




# A critical literature review on biosolids to biochar: an alternative biosolids management option

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**Abstract** The biosolids management is becoming an increased concern for the wastewater sector in recent times due to production of large volume of biosolids, their higher processing costs and the presence of emerging contaminants. The pyrolysis of biosolids is gaining significant interest in the industry sector as well as research community over the last decade due to its ability to reduce biosolids volume, produce high-value biochar product and minimise the risk associated with contaminants. This paper aims to critically review the literature on biosolids management techniques and their current challenges, biosolids characteristics and its suitability for pyrolysis, pyrolysis product characterisation from different reactor designs and biochar application as a soil amendment, adsorbent and catalyst. The efforts have also been made to critically summarise studies on the process modelling

activities and techno-economic assessments including some key pilot-scale demonstrations of recent time. The review concludes that biosolids to biochar can be an effective alternative to biosolids management; however, its commercial viability is limited in the current scenario. In the end, efforts have been made to highlight current challenges including research gaps and future perspectives in improving its commercial viability.

**Keywords** Biosolids · Sewage sludge · Wastewater · Biochar · Thermochemical conversion · Pyrolysis

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## 1 Biosolids management and its challenges

Biosolids are the solid by-product formed after the treatment of sewage sludge. Biosolids usually contain solids between 15 and 90% (Fonts et al. 2012; Furness et al. 2000; Kemmer et al. 1971; Patel et al. 2019c). Biosolids are composed of organic as well as inorganic materials including ash and minerals. Biosolids may contain a good amount of macronutrients, such as nitrogen, phosphorus, potassium, calcium, magnesium and sulphur as well as micronutrients such as copper, zinc, iron, boron, molybdenum and manganese (Khan et al. 2013b; Patel et al. 2018, 2019a).

Over the past two decades, biosolids production has increased worldwide as a result of population growth, new wastewater plants implementation and collection systems and strict regulations of environmental legislation (Arulrajah et al. 2011; O'Kelly 2005). The biosolids production in Australia reached to 329,000 dry tonnes in 2017, which was 6% more than the biosolids production (310,000 dry tonnes) noted in 2015 (ANZBP 2017). Majority of the biosolids in Australia are beneficiated to agricultural land due to high nutrient and organic carbon values. It is estimated that the municipal wastewater produced globally contains enough nutrients to replace 25% of the nitrogen and 15% of the phosphorus currently used to fertilise the agricultural land in the form of synthetic fertilisers (Andersson 2016). The other beneficial applications of biosolids are land rehabilitation, landscaping (compost) and incineration (Agrafioti et al. 2013; ANZBP 2017; Faria et al. 2018). Non-beneficiated biosolids are either stockpiled, landfilled or discharged in the ocean. In Australia, ~ 75% of the biosolids are used for agricultural land while 11% are used for land rehabilitation, and 14% are either landscaped in the form of compost, stockpiled, applied to landfills or discharged to the ocean (ANZBP 2017). Incineration is not considered as favourable in Australia due to the potential risk of harmful gas emissions (Barry et al. 2006; Darvodelsky and Bridle 2012). Biosolids production in United States in 2019 was ~ 4.27 million dry metric tonnes. In United States, ~ 51% of the biosolids are used for land application, 22% are used for landfilling, 16% are used for incineration, 10% are used under other management practices and 1% is used as surface disposal.

Biosolids can be used as wet or dry. Based on their wetness, biosolids are also referred to as wet or dried

biosolids. There are total of four types of biosolids based on their post-treatment method. Their information is provided below (Agency 1999).

1. Mainstream biosolids (i.e. biosolids obtained after dewatering, drying and/or stockpiling)
2. Pelletised biosolids
3. Lime amended biosolids
4. Composted biosolids

Mainstream biosolids can be further categorised into five types as below based on their origin in the Waste Water treatment plant (WWTP) (Agency 1999).

1. Non-digested biosolids (i.e. primary or activated)
2. Digested biosolids (obtained after digestion)
3. Lagoon biosolids (obtained after digestion in lagoon based treatment plants)
4. Dried biosolids (obtained after centrifuge, solar dryer and/or drying pan)
5. Stockpiled biosolids (obtained from stockpile)

Biosolids are generally classified according to their treatment (microbiological) and contamination grade. Biosolids are classified differently based on different State, Territory or National Guidelines. Environmental Protection Agencies (EPAs) are responsible for the development of guidelines for biosolids' application (McLaughlin et al. 2007). Different Australian State guidelines have specified three to four treatment grades and two to three contaminant grades. The overall combined grade (treatment and contaminant) helps in identifying where any particular biosolids can be applied (McLaughlin et al. 2007).

### 1.1 Presence of contaminants in biosolids

The increasing use of new chemicals in day to day life has caused concerns of them being ended up in biosolids and further contaminating soil and groundwater via biosolids' land application (Choudri et al. 2018; Dumontet et al. 2001; Haynes et al. 2009; Lu et al. 2012b; Pritchard et al. 2010; Scher et al. 2018). Particularly, the presence of micro-plastics, pharmaceuticals, pesticides, and per- and poly-fluoroalkyl substances (PFAS) (Choudri et al. 2018; Dorrance et al. 2017; Dumontet et al. 2001; Haynes et al. 2009; Pritchard et al. 2010; Scher et al. 2018) in biosolids cause a major concern for its land application in recent times. This is due to the fact that uptake of these

emerging contaminants by vegetation might cause human health risks and possible contamination in the food-supply chain.

### 1.2 Drawbacks of existing management options

Current biosolids management options have some shortcomings. For example, stockpiling of biosolids was considered as a favourable option by several water utilities in the past with large portion of free land availability. However, this is not recommended by EPA as a long-term option due to potential contamination, health risks and increasing production of biosolids (Agency 1999). Incineration, despite offering significant advantages such as low cost, high energy recovery and potential destruction of contaminants and pathogens; may emit harmful substances such as acid gases, dioxins, particulate matter and NO<sub>x</sub> (Chen and Yan 2012; Hjelmar 1996; Marani et al. 2003; Roy et al. 2011). Therefore, it is also not considered as a long-term option for waste management in Australia and many countries (Barry et al. 2006). Landfilling was a low-cost option for managing all types of wastes including biosolids in the past. However, increasing risk of contamination of groundwater in long-term, rise in the tipping fees and shortage in landfilling sites have made this option less favourable in recent times (AWA 2012; Barry et al. 2006; Haynes et al. 2009). Biosolids are rich in decomposable organic matter and they exacerbate CH<sub>4</sub>-rich greenhouse gases when they are landfilled or stockpiled (Lewis and Gattie 2002). They also generate significant CO<sub>2</sub> and other toxic gas emissions when incinerated. Therefore, under the current climate change scenario, landfilling, long-term stockpiling and incineration of biosolids are no longer acceptable (Agency 1999; Lu et al. 2012b). Composting of biosolids may be considered as one of the most favourable biosolids' management options. However, there is only a limited capacity of biosolids that can be used in this process (AWA. 2012; Barry et al. 2006).

### 1.3 Decentralised production of biosolids

Wide distribution of wastewater treatment plants poses the biggest challenge in biosolids management and transport. In Australia alone, there are 1200 wastewater treatment plants (Hill et al. 2012). These numbers for the United States are roughly 15,000

(CSS 2019). Biosolids, as described earlier, can have a very large amount of moisture. Moreover, the availability of the agricultural land in close proximity to the wastewater treatment plant is not always the case. Therefore, the biosolids land application may incur huge transportation costs and associated emissions (Barry et al. 2006; Pritchard et al. 2010). Average biosolids management costs, including monitoring and transporting in Australia alone may vary from \$120 to \$280/dry tonne (AWA 2012).

## 2 Pyrolysis: an alternative biosolids management option

Pyrolysis is gaining immense interest in recent times for biosolids management (Fonts et al. 2012). The principle goal of pyrolysis is to reduce the volume of biosolids and produce energy or value-added products from the organic fraction of the waste while having minimal environmental impacts. It has the potential to partially/completely destruct contaminants such as pharmaceuticals, micro-plastics and PFAS while may be able to immobilise heavy metals in the char matrix (Bioforcetech 2019a; GHD 2019; Patel et al. 2019b; Surti 2019).

Pyrolysis of biosolids produces three different products, such as biochar, pyrolysis-gas and pyrolysis-oil. Pyrolysis occurs in an oxygen-free environment (Fonts et al. 2012; Patel et al. 2019b). It is an endothermic process in contrast to other processes, like incineration or gasification, which are either exothermic or auto-thermal. Pyrolysis can reduce biosolids' volume up to 70%, stabilises organic content and produces fuel and valuable liquid chemicals (Fonts et al. 2012; Inganzo et al. 2002). Furthermore, pyrolysis of biosolids enables heavy metals to be encapsulated in the pyrolytic char, which are more resistant to leaching than those from the ash obtained through the gasification/incineration of biosolids (Caballero et al. 1997; Conesa et al. 1998; Fonts et al. 2012; Furness et al. 2000). Gas products from pyrolysis are low in NO<sub>x</sub>, and SO<sub>x</sub> as well as organic pollutants which makes pyrolysis process environmentally friendly. Moreover, it sequesters carbon in biochar, decreases greenhouse gas emissions and contributes to a cleaner environment (Fonts et al. 2012; Tripathi et al. 2016; Wang et al. 2012).

### 3 Novelty/originality of the current review

Pyrolysis of biosolids to produce biochar has been extensively studied in the last decade. Also, there are some recent review papers published on this topic (Callegari and Capodaglio 2018; Li et al. 2019d; Liu et al. 2018). For example, Callegari and Capodaglio's (Callegari and Capodaglio 2018) review on sludge pyrolysis mainly focused on identifying relationships between char, bio-oil, and syngas fractions in different process operating conditions, general char properties, and possible beneficial uses. Liu et al. (2018) focused on comparing the characteristics of biosolids' biochar to biomass' biochar and reviewed specific applications of biosolids' biochar. Paz-Ferreiro et al. (2018) critically reviewed options for the management of biosolids with a focus on pyrolysis and the application of the biosolids' biochar into the soil. In terms of originality and novelty, this paper for the first-time critically reviews and compare biosolids pyrolysis research carried out in lab- to bench and pilot-scale reactor designs (i.e. TGA, fixed bed, fluidised bed). Furthermore, this review is focused on summarising some advanced applications of biosolids' biochar as adsorbent and catalyst, energy storage and energy generation material in addition to soil amendment. Efforts are also made to critically review the work performed on process modelling and development, techno-economic analysis and large scale demonstrations on biosolids pyrolysis. Finally, current challenges, research gaps and future perspectives providing further directions to commercialise pyrolysis for converting biosolids to biochar are highlighted.

### 4 Pyrolysis process types and product distribution

In the pyrolysis process, biosolids are thermally decomposed without being exposed to oxygen environment where multiple reactions occur simultaneously (Fonts et al. 2012). The pyrolysis mainly consists of two stages of decomposition reactions called primary and secondary pyrolysis (Fonts et al. 2008a, b). Biosolids components are broken and devolatilised in the primary pyrolysis by heat effect into its main constituents. In the first stage, various groups of hydroxyl, carbonyl and carboxyl are formed. The devolatilisation process of biosolids consists of dehydration, dehydrogenation and decarboxylation

(Fonts et al. 2008a, b, 2012). Once the primary reactions are completed, the secondary pyrolysis process begins, and this is the critical step of pyrolysis process where cracking of large molecules takes place transforming biosolids into biochar or gases, such as CO, CH<sub>4</sub>, CO<sub>2</sub> and other gases. Some condensable volatiles components are produced and they are known as pyrolysis-oil. This secondary pyrolysis cracking is often called as thermal cracking or catalytic cracking, depending on the conditions of pyrolysis and catalysts used (Fonts et al. 2008a, b, 2012; Liu et al. 2017; Patel et al. 2019b).

Pyrolysis processes are mainly divided into slow and fast pyrolysis. The operating conditions such as reaction temperature, heating rate, particle size and residence time are different in each of these pyrolysis processes, which are summarised in Table 1.

#### 4.1 Slow pyrolysis

Slow pyrolysis has been used for biochar production for a long time. In slow pyrolysis, biosolids are pyrolysed at a temperature between 300 and 900 °C (Fonts et al. 2012; Manara and Zabaniotou 2012). The heating rate in the slow pyrolysis is generally kept between 0.1 and 1 °C/s and residence time vary from an hour to days, as shown in Table 1. Slow pyrolysis promotes the formation of biochar, but nearly an equal amount of pyrolysis-oil and gaseous products are also formed during the process (Fonts et al. 2012; Manara and Zabaniotou 2012; Patel et al. 2019b). Lower heating rates and longer residence times provide the

**Table 1** A summary of pyrolysis processes, operating conditions and their product distribution

Process	Slow pyrolysis	Fast pyrolysis
Reaction temperature (°C)	300–900	300–1000
Heating rate (°C/s)	0.1–1	10–1000
Residence time	Hour-days	< 2 s
Particle size (mm)	5–50	< 1
Feed stock size	Coarse	Finely ground
Py-oil yield (%)	20–50	60–75
Biochar yield (%)	25–35	10–25
Py-Gas yield (%)	20–50	10–30

(Bahng et al. 2009; Demirbas and Arin 2002; Fonts et al. 2012; Homagain et al. 2014; Li et al. 2013; Manara and Zabaniotou 2012; Yan et al. 2005)

appropriate atmosphere that is favourable for the secondary reactions. This eventually leads to increased biochar formation (Ábrego et al. 2009; Fonts et al. 2012; Patel et al. 2019b). In addition, longer residence time also enables the increase in vapour yield during the secondary reaction (Fonts et al. 2012; Patel et al. 2019b).

#### 4.2 Fast pyrolysis

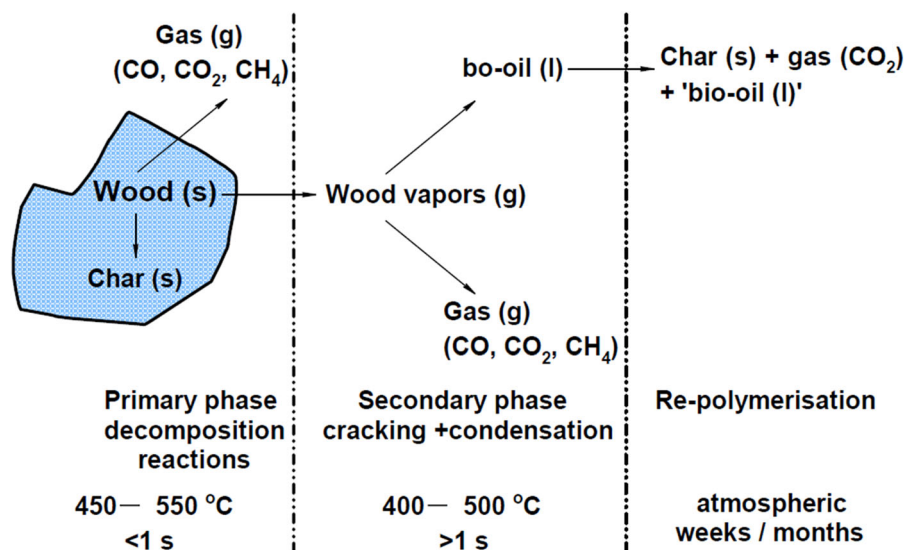
Fast pyrolysis is mainly used for pyrolysis-oil production. For fast pyrolysis, biosolids are generally heated up to 300–1000 °C with a high heating rate that ranges from 10 to 1000 °C/s and a short residence time, which is less than two seconds (Alvarez et al. 2015a). The oil produced from fast pyrolysis has a heating value of nearly half of heavy fuel oil, has higher oxygen and nitrogen content which makes it necessary to upgrade before using it in vehicles (Alvarez et al. 2015a). Fast pyrolysis can be divided into three stages. The first is primary pyrolysis, where the decomposition reaction is followed by secondary cracking and re-polymerisation, as shown in Fig. 1 developed for woody biomass.

#### 4.3 Pyrolysis products

Pyrolysis of biosolids produces solid (i.e. biochar), gas (i.e. pyrolysis gas) and liquid (i.e. pyrolysis oil)

products. Their average yields are summarised in Table 1.

The biochar is a carbonaceous solid product that is mostly used as an adsorbent, fuel, catalyst and/or soil amendment (Agrafioti et al. 2013; Fonts et al. 2012; Hossain et al. 2011; Manara and Zabaniotou 2012). The pyrolysis-gas consisting of CH<sub>4</sub>, CO<sub>2</sub>, H<sub>2</sub> and CO, as well as few other low-molecular gases, can be used for heat generation, in a gas engine, after proper cleaning, to produce electricity or can be converted into fuels via further processing such Fischer–Tropsch synthesis (Ail and Dasappa 2016; Conesa et al. 1998; Davis and Occelli 2007; Inguanzo et al. 2002; McNamara et al. 2016; Wright et al. 2008). Pyrolysis-oil, an extremely complex mixture of multiple small and long-chain hydrocarbons, is the third product of pyrolysis. Pyrolytic-oils comprising of four major chemical groups are formed after cooling in the condenser: (i) water, (ii) oxygen-containing compounds (e.g., alcohols, ketones, acids, phenols, sugars) (iii) aliphatic and aromatic hydrocarbons, and (iv) nitrogen containing compounds (e.g. pyrazine, pyridine and amines) (Alvarez et al. 2016; Fonts et al. 2012; Kim and Parker 2008; Manara and Zabaniotou 2012; Shen and Zhang 2003). Pyrolysis-oil is typically dark brown in colour with a high viscosity. It is corrosive in nature and has high water, sulphur and nitrogen content. It requires upgrading prior using as a fuel (Kim and Parker 2008; Shen and Zhang 2003; Xie



**Fig. 1** Representation of the reaction paths for fast pyrolysis of wood (Venderbosch and Prins 2010)



et al. 2014). In addition, pyrolysis-oil is also found to be chemically unstable since they consist of compounds containing reactive oxygen. Therefore, it needs proper storage or should be immediately used. The main use of pyrolysis-oil without upgradation could be in boilers to generate steam or heat (Fonts et al. 2012; Shen and Zhang 2003). The properties of pyrolysis-oil, such as density, miscibility and viscosity can be controlled by selecting pyrolysis conditions (Fonts et al. 2012; Manara and Zabaniotou 2012; McNamara et al. 2016). Of the major products produced from pyrolysis, biochar offers significant promises. Pyrolysis-oil can be used as a low-grade liquid fuel in boilers or require further upgradation for it to be used in transport sector.

## 5 Biosolids characteristics and its suitability for pyrolysis

The proximate and ultimate analysis is an important measure, especially in the assessment of the properties of the material for it to be used in the thermal conversion processes.

### 5.1 Proximate and ultimate analysis

Proximate analysis is the simplest and frequently used analytical technique. The proximate analysis provides information related to (i) moisture, (ii) volatile matter, (iii) fixed carbon and (iv) ash—generally referred to as inorganic residues remain after burning. The ultimate analysis provides more extensive information than proximate analysis. In this case, the material is characterised by its carbon (C), hydrogen (H), oxygen (O), nitrogen (N) and sulphur (S) contents. The ultimate analysis is also used to determine the feedstock's heating value.

The summary of the proximate analysis of the various biosolids is presented in Table 2. On average, the contents of C, O, H, N and S in biosolids are 33.8%, 25.4%, 5.4%, 4.8% and 1.2%, respectively. The nitrogen and sulphur contents in biosolids generally vary between 2 to 10 wt% and 0.5 to 2 wt%. Biosolids seem to have higher oxygen content ranging between 15 and 50 wt%. According to the Table 2, volatile matter in biosolids can vary between 46 and 75 wt% while ash can vary between 23 and 45%. The biosolids summarised in Table 2 present the average

ash content of 36.3%. Usually biosolids contain higher ash content than biomass such as wood and organic residues (Li et al. 2019a). This higher ash content in biosolids can promote higher yield and greater accumulation of mineral nutrients in biosolids biochar. Heavy metals in biosolids might be beneficial in making biochar catalyst. The heating values in biosolids vary between 12 to 22 MJ/kg which were estimated based on equation proposed by Channiwala and Parikh (Channiwala and Parikh 2002). These heating values suggest that biosolids if dried reasonably (up to 50–60% moisture) can be pyrolysed auto-thermally without the need of any external energy (Patel et al. 2019c).

### 5.2 Biosolids composition and thermal analysis

Li et al. (2015) observed that organic content in biosolids is mainly composed of protein (24–42%), carbohydrate (7–18%) and lipid (1–14%). Chen et al. (2007) also reported similar findings for biosolids with 61% protein, 11% carbohydrate, less than 1% lipid and remaining 27% to be unknown constituents.

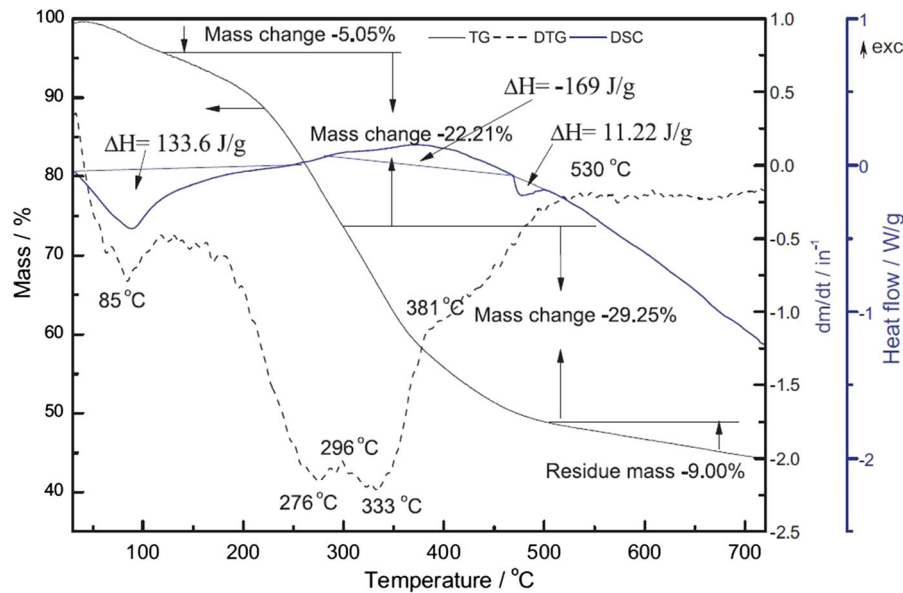
Thermogravimetric Analysis (TGA) is a standard thermal analysis method used by the researchers where mass changes are measured at a controlled increase in temperature. This technique is particularly useful for understanding the thermochemical conversion behaviour of polymeric materials and organic solids. TGA data are generally used for deriving reaction mechanisms, estimating kinetic parameters and determining inorganic (e.g. ash) and organic content in the sample (Urych and Smolinski 2016; Wang et al. 2018; Xu et al. 2017; Zhang et al. 2014b). Several researchers have studied the behaviour of biosolids during pyrolysis using the thermogravimetric technique (Alvarez et al. 2015a, 2015b; Font et al. 2001; Gao et al. 2014; Magdziarz and Werle 2014; Nowicki and Ledakowicz 2014; Shao et al. 2007).

TGA, DTG and DSC curves of biosolids sample reported by Gao et al. (2014) are highlighted in Fig. 2. The initial 5% of weight loss below 120 °C is mainly due to the evaporation of water. The second stage of weight loss, which is approximately 65% of the total weight, occurs in the range of 130–500 °C. In this temperature range, the components such as proteins and carboxyl groups char yield from 46 are decomposed. In the temperature range of 500–700 °C, generally referred to as the third stage, the decomposition of inorganic

**Table 2** Ultimate and proximate analyses of biosolids

Biosolids type	Collection point	Ultimate analysis (wt%)					Proximate analysis (wt%)				Heating value (MJ/kg)	References
		C	H	O	N	S	M	VM	FC	Ash		
Dried biosolids	After dewatering and drying using belt press and solar dryer shed	30.9	4.2	30.9	4.6	1.3	13.3	50.3	9.1	27.3	12.0	Patel et al. (2019a)
Dried biosolids	Unknown	26.5	4.6	19.4	4.3	0.6	–	48.8	6.6	44.6	11.7	Wang et al. (2018)
Air dried biosolids	After digestion	26.6	4.1	–	3.3	1.1	–	57.0	–	43.0	13.3	Hernández et al. (2017)
Dried biosolids	Unknown	32.8	10.2	24.5	5.4	1.7	–	55.8	10.5	33.7	20.3	Xu et al. (2017)
Dried biosolids	After stockpiling	25.3	5.3	15.8	4.1	1.6	10.5	45.9	7.9	35.6	12.8	Urych and Smolinski (2016)
Dried biosolids	Unknown	31.8	4.7	20.5	4.9	1.7	5.3	51.0	–	36.5	13.8	Nyakuma et al. (2016)
Dried biosolids	Unknown	51.7	6.8	30.8	9.8	0.8	6.8	50.4	5.9	36.8	22.0	Liu et al. (2015)
Raw biosolids	After dewatering via centrifuge	30.2	4.6	17.5	5.9	0.9	–	55.8	3.23	39.3	13.3	Zhang et al. (2014b)
Dried biosolids	Unknown	33.6	5.0	–	5.6	1.2	5.7	46.0	5.7	42.6	16.8	Xiaohua and Jiancheng (2012)
Activated biosolids	After activated sludge plant	40.5	5.5	47.9	4.1	1.9	6.1	54.7	3.8	35.4	15.0	Zhai et al. (2011)
Non-digested biosolids	After primary treatment	40.3	5.4	14.9	3.9	–	5.9	54.9	3.7	35.5	18.1	Ji et al. (2010)
Wet activated biosolids	After activated sludge plant	33.4	5.2	26.2	4.2	1.5	–	62.6	7.9	29.5	14.5	Gašparović et al. (2011)
Dried biosolids	Unknown	33	4.4	36.7	2.0	0.5	–	75.3	1.3	23.4	12.4	Shao et al. (2007)
Average		33.8	5.4	25.4	4.8	1.22	6.7	54.8	5.6	36.3	15.1	
Standard deviation		7.4	1.6	10.5	1.9	0.5	1.9	8.1	2.7	5.9	3.15	

M, moisture, VM, volatile matter, FC, fixed carbon



**Fig. 2** TGA, DTG and DSC curves for thermal analysis of biosolids (Gao et al. 2014)

materials occurs. Similar findings are also reported by Alvarez et al. (2015a, b). They reported stages of pyrolysis slightly differently than Gao et al. (2014). Alvarez et al. (2015a, b) concluded that pyrolysis of biosolids occurs mainly in three stages: (1) the first stage of thermal decomposition is carbohydrate pyrolysis that occurs at around 255 °C, after moisture evaporation at 100–200 °C; (2) the second stage is around 300 °C, which is linked to lipid breakdown and (3) third stage is related to protein and lignin breakdown between 360 °C and 525 °C. Font et al. (2001) suggested that decomposition of dead organisms and biodegradable organic matter may occur between 200 and 300 °C and organic polymers present in the biosolids may decompose in the temperature range of 300–450 °C. Patel et al. (2018) found that pyrolysis of Australian biosolids can be divided mainly into three stages such as: (1) dehydration, where around 12–13% mass is lost due to evaporation of water molecule from room temperature to 180 °C, (2) the second stage is devolatilisation stage in which 30% mass is lost between 180–350 °C. They observed two peaks in DTG profile in this temperature range and concluded that the first peak indicates the decomposition of the easily biodegradable carbohydrate-rich volatile fraction of organic matter and dead microorganisms (temperature range from 190 to 300 °C) and the second peak shows the decomposition of organic polymer fractions rich in lipids (temperature

up to 350 °C) and (3) third stage occurs between 360 and 900 °C, in which protein, lignin and polymers are decomposed.

Biosolids composition and understanding on its pyrolysis behaviour via thermal analysis is extremely useful in designing the pyrolysis process. From the published literature on composition and thermal analysis, it can be suggested that higher carbohydrate, lipid and protein contents over lignin may make biosolids a suitable candidate for low temperature pyrolysis. However, higher inorganic content (i.e. ash) may require high temperatures for biochar pores to be fully developed.

## 6 Effect of process parameters (i.e. reaction conditions) in different reactor designs

Reactor designs and reaction conditions in the pyrolysis process are extremely important. The product distribution and their quality in pyrolysis are primarily dependent on the process parameters such as temperature, heating rate and residence time. Literature has reported several experimental studies using TGA and different reactor designs such as fixed bed and fluidised bed reactors to understand the effect of process parameters on biosolids pyrolysis performance (Fonts et al. 2008a, b, 2009; Gao et al. 2014;



Kan et al. 2016; Kim and Parker 2008; Lu et al. 1995, 2013; Park et al. 2008; Patel et al. 2019a, b, 2018; Shao et al. 2010; Shen and Zhang 2003; Xie et al. 2014). TGA represents the highly controlled intrinsic environment. However, it must be noted that TGA use very little sample (in milligram) and operates at much lower heating rates than a large scale pyrolyser. Therefore, studies on lab and bench-scale fixed bed and fluidised bed rigs with higher amount of sample and desired heating rate are important to derive the information on physico-chemical transformations (including morphological changes) that may occur in a real pyrolysis environment. It should be noted that fixed bed and fluidised bed lab- and bench-scale rigs might be suffering from poor gas–solid contact, mainly due to diffusional barrier in the case of fixed bed while poor fluidisation in the case of fluidised bed. Therefore efforts should have been made to overcome them. In this section we have summarised and compared the literature related to slow pyrolysis of biosolids mainly in TGA, fixed bed and fluidised bed rigs.

## 6.1 TGA study

### 6.1.1 Effect of heating rate

In biosolids pyrolysis, heating rates play a significant role as the rate of heat affects the composition and nature of the final product. Generally, standard TGAs can only perform heating rates up to 10–50 °C/min, which can mimic only slow pyrolysis conditions. Non-flexibility of TGAs in changing the heating rates make them less favourable to study the effect of heating rate in pyrolysis studies. Therefore, limited literature is reported on this topic. The effect of heating rates (10, 40, 70, and 100 °C/min) in gas evolution, oil composition and functional groups of the pyrolytic biochar at a temperature of 500 °C is studied using TGA with micro-GC for online gas analysis by Kan et al. (2016). Dried biosolids after digestion obtained from Sydney Water were used in this study. The results indicated that at 300 °C and below, carbon dioxide is the major gas that is evolved, and its evolution rate increases steeply with increasing heating rate. Carbon monoxide and hydrogen formation also increase with the increase in heating rate, but carbon monoxide evolution becomes significant at temperatures above 600 °C. The investigated heating rates (10 and

100 °C/min) had no clear effect on the functional groups in the produced biochar and the composition of the pyrolysis oil. Xiaohua and Jiancheng (2012) found that the heating rate has a little effect on the activation energy and reaction rate in biosolids pyrolysis.

### 6.1.2 Effect of catalyst

Catalysts can reduce the activation energy as well as alter the distribution of pyrolysis products in solid, liquid and gaseous phases. The pyrolysis catalysts can be divided into mainly two groups: (i) primary catalysts, which are mixed with the biosolids before pyrolysis and (ii) secondary catalysts, which are not mixed with biosolids but are located next to the main pyrolysis reactor (Liu et al. 2017). For TGA study, catalysts are generally mixed with biosolids prior to experiments. Shao et al. (2010) performed TGA experiments on dried biosolids to evaluate the effect of catalysts on biochar yield in slow pyrolysis at a heating rate of 10 °C/min. The catalysts used in their study were Fe<sub>2</sub>O<sub>3</sub>, ZnO, Al<sub>2</sub>O<sub>3</sub>, CaO, and TiO<sub>2</sub>. They observed that Al<sub>2</sub>O<sub>3</sub>, CaO and TiO<sub>2</sub> encouraged organic matter degradation, which resulted in lower biochar amounts. In contrast, Fe<sub>2</sub>O<sub>3</sub> and ZnO inhibited organic matter decomposition in demineralized sludge samples and resulted in increased biochar production. The TGA study reported by Kim and Parker (2008) and Xie et al. (2014) found a decrease in the biochar yield with the use of zeolites and HZSM-5. In contrast, biochar yield was found to increase with CaO as a catalysts in their study. It was suggested that the increase in the char yield with CaO might be attributed to the potential capture of CO<sub>2</sub> by CaO in the lower pyrolysis temperature. Based on the literature available on biomass pyrolysis, it can be noted that the acidic catalyst generally increases the production of pyrolytic biochar and reduces oil yield while biochar yield increases in the presence of the basic catalysts (Stefanidis et al. 2011; Wang et al. 2006; Zhang et al. 2009). Patel et al. (2019a) conducted TGA-FTIR study in the presence of minerals such as zeolite, lime and one metal oxide based synthetic catalyst (5% Co/Al<sub>2</sub>O<sub>3</sub>). They observed that rate of devolatilisation (i.e. decrease in biochar yield) can be ordered as follows: 5% Co/Al<sub>2</sub>O<sub>3</sub> > zeolite > lime from TGA study. From FTIR data they found that the catalytic activity of 5% Co/Al<sub>2</sub>O<sub>3</sub> is higher compared to minerals for the production of light gases especially

**Table 3** Yield of biochar produced from the fixed bed pyrolysis of biosolids at different temperatures

Biosolids type	Location	Moisture (%)	Biochar yield (wt%)				References
			300 °C	400 °C	500 °C	600 °C	
Wet biosolids	Before digestion	83	12	10	10	9	Lu et al. (2013)
		79	15	14	13	13	
		85	10	8	8	7	
Dried biosolids	Primary sludge	7	72.3	63.7	57.9	–	Hossain et al. (2011)
Dried biosolids	After digestion, dewatering and drying	–	78	65	55	–	Kim and Parker (2008)

**Table 4** BET surface area (m<sup>2</sup>/g) of biochar produced from fast and slow pyrolysis of biosolids in fixed bed experiments at various temperatures

Temperature (°C)	BET surface area (S <sub>BET</sub> )	
	Fast pyrolysis (Chen et al. 2014)	Slow pyrolysis (Ábrego et al. 2009)
600	20.3	10.2
700	32.2	50.1
800	48.5	91.0
900	67.6	122.0

the concentration of CO<sub>2</sub> because at lower pyrolysis temperatures CO<sub>2</sub> capture in these minerals.

## 6.2 Fixed bed reactor

### 6.2.1 Effect of temperature

Pyrolysis temperature is the main parameter affecting the biochar yield. Fixed bed reactor offers great flexibility in terms of process operation and changing process parameters. Therefore, many studies are reported with fixed bed pyrolysis in the literature. Low pyrolysis temperature is generally suitable for achieving higher biochar yield. Heavy hydrocarbons from biosolids are cracked thermally due to secondary reactions at higher temperatures, resulting in increased oil and gas yields and decreased biochar yield. The effect of temperature on the biochar yield with different types of biosolids during the slow pyrolysis of biosolids in fixed bed is presented in the Table 3. Lu et al. (2013) studied wet biosolids with moisture content as high as 80%. The biochar yields with wet biosolids were found to be very low as the moisture evaporated during the pyrolysis. However, when the temperature was increased from 300 to 600 °C, the

biochar yield was found to be decreased by only 25%. Dried biosolids in comparison to wet biosolids yielded a higher amount of biochar. Nevertheless, only 30% drop in biochar yield was observed when the pyrolysis temperature was increased from 300 to 600 °C.

The surface area is an important characteristic of biochar when used as a soil amendment or adsorbent material. The surface area of biochar produced from slow and fast pyrolysis of biosolids in a fixed bed reactor at different temperatures is provided in Table 4. The surface area was found to increase with increasing pyrolysis temperature for both slow and fast pyrolysis. However, in the case of slow pyrolysis, the surface area increased rapidly compared to fast pyrolysis. For example, biochar surface area increased from 20.27 m<sup>2</sup>/g at 600 °C to 67.60 m<sup>2</sup>/g at 900 °C in fast pyrolysis while in slow pyrolysis surface area increased from 10.21 m<sup>2</sup>/g at 600 °C to 122.04 m<sup>2</sup>/g at 900 °C. The specific surface area of biochar is related to biosolids structure, pore size distribution and biochar density. The effect of temperature on the surface area of biochar was also observed by other researchers (Agrafioti et al. 2013; Ahmad et al. 2014; Bridle and Pritchard 2004; Carey et al. 2015; Cha et al. 2016; Chen et al. 2014; Hossain et al.

2010, 2011, 2009; Inguanzo et al. 2002; Khan et al. 2013a; Lu et al. 2013, 2012a; Méndez et al. 2012, 2013; Paz-Ferreiro et al. 2012; Shimabuku et al. 2016; Zielińska and Oleszczuk 2016; Zielińska et al. 2015). There is a general agreement in the literature that the increase in the temperature and holding time leads to an increase in the biochar's surface area. However, high temperatures and holding time can reduce the volume of mesopores due to sintering. In general, biochar has a narrow micropore distribution with a large surface area.

The overall weight percentage of C, N and H decreased in the solids with the increase in the pyrolysis temperature as a result of pyrogenic decompositions (Ábrego et al. 2009; Jin et al. 2017). A further explanation of the decrease in carbon and organic carbon is the formation of aromatic ring clusters. This is responsible for the low H/C ratio (Ábrego et al. 2009; Chen et al. 2014). Chen et al. (2014) reported in a fixed bed reactor experiment that the content of remaining H, N and O was 0.70%, 1.54% and 10.55% respectively at pyrolysis temperature of 500 °C while it rapidly decreased to 0.11, 0.53% and 2.44% at 900 °C in the biochar produced from biosolids. This further reduced H/C and O/C atomic ratio and increased amorphous carbon. The weight percentage of ash was found to increase with the increase in the temperature. However, as a result of pyrolysis, the overall bio-availability of heavy metals such as Ni, Co, Pb, Cr and As greatly reduced (Khan et al. 2013a; Méndez et al. 2012). Macro and micro-nutrients retention into biochar is expected to be in the range of 30–80%. For example, ~ 50% of N and > 70% of P and Fe were retained in the biochar matrix from biosolids. The temperature played an important role in retaining nutrients in the biochar matrix. Higher temperatures were responsible for their release into gas followed by their recovery in the condensed aqueous phase. Table 5 summarises the change in biochar composition with varying temperatures.

### 6.2.2 Effect of heating rate

Experiments conducted in the fixed bed reactor demonstrated that biochar yield decreased during the slow pyrolysis, when heating rate and temperature were increased from 5 to 60 °C/min and 450 to 850 °C, respectively (Inguanzo et al. 2002). Gao et al. (2014) investigated slow and fast pyrolysis of dried

biosolids in fixed bed reactor and found that biochar yield decreased from 53.60 to 33.24%, when the heating rate was increased from 8 °C/min (slow pyrolysis) to 100 °C/min (fast pyrolysis), with an increase in temperature from 450 to 650 °C (Fig. 3).

There is no study that reported on the effects of heating rate on the biosolids biochar pore structure. However, in biomass pyrolysis, Kambo and Dutta (2015) reported that very high heating rate (> 100 °C/min) adversely affected the pores of the biochar as it caused the natural biomass structures to fragment, which resulted in pores being drastically reduced.

### 6.2.3 Effect of solid residence time

Solid residence time was found to have a significant impact on products yield as well as quality. In slow pyrolysis experiments of biosolids conducted in a fixed bed reactor, it was observed that biochar yield decreased with the increase in the solid residence time and pyrolysis temperature (Agrafioti et al. 2013; Gao et al. 2017). The effects of solid residence time on BET surface area and micro-pore volume is highlighted in Fig. 4. BET surface area and micro-pore area were found to increase with the increase in the residence time, which reached to a maximum value at 2 h and then started dropping. The drop-in surface area and micro-pore area might be attributed to the possible sintering of the biochar particles due to extended residence time. A slow pyrolysis with high solid residence time and lower temperature is favourable to obtain biochar with the superlative properties and yield. This provides the best distribution of pores, evaporation of volatile and water molecules and the best possible adsorption properties (Agrafioti et al. 2013; Bandosz and Block 2006; Hossain et al. 2010; Lu et al. 2013).

## 6.3 Fluidised bed reactor

### 6.3.1 Effect of temperature

Shen and Zhang (Shen and Zhang 2003) conducted fluidised bed pyrolysis experiments using activated sewage sludge obtained from Western Australia. They studied the temperature range between 300 and 600 °C and observed that maximum oil yield was obtained at 525 °C. From the GC–MS analysis, it was concluded that carbohydrates and polyphenols are

**Table 5** Elemental composition and physical properties of biosolids derived biochar from fixed bed experiments

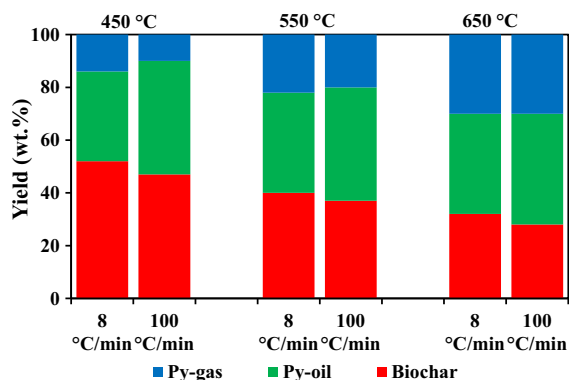
Compound	Unit	Biosolids	Biochar		
			Pyrolysis Temperature Range °C		
			300–500	550–650	700–900
Ultimate Analysis					
H	(%)	4.1–10.2	0.7–2.8	0.4–1.2	0.2–0.7
C	(%)	25.3–51.7	18.9–47	8.2–30.8	6.5–33
N	(%)	2–9.8	2.1–6.4	0.4–3.8	0.3–2.9
H/C		2.35–0.35	0.23–0.35	0.05–0.25	0.05–0.30
O/C		0.4–1.18	0.13–0.16	0.05–0.17	0.01–0.18
Proximate Analysis					
Ash	(%)	23.4–44.6	35–72	60.3–84.0	62.3–82.0
Volatile C	(%)	45.9–75.3	18.4–24.1	11.0–16.7	4.6–5.7
Fixed C	(%)	1.3–10.5	4.7–12.9	1.9–25.4	23.9–28.4
Macro-nutrients					
P	(%)	2.0–4.94	5.6	2.4–5.2	4.9–5.1
K	(%)	0.3–0.8	0.24	1.4–1.8	0.3
Ca	(%)	3.02–10	3.5–4.2	1.3–4.6	5.4
Mg	(%)	0.33–2.06	0.4–0.4	0.04–0.5	0.5
S	(%)	1.0–5.17	4.5–5	2.7–5	6.2
Micro-nutrients					
Fe	(%)	2.0–6.83	7.8–8.9	10.2	11
Zn	(mg/kg)	470–2398	1250–2980	845–3900	2175
Cu	(mg/kg)	370–1400	222–2600	163–2700	1500
pH		4.42–7.39	4.7–9.5	7.2–9	11.7–12
Physical properties					
Total surface area	(m <sup>2</sup> /g)		4–35.7	5.5–37.2	9.22–107
Pore volume	(cm <sup>3</sup> /g)		0.0326–0.0738	0.0144–0.0681	0.0321–0.0894

Agrafioti et al. (2013), Ahmad et al. (2014), Bridle and Pritchard (2004), Carey et al. (2015), Cha et al. (2016), Chen et al. (2014), Hernández et al. (2017), Hossain et al. (2010), Hossain et al. (2011), Hossain et al. (2009), Khan et al. (2013a), Liu et al. (2015), Lu et al. (2012a), Manara and Zabaniotou (2012), Méndez et al. (2013), Nyakuma et al. (2016), Patel et al. (2019a), Paz-Ferreiro et al. (2012), Shimabuku et al. (2016), Urych and Smolinski (2016), Xu et al. (2017), Zhang et al. (2014b) and Zielińska et al. (2015)

decomposed at this temperature and those are mainly responsible to produce pyrolysis oil. Park et al. (2008) studied dried biosolids obtained after digestion in a fluidised bed reactor and observed that pyrolysis gas was mainly composed of CO<sub>2</sub>, CO, CH<sub>4</sub>, and other light hydrocarbons whereas pyrolysis oils were mainly composed of acids, aldehydes, alcohols, and hydrocarbon compounds. The pyrolysis oil produced from biosolids pyrolysis is found to have a higher nitrogen content and with the increasing temperature, the nitrogen is converted into gas phase NH<sub>3</sub>.

Patel et al. (2019b) investigated the slow pyrolysis of biosolids with an aim to produce biochar using a bench-scale fluidised bed reactor which operated in

bubbling mode of fluidisation by varying pyrolysis temperatures (500, 700 and 900 °C) as well as the type of bed material (i.e. biochar, activated char and lime). The analytical instruments such as GC–MS, SEM, FTIR and BET were employed to characterise products. The activated char was found to be the most suitable bed material due to its ability to crack down unwanted aromatic compounds and to improve the morphology of the resultant biochar. It was observed in their study that biosolids pyrolysis might need a slightly higher temperature than biomass pyrolysis to open up the pore structure of biochar. Furthermore, it was observed that the production of NH<sub>3</sub> from organic nitrogen of biosolids, can be helpful in reducing NOx



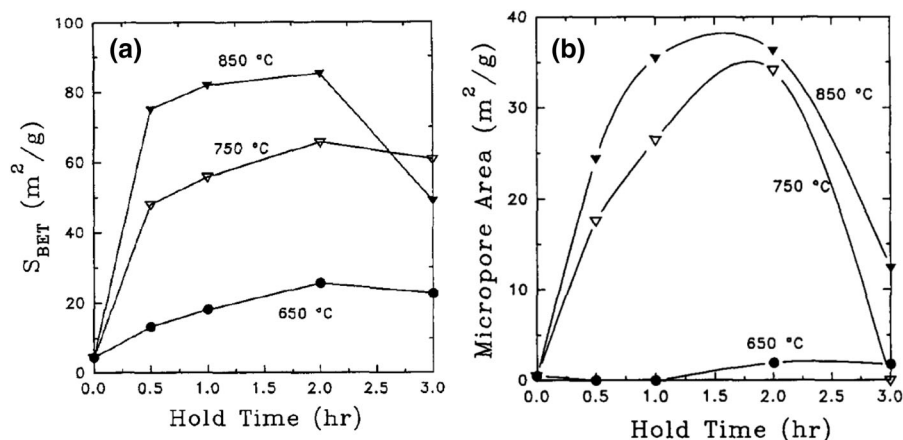
**Fig. 3** Effects of pyrolysis temperature and heating rate on pyrolysis product yields under fast pyrolysis (100 °C/min) and slow pyrolysis (8 °C/min) conditions in fixed bed (Gao et al. 2014)

in the flue gas generated from the combustion of gas and oil vapours. The BET surface area analyses suggested that the highest surface area of 91 m<sup>2</sup>/g was

obtained for biochar produced at 700 °C which was reduced to 40.3 m<sup>2</sup>/g at 900 °C possibly due to sintering effect.

### 6.3.2 Effect of vapour residence time

Table 6 shows the effect of vapour residence time on the product distribution. In fluidised bed reactor, vapour residence time is more controllable than the solid residence time in a bench-scale reactors. An increase in vapour residence time resulted in a higher biochar yield during the fast pyrolysis of biosolids in a fluidised bed reactor. For example, Shen and Zheng (Shen and Zhang 2003) in their study observed char yield increased from 43 to 69 wt% when vapour residence time increased from 1.5 to 3.5 s. Similarly, Fonts et al. (2008a, b, 2009) observed increase in char yield from 46 to 63 wt% with vapour residence time increasing from 1 to 3 s. This might be due to the re-



**Fig. 4** **a** BET surface area and **b** micropore area evolution with time at different temperatures in fixed bed (Lu et al. 1995)

**Table 6** Effects of vapour residence time in biosolids' pyrolysis in fluidised bed experiments

Biosolids type	Location	Temperature (°C)	Residence time (s)	Oil yield (wt%)	Char yield (wt%)	Gas yield (wt%)	References
Dried biosolids	After activated sludge plant	300–600	1.5–3.5	5–32	43–69	12–25	Shen and Zhang (2003)
Dried biosolids	Unknown	450–700	0.2–0.5	35–52	40–45	7–25	Park et al. (2008)
Dried biosolids	After anaerobic digester	450–650	1–3	23–41	46–63	7.4–28	Fonts et al. (2009), Fonts et al. (2008a) and Fonts et al. (2008b)

**Table 7** Summary of biosolids slow pyrolysis research conducted in TGA, fixed bed and fluidized bed

References	Type of experimental rig	Biosolids type	Process variables studied				Scope of work				
			Peak temperature/temperature range (°C)	Heating rate (°C/min)	Solids residence time (min)	Vapor residence time	Reaction kinetics and mechanism derivation	Char, oil and gas yield measurements	Morphological analysis and surface area measurement for biochar	Biochar characterization	Oil and gas characterization
Ji et al. (2010)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Gasparović et al. (2011)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Nyakuma et al. (2016)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Urych and Smolinski (2016)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Xiaohua and Jiancheng (2012)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Patel et al. (2018)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Xu et al. (2017)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Zhai et al. (2011)	TGA	Dried biosolids	✓	✓	x	x	✓	x	x	x	x
Nowicki and Ledakowicz (2014)	TG-MS	Dried biosolids	✓	✓	x	x	✓	x	x	✓	✓
Hernández et al. (2017)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	x	x	Δ
Kan et al. (2016)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	x	x	Δ
Gao et al. (2014)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	x	✓	✓
Patel et al. (2019a)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	x	x	Δ
Shao et al. (2007)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	✓	x	Δ
Wang et al. (2018)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	✓	x	Δ
Zhang et al. (2014b)	TGA-FTIR	Dried biosolids	✓	✓	x	x	✓	x	x	x	Δ
Ábrego et al. (2009)	FB	Dried biosolids	✓	✓	✓	x	x	✓	✓	✓	✓
Agrafioti et al. (2013)	FB	Dried biosolids	✓	✓	✓	x	x	✓	✓	✓	x
Cha et al. (2010)	FB	Dried biosolids	✓	x	x	x	x	x	✓	✓	x
Figueiredo et al. (2018)	FB	Dried biosolids	✓	x	x	x	x	x	x	✓	x
Gao et al. (2014)	FB	Dried biosolids	✓	✓	x	x	x	✓	x	✓	Δ
Gao et al. (2017)	FB	Dried biosolids	✓	x	✓	x	x	✓	x	✓	✓
Hossain et al. (2011)	FB	Dried digested biosolids	✓	✓	x	x	x	x	x	x	x
Hossain et al. (2010)	FB	Dried digested biosolids	✓	✓	x	x	x	x	x	✓	X
Inguanzo et al. (2002)	FB	Dried biosolids	✓	✓	x	x	x	✓	✓	x	✓
Khanmohammadi et al. (2017)	FB	Dried digested biosolids	✓	✓	✓	x	x	x	x	✓	x
Kim and Parker (2008)	FB	Dried digested biosolids	✓	x	x	x	x	✓	x	x	✓
Liu et al. (2016a)	FB	Dried biosolids	✓	✓	✓	x	x	x	I	✓	x
Lu et al. (1995)	FB	Dried biosolids	✓	✓	✓	x	x	x	✓	✓	x
Lu et al. (2013)	FB	Dried biosolids	✓	✓	✓	x	x	✓	✓	✓	x



**Table 7** continued

References	Type of experimental rig	Biosolids type	Process variables studied				Scope of work				
			Peak temperature/temperature range (°C)	Heating rate (°C/min)	Solids residence time (min)	Vapor residence time	Reaction kinetics and mechanism derivation	Char, oil and gas yield measurements	Morphological analysis and surface area measurement for biochar	Biochar characterization	Oil and gas characterization
McNamara et al. (2016)	FB	Dried biosolids	✓	✓	✓	x	x	✓	x	✓	✓
Méndez et al. (2013)	FB	Dried biosolids	✓	✓	✓	x	x	x	x	✓	x
Patel et al. (2019b)	FBR	Dried biosolids	✓	✓	✓	✓	x	✓	✓	✓	✓
Shen and Zhang (2003)	FBR	Dried biosolids	✓	x	✓	✓	x	✓	x	x	✓

FB = Fixed bed, FBR = Fluidised bed reactor, “✓” means studied, “x” means not studied, “I” means only BET analysis study, “Δ” means only gas analysis study

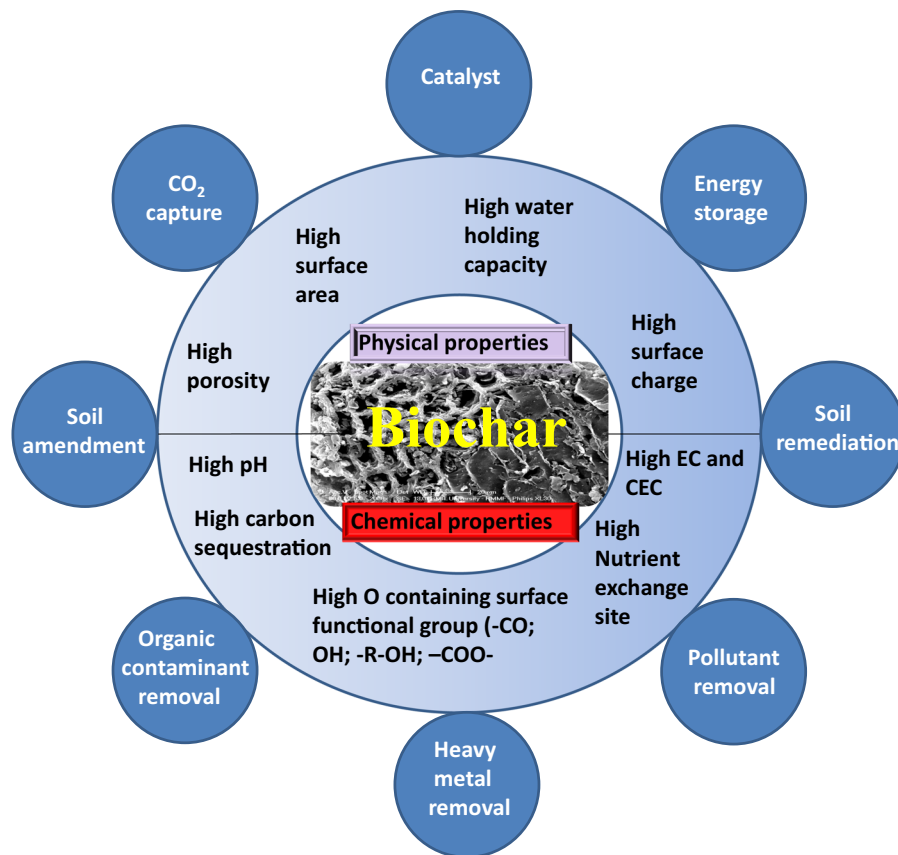
polymerisation and condensation of gas and oil on the char surface. However, when the vapour residence time is lower, re-polymerisation will not occur, thus the biochar yield will be lower. Vapour residence time can not only alter the pyrolytic biochar yield, but also may affect biochar quality including surface area and micro-pore volume. However, there is no study reported in the literature which provides information about the effect of vapour residence time on biochar quality. The effect of vapour residence time on pyrolysis oil yield is opposite to the char yield. In pyrolysis, pyrolysis oil yield decreased with the increasing vapour residence time.

#### 6.4 Comparison between TGA, fixed and fluidised bed experimental observations

Literature shows highest number biosolids pyrolysis studies in the order of TGA > fixed bed > fluidised bed (Table 7).

TGA studies mainly used to derive intrinsic kinetic parameters and reaction mechanisms. As TGA experiments use very small quantity of biosolids sample, it is very hard to conduct comprehensive physico-chemical property analysis of the resultant biochar. Also, TGA coupled with MS or FTIR can be used for gas analysis. However, such studies on biosolids are not reported extensively in the literature. TGA study cannot help estimating char, oil and gas yield as it is hard to measure the quantity of gas and oil in TGA or TGA-MS/FTIR experiments. Some TGA studies looked at the effect of catalysts mainly on the biosolids' biochar yield (Patel et al. 2019a; Zhang et al. 2014b) in conjunction with reaction kinetics.

Fixed bed lab- or bench-scale studies were mainly focused on understanding the effect of process parameters such as temperature, heating rate and solids and vapour residence time on the yield and morphological changes in the biochar. It is generally difficult to vary vapour residence time in fixed bed rigs as they suffer from diffusional barriers and channelling at lower and higher gas velocities respectively. Fluidised bed lab- or bench-scale studies were mainly focused on understanding the effect of temperature and vapour residence time. In fluidised bed, vapour residence time is relatively easy to control given the fact that particles will still be fluidising uniformly at lower (or minimum) and higher gas velocities and



**Fig. 5** Potential applications of biochar produced from the pyrolysis of biosolids

hence the chances of diffusional barrier or channelling are relatively less.

In applied terms, the fluidised bed offers better performance as mass and heat transfer are greatly enhanced in the fluidised bed. However, the capital cost of building fluidised bed may be significantly higher than the fixed bed. Therefore, there is a need to quantify the benefits of fluidised bed over a fixed bed. For biosolids, there is no study reported in the literature comparing results from TGA, fixed bed and fluidised bed experiments on a single biosolids feedstock. Such a study would be able to quantify the benefits and drawback different methods employed at research and commercial scale.

## 7 Applications of biosolids pyrolytic biochar

Biochar can be used for various applications as highlighted in Fig. 5. The main applications of biochar

reported in the literature are soil amendment, water and soil remediation, catalysts and energy storage and generation materials (Wang et al. 2017b; Yuan et al. 2013). Biochar produced from biosolids might have different properties than biochar produced from biomass. Therefore, the applications of biosolids derived biochar may slightly vary. The following sub-sections have critically reviewed efforts being made in the open literature.

### 7.1 Use as soil amendments

Table 8 summarizes studies that focused on agricultural use of biochar derived from biosolids/sewage sludge. In more than 90% of publications, it is noticed that biochar increased the yield of a wide variety of agricultural crops. Hossain et al. (2010) demonstrated that the yield of cherry tomato could be improved by 64% with the application of biosolids derived biochar produced at 550 °C. This yield gain is a result of the

**Table 8** Summary of previous research on the effect of biosolids biochar on soil chemical properties and crop yields

References	Pyrolysis temperature (°C)	Biochar rate	Crop	pH	N	P	K	S	Ca	Mg	CEC	Mic	Yield
Gondek et al. (2019)	300	1%	Grass	↑		↑			↑	↑			↑
Yuan et al. (2016)	500	1%				↑	↑						
Hossain et al. (2010)	550	10 t ha <sup>-1</sup>	Tomato	↑	↑	—	—		↑	—	↑		↑
Faria et al. (2018)	300–500	15 t ha <sup>-1</sup>	Corn	—		↑	—		—	—	↑		↑
Sousa and Figueiredo (2016)	300	3%	Radish	—		↑	—		↑	↑	↑		↑
Gwenzi et al. (2016)	300–500	15 t ha <sup>-1</sup>	Corn	—	—	—	—		—	—	—	—	↑
Zong et al. (2018)	—	2%	-	↑					↑	↑			
Khanmohammadi et al. (2017)	350	14,5 t ha <sup>-1</sup>	Corn		—	↑	—					↑	—
Méndez et al. (2012)	500	4%	-	—							—		
Fathi Dokht et al. (2017)	350	20 t ha <sup>-1</sup>	Soybean		↑	↑	↑						↑
Shao et al. (2019)	800	20 t ha <sup>-1</sup>	Rice	—	—	—	—				—		↑
Tian et al. (2019)	200	5%	Grass	↓	↑	↑	↑					↑	↑
Yue et al. (2017)	500	10%	Grass	↑	↑	↑	↑					↑	↑
Khan et al. (2013a)	550	5%	Rice	↑	↑	↑	—	↑				↑	↑
Luo et al. (2018)	500	5%	Alfafa	↑	↑	↑	↑						↑
Waqas et al. (2014)	550	5%	Cucumber	—	↑	↑		↑				—	↑
You et al. (2019)	700–850	40 t ha <sup>-1</sup>	Peanuts	↓	↑	↑	↑						↑

Mic = micronutrients; ↑, ↓ and (—) mean increase, decrease and maintain values, respectively, in relation to the control treatment without biochar

improvement of the soil chemical, physical and biological properties promoted by the biochar application.

In most studies, biochar increased the soil phosphorus, nitrogen, calcium and magnesium contents. It is well known that biochar from biosolids is a source of P, with the potential to replace soluble mineral fertilizers (Faria et al. 2018). Application of biosolids biochar to soil can increase P availability by up to 38 times (Yue et al. 2017), thus providing higher crop yields (Faria et al. 2018). In general, higher pH values in biochar may be related to increased concentrations of alkali elements, such as Ca and Mg oxides and their alkaline reactions (Figueiredo et al. 2018). However, the effect of biochar on soil pH depends on the applied dose and pH of the biochar, the initial soil pH and the soil buffering capacity of both biochar and soil (Biederman and Harpole 2013). Therefore, biochar application can also reduce or not affect the soil pH. In 50% of the studies summarized in Table 8, the soil cation exchange capacity (CEC) was increased by biochar application. The large specific surface area and the abundance of functional groups can increase

the CEC of biosolid biochar (Luo et al. 2018). Therefore, when applied to the soil, biosolids biochar can improve soil CEC (Faria et al. 2018; Hossain et al. 2010; Lehmann et al. 2006; Sohi et al. 2010).

Biosolids biochar presents great potential to store C in the form of stable structures over a long period of time, from centuries to millennia (Zimmerman et al. 2011). This is because pyrolysis, which catalyzes the transformation of biomass, promotes C retention in the form of inert carbon (Novotny et al. 2015). Therefore, in agriculture, biochar can mitigate greenhouse gas emissions. It has been estimated that global sustainable biochar production could offset up to 12% of current anthropogenic CO<sub>2</sub> emissions (Woolf et al. 2010). Recent estimation indicates that the application of biochar to the soil can promote a C sequestration rate of 0.54 Mg C ha<sup>-1</sup> year<sup>-1</sup> (Lal et al. 2018). In the short term, residual bio-oils and those adsorbed on the biochar surface, immediately after pyrolysis, serve as the labile support of C for microbial metabolism (Steiner et al. 2004). In the long term, the sorption of biochar with soil organic matter (SOM) may increase C storage in the soil, providing organic matter with

physical protection against microbial decomposition (Zimmerman et al. 2011). Figueiredo et al. (Figueiredo et al. 2019) demonstrated that the increase of organic C in the soil promoted by biochar varies with the pyrolysis temperature employed. While biochar produced at 300 °C increased the levels of labile and microbial C that obtained at higher temperature (500 °C) favored the accumulation of C in more recalcitrant fractions of the SOM. When combined with NPK, the application of biosolids biochar can increase total organic C levels in the soil by up to 40%. When produced at a lower temperature (300 °C) biosolids biochar appears to promote a more balanced between labile and stable fractions of the SOM. However, long-term studies are needed to evaluate the actual contribution of biosolids biochar on soil C accumulation.

One of the major concerns of using biosolids derived biochar for agriculture land application is the presence of high levels of heavy metals. Majority of the heavy metals may remain in the char phase after pyrolysis (Choudri et al. 2018; Dorrance et al. 2017; Dumontet et al. 2001; Haynes et al. 2009; Pritchard et al. 2010; Scher et al. 2018). In fact, the concentration of heavy metals will increase in the biochar compared to biosolids (Lee et al. 2017; Paz-Ferreiro et al. 2018). Levels of some of the heavy metals in biochar may even cross the limits suggested by biosolids guidelines for agricultural land application (Darvodelsky and Bridle 2012); however, they may be still well below the limits suggested by international biochar guidelines (Paz-Ferreiro et al. 2018). The biochar produced from the biosolids is expected to have higher levels of heavy metals than the biochar produced from other biomass feedstock (Chen and Yan 2012). However, bio-availability of most of the heavy metals from biosolids biochar is suggested to be very low (Lee et al. 2017; Paz-Ferreiro et al. 2018; Yang et al. 2018b). Unfortunately, the bio-availability of heavy metals are not discussed in the biosolids or biochar guidelines (Darvodelsky and Bridle 2012). Méndez et al. (2012) reported that compared to direct agricultural reuse of raw biosolids, the use of biosolids derived biochar as a soil amendment could lower the leaching of Cu, Ni, Cd and Zn, as well as reduce the plant availability of Ni, Zn, Cu and Pb. Waqas et al. (2014) reported that biochar produced from biosolids at 550 °C was effective in reducing the polycyclic aromatic hydrocarbon (PAH) concentrations and the

availability of potentially toxic elements (PTEs) such as As, Pb and Cu. Song et al. (2014) revealed that proper pyrolysis temperature choice could prevent the leaching of heavy metals and inhibit the heavy metals accumulation in the plants.

## 7.2 Use as adsorbents

The use of biosolids derived biochar as low-cost adsorbents can be considered as a reasonably profitable route in biosolids management. The production of biochar based adsorbents from biosolids via pyrolysis, generally referred to as biosolids-based adsorbents, was first proposed by Kemmer et al. (1971). In the same year, Beeckmans and Ng (Beeckmans and Ng 1971) prepared biosolids derived biochar based adsorbent with 14.1% carbon content from the pyrolysis of biosolids. They demonstrated that the adsorbing capacity of such adsorbents was between fly ash and activated coconut charcoal. After these studies, a wealth of research on the production and application of biosolids derived biochar based adsorbents has been carried out in the subsequent years. The surface area and functional group were considered as two of the most important characteristics of the biochar produced from biosolids for their use as adsorbents. Generally, the studies indicated that the BET surface area and porosity of biosolids derived biochar may be affected by many factors such as residence time, temperature, heating rate, functional group and inlet particle size (Agrafioti et al. 2013; Otero et al. 2009; Tang et al. 2013; Zou et al. 2013). Different approaches have been used in order to obtain biochar with a high surface area, including (i) physical activation under the treatment of CO<sub>2</sub>, air and steam (Jindarom et al. 2007; Méndez et al. 2005; Rio et al. 2006); (ii) chemical activation with the addition of activation reagents, such as ZnCl<sub>2</sub> (Lu and Lau 1996; Wen et al. 2011; Zhang et al. 2005; Jeyaseelan and Qing 1996), KOH (Cha et al. 2010), H<sub>2</sub>SO<sub>4</sub> (Lu and Lau 1996; Martin et al. 2002; Otero et al. 2003; Zhang et al. 2005) and H<sub>3</sub>PO<sub>4</sub> (Zhang et al. 2005) as shown in Table 9.

The biosolids based adsorbents can be used for organic contaminant removal, heavy metal removal, polluted substance removal and nutrient adsorption. The summary of literature related findings is provided below, as shown in Table 10.

In recent years, extensive research has been conducted on the use of biosolids derived biochar to

**Table 9** Effects of biochar's activation methods on surface area

Biosolids type	Location	Activation method	Activation agent	BET surface area (m <sup>2</sup> /g)	References
Dried biosolids	Unknown	Physical	CO <sub>2</sub>	61	Jindarom et al. (2007)
Dried biosolids	After aerobic digestion	Physical	Air	102	Méndez et al. (2005)
	After anaerobic digestion			105	
Dried biosolids	After digestion	Physical	Steam	96	Rio et al. (2006)
Dried biosolids	After dewatering	Chemical	ZnCl <sub>2</sub>	309	Lu and Lau (1996)
			H <sub>2</sub> SO <sub>4</sub>	220	
Dried biosolids	After dewatering	Chemical	ZnCl <sub>2</sub>	509	Wen et al. (2011)
Dried biosolids	After activated sludge plant	Chemical	KOH	783	Cha et al. (2010)
Dried biosolids	After anaerobic stabilisation	Chemical	H <sub>2</sub> SO <sub>4</sub>	390	Otero et al. (2003)
Dried biosolids	After dewatering via centrifuge	Chemical	H <sub>2</sub> SO <sub>4</sub>	253	Martin et al. (2002)
Dried biosolids	Unknown	Chemical	H <sub>3</sub> PO <sub>4</sub>	289	Zhang et al. (2005)
			H <sub>2</sub> SO <sub>4</sub>	408	
			ZnCl <sub>2</sub>	555	

capture liquid phase organic pollutants (Jindarom et al. 2007; Otero et al. 2003; Rozada et al. 2007). It was revealed that higher mesopore volume and BET surface area of biosolids derived biochar are beneficial for the uptake of pollutants with large molecular sizes, such as dyes (Jindarom et al. 2007; Otero et al. 2003; Rozada et al. 2007). There are several studies reported in the literature about an uptake of anionic and cationic dyes by biochar produced from biosolids (Jindarom et al. 2007; Martin et al. 2003, 2002; Otero et al. 2003; Rio et al. 2006; Zou et al. 2013). It was found that basic (cationic) dye was adsorbed in greater quantities than acid dye and/or reactive dye on the biochar produced by carbonisation with N<sub>2</sub> and CO<sub>2</sub> activation (Jindarom et al. 2007). The high concentration of carboxyl groups (R-COOH), phenolic groups (R-OH) and oxygen-containing functional groups on the surface of biochar are beneficial for the uptake of dyes (Martin et al. 2002).

Except for the uptake of organic contaminants, the biochar was also used as adsorbents for the removal of heavy metals such as Cd<sup>2+</sup>, Ni<sup>2+</sup>, Pb<sup>2+</sup>, Cu<sup>2+</sup> and Zn<sup>2+</sup> in aqueous solutions (Agrafioti et al. 2013; Chen et al. 2014; Lu et al. 2013; Smith et al. 2009). Biosolids derived biochar have shown that cation exchange ability in the negatively charged biochar surface can efficiently bind to positively charged heavy metal ions in the solution (Agrafioti et al. 2013; Chen et al. 2014; Lu et al. 2013; Smith et al. 2009). Rio et al. (2006) investigated the Cu<sup>2+</sup> ion uptake by commercial

activated carbon (AC) and activated biosolids biochar (ABB) carbonised at 800 °C. Approximately 182 mg/g and 227 mg/g of Cu<sup>2+</sup> removal was achieved by AC and ABB, respectively Rio et al. (2006). and Chen et al. (2014) found that the adsorption of Cu<sup>2+</sup> and Cd<sup>2+</sup> on biochar was mainly through an ion-exchange mechanism with the Ca<sup>2+</sup> ions present in the biochar. Lu et al. (2012a) found that the presence of functional groups, such as carboxyl group and hydroxyl, were beneficial for the higher cation exchange capacity of biosolids derived biochar. Agrafioti et al. (2013) reported that biochar produced from biosolids removed ~ 70% of Cr (III) and ~ 30% of As (V) from aqueous solutions. Biosolids' biochar was also capable of removing 40% and 65% of Pb (II) and Cr (VI), respectively, from the aqueous solution (Zhang et al. 2013). In this case, the presence of organic functional groups on the biochar surface and biochar's higher surface area were found to be responsible for such high removal efficiency.

Biochar was also found to be a very efficient media for the removal of some contaminants generated in pyrolysis and gasification, such as hydrogen sulfide, sulfur dioxide or nitric oxides. Bashkova et al. (2001) investigated the adsorption of SO<sub>2</sub> on biosolids derived biochar and found that the capacity of the adsorbents increases with the increasing temperature of carbonisation, and the maximum adsorption capacity of SO<sub>2</sub> was 30 mg/g. The development of adsorbent/catalyst from municipal wastewater sludge for

**Table 10** Applications of biosolids derived biochar or activated char as adsorbent

Biosolids types	Location	Biochar/ activated char	Pyrolysis temperature (°C)	Contaminated stream	Target material	Adsorption capacity (all unit in mg/g or defined if different)	References
<i>Biochar used for dye removal</i>							
Dried biosolids	Unknown	Biochar	750	Dyes	Acid yellow 49 Basic blue 41 Reactive red 198	116 588 25	Jindarom et al. (2007)
Dried biosolids	After anaerobic stabilisation	Activated char	625 and activated by H <sub>2</sub> SO <sub>4</sub>	Pharmaceutical, food industries and wastewater	Crystal violet Indigo carmine Phenol	263 60 42	Otero et al. (2003)
Dried biosolids	After digestion	Activated char	600 and activated by steam	Wastewater	Phenol	50	Rio et al. (2006)
Dried biosolids	After aerobic digestion and dewatering	Activated char	700 and activated by H <sub>2</sub> SO <sub>4</sub>	Dyes	Direct red Direct black Acid brown Basic red Rhodamine-B	49 29 21 188 28	Martin et al. (2003)
Dried biosolids	Unknown	Biochar	1000	Dyes			Zou et al. (2013)
Dried biosolids	After dewatering via centrifuge	Activated char	700 and activated by H <sub>2</sub> SO <sub>4</sub>	Wastewater	Phenol value Iodine number Methylene blue number Tannin value	83 (g dm <sup>-3</sup> ) 599 14 2500 (g dm <sup>-3</sup> )	Martin et al. (2002)
<i>Biochar used for heavy metal removal</i>							
Dried biosolids	After dewatering	Biochar	550	Acid mine drainage	Pb <sup>2+</sup>	31	Lu et al. (2012a)
Dried biosolids	After activated sludge plant	Activated char	650 Activated by ZnCl <sub>2</sub>	Wastewater	Cr (III) Pb (II) Cu (II) Hg (II)	6 40 6 63	Otero et al. (2009)



Table 10 continued

Biosolids types	Location	Biochar/ activated char	Pyrolysis temperature (°C)	Contaminated stream	Target material	Adsorption capacity (all unit in mg/g or defined if different)	References
Dried biosolids	After dewatering	Activated char	850	Aqueous waste solution	Cd <sup>2+</sup>	16	Zhai et al. (2004)
Dried biosolids	After digestion	Activated char	Activated by ZnCl <sub>2</sub> 600 and activated by steam	Raw water sludge	Ni <sup>2+</sup> Cu <sup>2+</sup>	9 80	Rio et al. (2006)
Dried biosolids	After dewatering	Biochar	650–950	Aqueous waste solution	Phenol Cu <sup>2+</sup>	50 70	Seredych and Bandosz (2006)
Dried biosolids	After biophysical and drying	Biochar	500–900	Wastewater	Cd <sup>2+</sup>	15	Chen et al. (2014)
Aerobically digested dried biosolids	After aerobic digestion	Biochar	450	Saline water	Na <sup>+</sup> K <sup>+</sup> Ca <sup>2+</sup> Mg <sup>2+</sup>	77% 66% 35% 45%	Gascó et al. (2005)
Dried biosolids	After aerobic digestion and drying	Biochar	650	Wastewater	Cr (III) Pb (II) Cu (II) Hg (II) Hg (II)	3 40 7 65 60–80%	Rozada et al. (2008) Zhang et al. (2005)
Dried biosolids	Unknown	Activated char	650 Activated by H <sub>2</sub> SO <sub>4</sub> , H <sub>3</sub> PO <sub>4</sub> and ZnCl <sub>2</sub>	Aqueous waste solution	As (V) Cr (III) Pb (II) Cr (VI)	30% 70% 18 30	Agrafioti et al. (2013) Zhang et al. (2013)
Dried biosolids	After anaerobic digestion and belt-filter-press dewatering	Biochar	300–500	Wastewater			
Dried biosolids	After dewatering via pressed filtration and drying	Biochar	300–600	Wastewater			
<i>Biochar used for pollution control</i>							
Dried biosolids	After activated sludge plant		450	Biosolids filtrate	NH <sub>4</sub>	5.3	Carey et al. (2015)
Dried biosolids	After activated sludge plant	Activated char	500 activated by KOH	Unknown	NO <sub>x</sub>	55%	Cha et al. (2010)

Table 10 continued

Biosolids types	Location	Biochar/ activated char	Pyrolysis temperature (°C)	Contaminated stream	Target material	Adsorption capacity (all unit in mg/g or defined if different)	References
Dried biosolids	Unknown	Biochar	400–950	Dry and moist air	SO <sub>2</sub>	47	Bashkova et al. (2001)
Dried biosolids	After dewatering	Activated char	400–950—activated by H <sub>2</sub> SO <sub>4</sub> and ZnCl <sub>2</sub>	Wastewater	H <sub>2</sub> S	25%	Lu and Lau (1996)
Activated dried biosolids	After activated sludge plant	Biochar	800–	Wastewater	Triclosan	239 µg/g	Tong et al. (2016)
Dried biosolids	Unknown	Biochar	500–800	Hydrophobic organic pollutant (industrial material)	1-Naphthol	666	Liu et al. (2016a)
Dried biosolids	After dewatering	Activated char	450 and activated by ZnCl <sub>2</sub>	Wastewater	Formaldehyde	74	Wen et al. (2011)
Dried biosolids	After dewatering	Biochar	550	Antibiotics	Fluoroquinolone	20	Yao et al. (2013)
<i>Biochar used for nutrient recovery</i>							
Dried biosolids	After anaerobic digestion and drying	Biochar	450 550	K <sub>2</sub> HPO <sub>4</sub> solution	Phosphorous	1.06 0.97	Shepherd et al. (2016)
Dried biosolids	After anaerobic digestion and belt-filter-press dewatering	Modified biochar	300 and conditioned with Fenton's reagent	KH <sub>2</sub> PO <sub>4</sub> solution	Phosphorous	1.84	Wang et al. (2020)
Dried biosolids	After anaerobic digestion and drying	Modified biochar	700, Modified by CaCl <sub>2</sub>	K <sub>2</sub> HPO <sub>4</sub> solution	Phosphorous	153.85	Saadat et al. (2018)
Dried biosolids	After dewatering	Modified biochar	800, Modified by dolomite powder	Phosphate stock solution	Phosphorous	29.18	Li et al. (2019b)
Dried biosolids	Unknown	Biochar	700	Phosphate stock solution	Phosphorous	16.5	Fang et al. (2020)
Dried biosolids	After dewatering	Biochar	550	Phosphate stock solution	Phosphorous	15.88	Yang et al. (2018a)
Dried biosolids	After dewatering	Modified biochar	550, FeCl <sub>3</sub> impregnation			111	
Dried biosolids	Unknown	Biochar	700, mixed with brewers spent grain	NH <sub>4</sub> <sup>+</sup> -N solution solutions	Nitrogen	79.2%	Zhang and Wang (2016)

H<sub>2</sub>S removal was firstly proposed by Lu and Lau (1996). They prepared activated biosolids derived biochar (ABB) from dewatered biosolids via chemical activation by ZnCl<sub>2</sub>. The BET surface area of ABB was ~ 309 m<sup>2</sup>/g BET and it achieved 1.25 times higher H<sub>2</sub>S adsorption when compared with commercial activated carbon. The high BET surface area, high hydrophilicity and the presence of functional group and mineral phases are considered to be beneficial for the adsorption of NH<sub>4</sub> (Carey et al. 2015), NO<sub>x</sub> (Cha et al. 2010), SO<sub>2</sub> (Bashkova et al. 2001), H<sub>2</sub>S (Lu and Lau 1996). These features of biosolids' biochar are also favourable for the adsorption of other polluting substances such as triclosan (Tong et al. 2016), 1-naphthol (Liu et al. 2016a), formaldehyde (Wen et al. 2011) and fluoroquinolone (Yao et al. 2013).

The biochar can also be used for the adsorption of inorganic N and P. To date, various materials such as zeolite, opoka, ochre, Polonite, blast furnace slags and Filtra P, have been suggested for removing P from wastewater effluent (Cucarella et al. 2008; Dobbie et al. 2009, 2005; Heal et al. 2005). The biochar derived from biosolids for capturing P from wastewater can demonstrate environmentally sustainable, cost-effective and circular economy approach. Shepherd et al. (2016) demonstrated phosphate adsorption by biochar produced from anaerobic digested biosolids but adsorption capacity was found to be extremely low (i.e. ~ 1 mg/g) when compared with other commercially available adsorbent. This might be attributed to poor physico-chemical properties of the biosolids derived biochar. However, Li et al. (2019b) modified biosolids' derived biochar with dolomite and demonstrated improved adsorption capacity of 29.18 mg/g. He reported that Ca containing nanoparticles play significant roles in P adsorption via electrostatic attractions. Saadat et al. (2018) also reported that the maximum P sorption capacity of Ca-rich biochar derived from biosolids could be as high as 153.85 mg/g. Yang et al. (2018a) improved the P adsorption capacity of waste activated sludge-based biochar through chemical co-precipitation of Fe<sup>3+</sup>/Fe<sup>2+</sup> or FeCl<sub>3</sub> impregnation and showed that the adsorption capacity increased from 15.88 mg/g to 111 mg/g in iron-modified biochars. There is no study available for N adsorption for biosolids derived biochar. However, Zhang and Wang (2016) demonstrated 62.3–79.2% adsorption of N using biochar

derived from co-pyrolysis of Brewers spent grain (Mg enrich) and Sewage sludge (P enrich).

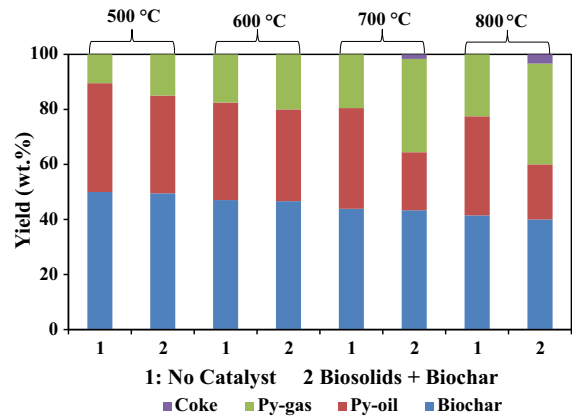
Biochar is regarded as an efficient and cost-effective adsorbent. Nevertheless, adsorption only achieves the transformation of pollutants from aqueous solution to adsorbents, but cannot destroy pollutants, so the contaminant-loaded biochar becomes hazardous waste that must be disposed (Quintanilla et al. 2010; Tang et al. 2018).

### 7.3 Use as activation agent

Due to its abundant oxygen-containing functional groups (OFGs) and persistent free radicals (PFRs), biochar can effectively catalyze to form reactive oxygen species (ROS) or sulfate radicals and then degrade pollutants from aqueous solution. The metal ions existed in pyrolytic biochar derived from biosolids can be effective catalyst for activation of persulfate (PS), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) and Ozone (O<sub>3</sub>). Table 11 provides the summary of Biosolids derived biochar application for the activation of PS, H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub>. Huang et al. (2018) prepared biosolids biochar based catalyst to activate peroxymonosulfate (PMS) for Bisphenol-A (BPA) degradation and found 80% degradation in 30 min. Wang et al. (2017a) found that sludge-derived biochar can effectively activate PS for the degradation of 4-chlorophenol. The removal efficiency of 4-chlorophenol reached 92.3% within 100 min. In the presence of biosolids derived biochar, 9.8 mg/L of triclosan could be completely degraded by PS within 240 min. In comparison, no obvious degradation of triclosan was observed in the absence of biochar (Wang and Wang 2019). Luo et al. (2019b) reported that H<sub>2</sub>O<sub>2</sub> activated by pyrolytic biochar from biosolids can degrade upto 70.19% ciprofloxacin. Moreover, > 80% ciprofloxacin was removed by synthesis of heterogeneous Fenton-like catalyst using sewage sludge biochar (Li et al. 2019c). Zhang et al. (2018) found that 95% of phenol was removed from aqueous solution via O<sub>3</sub> within 30 min in the presence of biosolids derived biochar synthesized at 700 and 900 °C. The Humic Acid (HA) activation by biosolids derived biochar has demonstrated ~ 65% degradation of ciprofloxacin (CIP) (Luo et al. 2019a). These studies highlight that besides adsorption, degradation induced by biochar is also an important pathway in minimizing the fate of organic pollutants.

**Table 11** Applications of biosolids derived biochar as activating agent

Biosolids types	Location	Biochar/Activated char	Pyrolysis temperature (°C)	Contaminated stream	Target material	Degradation capacity	References
<i>Biochar used for activation</i>							
Dried biosolids	After dewatering	Biochar	500, activate hydrogen peroxide	Aqueous solution	Ciprofloxacin	70.60%	Luo et al. (2019b)
		HNO <sub>3</sub> modified biochar				93.00%	
Dried biosolids	Unknown	Biochar	400, activate persulfate	Aqueous solution	Phenol	36.3%	Liang et al. (2020)
			700			57.8%	
Dried biosolids	After dewatering	Biochar	activate peroxymonosulfate	Aqueous solution	Bisphenol-A (BPA)	100%	Huang et al. (2018)
Dried biosolids	Unknown	Biochar	400, activate persulfate (PS)	Aqueous solution	4-chlorophenol	92.3%	Wang et al. (2017a)
Dried biosolids	Unknown	Biochar	700, activate peroxydisulfate (PDS)	Aqueous solution	Sulfamethoxazole (SMX)	94.6%	Yin et al. (2019)
Dried biosolids	After dewatering	Biochar	450, activate PMS (available as Oxone)	Aqueous solution	Triclosan (TCS)	98.9%	Wang and Wang (2019)
Dried biosolids	Unknown	Biochar	300–900, activating O <sub>3</sub>	Aqueous solution	Phenol	95%	Zhang et al. (2018)

**Fig. 6** Effects of biochar as catalyst on product yields under slow pyrolysis conditions in a fixed bed reactor: sample (1) no catalyst (2) biosolids and biochar mixture (Liu et al. 2017)

#### 7.4 Advance applications as catalysts and energy storage and energy generation materials

The literature on the use of the biosolids derived pyrolytic biochar as catalyst or catalyst support is scarce when compared to their use as an adsorbent. Liu et al. (2017, 2016b) investigated biosolids derived pyrolytic biochar as a catalyst and found that metal content (i.e., Ca and Fe) present in the biochar might be responsible in decreasing bio-oil yield and increasing py-gas yield as shown in Fig. 6. The use of biochar in a microbial fuel cell (MFC) as a catalyst is studied by Yuan et al. (2013). They built a catalytic layer air cathode coated with biosolids' biochar. The cathode power density of the biosolids' biochar catalyst layer was comparable with Pt/C-layer of the cathode, reached  $500 \pm 17 \text{ mW/m}^2$ . Wang et al. (2017b) demonstrated the use of Ni loaded biosolids derived biochar as an electrode for supercapacitors and found that loss in capacitance was less than 2% after 1000 charge–discharge cycles.

A number of lignocellulosic precursors, such as coconut shell (Jin et al. 2013; Kim et al. 2004; Muradov et al. 2005; Prasad et al. 2010), palm shell (Abbas and Daud 2010) and olive stones (Mahmoudi et al. 2017) and hardwood (Bai et al. 2005; Prasad et al. 2010) were used as raw materials for the production of biochar or activated char which later applied in catalytic methane decomposition as shown in Table 12. The work on the use of biochar and activated char produced from biosolids for catalytic methane decomposition was first demonstrated by

**Table 12** Catalytic methane decomposition (CMD) by biochar reported in the literature

Catalyst	Reactor	Methane concentration (%)	Methane conversion (%)	Reaction temperature (°C)	Reaction time (h)	Conversion before drop (%) / time interval	Final conversion (%) / time interval	WHSV ( $\text{L gm}^{-1} \text{ h}^{-1}$ )	References
Biosolids biochar	Fixed bed	10	68–42	900	6	68	42	6	Patel et al. (2020)
		50	57–32			57	32		
		90	52–25			52	25		
		10	35–10	700		35	10		
		50	32–2			32	2		
		90	25–0			25	0		
Biosolids Activated char	Fixed bed	10	72–40	900	6	72	40	6	
		50	63–24			63	24		
		90	60–20			60	20		
		10	43–7	700		43	7		
		50	32–0			32	0		
		90	29–0			29	0		
Coconut shell activated carbon	Fixed bed	20	22–15	850	4	22	15	15	Jin et al. (2013)
		99.99	52–10	850	4	35/5 min	55/5 min	1.62	
Hard wood	Fixed bed		45–12						Prasad et al. (2010)
Vegetable based									
Coconut activated carbon			52–17						
Coconut activated carbon	Fixed bed	99.99	35–20	850	1.5	35	20/30 min	0.1	Muradov et al. (2005)
Coconut shell activated carbon	Fixed bed	99.99	9–3	850	2	9	3/20 min	15	Kim et al. (2004)
Palm shell activated carbon	Fixed bed	99.99	45–5	850	3	45	5/30 min	0.35–2.1	Abbas and Daud (2010)
Olive stones activated carbon	Fixed bed	99.99	6–0.2	850	1	6	0.2/60 min	0.9	Mahmoudi et al. (2017)
Hardwood activated carbon	Fixed bed	99.99	33–10	900	2	33	10/120 min	3	Bai et al. (2005)

Patel et al. (2020). He observed that biosolids' biochar and activated char performed slightly better than biomass biochar and activated char. He concluded that better conversion rates with biosolid's biochar and activated char might be attributed to catalytic activity enhanced by heavy metals present in them.

Biochar use in advance applications as mentioned above can generate higher revenue and expected to improve the commercial viability of the biosolids to biochar route. However, more scientific and demonstration work is needed in this area to prove that these applications are techno-commercially feasible.

## 8 Process modelling, techno-economic assessment and large scale demonstrations

Process modelling of pyrolysis using ASPEN Plus or Hysys is a challenging task due to varied solid's feedstock, complicated primary and secondary reactions involved in the pyrolysis process and complex pyrolysis-oil composition comprising more than 100 compounds. There are very limited number of process modelling related studies published so far for the slow pyrolysis of biosolids. McNamara et al. (2016) developed a simple energy balance code and concluded that pyrolysis of dried biosolids can be energy positive and the energy required for pyrolysis of dried feedstock can be five times lesser than the energy required for drying biosolids Patel et al. (2019c). developed ASPEN Plus process model and performed detailed energy balance to study the feasibility of the production of biochar from biosolids. The modelling suggested that moisture content lower than 50 wt.% in biosolids can make pyrolysis process energy neutral.

The article published in 2008 focused on the preliminary techno-economic analysis to produce pyrolysis-oil from primary (collected from the primary settling tank), thickened waste activated sludge (TWAS) and digested biosolids (collected as cake after anaerobic digestion and dewatering). The results concluded that primary sludge pyrolysed at 500 °C produced the greatest economic benefit (i.e., 9.9 Canadian ¢/kg-dry solids) (Kim and Parker 2008). Patel et al. (2019c) performed a techno-economic assessment of the slow pyrolysis of 6 tonnes/day biosolids with an aim to produce biochar. The Net Present Value (NPV) calculations suggested that biosolids management cost and biochar sale price

were the most critical factors in establishing commercial viability.

There are significant moves afoot with demonstration/commercial-scale process development of biosolids pyrolysis. The summary of the key demonstrations is provided below.

### 8.1 EnerSludge (Bridle et al. 2000; Bridle and Pritchard 2004; Bridle and Skrypski-Mantele 2004)

The EnerSludge process developed by Environmental Solutions International Ltd (Australia) utilized temperatures near 450 °C, used catalysts and focused on the production of oil. The process has also been referred to as the oil-from-sludge process. The Ener-sludge process was first demonstrated in Subiaco wastewater treatment plant, Perth, Australia. Dewatered undigested sludge between 26 and 28% solids was first fed into a dryer, which produced pellets at 95% solids that were later fed into the reactor. The EnerSludge system had two pyrolysis reactors zones (i) volatilisation Zone, and (ii) reaction zone. In the first zone, approximately 50% of the sludge was vapourised. The oil and char from the first reactor were directed to a second reactor. In the second reactor, catalysts (alumina silicates and heavy metals) were used to convert the generated oil vapours into short chain hydrocarbons. These vapours were condensed in a second reactor and then an oil/water separator was used to refine the product. The oil product was further refined in a hydro-cyclone to diesel fuel. The energy requirements for the EnerSludge process were satisfied by burning char, non-condensed gas and reaction by-products in Hot Gas Generator (HGG). The Perth's EnerSludge facility was operated from September 2000 to December 2001 and then stopped when a more cost-effective lime conditioning process was installed.

### 8.2 BioForceTech (Bioforcetech 2019b)

The BioForceTech (USA) plant is composed of multiple biodryer units coupled with a pyrolysis reactor. Pyrolysis process, which is a screw type reactor operates between 350 and 600 °C. The biodryer coupling with pyrolyser allow high mass reduction rate; ~ 78 kg of char is obtained from one ton of initial biosolids (20% solid content). If the biosolids' moisture content is equal or higher than



20%, no external fossil fuel is needed for the drying process because the oil and gas vapours produced during pyrolysis are combusted and used to generate heat, which is used in the drying process. The first plant is currently under operation at Redwood City, CA, USA since 2017.

### 8.3 Anaergia (Josse et al. 2016)

The Anaergia pyrolysis process is developed by Anaergia Inc. (USA). The process is performed in a screw type reactor. The operating temperature is found to be in the range of 300–550 °C and the focus is on the production of biochar. The processes can handle 500 kg/h of dried biosolids pellets. A pyrolysis liquid and gas are used in an anaerobic digester to enhance biogas production. It can handle biosolids with < 45% moisture. The first plant is currently under operation at Carlsbad, CA, USA since 2013.

## 9 Conclusions, current challenges, research gaps and future perspectives

The biochar production from biosolids has been extensively reported in the literature. By and large, the literature has concluded that biosolids to biochar via pyrolysis can be an effective option for biosolids management. It helps reducing biosolids volume as well as add value to them by converting them into high-quality biochar. It is also established that biochar produced from biosolids, can have applications as soil amendment materials, adsorbent and catalysts due to its excellent physico-chemical properties. Despite this fact, the commercial viability of the existing pyrolysis process for converting biosolids to biochar is still very limited in the current scenario. The major research gaps and challenges that need to be addressed for this route to be commercially viable are highlighted below with some recommendations.

### 9.1 Requirement of biosolids drying

The biosolids obtained at different treatment levels in wastewater treatment plants can have a moisture content of 75–80 wt%. Therefore, they require drying prior to their introduction in the pyrolysis process. The moisture can have a significant impact on the pyrolysis process efficiency and performance. For example, wet

biosolids are difficult to grind or sieve. Moreover, wet biosolids, if used in pyrolysis processes without drying, can agglomerate or sinter the reactor bed. It can also increase exergy losses of the system as moisture will take a significant amount of heat away from the system without it being fully recovered. Drying of biosolids with such high moisture content can be highly energy-intensive as well. Newer designs of slow pyrolysis process have introduced an integration step where oil and gas vapours can be combusted in combustor while keeping biochar as the main product. The energy generated from the combustor can be used for the energy required for drying as well as pyrolysis. It has been reported by Patel et al. (2019c) that greater than 50 wt.% moisture in biosolids would require external energy even for such integrated pyrolysis process. Therefore, solar, air convection or bio-additive based drying methods should also be researched in future for biosolids drying, which can make the process self-sustainable without the requirement of any external fossil or renewable fuels including biogas.

### 9.2 Transport of biosolids and requirement of mobile pyrolysis processes

Wider distribution of wastewater treatment plants is one of the major bottlenecks of biosolids management or even for its conversion to biochar via pyrolysis. High moisture content in biosolids and their decentralised production may incur a significant transport costs and associated emissions if they are required to be sent to a centralised waste to resource facility such as pyrolysis. Therefore, a decentralised small scale pyrolysis systems would be more preferable as it can avoid transport of biosolids. However, majority of the existing pyrolysis processes become commercially viable only at medium to large scale. Therefore, future efforts should be made in designing a novel but simple and cost-effective small scale mobile pyrolysis units so that biosolids transport can be avoided.

### 9.3 Secondary emissions/risks from biosolids pyrolysis

Biosolids pyrolysis can generate emissions of SO<sub>x</sub>, NO<sub>x</sub>, mercury, polyaromatic hydrocarbons (PAHs) dioxins and furan. It has been found that NO<sub>x</sub> emissions with biosolids pyrolysis is found to be

relatively lower when compared with biomass. This is due to the fact that organic nitrogen in biosolids may generate ammonia at medium-higher pyrolysis temperature, which can act as a NO<sub>x</sub> reducing agent during the combustion of oil and gas vapours (Jensen et al. 1995; Patel et al. 2019b). It has also been suggested that fluidised bed reactors reduced the PAHs formation due to the effective heat and mass transfer (Rollinson 2016). However, dedicated studies on secondary emissions/risks are still limited in the current literature, and therefore, future research should target on the comprehensive long-term measurement and reduction of the secondary emissions generated from the biosolids pyrolysis process.

#### 9.4 PFAS, micro-plastics and pharmaceuticals destruction in biosolids pyrolysis

Pyrolysis is a thermal process. The temperature (i.e., 400–700 °C) at which the pyrolysis operates is expected to completely destroy pathogens, pharmaceuticals, pesticides and micro-plastics from biosolids. It is also hypothesized that PFASs from biosolids at this temperature will be vaporised and destructed partially/completely in the subsequent combustor where gas and oil vapours are combusted (Bio-forcetechnology 2019a; GHD 2019; Surti 2019). There is a possibility that, if PFASs not fully destroyed, at least long-chain PFASs might be converted into shorter chains. However, no scientific studies on biosolids are published yet demonstrating this. Also, the fate of fluorine post-pyrolysis is not fully understood with biosolids being the feedstock. More research should be carried in the future demonstrating the ability of pyrolysis to destruct some of the key emerging contaminants from biosolids and reduce the secondary risk from them polluting air, water and land.

#### 9.5 Nutrient recovery from biosolids pyrolysis

Biosolids derived biochar is expected to keep nutrients into their matrix and act as a slow release fertiliser when applied to the soil. However, the literature suggests that some macro and micro-nutrients from biosolids will partly release (i.e., 30–50%) into the gas phase during pyrolysis and might be obtained in the aqueous stream during the scrubbing of flue gas (Hossain et al. 2011; Zielińska et al. 2015). The research on aqueous stream and its potential to be used

as a source of the nutrient is not reported in the literature. In the future, more efforts should be made to evaluate the potential of nutrient recovery from the aqueous stream of the integrated pyrolysis-combustion process.

#### 9.6 Effect of biosolids derived biochar on soil biota

It has been established in the literature that microbial biomass increases as a result of biomass derived biochar additions. This is due to significant changes in microbial community composition and enzyme activities that may explain the biogeochemical effects of biochar addition on element cycles, plant pathogens and crop growth (Zhang et al. 2014a). However, similar studies on the biosolids derived biochar are currently lacking in the literature and therefore, should be encouraged in the future.

#### 9.7 Lack of policy framework and guidelines for biosolids derived biochar

The consistency in policy framework and guidelines for both biochar production and use lacks at state, national and regulatory body levels. The biochar market is also not regulated currently neither by price nor by quality/standards. Same applies to biosolids derived biochar. More collaborative research in the future should be encouraged at state, national and regulatory levels to ensure consistency in policy framework and guidelines for both biochar production and use.

#### 9.8 Benchmarking of biosolids derived biochar with biomass derived biochar

The biomass derived biochar can be significantly different from biosolids derived biochar. This is due to the fact that biomass and biosolids are quite different in their physico-chemical properties. For example, the biosolids have higher ash content compared to biomass. The biosolids, unlike biomass, may have polymers present in them, which are used as sludge thickening agents during wastewater treatment. Different structures and composition of biosolids make them distinctively different feedstock for pyrolysis. For example, Patel et al. (2019b), in their recent study, identified that pyrolysis of biosolids required higher

temperature (i.e., 700 °C) compared to biomass pyrolysis (i.e., 500 °C) to produce high surface area biochar. More detailed comparison/benchmarking of biomass and biosolids derived biochar should be carried out in the future to explore the potential use of biosolids in the pyrolyser designed for biomass and co-pyrolysis.

#### 9.9 Increased concentration of heavy metals in biosolids derived biochar

Biosolids generally have higher heavy metal content, which are expected to further increase in the resultant biochar after pyrolysis. However, it has been found in the literature that heavy metal content in biosolids derived biochar are immobilised, remain bio-unavailable and their values are still under the regulatory limits proposed by international biochar guidelines (Paz-Ferreiro et al. 2018; Yang et al. 2018b). However, it is not clear for how long the heavy metals will remain immobilised in the biochar. More research should be conducted to answer this question. Also, co-pyrolysis of biomass (and biosolids) should be considered in the future to reduce heavy metal concentration in the resultant biochar. If bio-availability still remains as a major concern for biosolids' biochar for land applications then, in future, more efforts should be made to develop non-agriculture applications such as their use as adsorbents, catalysts and energy storage and generation materials. Particularly, biosolids' biochar role in catalytic methane decomposition can be more attractive as metals can be seen highly favourable in catalysing the reaction.

#### 9.10 Lack of pilot scale demonstrations and life cycle assessments

Despite extensive publications in this area, so far, there are only three successful pilot-scale demonstrations of biosolids pyrolysis reported in the literature. More such demonstrations should be supported in the future. Also, the majority of the experiments reported in the literature are batch or semi-continuous type. Continuous flow experiments are essential in understanding the process performance and efficiency before going to large scale pilot-scale demonstration.

It has been established that compared to other biosolids management options, pyrolysis achieves a significant reduction in the life cycle emissions

(i.e., > 25%) (Miller-Robbie et al. 2015; Peters and Rowley 2009). However, more life cycle assessment studies including different scenarios such as mobile (decentralised) versus centralised pyrolysis unit, urban versus regional wastewater treatment plant, dried versus wet biosolids, biochar's use as soil amendment versus adsorbent/catalyst should be studied in the future to realise the potential of biosolids pyrolysis over the entire life cycle.

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