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REVIEW PAPER

The fate of total petroleum hydrocarbons during oily sludge composting: a critical review

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Abstract In this review, studies on petroleum sludge composting have been investigated in the aim to understand the sludge biotransformation and highlighting the points that need more attention. Total petroleum hydrocarbons (TPH) are the major contaminants present in the petroleum sludge they ranged from 0.204 to 521.12 g/kg of dry matter, indicating a potential risk to the environment if not correctly managed. In this case, composting of petroleum sludge with different bulking agents (soil, wood, green wastes and manure) have been used as an environmentally friendly treatment. The TPH removals ranged between 47 and 95% during the composting process. This difference between the TPH removals is due to divergence in TPH concentration, chemical characterization, mixing ratio and applying or not of ameliorated treatment. The enhanced treatment like the bioaugmentation and the biostimulation increased significantly the TPH degradation kinetic which ranged between 0.8 and 2%/day of TPH degradation, while in the unimproved composts it was below 0.66%/day of TPH degradation. Moreover, lack information on physico-chemical parameters in totally or partially of some studies restrain the better understanding of the whole petroleum sludge composting. The humic substances can absorb some hydrocarbons during their formation process, it is required to assess the interactions between these compounds throughout the composting. It is also important to know about the finality of the product from the composting process. In all, more researches are needed on the interactions between TPH and other organic matters, removal mechanisms, the toxicity and the stability of the final products.

Keywords Petroleum sludges · Total petroleum hydrocarbons · Composting · Biodegradation · Bioremediation · Humification

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1 Introduction

Petroleum sludge is produced throughout the petrochemical industry chain during the extraction, the transportation, the storage and the refining. At the storage tank level, sediments are formed in the bottom and these sludges are the most studied type in the literature (Hu et al. 2013). They are composed of oil, solid particles and water, and are commonly known as oily sludges (Hu et al. 2013). Sludge production in the petroleum refining industry also comes from oil/water separation operations at the wastewater treatment plant, especially the coagulation-flocculation treatment, the dissolved air flotation and the biological treatment (Van Oudenhoven et al. 1995; IPIECA 2010).

Sludge produced from oil refining continues to increase year after year as the global demand for fossil energy grows (Bhattacharyya and Shekdar 2003). As reported in the BP statistical review of world energy the global energy demand raised up in recent years, increasing the oil refining production all over the world (Fig. 1). It has been estimated that for each ton of crude oil, 3–5 kg of sludge is produced by refineries (Van Oudenhoven et al. 1995). Jean et al. (2001) reported that annually a petroleum refinery produces about 28,000 tons of sludges.

Total petroleum hydrocarbons are one of the most abundant constituents in petroleum sludges. These compounds are responsible for the toxic nature of the sludges and can represent from 5 to 86% of the total sludge masse (da Silva et al. 2012; Hu et al. 2013). These hydrocarbons are rich in sulfur, nitrogen, oxygen, phosphorus and metallic elements. Hydrocarbons can be utilized as a source of carbon and energy and then degraded through the action of microorganisms (Sierra-Garcia and Oliveira 2013). Nevertheless, this biodegradation is influenced by several factors such as the concentration of hydrocarbons, the nutrients, the temperature, the oxygen and the humidity. Composting can improve the biodegradation by modifying some parameters such as the carbon/nitrogen/phosphorus ratio (C:N:P), the mixing ratio, and the moisture to maintain a high microbial activity (Ball et al. 2012).

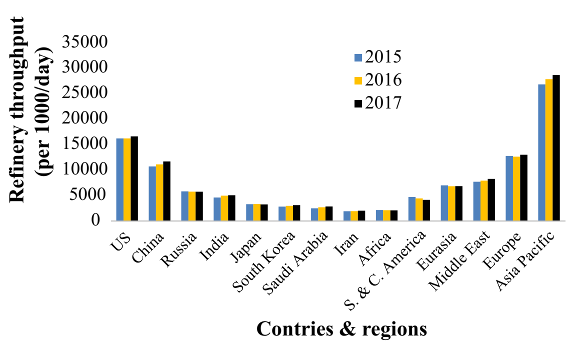

Fig. 1 Global oil refining throughput in recent years (BP 2018)

Composting involves microbial strains that feed on waste while transforming organic substances into stable and humified products. In general, the composting takes place in two phases: a stabilization phase recognized by an intense microbiological activity which is accompanied by a rise in temperature (thermophilic phase), afterward the temperature gradually decreases and stabilizes indicating the depletion of the medium in simple molecules and is dominated by the humification process (maturation phase) and may last for months (Hassen et al. 2001). Composting can be done outdoors in the form of a pile or in a semi-closed system called bioreactor and the sludge is mixed with a bulking agent, e.g. green waste, straw, wood waste, household waste or other sources of easily biodegradable organic matter.

In this review, we have investigated the most relevant studies on petroleum sludge composting, in order to compare between the initial conditions, the processing system and the treatment enhancements. In addition, some points not stated in these studies have been highlighted to better assess the pollution transfer, in the aim to maximize preservation of the environment.

2 Composition and toxicity of the petroleum sludges

Due to their varied origins, which depend on the crude oil quality, the process refinery configuration and the wastewater treatment system, the composition of the petroleum sludge is very diverse and complex. Table 1 summarized the physico-chemical proprieties of the petroleum sludges and highlighting their content diversity. If the biological way of treatment is adopted for this kind of sludges, the carbon and the nitrogen amount can restrain the microbial growth and thus the all biological process. In this case, the addition of co-substrates or nutrition

Table 1 Petroleum sludge physico-chemical characteristics

References	Sludge characteristics					Heavy metals (mg/kg)				
	pH	TOC	TKN	TPH	PAH	Nickel	Chrome	Zinc	Pb	Cu
Ayotamuno et al. (2007)	5.8	4.9	1.3	69.372	–	–	–	–	–	–
Koolivand et al. (2013)	6.9	374.17	1.5	104.3	–	–	–	–	–	–
Koolivand et al. (2017)	6.4	412.58	1.58	131.72	–	37.6	70.7	15.4	122.4	537
Asgari et al. (2017)	1.35	542.44	9.96	521.12	–	56	119	12,248.7	132.7	4420.4
Marín et al. (2006)	7.81	391	–	281	–	175	121	1299	112	115
Kirchmann and Ewnetu (1998)	6.1	499	–	200	–	–	–	–	–	–
Kriipsalu et al. (2007)	8	265	16	130	0.134	–	–	–	–	–
Ouyang et al. (2005)	7.2–7.6	–	–	371.2	–	–	–	–	–	–
Atagana (2014)	–	395	–	300.592	–	175	165	1445	123	105
Milne et al. (1998)	–	–	–	300	–	–	–	–	–	–
Fountoulakis et al. (2009)	7.5	125.1	5.1	0.204	0.051	–	–	–	–	–
Ayotamuno et al. (2010)	5.4	94.6	0.02	98.032		–	–	–	–	–
Kriipsalu and Nammari (2010)	7.3	171	9	160	0.171	–	–	–	–	–
John (2007)	–	–	–	–	–	15.2	–	288.7	66	22.6
Roldan-Carrillo et al. (2012)	7.82	234.3	4.08	334.7	–	100	200	8200	–	–
Kriipsalu et al. (2008b)	7.3	171	9	160	0.168	130	14	840	40	43
Aguelmous et al. (2016, 2018)	8.02	415	22.9	470	–	167.9	53.6	4238.5	45.8	89.3
da Rocha et al. (2010)	–	–	–	–	–	125	119	769	565	213.5

–, no information; TOC, KTN, TPH and PAH presented in g/kg

containing nitrogen and phosphorus can help to achieve an optimum C/N ratio and thus enhance the bioremediation. In addition to organic matter, petroleum sludges may contain from 4 to 7% of mineral sediments such as halite, calcite, kaolinite, quartz, silica, carbonate and hercynite (Monteiro et al. 2007; Aguelmous et al. 2016). Hu et al. (2013) reported that the most common forms of heavy metals in this type of sludge are zinc, lead, copper, nickel and chromium. Their concentrations vary from sludge to another and can reach very high values making this type of waste harmful to the environment (Table 1). However, microorganisms are capable to remove the heavy metals by different mechanisms such as biotransformation, bioaccumulation, biomineralization, bioadsorption, and bioleaching (Dixit et al. 2015). Microbial species have been reported to have the capacity to adsorb high amount of heavy metals, e.g. 567, 467, 270, 166 and 116 mg/g of Pb can be degraded, respectively by *Corynebacterium glutamicum*, *Bacillus firmus*, *Pseudomonas putida*, *Rhizopus nigricans*, *Penicillium chrysogenum* (Choudhary et al. 2017).

The total petroleum hydrocarbons (TPH) are the major toxic fraction in the oily sludges and can reach high values (Table 1). They are composed of carbon (83–90%), hydrogen (10–14%), sulfur (0.1–5.5%), nitrogen (0.05–2%), oxygen (0.1–4%) and traces of phosphorus and metallic elements (Okoh 2006; Zhang and Fan 2010). They are classified into four categories, aliphatic ($>50\%$), aromatic (26–30%), resins and asphaltenes ($<20\%$) (Lal and Khanna 1996; Kadali et al. 2012). Aliphatic hydrocarbons are the most appreciable TPH fraction by microorganisms. Fraction from C_{10} to C_{24} are the most easily biodegradable, while those with long or branched-chain are harder to degrade (Kunihiro et al. 2005). Dumped into the environment, this aliphatic fraction can easily penetrate the ground and reach the groundwater, but can also evaporate quickly (Alvarez and Illman 2005). Aromatic hydrocarbons are generally less volatile and less water-soluble than the aliphatic (Seo et al. 2009). The volatility and the solubility of the aromatic hydrocarbons decrease as the number of benzene rings increases (Mrozik et al. 2003). Polyaromatic hydrocarbons (PAH) are considered ubiquitous and persistent in the environment (Baklanov et al. 2007; Abdel-Shafy and Mansour 2016). The PAH can be photo-oxidized, chemical oxidized, biodegraded or bioaccumulated (Alebic-Juretic et al. 1990; Nadarajah et al. 2002). The PAH are probably mutagenic and carcinogenic to humans and can inflect lung cancer (Kim et al. 2013; Abdel-Shafy and Mansour 2016). Resins and asphaltenes are polar compounds with a high molecular weight (Viñas et al. 2002). Resins are generally dissolved compounds of nitrogen, oxygen and sulfur, while asphaltenes are large and complex colloidal molecules (Varjani and Upasani 2017).

Behind these physico-chemical parameters, the TPH can present a certain toxic degree. The contamination of the water sources, the marine environment, the soils and the air by TPH is one of the major environmental concerns. Water decontamination is complex and difficult given the low solubility of hydrocarbons and their relatively limited mass transfer rate (Paria 2008). Soil contamination can lead to a potentially long source of pollution, and subsequently destroys the plants, the microbial community, and the soil can lose its fertility (Osuji et al. 2006). Trapp et al. (2001) reported that the most biologically active substances in terms of toxicity have a logarithmic value of the octanol/water partition coefficient (Kow) between 2 and 6. Thus, several petroleum hydrocarbons fall into this category, such as hexane and decane which respectively have a Kow of 2.91 and 5.58 (Trapp et al. 2001).

Microorganisms are also affected by the toxic effect of petroleum hydrocarbons. Old previous studies have shown the toxic effect of the aliphatic and the aromatic hydrocarbons on microorganisms (Gill and Ratledge 1972; Calder and Lader 1976; Andrews et al. 1980). Uribe et al. (1990) have shown that this toxic effect on microorganisms is represented by the inhibition of oxygen and potassium uptake by cells, the isolation of mitochondria and the alteration of ATP synthesis. Petroleum hydrocarbons can also alter the cell membrane structure by changing their fluidity and their protein conformation, resulting in barrier disruption and energy transduction, in addition to enzymatic activity disruption (Van Hamme et al. 2003).

Light petroleum hydrocarbons can inhibit or decrease seed germination by easily entering the seeds cells or even by blocking the passage of oxygen and water (Adam and Duncan 2002). Sharifi et al. (2007) observed the phytotoxic effect of different concentrations ranging from 25 to 100 g/kg of hydrocarbons on the germination of certain seeds, the results showed germination between 2.7 and 63.5%. The hydrocarbon toxicity for plants has been demonstrated by several authors proving that at high concentration ($>10\%$) germination can be inhibited while specifying that some plant species have the capacity to withstand at high hydrocarbons percentages (Ogbo 2009; Saadoun and Al-Ghazawi 2010).

Given the composition of petroleum sludges in toxic elements, their disposal in nature may contaminate and pollute the receiving environment. Viscous compounds in petroleum sludges can alter and change the properties of the medium (Robertson et al. 2007), obstruct the soil pores, can be absorbed by mineral elements or form a film on the surface (Al-Mutairi et al. 2008). Tang et al. (2012) reported that residual chemical compounds that have been in the environment for a long time appear to be resistant to sorption and degradation. Generally, in oil refineries, the sludges are placed in drying beds and subsequently stored in an open space which can be seen as a source of atmospheric pollution by volatile organic compounds (Cheremisinoff and Rosenfeld 2009). Thus, to limit the environmental risks associated to the management of the petroleum sludges, several regulations have emerged such as the Resource Conservation and Recovery Act (RCRA) in the United States, which sets strict standards for the handling, storage and safe disposal of these wastes.

3 Petroleum sludge treatments

Petroleum sludge treatment is essential before their disposal in the natural environment. Different approaches can be used for treating the petroleum sludges: we can act at the production source for the reduction of the sludge produced volume, the extraction and reuse of the hydrocarbons contained in the sludges, and finally the disposal after the degradation of organic matter (Pinheiro and Holanda 2009; da Silva et al. 2012).

There exist various ways of petroleum sludge treatments. In general, the physical and chemical methods are applied in the main interest to recover the hydrocarbons present in the sludges, and thus to reduce the released wastes outside the company. The most used methods are solvent extraction, froth flotation, ultrasonic and microwave irradiation, pyrolysis, electrokinetic method, centrifugation and freeze/thaw (Hu et al. 2013). The oil recovery treatments are generally easy and short. However, these types of treatment are quite expensive given the machines applied, the large amount of solvent and water, persistence of heavy metals and the energy consumption, as well as the generation of another waste that will have to be treated later (Fig. 2). Moreover, residues from hydrocarbon recovery processes must undergo a stabilization treatment prior to the final discharge.

The disposal of petroleum sludge can be done by incineration, biodegradation, oxidation and

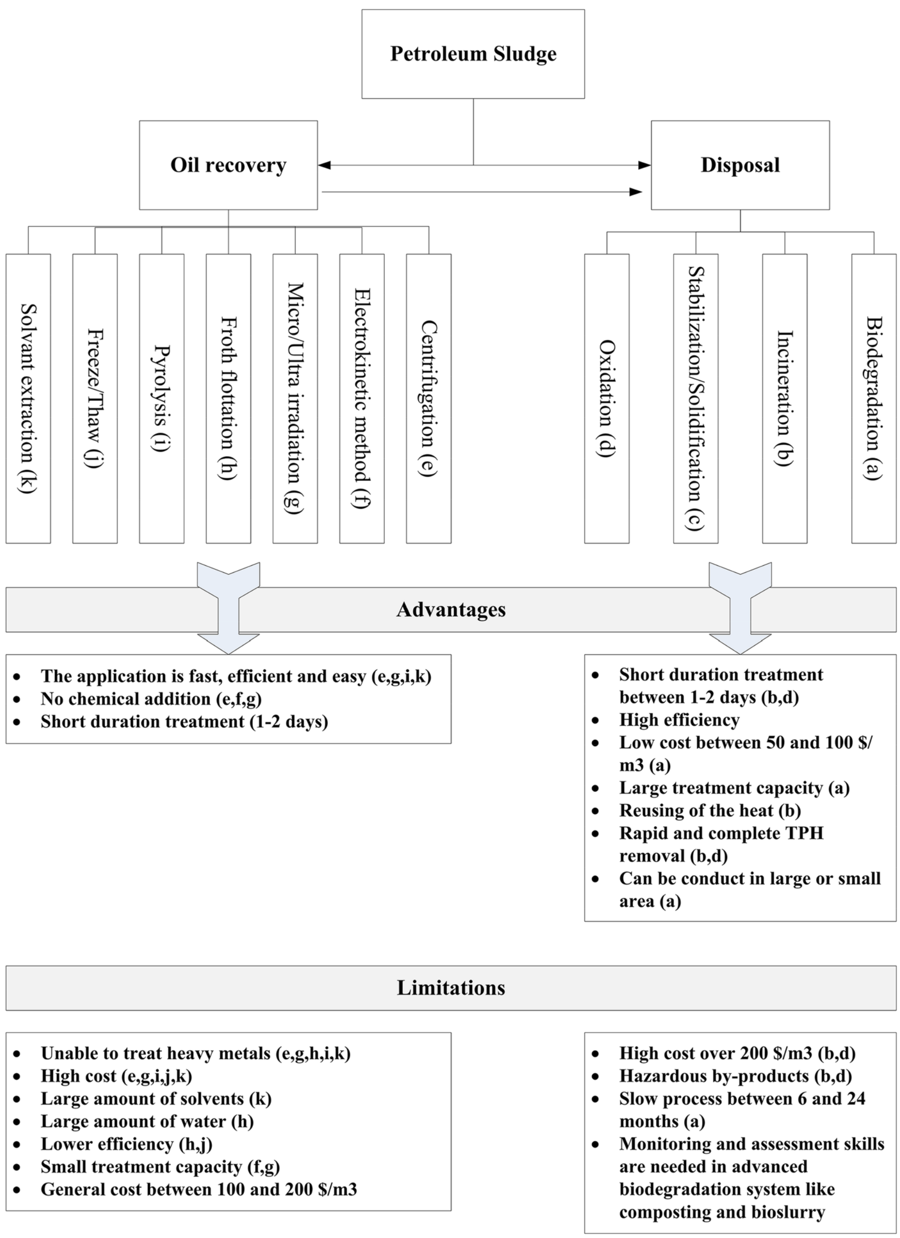

Fig. 2 Overview of petroleum sludge treatments, with respective advantages and limitations (Hu et al. 2013)

stabilization/solidification (Udotong et al. 2011; Hu et al. 2013). The incineration consists in the complete combustion of the petroleum sludge in incinerator over 700 °C (Scala and Chirone 2004). Even if the incineration is effective (99%), its feasibility is affected by the sludge humidity ($>51\%$) (Sankaran et al. 1998). In addition, hazardous gas emissions cause atmospheric pollution in plus of ash residue (Li et al. 1995). Stabilization/solidification treatment is applied to reduce the availability of contaminants (Malviya and Chaudhary 2006). However, some limitations of this method have been noticed, as some studies revealed that the product from this treatment may leach (Karamalidis and Voudrias 2007). The oxidation treatment is aimed to introduce chemical reactive to oxidize organic matter (Ferrarese et al. 2008). This treatment method will require a large quantity of chemical products with increasing quantity of petroleum sludge, in plus of some advanced equipment to enhance the TPH removal (Hu et al. 2013). The choice of the treatment method can be determined by the feasibility, the desired duration, the appropriate space, the targeted contaminants and the global cost (Fig. 2).

Bioremediation has been the most studying treatment of petroleum hydrocarbons, taking into account its low cost, large capacity and high efficiency. Biodegradation is a process by which microorganisms transform or mineralize organic compounds through metabolic or enzymatic processes into less harmful products, which are subsequently integrated into natural biogeochemical cycles (Maletić et al. 2013). Under aerobic conditions, microorganisms use hydrocarbons and thus form CO_2, H_2O and biomass. For more than a century, it has been reported that certain types of microorganisms have developed the ability to use aliphatic and aromatic hydrocarbons as a source of carbon and energy (Sierra-Garcia and Oliveira 2013). Some species such as *Pseudomonas aeruginosa*, *Pseudomonas putida*, *Bacillus subtilis*, *Bacillus cereus*, *Bacillus licheniformis* and *Bacillus laterospor* can modify the membrane surface of their cells to increase their affinity for hydrophobic substances and facilitate their absorption (Cybulski et al. 2003). Two mechanisms can be undertaken to activate the biodegradation of petroleum hydrocarbons: the interfacial adhesion by direct contact with the hydrocarbons, and by the adhesion to emulsified hydrocarbons via biosurfactants (Kaczorek et al. 2011; Varjani and Upasani 2017). The biodegradation of petroleum hydrocarbons involves metabolic reactions catalyzed by a multitude of enzymes. The enzymes that play an important role in this process are oxygenase, peroxidase, reductase, hydroxylase and dehydrogenase (Abbasian et al. 2015). In general, during the hydrocarbon biodegradation, the oxygen serves as an external acceptor of electrons, while an organic element from the substrate functions as an electron donor or energy source (Braddock et al. 1997). In the literature, several species of bacteria have proven their ability to degrade the petroleum hydrocarbons, such as *Pseudomonas spp*, *Alcaligenes* spp., *Stenotrophomas maltohilia*, *Yersinia* spp., *Bacillus* spp., *Micrococcus* and *Enterobacteria* (Lazar et al. 1999; Van Hamme et al. 2000). However, the *Pseudomonas* spp. species was the most reported in the TPH biodegradation studies, especially *P. aeruginosa* (Rocha and Infante 1997). On the other hand, fungal strains are also frequently used in the TPH biodegradation. These include *Aspergillus* spp., *Penicillium* spp., *Mortierella* spp., *Trichoderma* spp., *Candida*, *Rodoturela*, *Aureobasidium* and others (Riser-Roberts 1998; Bento and Gaylarde 2001).

4 Petroleum sludge composting

The composting helps to achieve an optimum C/N ratio around 20 which is not the case of petroleum sludge (Table 1), to enhance the growth of microorganisms. The composting takes place in two phases, the first called thermophilic is the short one (3–15 days) and it is characterized by the rise of temperature and degradation of light/micro-molecules; the second called maturation characterized by the long duration (3–12 months) in which degradation of heavy/macro-molecules occurs. The rise of the temperature over than 60 °C allows the sanitization but also the application of this treatment in extreme climatic conditions (Delille et al. 2007; Ball et al. 2012). The composting process can be done in piles/windrows, bioreactors or both successively (Fig. 3). The choice of the engineered composting system can be done by referring to the physico-chemical parameters of the petroleum sludge, area, duration and cost. The bioreactor is the most desirable composting mode since it allows the optimization of oxygen, temperature, humidity, nutrients and the control of

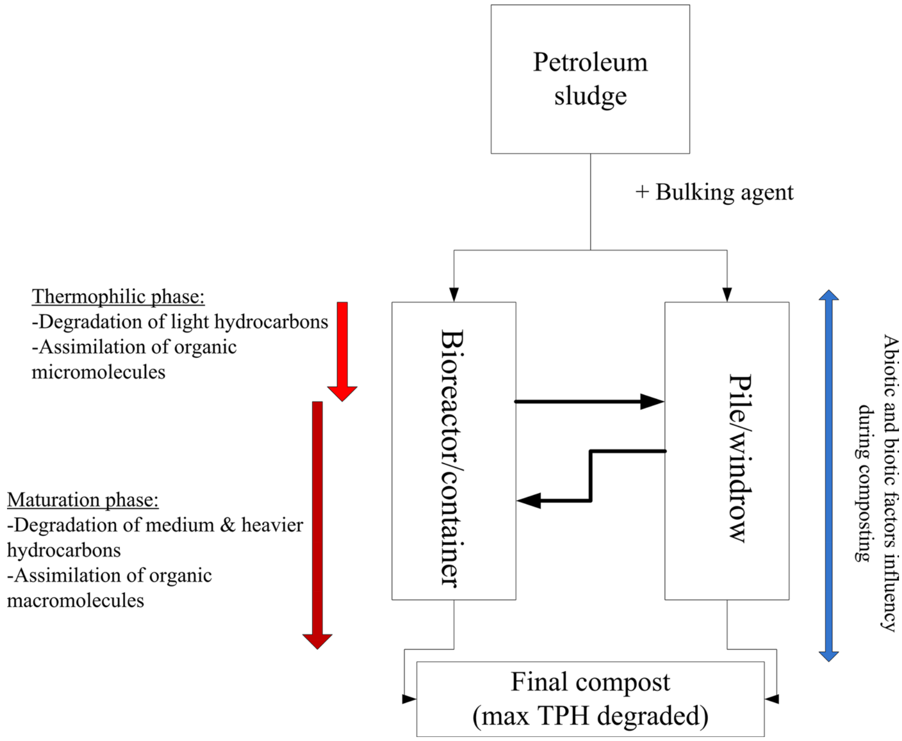

Fig. 3 Overview of petroleum sludge composting process

gas emissions. In a novel arrangement of petroleum sludge treatment consisting in primary composting in piles followed by a secondary composting in bioreactor, revealed an improvement of 10% in TPH removal than composting in piles only (Koolivand et al. 2017). The removal of TPH in composting process can be affecting by several factors such as the physico-chemical parameters and the microbial community and their specific enzymes.

4.1 Abiotic factors

Varjani and Upasani (2019) recorded between 3.4 and 4.6% of TPH degradation in abiotic test during petroleum sludge composting with disappearance of lighter hydrocarbons, indicating the role of abiotic factors (i.e. physico-chemical proprieties) in this process.

The pH range between 6.5 and 8.5, may enhance microbial activity during the composting process of petroleum sludges (Koolivand et al. 2017). However, there are some microbial strains that can resist and adapt to extreme conditions, e.g. *Candida digboiensis* which was reported to biodegrade 95.7% of TPH in a pH at 3 (Sood et al. 2010).

Aerobic biodegradation is the most effective way for TPH degradation, which means that the presence of oxygen for this process is the key parameter for hydrocarbon catabolism (Van Hamme et al. 2003). The initial step in the degradation of aliphatic, cyclic and aromatic hydrocarbons by bacteria and fungi involves oxidation of substrates by oxygenase, for which the presence of oxygen is paramount (Leahy and Colwell 1990).

According to Viñas et al. (2005), the degradation rate of petroleum sludge compounds is also affected by the moisture content. However, very high humidity can disadvantage the microbial growth by limiting the oxygen supply. Otherwise, a study on the TPH bioremediation reported that moisture of 10% is sufficient to reach more than 70% of TPH removal in 270 days (Haghollahi et al. 2016).

The TPH characteristics may influence on the petroleum sludge composting. Minai-Tehrani et al. (2015) reported that in 31% of TPH degradation, aliphatic, aromatic, resin and asphaltene fractions decreased, respectively by 60, 42, 6 and 4%. In another TPH bioremediation study, the aromatic compounds increased by 2% while the aliphatic ones decreased by 28.7% (Wu et al. 2017b). PAH and chlorinated organic compounds present in petroleum sludge are very resistant to microbial attack (Arun et al. 2011). The TPH properties that may affect their biodegradation are chemical structure, substitution degree, viscosity, solubility, and concentration (Seo et al. 2009; Jain et al. 2011). Margesin et al. (2007) reported that increasing TPH concentration by 8 times can reduce the TPH removal by 6.75 times. However, decreasing the TPH concentration will not automatically rise up the removal rate. Ramadass et al. (2015) recorded the lowest removal rate in 27.8 g/kg of TPH, and assigned this to the fact that this sample was composed by 70% of recalcitrant hydrocarbons (C_{28}–C_{40}) and only 2 g/kg of bioavailable hydrocarbons. Even if Admon et al. (2001) and Huesemann et al. (2004) recommended a TPH concentration between 0.2 and 5.5% for better microbial activity, many studies have reached more than 90% of TPH removal with a TPH concentration exceeding 160 g/kg (Sood et al. 2010; Adetutu et al. 2015). In petroleum sludge composting the initial TPH concentration plays a role in the treatment effectiveness; as much as the concentration is high the TPH removal is low (Koolivand et al. 2017). However, a relatively low TPH concentration may have the opposite effect by limiting the carbon input required for the growth of microorganisms (Sayara et al. 2010).

Nutrients such as nitrogen (N) and phosphorus (P) limit the TPH biodegradation due to their limited availability (Wu et al. 2017a). Several authors have suggested biostimulation by adding N and P to improve the TPH biodegradation (Wang et al. 2012; Silva-Castro et al. 2013). Wu et al. (2016) recorded 60% of TPH removal in biostimulated treatment with an addition of ammonium sulfate ($(NH_4)_2SO_4$) and dipotassium phosphate (K_2HPO_4), against only 16% of TPH removal for natural attenuation treatment. Roy et al. (2018) reported that addition of N and P simultaneously conduct to 55% of TPH removal, while adding N or P separately recorded respectively, 50 and 46% of TPH removal. Nevertheless, an excessive concentration of nutrients in the medium can inhibit the hydrocarbon biodegradation (Souza et al. 2014).

The temperature plays a vital role in the biodegradation process. It affects the physical properties of the petroleum hydrocarbons and the type of microorganisms responsible for the degradation (Chandra et al. 2013). Gibb et al. (2001) reported that the rise of temperature is proportional to the solubility of the contaminants and induces a high metabolic activity. High temperature increases the solubility of hydrophobic pollutants, decreases the viscosity, improves the diffusion, and transfers long-chain alkanes from solid to liquid phase (Aislabie et al. 2006). At high temperature, the enzymatic kinetic involved in the petroleum sludge biodegradation increases and thus affects positively the TPH removal efficiency (Koolivand et al. 2017). The TPH bioremediation can occur even in extreme cold-climate. As reported by Kim et al. (2018), TPH was removed by 58% in cold seasonal temperatures ranging from −10 to 19 °C, and highlighted that *Arthrobacter*, *Rhodococcus* and *Polaromas* abundance was better throughout the wintry season than in summer.

4.2 Biotic factors

Firstly, microbial diversity and growth may influence positively on the petroleum sludge composting. It has been well reported that a microbial consortium leads to better biodegradation of complex hydrocarbons (Cerqueira et al. 2011; Shen et al. 2015). Ghazali et al. (2004) reported that a consortium constituted by three *Bacillus*, two *Pseudomonas* and one *Micrococcus* strains helped degrading more recalcitrant hydrocarbons than consortium of two *Bacillus* and one *Pseudomonas*. Otherwise, the bioaugmentation in petroleum hydrocarbons studies has revealed more TPH biodegradation against natural attenuation or biostimulation (Bidja Abena et al. 2019; Varjani and Upasani 2019). Moreover, hydrocarbon-degrading bacteria isolated from petroleum sludge compost, have been able to degrade toxic elements such as PAH (Ubani et al. 2016).

Bioavailability of hydrocarbons is such a key factor for biological uptake. For petroleum substrates, biological uptake can be done by direct contact with soluble fraction or by biosurfactant-mediated contact with heavier and insoluble hydrocarbons (Kapellos 2017). Biosurfactants consist of hydrophilic and hydrophobic segments and are classified in glycolipids, phospholipids, lipopeptides, lipoproteins and membranous vesicles (Desai and Banat 1997). These compounds are produced by microorganisms during the stationary phase (Yan et al. 2012), and enhance solubilization, desorption or emulsification of hydrocarbons (Dias et al. 2012; Kavitha et al. 2014). The main role of biosurfactants in petroleum biodegradation is reducing the repulsive force between hydrocarbons and microorganisms, helping mixing and interaction between them (Souza et al. 2014; Hua and Wang 2014). Microorganisms involved in petroleum hydrocarbons bioremediation have been largely reported to have the capacity of producing biosurfactants (Waigi et al. 2015; Varjani and Upasani 2016), which certainly affects the composting process. In plus of lipoprotein biosurfactant, a consortium of *Shewanalla chilikensis*, *Bacillus firmus* and *Halomonas hamiltonii* produced biocatalysts (lipase, catalase and oxidoreductase) which rise the TPH removal at 96% (Suganthi et al. 2018).

4.3 Co-substrates in petroleum sludge composting

Different natures of co-substrates were used in petroleum sludge composting. In general, these co-substrates called bulking agents are applied to increase porosity and oxygen diffusion and to form water-stable aggregates (Zahrim et al. 2015). This for interest to increase the aeration and the microbial activity (Eftoda and Mccartney 2004; Malińska and Zabochnicka-Świątek 2013). To enhance the TPH remediation by composting, researchers seem that they have focused more on the aeration aspect, as most of the studies applied a mixture of petroleum sludge with soil, sand or wood chips. These co-substrates decrease the TPH concentration in the mixtures, increase microorganisms/sludge surface contact and ameliorate the bacterial activity. However, mixing the petroleum sludge with soil can be also in interest to bring an exo/indi-genous microbial consortium. In some case, the soil has been contaminated by hydrocarbons and so the microbial consortium has already been adapted to degrade the targeted contaminants (Ling and Isa 2006). In the same way, other authors have chosen to introduce more extraneous biomass by adding animal manures, while in some studies, we added directly a microbial consortium, which is known as hydrocarbon degrading (Milne et al. 1998; Ayotamuno et al. 2007). As reported in some studies, the carbon of TPH is more assimilated than the one from the co-substrates (Kirchmann and Ewnetu 1998; Koolivand et al. 2013; Asgari et al. 2017). This confirmed that the choice of the co-substrates in petroleum sludge composting is not like the one for the composting of sewage sludge or another kind of biodegradable waste. In the latter one, the choice of the co-substrates is based on the intake of a suitable assimilable carbon for microorganisms. In addition, the main aim of the petroleum sludge composting is to reduce or eliminate the TPH.

In this case, we can understand that composting of petroleum sludge needs a consensus between different parameters. The choice of the bulking agent for suitable aeration, not competing as the main carbon source, and the activation of microbial growth.

4.4 Sludge characteristics and composting system

Petroleum sludges used in composting studies are very diverse. The pH ranged from 5.8 to 8, except for the study of Asgari et al. (2017) which applied an acidic petroleum sludge with a pH of 1.35 (Table 1). These pH values are adequate with the optimum range of pH for the biodegradation of the TPH, and does not constitute an obstacle for the good running of the composting process. This represents an advantage for treating the petroleum sludge by composting process, with no adding of commercial products to maintain the pH in the range between 6.5 and 8.5. However, for acidic petroleum sludges, adding of a co-substrate with high pH can rise the composting mixture pH, as applied by Asgari et al. (2017).

The range diversity of the total organic carbon (TOC) and the total Kjeldahl nitrogen (TKN) of the petroleum sludges was too consequent. As the TOC concentration ranged between 4.9 and 542.44 g/kg, and for the TKN it varied between 0.02 and 16 g/kg (Table 1). This diversity in the TOC and the TKN concentrations is generally due to the initial oil quality and the refining system. This induces a certain vigilance against the composting method, as each type of petroleum sludge must be considered separately. Taking into account the C/N ratio, some type of petroleum sludge has to be supplemented with a nitrogen source and other with a carbon source, for redressing or diminishing the C/N value. It is known that a C/N value between 25 and 30 enhance the biodegradation (Huang et al. 2004; Ma et al. 2016).

The composting process for petroleum sludges was mainly conducted in a semi-closed system (reactor).

Table 2 Composting conditions and physico-chemical changes

References	Ratio	Co-substrate	Composting system and capacity	Composting conditions	Compost mixture			
					Initial		Final	
					pH	C/N	pH	C/N
Ayotamuno et al. (2007)	1:6	Soil	Container 1.75 kg	Addition of *Bacillus* and *Pseudomonas* consortium (1.5×10^{12} UFC/ml)	–	–	7.7	4
Koolivand et al. (2013)	1:8	Immature compost	Container 0.5 kg	PE container, air flow at 10 mL/min and humidity set at 55%	7	20″	7.1	25.1
Koolivand et al. (2017)	1:6	Immature compost	Pile + reactor 2 m^3 & 2 kg	First 6 weeks in pile turned every 5 days, followed by 6 weeks in container with continuous airflow and humidity 50-60%	7.4	20″	6.8	10
Asgari et al. (2017)	1:5	Immature compost	Container 0.5 L	20 mL/min of air flow and moisture 45-65%.	6.2	20″	6.5	26
Marín et al. (2006)	1:3	Shaving wood	Pile 2 m^3	Piles turned every week	7.8	–	7.4	–
Kirchmann and Ewnetu (1998)	1:55	Horse manure	Bins 280 L	Bins were insulated with rockwool (3 cm)	–	–	–	–
Ling and Isa (2006)	0.1:1:0.5*	Soil + grass	Container 5 kg	Containers are set at room temperature (23-25 °C)	7	20″	7.1	–
Kriipsalu et al. (2007)	1:6	Sand	Container 1000 kg	PE container, approximately 7 m^3/min of airflow	8.1	7.14		6.94
Ouyang et al. (2005)	4:1	Straw + sawdust + sand	Pile 350 kg	Mixing and watering every day, with addition of 0.3 g of Rhodococcus strains per 1 kg of sludge	7.7	–	7.8	–
Atagana (2014)	3:1	Poultry manure + sawdust	Container 1 m^3	Compost turned every week and moisture kept at 60–70%	7	4.34	7.2	2.43
Milne et al. (1998)	1:12	Peat-moss	Reactor 0.7 kg	The reactor is kept in temperature-controlled chamber. Peat-moss is enriched by nutrient and oil-degrading bacteria	–	–	–	–
Fountoulakis et al. (2009)	1:3	Green wastes	Container 1 m^3	Container enveloped by rockwool (20 mm). Compost watered occasionally and turned every 1 or 2 weeks	6.6	56.58	6.7	17.5
Ayotamuno et al. (2010)	4:2:1*	Soil + sawdust + poultry manure	Container 49 kg	Plastic container exposed to solar radiation	5.2	565.00	5.8	166
Kriipsalu and Nammari (2010)	1:2:2*	Immature compost + soil	Reactor 21 tons	Reactor insulated by glass wool. Airflow set at 30 m^3/min for 92 days then every 4 h, after day 140 1 h morning and 4 h afternoon	7.9	14.9	–	–

*, sludge:soil:co-substrate; ″, C/N adjusted; PE, polyethylene

This is generally to prevent the exhaust of the volatile compounds. Most of the studies have been applied at laboratory scale (0.5–49 kg) or semi-industrial scale (> 1000 kg), except Kriipsalu and Nammari (2010) who made a composting process at a very large scale (Table 2). The purpose of making compost at the laboratory or semi-industrial scale in the reactor system is the perfect control of the process (tilling, aerating, watering and sampling). It also helped to understand and ameliorate the biodegradation mechanism of TPH by adding nutrients or a microbial consortium and applying a different co-substrate in many ratios. In most of the studies, the ratios of petroleum sludge/co-substrate applied ranged between 1/1 and 1/100 in order to decrease the TPH concentration in the composting mixture. Except for the studies of Ayotamuno et al. (2007, 2010) and Atagana (2014) in which the petroleum sludge was 2 to 6 times the co-substrate. For Ayotamuno et al. (2007, 2010) the reason for applying much more petroleum sludge than the co-substrate can be explained by the low TPH concentration of 69.37 and 98 g/kg. For Atagana (2014) the reason can be the wish to keep a certain TPH concentration in the composting mixture after the successive dilution by applying five consecutive co-substrates.

4.5 Physico-chemical transformation during the composting process

Table 2 represents the physico-chemical parameters at the initial and the final stages of the mixture that indicates the highest TPH removal. In this case, we noted a certain lack of information. Most of the authors were not focalized on the assessment of the physico-chemical parameters during the composting process. This makes the comparison between the studies of petroleum sludge composting really difficult. In addition, the C/N ratio reported in some studies has evolved differently, in some its decrease and in others its increase (Table 2). Generally, the C/N ratio decreases in the composting process due to the assimilation of carbon by microorganisms (El Fels et al. 2014). However, in the petroleum sludge composting it has been reported that the C/N ratio can increase due to the preferential assimilation of carbon from TPH and quick consumption of nitrogen than carbon (Koolivand et al. 2013; Asgari et al. 2017). From this, we can afford to stipulate that in the petroleum sludge composting, the microorganisms can even target the main contaminant (TPH) or the organic matter from the co-substrate. That induces that the choice of the co-substrate, the addition of nutrition (biostimulation) or extraneous microorganism is critical for the rest of the composting process.

In general, the C/N ratio in the composting process of organic wastes is applied as an indicator of the maturity and stability of the final product. A C/N ratio between 10 and 15 designates a good degree of maturity and stability (El Fels et al. 2014). In petroleum sludge composting, all the authors have not extended their discussion on the maturity and stability of the final products. As reported in Table 2 most of the final C/N ratios were out of the range 10 and 15. Thereby, the final products from these studies are not stable and mature, and their storage in nature may cause a certain biological activity resumption.

4.6 TPH degradation during the composting process

In composting studies, the amount of TPH varied between 0.204 and 521.12 g/kg (Table 3). Comparing these studies, or in another term stating that a study is better than another isn't an easy thing. This difference in the initial TPH concentration influence on the TPH removal and the process duration. Less TPH concentration or quantity doesn't automatically mean that the biodegradation will be easy or more effective. Asgari et al. (2017) and Koolivand et al. (2017) observed a better TPH degradation kinetic and total removal in high mixing ratio compared to very low ones. As seen in some studies, the TPH removal rate reached more than 80% in 42 days, while in others it is just around 47% in 84 days (Table 3). However, by referring to the TPH degradation kinetic, the group of studies which haven't received any enhanced treatment expressed less than 0.66%/day of TPH degradation. Except for Vasudevan and Rajaram (2001) and Ouyang et al. (2005) who applied the bioaugmentation and obtained just 0.8%/day of TPH degradation, the other ameliorated treatment recorded more than 1%/day of TPH degradation (Table 3). This can be related to the small amount of the bacterial consortium or also, the thing that this consortium was an extraneous inoculum to the TPH biodegradation. In contrary, the other studies which

Table 3 Removal of TPH during petroleum sludge composting

References	TPH in sludge (g/kg)	Initial TPH in compost (g/kg)	R (%) K (% of TPH degradation/day)	TPH removal in control (%)	Treatment amelioration and time (days)	Comments
Ayotamuno et al. (2007)	69.37	–	R: 84.5 K: 2.01	12.8	Bioaugmentation (42)	The addition of a bacterial consortium (Pseudomonas/Bacillus) in plus of indigenous microbes (from soil) was more effective than the application of one strain, and that explains the high TPH degradation kinetic. The dilution effect was not taken into consideration, as the TPH removal was calculated on the basis of the initial TPH concentration in the sludge not in the initial mixture stage (sludge/soil)
Koolivand et al. (2013)	104.3	–	R: 80.2 K: 1.15	2.7	Biostimulation (70)	The same TPH removal rate was recorded in the first 5 weeks for all composting ratios, then a significant difference was observed until the 10^{th} week. The highest TPH removal rate recorded in the 1:8 ratio is due to the low TPH level and more aeration, which result in a high temperature that increases the enzyme kinetic and the progress of the biodegradation
Koolivand et al. (2017)	131.72	19.75	R: 95.2 K: 1.13	–	Biostimulation (84)	Re-addition of nutrients and microorganisms enhance the TPH removal by more targeting the remaining recalcitrant compounds. Decreasing the mixing ratio more than it need can have an opposite effect on the TPH removal
Asgari et al. (2017)	521.12	85.4	R: 76.2 K: 1.09	1.22	Biostimulation (70)	This study shows that a further increase of the co-substrate influences negatively the efficiency of the petroleum sludge composting by targeting other organic amendments instead of the TPH
Marín et al. (2006)	281	–	R: 59 K: 0.66	32	n.a.t. (90)	Addition of organic fertilizer has not improved the TPH degradation more than just adding a bulking agent
Kirchmann and Ewnetu (1998)	200	17.6	R: 71 K: 0.51	–	n.a.t. (139)	The carbon decrease by just 11%, which indicate that the organic amendments didn't compete with the targeted TPH. Aliphatic TPH was the most biodegraded fraction
Ling and Isa (2006)	–	–	R: 65.6 K: 1.04	45	Biostimulation (63)	Even if it is a study on the bioremediation of oil sludge, no further information on this last was reported. Some anaerobic microorganisms can be a part of the bioremediation process. Supplementing the compost by nutrients lead to change its physico-chemical parameters and thus result in an acclimatization period that can interrupt the TPH removal
Kriipsalu et al. (2007)	130	13	R: 74 K: 0.20	–	n.a.t. (373)	The calculation of the TPH removal rate was based on the TPH mass loss, not the concentration. If it is not at the optimal ratio, the organic amendment can compete with the targeted TPH in the first bioremediation steps

Table 3 continued

References	TPH in sludge (g/kg)	Initial TPH in compost (g/kg)	R (%) K (% of TPH degradation/day)	TPH removal in control (%)	Treatment amelioration and time (days)	Comments
Ouyang et al. (2005)	371.2	121.2	R: 47 K: 0.81	–	Bioaugmentation (58)	The mixture with the highest temperature recorded doesn't reach the highest TPH removal rate. The bioaugmentation by oil-degrading microbes worked much better than that by extraneous ones (manure). The percentage of lighter hydrocarbons (C_{13}-C_{21}) decrease while others from C_{22}-C_{29} increase. The low TPH removal rate in this study compared to other with bioaugmentation can be explained by the high initial level of TPH and the mixing ratio (4:1)
Atagana (2014)	300.59	–	R: 91 K: 0.51	–	n.a.t. (180)	Application of sawdust as a bulking agent was more effective than the woodchips or the hay. All the trials supplemented with manure showed more TPH removal than those without manure. The high initial TPH concentration and mixture ratio (3:1) explain the long treatment duration
Milne et al. (1998)	300	50	R: 55 K: 1.83	–	Bioaugmentation (30)	The sludge concentration is an important thing, it became toxic at a level beyond 250,000 ppm in the compost. It was shown that microorganisms taken from a contaminated refinery soil were far more effective than those from animal manure.
Fountoulakis et al. (2009)	0.204	–	R: 62.1 K: 0.52	23.2	n.a.t. (120)	Authors assume that independently from the limit values for the landfill disposal, the ratio 1:1 even if it recorded the low TPH removal rate, can be much more economical when it comes to the amount of TPH treated
Ayotamuno et al. (2010)	98.03	–	R: 47 K: 0.56	12	n.a.t. (84)	The TPH removal rate is influenced by the initial C/N ratio as it affects microbial growth. This study revealed that proceeding simultaneously with composting and phytoremediation doesn't enhance the treatment
Kriipsalu and Nammari (2010)	160	15	R: 68.7 K: 0.19	–	n.a.t. (370)	In a large scale, the measurements can be skewed, as the temperature, the moisture and the airflow are not equally propagated into the compost and this result in a varied TPH reduction through the mixture
Vasudevan and Rajaram (2001)	240	–	R: 72 K: 0.80	0	Bioaugmentation (90)	In the composting process of petroleum sludge, it is preferable to use a bulking agent rather than inorganic nutrients, it increases the oil bioavailability and also serves as an additional source of energy for microorganisms

–, no information; R, TPH removal rate; K, TPH degradation kinetic; n.a.t., non ameliorated treatment performed the bioaugmentation with an inoculum of hydrocarbon-degrading bacteria and a large amount between 7.4×10^{11} and 1.5×10^{12} CFU/g, have recorded 1.8 and 2%/day of TPH degradation (Table 3). It seems that the bioaugmentation affects more positively the TPH biodegradation and help to achieve quickly a high rate of TPH removal if it is done by inoculating hydrocarbon-degrading bacteria. The biostimulation enhanced also the biodegradation, as the four studies which applied it recorded between 1.04 and 1.15%/day of TPH degradation (Table 3). In addition, TPH consist of a mixture between aliphatic, aromatic, resin and asphaltene hydrocarbons, this chemical diversity can play an essential role in the biodegradation. More aliphatic hydrocarbons in a petroleum sludge than in another will facilitate furthermore the growth of the microbial community, helping to degrade more recalcitrant contaminants (cyclic alkanes, PAH or asphaltenes), and achieving a better TPH degradation result.

The rise of the temperature in petroleum composting studies doesn't seem to be an indicator of the good process progress like for composting of other organic wastes, and in which we can base ourselves to affirm or predict the highest removal rate of TPH. In this case, four studies with the highest temperature peaks haven't exceed 0.66%/day of TPH degradation. In addition to that, the study with the highest peak temperature of 77 °C has reached the less kinetic of TPH degradation (Table 3). It is recognized that high temperature increases the solubility of the TPH and facilitate their uptake by microorganisms, however surpassing an optimal value affects negatively the TPH degradation. Several studies have been focused on the relationship between temperature and hydrocarbons biodegradation. Margesin and Schinner (1997) studied the influence of temperature ranged between 4 and 30 °C on oil degradation by yeast, and found that the oil biodegradation was higher in 20 °C than in 30 °C. In another study, increasing temperature over the optimum has a restrained effect, as Coulon et al. (2005) reported that increasing temperature from 4 to 10 °C had a significant result while increasing from 10 to 20 °C slowed the TPH degradation kinetic. It is also reported that oil-degrading bacteria have a specific preferential temperature. Development of *Pseudoalteromonas, Reinekea, Cycloclasticus* and *Sulfitobacter* is favored at an incubation temperature of 4 °C, while a temperature of 24 °C is more suitable for *Oleibacter, Thalassobius, Phaeobacter,* and *Roseobacter* (Liu et al. 2017). By the way, even if the maximal value of temperature isn't an indicator of a high TPH degradation, it affects it by influencing the microbial activity and also the physico-chemical structure of the TPH (Brakstad et al. 2004).

Some reported studies on petroleum sludge composting have addressed the effect on PAH, and found that PAH can be removed between 60 and 88% (Kirchmann and Ewnetu 1998; Kriipsalu et al. 2007; Fountoulakis et al. 2009; Atagana 2014; Ubani et al. 2016). *Bacillus* has been reported to be the most abundant bacteria in petroleum sludge composting and responsible for most PAH biodegradation (Ubani et al. 2016). Kirchmann and Ewnetu (1998) reported that aromatic hydrocarbons decreased slightly compared to aliphatic. However, all PAH decreased and were under detection limit (< 0.1 mg/kg), except pyrene, chrysene and anthracene. Atagana (2014) reported that lighter PAH were removed easily, whereas heavier ones were recalcitrant without manure addition. This fact was also reported by Kriipsalu et al. (2008a) which recorded an effective reduction of 2–4 ring PAH over 5–6 ring PAH. However, the same authors reported that biodegradation of 5–6 ring PAH can be enhanced by 69% in case of secondary thermophilic phase.

In the degradation of hydrocarbons, the biological activity can be assisted by chemical oxidation or volatilization. In this case, the control test helps to distinguish between the real part of TPH bioremediation and natural oxidation. In petroleum sludge composting, 50% of the studies have applied a control test, but not all of them with the aim to determine the percentage of the volatilization in the TPH removal. Four studies have applied the control test for estimating the volatilization fraction, three of them found a negligible value between 0 and 2.7% while for the other study 23% of volatilization was recorded (Table 3). In the last study, Fountoulakis et al. (2009) acted differently in their abiotic test, they applied it in a drying oven. According to them, direct exposition of the mixture to the heating from the oven with no physical barrier has increased the volatilization; while in reality the high temperatures are reached in the core of the mixture and the outer surface can reduce the volatilization (Fountoulakis et al. 2009). No biocide was applied in the other composting studies. The purpose, in this case, was to compare the treatments applied (bulking agents, bioaugmentation, biostimulation) and sludge biotransformation with no treatment practiced. Appreciable TPH removal rates between 12 and 45% were recorded in these studies (Table 3), considering that petroleum indigenous microorganisms have a great adaptation capacity.

In general, when it comes to talking about composting, we think about the humus which is the product from the natural organic matter biotransformation called the humification process. More the compost is rich in humic substances, more is benefic for soil and plants growth (El Fels et al. 2014). Reviewing the studies of petroleum sludge composting no one addressed the subject of the humification process. A study that focused on the bioremediation of waste mazut, had revealed the formation of the humic substances during the biodegradation of petroleum hydrocarbons (Jednak et al. 2017). Hafidi et al. (2008) and El Fels et al. (2016) have stipulated that the decrease of hydrocarbons in composting process was not at all due to biodegradation, as some of them can be assimilated into non-accessible sites or can form another type of hydrocarbons. A recent study on the distribution of aliphatic and aromatic hydrocarbons in soil found that these compounds can be bound to humic substances (Zhang and Fan 2016; Zhang et al. 2018). They reported that in soils 46% of the aliphatic hydrocarbons were HA-bound (Humic Acids), and 36% of the aromatic hydrocarbons were Humin-bound and FA-bound (Fulvic Acids). This interaction between hydrocarbons and humic substances is an essential part of the bioremediation process, and their assessment in petroleum sludge composting is important.

5 Conclusion

Bioremediation of petroleum sludge by composting has been widely used to reduce TPH quantity and minimize their impact. This biological process achieved between 47 and 95% of TPH removal during treatment from 30 to 373 days. This review underlined the importance of bioaugmentation and biostimulation as an effective way to ameliorate the composting process. In the case of bioaugmentation, it is recommended to apply a microbial consortium instead of one microorganism species, and even more indigenous than extraneous ones. In regards to initial compost parameters, the authors suggested adjusting the C/N ratio to 20 as biostimulation to the microbial community to enhance TPH removal. To our knowledge, a C/N ratio between 25 and 30 is more required, as in the compost not only the TPH are targeted. The diversity of the petroleum sludges has led to better assess the response of different TPH concentrations to biodegradation. Lack of information in the majority of studies on other physico-chemical parameters than just TPH restrained the full organic matrix assessment. Nevertheless, the majority of these studies were realized in lab-scale under controlled conditions, suggesting that pilot-scale studies are recommended to fully assess the composting process under the influence of both biotic and abiotic factors. As no one of the studies on petroleum sludges composting has addressed interest in evaluating the stability and the maturity degree of the final product. We think that this part is required to identify the natural obstacles that interrupt the TPH removal process, and also the plan for the product from the petroleum sludge composting. All of this, in order to overcome the negative impact of persistent hydrocarbons on soil and environment.

6 Scope of future researches

With the increasing energy demand and the unsatisfying of the green energies, the petroleum industry still has a bright future. This may engender more hazardous wastes which can contaminate the soil, the water and the air. The consequences on the environment and by the way human health can be disastrous. The researches on the treatment of petroleum wastes have taken a great interest and still in progress. Although, studies on the mechanisms, the persistence and the transfer of hydrocarbons are highly recommended to better assess their impact on the environment. Indeed, more research areas are needed:

- Physico-chemical parameters (e.g. C/N ratio) that help better biodegradation of both targeted TPH and also all organic matrix.
- Identification of microbial consortia capable to degrade the TPH and other organic matters at the same time to reduce the process duration.

- Isolation of microorganisms able to assimilate recalcitrant TPH compounds and their introduction at the final stage of composting.
- Discover the formation of humic substances (e.g. humic and fulvic acids) during the petroleum sludge composting, and their potential interactions with the TPH.
- Highlighting the quality of the final compost in term of maturity and stability, which can eventually give an idea about the whole biotransformation leading to better find the right disposal into the environment.

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