mW/m^2
(Xia et al., 2015).	SMFC	Sediment	PAHs, cylohexanes, cyclopentanes	ca. 3 µg/g sediment (PAHs)	< 15 %	Mainly < 10 % (open circuit)	20 days	0.6 V 4.32 mW/m^2 9.29 mA/m^2
(Yan et al., 2012)	SMFC	Sediment	Phenanthrene, pyrene	10 and 5 mg/kg dry sediment for phenanthrene and pyrene	94.8 (Pyrene), 99 .5 (phenanthrene) (SMFC + FeOOH); combined treatment best for all anaerobic conditions except methanogenic	Open circuit: 80.3 (phenanthrene), 74.5 (pyrene); ferric hydroxide (FeOOH): 87.6 (phenanthrene) , 79.9 (pyrene); SMFC: 96.1 (phenanthrene), 92.1 (pyrene)	240	16-17 mV, average CD not reported (average per anode surface: 170 mA)
(Yan et al., 2015)	SMFC	Sediment	Benzo(a)pyrene, pyrene	2 mg/kg dry sediment (benzo(a)pyrene), 4 mg/kg dry sediment (pyrene)	87 (Macrophyte +SMFC) Combined treatment degradation rates are 70% higher than control treatments	68 (Macrophyte alone), 56 (SMFC alone), 27 (control)	367	61 -65 mV, average CD not reported directly (average given with: 650 mA/anode surface, 17 A/m^2)

(Yang et al., 2015)	SMFC	Sediment	PAHs	PAH: 1.2 mg/kg wet weight, PBDE: 178µg/kg wet weight	22.1 (TOC removal in closed circuit)	3.8 (TOC removal in open circuit)	2 years	0.4 -0.6 V
(Yeruva et al., 2015)	MFC/ MFC + anoxic cathode	wastewater	Petrochemical wastewater (BOD 5 /COD: 0.36).	9.68 kg COD/m^3*day	55 (COD in anoxic SBR-BET), 49 (COD in aerobic SBR-BET) SBR: Sequential batch reactor BET: Bioelectrochemical treatment	34.6 (COD anoxic SBR), 30 (COD removal in aerobic SBR); both of these without BET	HRT: 48 h	17.1 mW/m^2 (anoxic), 14.3 mW/m^2 (aerobic) OCV: 248 and 280 mV
(Zhang et al., 2010)	MEC	Sediment	Naphtalene	100 µM (naphthalene) mixed with seawater and sediment	100 (Naphthalene)	0 (Naphtalene if heat treated) 0 (Naphtalene without electrode)	9	-
(Zhang et al., 2015)	MFC	Soil	TPHs, 16 PAHs, C8-C40 n-alkanes	25700 mg/kg dry soil	12.5 (Horizontal anodes HA),	8.3 (Vertical anodes VA) 6.4 (open circuit)	135	HA max. 0.282 V, VA max. 0.285 V, 833 coulomb accumulated for VA over the whole duration: 9 % more than in VA, current and power densities not indicated
(Zhou et al., 2016)	MFC	Soil	Petroleum hydrocarbons	20 mM	-	-	7	600 µA/cm^2 (6 A/m^2), 220 mW/m^2

Table 1: MFCs and MECs that are used to degrade aromatic hydrocarbons, total petroleum hydrocarbons and diesel (If not indicated explicitly in the cited article, power and current densities (P/m^2) are calculated by multiplication of maximum voltage and current density (P/m^2=U x I/m^2 x 1/1000) and by the quotient of maximum voltage and resistance per surface of electrode [I=U/R x A])

References	Species	Bacteria (Phylum, Class, Order, Family, Genus)	Hypothetical role proposed by author
(Li et al., 2014)	*Geobacteraceae* sp.	*Proteobacteria*, *δ- Proteobacteria*, *Desulfuromonadales*, *Geobacteraceae*	Electricity generation, hydrocarbon degradation
(Li et al., 2014)	*Escherichia sp.*	*Proteobacteria*, *γ-Proteobacteria*, *Enterobacteriales*, *Enterobacteriaceae*, *Escherichia*	Electricity generation, hydrocarbon degradation
(Li et al., 2016b)	*Negativicutes* *δ- Proteobacteria* *Bacilli*		Hydrocarbon degradation (TPH, alkane, PAH)
(Li et al., 2015)	*Alkanivorax*	*Proteobacteria*, *γ – Proteobacteria*, *Oceanospirillales*, *Alcanivoracaceae*	Hydrocarbon degradation
(Li et al., 2015)	*Proteobacteria* (88%, *γ – Proteobacteria)*		Hydrocarbon degradation
(Li et al., 2015)	*Escherichia* sp.		Hydrocarbon degradation
(Lu et al., 2014a)	*Enterobacter cloacae*	*Proteobacteria*, *γ-Proteobacteria* *Enterobacteriales*, *Enterobacteriaceae*, *Enterobacter*	Oxidation of hexadecane and diesel
(Lu et al., 2014a)	*Pseudomonas aeruginosa*, *Pseudomonas putida*	*Proteobacteria*, *γ-Proteobacteria*, *Pseudomonadales*, *Pseudomonadaceae*, *Pseudomonas*	Electricity generation, hydrocarbon degradation
(Morris et al., 2009)	*Shewanella* sp.	*Proteobacteria*, *γ-Proteobacteria*, *Alteromonadales*, *Shewanellaceae*, *Shewanella*	Diesel range organics degradation
(Sabina et al., 2014)	*Bacillus subtilis moh3*	*Firmicutes*, *Bacilli*, *Bacillales*, *Bacillaceae*, *Bacillus*	Waste engine oil: long-chain, branched and cyclic hydrocarbons

(Venkidusamy et al., 2016)	*Shewanella*, *Rhodopseudomonas*, *Bacteriodetes*, *Clostridium*		Electricity generation
(Venkidusamy et al., 2016)	*Geobacillus* sp.	*Firmicutes*, *Bacilli*, *Bacillales*, *Bacillaceae*, *Geobacillus*	Diesel degradation, current generation in mediator-less MFC
(Venkidusamy et al., 2016)	*Megasphaera* sp.	*Firmicutes*, *Negativicutes*, *Selenomonadales*, *Veillonellaceae*	Diesel degradation, current generation in mediator-less MFC
(Venkidusamy et al., 2016)	*Flavobacterium*, *Stenotrophomonas*, *Geobacillus*, *Gordonia*, *Actinobacterium*	*Flavobacteria*, *Flavobacteriaceae*, *Flavobacterium*; *Proteobacteria*, *γ – Proteobacteria*, *Xanthomonadales*, *Xanthomonadaceae*; *Firmicutes*, *Bacillis*, *Bacillales*, *Bacillaceae*; *Actinobacteria*, *Actinomycetales*, *Gordoniacea*	Diesel degradation
(Zhang et al., 2010)	*Geobacter metallireducens*	*Proteobacteria*, *δ- Proteobacteria*, *Desulfuromonadales*, *Geobacteraceae*	Toluene degradation
(Zhou et al., 2016)	*Ochrobactrum cicero* *Ochrobactrum intermedium*	*Proteobacteria*, *α– Proteobacteria*, *Rhizobiales* *Brucellaceae* *Ochrobactrum*	Electricity generation, petroleum hydrocarbon degradation
	Geobacter sulfurreducens	*Proteobacteria*, *δ- Proteobacteria*, *Desulfuromonadales*, *Geobacteraceae*	Electricity generation, petroleum hydrocarbons degradation

Table 2: MFC microbial communities involved in PAH removal

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