A review on wastewater sludge valorisation and its challenges in the context of circular economy

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Abstract: The use of wastewater sludge as a source for energy and resource recovery is a good alternative for its management considering the legislation requirements and the circular economy principles. Recognizing sludge as a resource, not as a waste, has made researchers consider the recovery of valuable components from sludge, such as carbon and nutrients. The energy that can be obtained from wastewater sludge may be a sustainable solution to fulfill present and future energy requirements. This review discusses about the types of sludge produced by wastewater treatment plants (WWTPs), the technologies that can be implemented in the water and sludge line to reduce the sludge amount, as well as the conventional treatment and disposal methods. Moreover, the technologies that can be used to recover resources and energy in the context of cicular economy are also presented. Finally, a detailed description of some urban biorefineries aimed at the recovery of cellulose and nutrients and the production of bioplastics is reported. The study ends with conclusions and future research directions.

Dear Editors of the Journal of Cleaner Production,

We would like to submit the attached manuscript, entitled **"A review on wastewater sludge valorisation and its challenges in the context of circular economy"** by *Andreea Gherghel, Carmen Teodosiu* and Sabino De Gisi* for* your consideration for possible publication as a Review in ***Journal of Cleaner Production.***

This study presents a comprehensive review that structures the research efforts realised so far related to the types of sludge produced by wastewater treatment plants (WWTPs), the technologies that can be implemented in the water and sludge line to reduce the sludge amount, as well as the conventional treatment and disposal methods. Moreover, the technologies that can be used to recover resources and energy in the context of cicular economy are also presented. Finally, a detailed description of some urban biorefineries aimed at the recovery of cellulose and nutrients and the production of bioplastics is reported.

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Carmen Teodosiu (on behalf of all authors)

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A review on wastewater sludge valorisation and its challenges in the context of circular economy

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There are no interests to declare.

Highlights (for review)

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- Wastewater sludge can be used for materials and energy recovery in circular economy
- Possible biorefineries based on sludge valorization were identified and described
- The recovery of short-chain fatty acids, phosphorus and bioplastics was disscussed
- More than 180 references about wastewater sludge valorization routes were analysed
- Future directions for wastewater sludge management were investigated.

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1. Introduction

The increase of the quantity of wastewater sludge is a global problem in the context of population growth and adequate sanitation in large wastewater treatment plants. Sludge is the solid residue that remains after wastewater treatment (Abelleira et al., 2012), being produced by processes such as activated sludge, aerobic-oxic, anaerobic-anoxic-oxic, oxidation, cyclic activated sludge and up-flow anaerobic sludge bed processes (Gong et al., 2014). The quality of the raw sewage sludge and the treatment technologies used in wastewater treatment plants (WWTPs) can influence the final characteristics of sewage sludge (Kacprzak et al., 2017). Meanwhile, the combination of various physical, mechanical, chemical and biological processes used in a WWPT, is the key for achieving the removal of pollutants from sludge (Anjum et al., 2016). A short review of literature shows an increase of sewage sludge production in Europe and development of new wastewater treatment technologies, due to stringent legislative requirements for wastewater discharges or reuse (Praspaliauskas and Pedišius, 2017). The mains directives regarding the wastewater sludge management in Europe are presented in Table 1 (Manara and Zabaniotou, 2012).

Table 1. The European Union (EU) legislation on wastewater sludge management

Directives	Highlights
Directive 1986/278/EEC (Sewage Sludge Directive)	Refers to the environmental protection and in particular of the soil, when sewage sludge is used in agriculture;
Directive 1975/442/EEC with its amendments: 1991/156/EEC, 2006/12/EC and 2008/98/EC	The Waste Framework Directive which incorporates the *Polluter Pays Principle* along with the waste hierarchy (article 4 of the Directive is pertinent to the land spreading of wastes);
Directive1991/271/EEC with its amendment 1998/15/EEC (Urban Water Treatment Directive)	Considers the improvement of wastewater treatment processes, increasing the number of existing plants;
Directive 1999/31/EEC (Landfill Directive)	Increases the restrictions on quantities of biodegradable waste that can be landfilled, due to concerns over methane generation under anaerobic digestion;
Directive 2003/33/EEC	Establishes the criteria and procedures for the acceptance of waste at landfills; pursuant to the Article 16 and Annex II of Directive 1999/31/EC;
Directive 2000/76/EEC	Refers to the incineration of waste;
Directive 1989/369/EEC	Refers to the prevention of air pollution from municipal WWTPs;
Directive 1991/676/ EEC	Considers water protection regarding pollution with nitrates from agricultural sources;
Directive 1991/689/EEC	Refers to the controlled management of hazardous waste;
Decision 2000/532/EC	Establishes a list of wastes, as amended.

Although, considered a residue, sewage sludge can be used as a source of energy or resources, thus replacing an equivalent amount of materials/energy that would otherwise need to be produced from non-renewable resources with considerable environmental impacts (Fijalkowski et al., 2017). The concept of circular economy appeared in 1970 and is attributed to Pearce and Turner (1989). Investigating the linear and open-ended characteristics of contemporary economic systems, the researchers described how the natural resources can influence the economy by providing inputs for production and consumption as well as serving as a sink for outputs in the form of wastes. The circular economy concept emerged as an alternative to the "*Take-Make-Dispose*" (linear) economic model and is based on the principles of: cradle-to-cradle, regenerative design, industrial ecology, laws of ecology, biomimicry, looped and performance economy and the blue economy (Geissdoerfer et al., 2017). According to Ellen MacArthur Foundation (EMF) the circular economy is: "an industrial system that is restorative or regenerative by intention and design. It replaces the 'end-of-life' concept with restoration, shifts towards the use of renewable energy, eliminates the use of toxic chemicals, which impair reuse, and aims for the elimination of waste through the superior design of materials, products, systems, and, within this, business models" (EMF, 2010); for short: "*circular economy is the one that is restorative and regenerative by design, and which aims to keep products, components and materials at their highest utility and value, at all times*" (Webster, 2015). The concept of circular economy is presented in Fig. 1.

The moving to circular economy is more than "*A zero waste programme for Europe*", while achieving the European strategies established until 2020, needs to be accomplished by reducing the resources escaping from the circle so that the system functions in an optimal way (Smol et al., 2015).

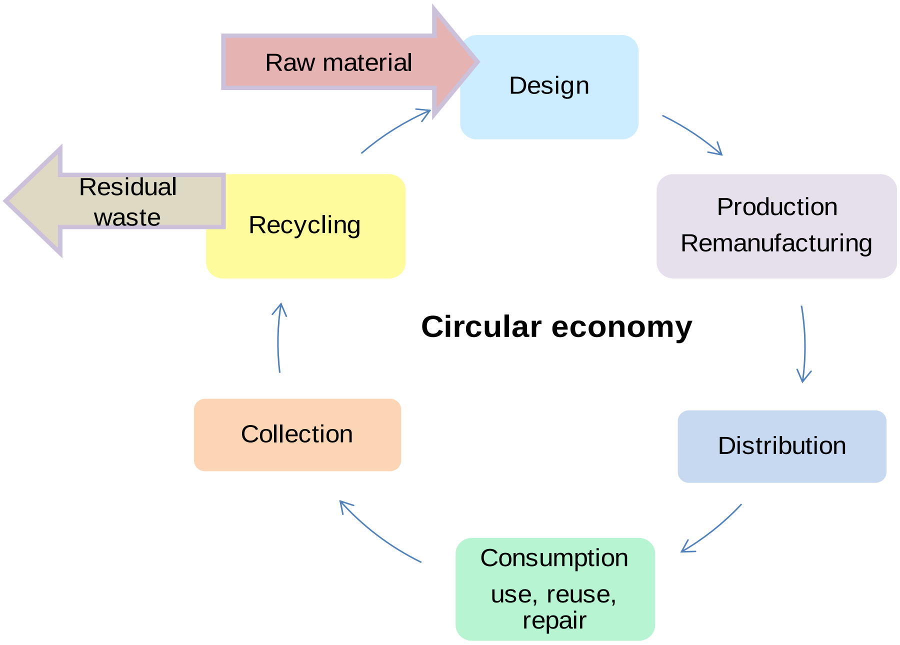


Fig. 1. The concept of circular economy.

Sludge reuse as raw material in different industries represents a good possibility of waste management considering the circular economy concept (Eliche-Qusada et al., 2011). Due to the legislation that limits the landfilling and land application as sludge disposal methods, many studies approached the sludge reuse and recycling to achieve its environmental sustainable waste management. In relation to this, European Commission (2011) considers that "if waste is to become a resource to be fed back into the economy as a raw material, then much higher priority needs to be given to re-use and recycling". Moreover, the combination of policies would help create a full recycling economy and the life cycle approach will be considered in product design, cooperation between markets actors will be improved, the regulatory framework will be appropriate and public investments will grow.

Furthermore, waste material incineration, is also not compatible with the concept of circular economy, due to significant greenhouse gas emissions resulting from this process (Nghiem et al., 2017). Taking into account the fact that organic component from sludge are a rich vein of resources in terms of energy and nutrient waiting to be tapped, a study realized in 2015 by the International Solid Waste Association (ISWA), shows that, in the context of circular economy, an important benefit of the energy and fuels obtained from waste is that they can replace other energy resources and thereby their associated emissions of CO_2.

The main objective of this study is to perform a comprehensive literature assessment regarding the wastewater sludge treatment processes and management used for its valorization in order to recover resources and energy, while taking into account the circular economy concept, and to identify the research issues that need further investigation. In detail, this review approaches the following research questions: (i) the sludge characteristics, its traditional treatment and disposal processes and the legislation changes limiting their use; (ii) the new technologies that may be used to reduce the amount of sludge; (iii) the valuable components/materials which can be recovered from sludge as resources; (iv) the integrated recovery of resources and energy from sludge in urban biorefineries.

2. Methodology

The selection and analysis of the scientific literature was made considering the following criteria:

a) *Relevant international information databases*. Bibliometric resources such as: Science Direct, Scopus, Web of Science were used to retrieve articles, book-chapters and international proceedings. European Commission or other organisations databases were also consulted; the relevant content included 183 articles (in journals or conference proceedings), reports and legal documents;

b) *Publication period*. The majority of references (95.62%) are from 2008 to 2018 (175 references from a total of 183 references), the rest of 4.38 % references belonging to 1989-2007;

c) *Relevant keywords*. The following keywords have been used in different combinations: *WWTP*, *circular economy*, *sludge valorization*, *treatment technologies*, *resource recovery and energy recovery*. About 63% from the references, were used to describe the methods for recovery of valuable components from sludge (40% for resource recovery and 23% for energy recovery);

d) *Selection of references based on content analysis.* After eliminating the articles referring to industrial sludge or other types of waste valorization (biomass, fly ash based wastes, etc.) the remaining articles/book chapters were analyzed thoroughly. Abstracts of all references left after this screening process were analyzed;

e) *Analysis of the data selected and structure of the review*. The selected scientific literature was presented based on the concept depicted in Fig. 2, considering the main resources that can be recovered from sludge and the technologies used. The data presented in this study gives an overview of all the stages of the sludge stream, from sludge production until its landfilling or valorisation. In other words, we can say that this study describes the "*sludge life cycle*" from the circular economy point of view.

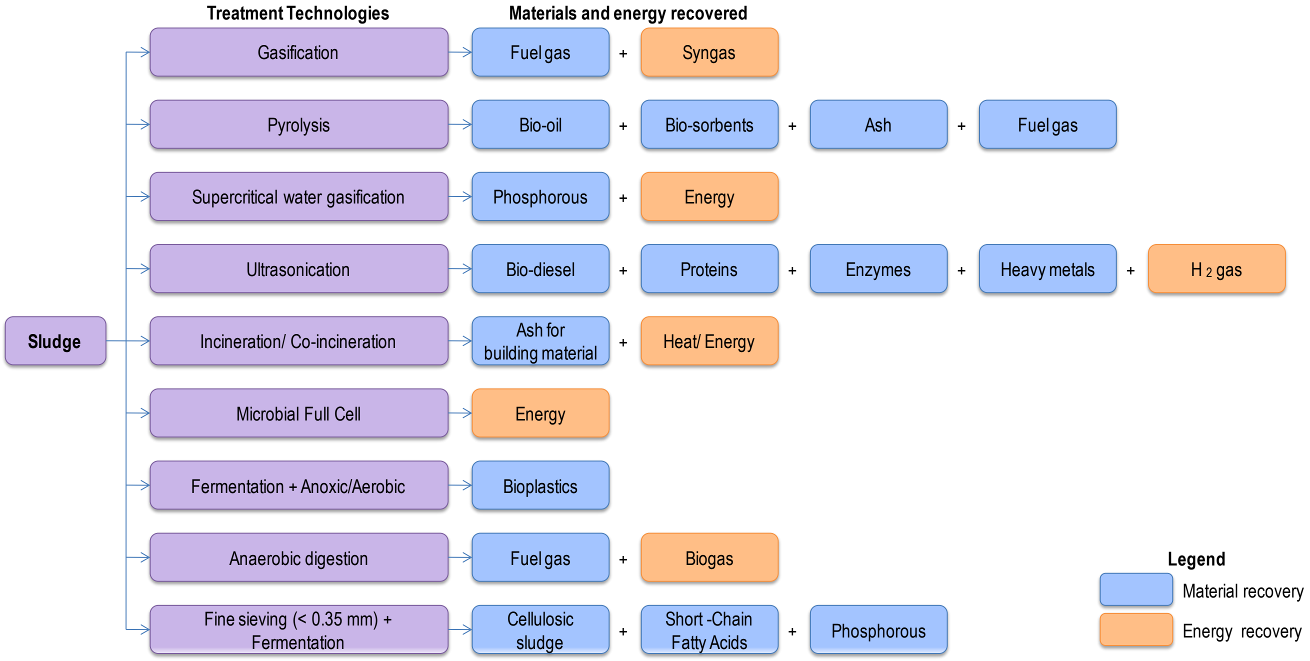


Fig. 2. Possibilities to recover material resources and energy from wastewater sludge.

Therefore, after the methodology (Section 2), the types of sludge and traditional treatment and disposal processes are presented (Section 3), along with the technologies used in wastewater and sludge lines, for reduction of sludge production. In the next three sections there are described the technologies that can be used for resources (Section 4) and energy (Section 5) recovery from sludge, as well as the integrated integrated recovery of resources and energy from sludge in urban biorefineries (Section 6). The paper ends with conclusions and future research directions.

3. Wastewater sludge characterization, treatment and disposal practices

3.1. Sludge caracterization

In a municipal WWTP, depending on the treatment stage, several types of sludge are generated, as follows:

1. **Primary sludge** is produced during the primary treatment (screening, grit removal, flotation, precipitation and sedimentation), when heavy solids, grease and oils are separated from raw wastewater (Manara and Zabaniotou, 2012; Tyagi and Lo, 2013; Suárez-Iglesias et al., 2017). Usually, primary sludge contains 2% to 10% solids, the remaining 90%, (sometimes even 99.5%) being water (Tyagi and Lo, 2013; Moran, 2018);
2. **Secondary sludge** (waste activated sludge) is produced during biological treatment, when the microorganisms decompose the biodegradable organic content from wastewater (Devi and Saroha, 2017). The total solids concentration is between 0.5-1.5%, depending on the type of biological treatment process employed (Tezel et al., 2011; Moran, 2018), the rest being water. The organic portion from waste activated sludge contains: carbon 50–55%, oxygen 25–30%, nitrogen 10–15%, hydrogen 6–10%, phosphorus 1–3% and sulfur 0.5–1.5% (Tyagi and Lo, 2013);
3. **Tertiary sludge** is obtained in the advanced wastewater treatment stages, when nutrients (nitrogen and phosphorus) removal is required (Manara and Zabaniotou, 2012).

According to Gianico et al. (2015), the caracteristics of primary and secondary sludge, in terms of pollutants, nutrients, water and energy contents are different. Because the primary sludge is more polluted, it needs a thermal treatment before disposal, while secondary sludge may be used in agriculture after stabilization, due to its rich nutrients content. However, in order to increase the dewatering potential of secondary sludge, the former is mixed with primary sludge characterized by a higher biodegradability (Carrere et al., 2010; Devi and Saroha, 2017). Table 2 sums up some characteristics of primary and secondary activated sludge (Tyagi and Lo, 2013; Suárez-Iglesias et al., 2017). The main features of different types of sludge from WWTPs were also presented by Bougrier et al. (2008), Manara and Zabaniotou (2012), Anjum et al. (2016) and Nazari et al. (2017).

Table 2. Characteristics of primary and secondary wastewater sludge

Parameter	**Sludge**	
	Primary	**Secondary biological**
Total solids (% TS)	2.0-9.0	0.8-3.3
Organic solids/volatile solids (% TS)	60-80	59-88
Nitrogen (N, % TS)	1.5-4.0	2.4-5.0
Phosphorus (P, % TS)	0.17-2.8	0.5-2.3
Potash (K_2O, % TS)	0-1	0.5-0.7
Cellulose (% TS)	8.0-15.0	7.0-9.7
Iron (Fe g/kg)	2.0-4.0	n.a.
Silica (SiO_2, % TS)	15-20	n.a.
pH	5.0-8.0	6.5-8.0
Grease and fats (% TS)	7.0-65	2-12
Protein (% TS)	20-30	32-41
Alkalinity (mg/L as $CaCO_3$)	500-1,500	580-1,100
Organic acids (mg/L, as acetate)	200-2,000	1,100-1,700
Carbohydrates (% TS)	n.a.	6.1-9.8

Energy content (kJ/kg TS)	2,900-23,000	19,000-23,000

n.a. - not available.

3.2. Sludge treatment and disposal practices

Sludge treatment and disposal are important stages in the context of environmental protection because of its content of residual organic pollutants, toxic metals and pathogenic microorganisms which can cause health problems (Anjum et al., 2016). Sludge treatment requires high amounts of energy and has associated environmental impacts, the cost of sludge treatment representing approximately 50% of the total running cost of WWTPs (Collivignarelli et al., 2015; Qian et al., 2016). It was determined that sludge disposal processes are responsible for 40% of the total greenhouse gas emissions from WWTPs, this percentage could be decreased if the circular economy concept would be applied (Brown et al., 2010; Pilli et al., 2015).

The amount of sludge has increased due to population growth and rapid development of industry (Praspaliauskas and Pedišius, 2017). According to the European Commission Report (EC, 2008), more then 10 million tones of dry solids of sludge were produced in Member States (26 EU) in 2008, and the sludge amount is expected to continue to grow up to 13 million tones by 2020 (Kelessidis and Stasinakis, 2012).

Therefore, many physical, chemical and biological processes have been developed to treat or minimize sludge production (Xu et al., 2014; Praspaliauskas and Pedišius, 2017). The most frequently used methods for the disposal of excessive sludge are: incineration, landfilling, ocean-dumping (Anjum et al., 2016; Qian et al., 2016), reuse in agriculture (directly or after composting) and reuse for production of cement, bricks and asphalt (Zhen et al., 2017).

Even if approximately 40% of the total sludge produced in EU is used in agriculture (Eurostat, 2015), some EU countries adopted strict limit values for contaminants, than those reported in the Sewage Sludge Directive (SSD). Each country has made its decisions, some of them have added new contaminants on the SSD list, while others, considering at the environmental risks of using sludge in agriculture, abandoned this method of sludge disposal (Kacprazak et al., 2017). For example, in 2010, several EU countries such as: United Kingdom, Denmark, France, Belgium and Spain, used more than 50% of sludge in agriculture (EC, 2008), while countries like Netherlands, Greece, Romania, Slovenia and Slovakia didn't used it at all. Smith (2002) argued that the utilization of Best Practicable Environmental Option (BPEO) approach can assess the impacts on all environmental media. Kacprzak et al. (2017) affirm that the agricultural use of sludge is considered a BPEO, but it depends on the agreement of farmers. When the land is used for food production, the specific analysis of sludge and the adoption of measures to stop the migration of contaminants are imperative. Considering the organic compunds and inorganic nutrients that can be taken up from sludge, the agricultural use remained one of the prefered options for sludge disposal (Fijalkowski et al., 2017).

Regarding the landfill disposal of sludge, it is less used due to leachate production and CO_2 emissions which affect the air (Kacprzak et al., 2017) and also due to EU legislation which became more stringent (Manara and Zabaniotou, 2012). According to Kelessidis and Stasinakis (2012), who have studied the methods used for treatment and final disposal of sludge in European countries, between 2000 and 2009 only three countries have reported an increase of landfill use (Italy, Danmark and Estonia).

Instead, the use of incineration increased in many European countries, due to large volume reduction of sludge and thermal efficiency (Manara and Zabaniotou, 2012). Germany and The Netherlands were the countries which developed more this technology, with a percentage of 28% and 16%, respectively (Kelessidis and Stasinakis, 2012). The choice and application of the best sludge management strategy should take into account: a) the costs of gas scrubbing for air pollution control are higher (Manara and Zabaniotou, 2012), b) heavy metals emissions (Kelessidis and Stasinakis, 2012) and c) the indication for incineration in the case of large WWTPs or when the quality of sludge is not suitable for its use on land, according to the law (Kacprzak et al., 2017). Therefore, the increased care for environmental protection, stricter legislation and circular economy implementation has led to the consideration of other methods for sludge minimization and treatment. Biological methods, such as, composting, aerobic and anaerobic digestion, replaced with success the traditional methods because of their benefits: reduction of sludge volume, removal of pathogens and volatile solids and conversion of sludge into stable biosolids (Semblante et al., 2015).

Nowadays, anaerobic digestion (AD) represents one of the most used methods for sludge stabilization because it can reduce odors, pathogenic microorganisms and, volatile solids and obtain biogas from the organic part of the sludge (Nazari et al., 2017). In terms of costs, it is an expensive technology, but considering its energy efficiency to recover methane from sludge, many countries/regions apply it on a large scale, an example being California, with 82% of the total WWTPs that are operating anaerobic digestion for sludge stabilization (Anjum et al., 2016). The AD process consists of several successive biochemical processes such as: hydrolysis, acidogenesis, acetogenesis and methanogenesis, presented in detail by Zhen et al. (2017). According to literature data (Gianico et al., 2015; Nazari et al., 2017), the hydrolysis step controls the anaerobic digestion rate. To overcome this limitation, a number of pre-treatments are required, such as: thermal hydrolysis, ozonation, alkcaline hydrolysis, enzymatic lysis, freezing and thawing, mechanical desintegration, high pressure homogenizers, ultrasound, microwave irradiation and photocatalytic pre-treatment (Zhang et al., 2010; Chang et al., 2011; Liu et al., 2013). The aim of these pretreatments is to destroy the microbial cell walls, release the extracellular and intracellular organic compounds which lead further to an accelerated subsequent biological treatment and a smaller solid retention time required for the digestion sludge proccess. Anjum et al. (2016) presents each of these pretreatments used to improve sludge AD preformance, which can be also combined between them.

Aerobic digestion is another method used for sludge (dewatered or thickened) stabilization, that takes place in a completely aerated reactor and is influenced by the system temperature and the retention time (Semblante et al., 2015). The proccess is characterized by mesophilic or thermophilic temperatures, the last being more used (Anjum et al., 2016). For example, Jin et al. (2015), studying the efficiency of autothermal thermofilic aerobic sludge digestion by chemical approach, reported that after 10 days of agitation of sludge with oxigen at 50 ^{o}C, volatile solids were removed in a percentage of 38%. Liu et al. (2012), expressed that the aerobic digestion process provided a rapid degradation of biomass in a short retention time at a high temperature, with inactivation of pathogenic microorganisms. However, when the temperature is higher than 35 ^{o}C, the accumulation of ammonium nitrogen takes place in the system due to inhibition of nitrification and denificatrion processes, which will reduce the bacterial activity and in the end the sewage sludge stabilization (Yuan et al., 2014).

Composting processes involve treatment and conversion of sludge into a stabilized product, which can be used as organic fertilizer or value added product (Anjum et al., 2016). The main factors that influence the microbial development and the organic matter stabilization are: pH, C/N ratio, and moisture (Ezzariai et al., 2018). In other words, in aeration conditions, the complex substances from sludge can be transformed in simple materials due to production of hydrolytic enzyme and increase of specific growth rate of microorganis. The sludge composting process takes place in three stages, being influenced by temperature. In the first stage, mesophilic microbiota grows with increased system temperature, following by the activation of thermophilic microbiota, where pathogenic organisms die due to high temperatures in the second stage. Finally, in the last stage, the temperature decreases and the mesophilic population is reactivated (Anjum et al., 2016). Similarly with AD, to guarantee the stability of composting, the process requires the addition of a bulking agent, such as sawdust (Elia Ruda et al., 2013).

The composting process has some limitations such as: the sludge complex characteristics, unavailability of microorganisms, temperature loss or the presence of pathogens. Despite such disadvantages, composting of sludge is still applied with succes at laboratory scale and at full scale (Elia Ruda et al., 2013; Anjum et al., 2016). Countries like Estonia, France, Slovakia, Sweden, Hungary and Czech Republic are composting the major quantity of their sludge (Praspaliauskas and Pedišius, 2017). A limitation in utilization of sludge composting is the compliance with the requirements for organic fertilizers (Fijalkowski et al., 2017).

With all this traditional and biological processes used for sludge disposal, sludge management is still a concern at global level. Therefore, because the use of sludge disposal methods at the end of WWTPs didn’t have favorable results, researchers tried to solve the problem from inside the plant, according to the Waste Framework Directive (Directive 2008/98/CE), by applying new process technologies.

3.3. Reduction of sludge in the wastewater/sludge treatment line

The technologies used to reduce the amount of sludge may be applied in the wastewater treatment line or sludge treatment line of WWTPs (Fig. 3). According to the United States Environmental Protection Agency (US EPA) (2012), the sludge reduction technologies on the wastewater line can be applied in WWTPs where AD is lacking; with respect to sludge line, the technologies can be applied in large WWTPs where AD is present. Regarding the wastewater treatment line, a number of mechanical, thermal and chemical treatment technologies have been developed. Among the chemical treatments, ozonation (Gardoni et al., 2011; Romero et al., 2015), Fenton oxidation (He and Wei, 2010), catalytic wet oxidation (Jing et al., 2012; Ureea et al., 2014) and free nitrous acid (Pijuan et al., 2012; Wang et al., 2013) are proposed. He et al. (2011) and Mohammadi et al. (2011) have applied in their studies ultrasonic treatment to reduce sludge production. Instead, Abelleira et al. (2012) and Heinz (2007) used thermal treatment and electrical treatment, respectively. Other treatments applied on the wastewater treatment line are: the addition of a chemical un-coupler (Guo et al., 2014; Zuriaga-Augusti et al., 2016) and combined process (Semblante et al., 2014), with physical and biological treatment stages. Some others researchers have proposed an organism-based treatment (protozoa and metazoa) (Khursheed and Kazmi, 2011; Zhang et al., 2013), while others the replacement of conventional activated sludge process (CAS) with new biological processes such as the sequencing batch biofilter granular reactor (SBBGR) (Di Iaconi et al., 2010; Lotito et al., 2012). Di Iaconi et al. (2010) mentioned that SBBGR sludge production was equal to 0.1 kgSS per kg of removed organic matter (expressed as chemical oxygen demand, COD), 5-6 times lower than the sludge quantity resulted from a CAS system.

Referring to the sludge treatment line, some of the technologies used in the wastewater treatment line, can also be applied here such as: ultrasonic pretreatment (Donoso-Bravo et al., 2010; Martinez-Guerra and Gude, 2015), thermal pretreatment (Perez- Elvira and Fdz-Polanco, 2012; Albelleira- Peraiva et al., 2015) and ozonation (Erden and Filibeli, 2011; Silvestre et al., 2014). The physical treatments used in sludge treatment line are: microwave pretreatment (Uma Rani et al., 2013; Yeneneh et al., 2015), focused pulsed technology (Lee et al., 2010), lysis-thickening centrifugation (Wang et al., 2017), high-pressure homogenization (Zhang et al., 2012) and stirred ball milling (Anjum et al., 2016). Others researchers focused in their studies on chemical pretreatment. For example, Zhang et al. (2010) applied alkaline pretreatment, while Zahedi et al. (2016) used the free nitrous acid pretreatment. Instead, Bolzonella et al. (2012) applied the biological pretreatment to increase the efficiency of AD. In his review, Wang et al. (2017), presented all the technologies used for sludge reduction, with their advantages and disadvantages, together with a comparison analysis between them.

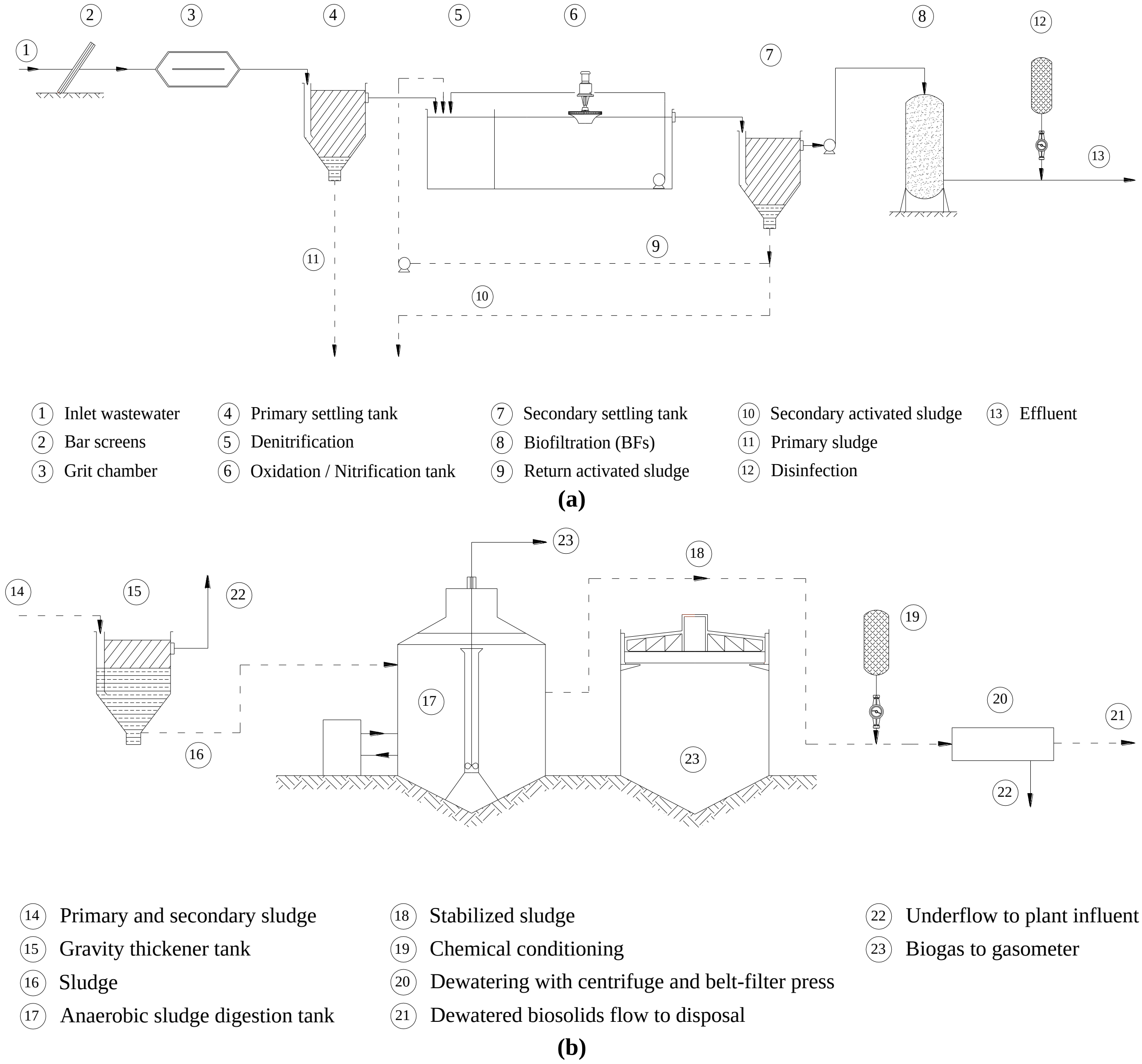


Fig. 3. Possible locations for sludge reduction technologies of a municipal WWTP: in the wastewater line (**a**) and in the sludge line (**b**).

4. Resources recovery from wastewater sludge

4.1. Nutrients recovery

Considerable quantities of nutrients (approximately 0.5-0.7% phosphorus and 2.4-5.0% nitrogen) are contained in the sewage sludge, in form of proteinaceous material that can be used to produce plant fertilizers (Tygi and Lo, 2013). Due to the fact that phosphorus is no longer an inexhaustible resource, along with the higher cost of commercial fertilizers and more demanding legislative requirements, many biological and chemical processes have been developed to recover nutrients from wastewater and sludge (Kleemann et al., 2015; Zhou et al., 2016). Recovery and recycling of phosphorus is considered a possible circular economy pilot, i.e. a potential case to "demonstrate that circular principles work in practice" (Ellen MacArthur Foundation, 2015).

Crystalization is a process used to recover phosphorus from WWTPs, in form of struvite (magnesium ammonium phosphate hexahydrate), which can be used like fertilizer and binding material (Kumar and Pal, 2013; Guadie et al., 2014). Nowadays, there are few such processes commercially available. For example, AirPrex® is a process which precipitates struvite from a mixture of water and sludge in an upstream process of dewatering by dosing $MgCl_2$ and increasing pH (Zhou et al., 2016). Struvia™ and Pearl® are two other technologies that are based on the same principle by using crystallization reactors. Even if struvite crystalization is a good alternative for P recovery, it is still not widely adopted (Pastor et al., 2010), because of some limiting operating factors such as: pH, temperature, supersaturation and foreign ions (Guadie et al., 2014).

To improve the production of struvite, different types of reactors were applied, such as a fluidized-bed reactor (FBR) (Le Corre et al., 2007; Bhuiyan et al., 2008; Guadie et al., 2014) and mechanical stirring reactor (MSR) (Pastor et al., 2008), which over the years have supported a number of improvements. PHOSPAQ® technology was used for the first time to recover phosphate via struvite in an aerated continuous stirred tank reactor (Driessen et al., 2009). In 2006, this technology was used at plant-scale in Olburgen (The Netherlands) to recover phosphate from a mixed influent (anaerobically treated and reject water from an industrial WWTP) (Desmidt et al., 2014). The amount of struvite that can be obtained daily is 1.2 tons (Abma et al., 2010).

Phosphate can be also recovered by adsorption, in two stages: adsorption and desorption of phosphate. After desorption the resulted ash or elution solution, rich in P, can be used in land application. Also the desorption solution can be processed by chemical precipitation to obtain phosphate precipitates (Ye et al., 2017). Metal-based adsorbents are the most studied for the P adsorption, due to their accessibility. Li et al. (2013) had applied a solid-state nuclear magnetic resonance (NMR) spectroscopy to investigate the mechanism of phosphate sorption on aluminum hydroxides under different environmental conditions, and reported that this can be a useful analytical tool for studying phosphorus chemistry at environmental interfaces.

Also wet-chemical treatment and thermochemical treatment can be used for phosphate recovery from sludge. Wet-chemical technology can release the phosphate from sewage sludge and sewage sludge ash by adding strong acids or alkalis to the liquid phase, removing at the same time the heavy metals and pathogens from the supernatant (Ye et al., 2017). The performance of this process is influenced by pH (Cokgor et al., 2009) and temperature (Xie et al., 2011). Seaborne® process is a wet-chemical technology, in which pH of the digested sewage sludge is adjusted to 4 by using H_2SO_4, when phosphate, organic matter and some heavy metals are simultaneously dissolved. Regarding the thermochemical treatment, AshDec® process was used to recover phosphate from sewage sludge ash in a rotary kiln (Ye et al., 2017). Egle et al. (2016), present a classification of the P recovery technologies depending on the source of the phosphorus: aqueous phase, sewage sludge and sewage sludge ash. Even if the amount of phosphorus recovered with all these technologies is high, the costs involved represent an important impediment for their application.

4.2. Heavy metals recovery

The presence of heavy metals (i.e., Zn, Ni, Pb, Hg, Cr, Cu and Cd) in sewage sludge restrict its use for land application due to probable soil and groundwater contamination, which can further affect the human and animal health (Tyagi and Lo, 2013). Therefore, a series of physical, chemical and thermal processes have been applied to remove heavy metals from sludge. Jamali et al. (2009), used microwave treatment to extract heavy metals (Zn, Pb, Ni, Cr, Cu and Cd) from sewage sludge, obtaining a recovery of 95.3-100%. Wu et al. (2009) combine H_2SO_4 with microwave treatment and reported that 90% of Cu can be extracted from sludge.

Utrasonication-assisted acid leaching method was used by Li et al. (2010), to recover some heavy metals from sludge. High recovery rates (Cu: 97.42%, Ni: 98.46%, Zn: 98.63% and Cr: 98.32%) have been reached. Xie et al. (2009) by using the same process reported that recovery of heavy metals was efficient, with low costs and great end product quality and the most important, no waste emission. Wet-chemical treatment (Ye et al., 2017), and ion exchange (Donatello et al., 2010) may be applied also to remove heavy metals from the P-rich leachate.

In addition, most of the heavy metals from sludge can be removed by thermo-chemical treatment, due to the formation of volatile heavy metal chlorides (Herzel et al., 2016), which can be captured in the flue gas, that can be further treated by filtration (Vogel et al., 2016). He et al. (2010) analyzing the potential bioavailability of Cu, Cd, Pb and Zn in sewage sludge, reported that pyrolysis enhances the stability of these metals, when the temperature rises up to certain values. Regarding the sludge gasification process, the major concern is the content of heavy metals remaining in ash dust (Manara and Zabaniotou, 2012). Saveyn et al. (2010) reported that after sewage sludge gasification, some metals (Cu, Zn, and Pb) can be retrived in the char, while others (Hg and Cd) are depleted from the sewage sludge and end up in different downstream fluxes.

4.3. Adsorbents

Pyrolysis treatment of sewage sludge is an attractive process because it can reduce the sludge volume and in the same time it can produce sewage sludge-based adsorbents (SBAs) (Lin et al., 2012). The preparation, characterization and utilization of SBAs have been reviewed by Smith et al. (2009) and Xu et al. (2015). The preparation of SBAs involves first a pyrolysis process and then an activation process. Pyrolysis of sewage sludge, takes place under inert atmosphere, at high temperatures (400-1000°C) with release of volatile matters, in order to obtain char as final product. After the pyrolysis process, the SBAs activation step takes place, which can be physical or/and chimical. If physical activation refers to carbonization of sewage sludge followed by activation with CO_2 or steam, chemical activation may be realised together with pyrolysis process in presence of dehydrating reagents like NaOH, KOH, H_2SO_4, H_3PO_4, K_2CO_3 and $ZnCl_2$.

Alvarez et al. (2015; 2016) focused on sludge valorization, obtaining adsorbents from sewage sludge pyrolytic char by carbon dioxide activation in a conical spouted bed reactor at 500°C. Villamil et al. (2016) when analyzing the potential of powdered activated carbon (PAC) to mitigate membrane fouling used air for activation. Chemical activation using K_2CO_3 was used by Cheng et al. (2016) to enhance the porosity and surface area of carbonized sludge. The literature data confirm that sludge is a promising feedstock for the production of adsorbents (Smith et al., 2009), their conversion representing an attractive alternative for safe sludge management (Xu et al., 2015).

Due to limitations like long processing time and high energy consumption, the pyrolysis process started to be less used, in favor of microwave heating. Yuen and Hameed (2009) reported that in comparison with the pyrolysis process, microwave heating has the advantages of higher heating rates, greater control of the heating proces and most important, a part of energy can be saved. Lin et al. (2012) used in their study a pilot-scale microwave heating equipment to prepare carbonaceous adsorbents in order to remove Cu^{2+} and Pb^{2+} ions from aqueous solutions. For the activation treatment, KOH, H_3PO_4 and $ZnCl_2$ were used, whereas the SBAs prepared via H_3PO_4 activation was the best adsorbent to remove the heavy metals from aqueous solution.

All these applications confirm that sludge can be a promising feedstock for the production of adsorbents and its conversion represents a good solution for sludge disposal methods and reuse routes. Furthermore, SBAs prepared from sludge have a great potential, competing with commercial activated carbon for pollutant removal from wastewaters (Lin et al., 2012).

4.4. Construction materials

The organic carbon-containing complexes and inorganic composites from sewage sludge reprezent a source of valuable materials, that by thermal treatment (Tyagi and Lo, 2013), can be transformed in products like artificial lightweight aggregates, slags and bricks (Wang et al., 2008).

Świerczek et al. (2018) affirm that the use of sewage sludge in mortars or construction materials eliminates some of the expensive and energy-intensive stages of their disposal and more important, environmentally harmful wastes are transformed in safe and stable products. Furthermore, Paris et al. (2016) reported that the addition of sludge in raw form to the production of cement and mortar products can be an alternative to the existing methods of its management.

Nowadays, the combustion of excess sewage sludge becomes a frequent solution due to the possibility of sludge hygienisation and, at the same time, reduction of its volume. Moreover, the ash that results after combustion can be used as an additive to mineral construction materials, cements or concretes (Tantawy et al., 2012). The use of sludge ash for the production of construction materials is a way to circular economy and can bring large benefits such as a reduction of sludge treatment costs, avoiding the transfer of ashes to landfill and the environmental problems derivated from leaching of their soluble constituents (Smol et al., 2015). According to Świerczek et al. (2018), the use of raw sewage sludge instead of water, during the production of cement mortars and concretes can be an interesting concept. Roccaro et al. (2015), by using aerobically and anaerobically stabilized sewage sludge instead of water, reported that the compressive strength of the concrete decreased from 44 to 39 MPa. Wang et al. (2011) and Yamuna Rani et al. (2015) studied the possibiliy of producing brick with the addition of sludge from industrial WWTP and pharmaceutical WWTP, respectively. Instead, Zhang et al. (2016), focused on bricks made with lake sediments, slag and sewage sludge.

In addition, sludge can be an alternative material for covering landfills by dewatering sewage sludge through a filter press (Chen et al., 2014) or it can be used as component of controlled low-strength material (CLSM), capable of self-compacting, used to fill hard to reach places (Hwang et al., 2017). Yang et al. (2013), by using the autoclaving innovative process, demonstrated that sewage sludge can be a good additive in a mixture of cement, ashes and slag, improving the long-term strength of the obtained materials. The production of floor tiles (Amin et al., 2017) and lightweight aggregates (Lau et al., 2017) are two other possibilities for sludge valorization. Suchorab et al. (2016) analized at laboratory scale the use of sewage sludge as an additive for the production of lightweight aggregate and concrete. The researchers mixed clay with sewage sludge (addition of 10%) and after the precipitate was dried, grounded and mixed it with another quantity of clay and water (to obtain the right consistency), the balls being formed and then dried at 1150°C for half an hour. The results showed that the concrete obtained by using the lightweight aggregate had a higher porosity and a lower density as compared to the concrete from commercial lightweight aggregate.

Furthermore, Ruiken et al. (2013) emphasized the importance of recovering the primary cellulosic sludge (PCS) from the inlet wastewater, as it will be described in detail in Section 6.

4.5. Bio-plastics

An alternative for petroleum plastics are polyhydroxyalkanoates (PHA), which are produced in nature by bacterial fermentation of sugar and lipids (Akaraonye et al., 2010). The PHA produced by bacteria has similar properties with conventional plastics for which, the production can affect the environment and human health (Balasubramanian and Tyagi, 2017). The use of municipal wastewater sludge as a raw material for bioplastics production could be an alternative sustainable solution. According to literature data (Tyagi and Lo, 2013), activated sludge is a source of PHA accumulating microorganisms, that take up the volatile fatty acids under anaerobic condition. The use of waste activated sludge as a source for PHA accumulation can reduce the cost of PHA production and the volume of sludge (Khardenavis et al., 2007). PHA accumulation from sludge takes place under anaerobic-aerobic condition, being influenced by temperature, pH, retention time and process configuration. Tyagi et al. (2009) sugested that sequencing batch reactor (SBR) is a good alternative for higher PHA production due to its highly flexible operation, easiness of control and biomass growth under transient conditions. Frison et al. (2015), by introducing an acidogenic fermentation phase before AD, demonstrated the possibility of recovering PHA by using two SBR reactors in series, as it will be better described in Section 6.

Yan et al. (2008) used as a source of microorganisms, pulp and paper mill waste activated sludge, obtained a PHA acumulation of 39.6% w/w of dry sludge suspended solids. On the other hand, Morgan-Sagastume et al. (2014) working with sludge and water from a municipal WWTP, achieved a PHA biomass content of 52%. Due to their biodegradability, PHA are used as packaging films and disposable products and have many applications in the medical field (for soft and hard-tissue repair and regeneration, carrier scaffolds for nerve repairs, cardiovascular applications and as functionalized beads for diagnosis and therapeutic applications). Germany, Brazil, China, Italy, UK, Canada and USA are only few countries that are using PHA in different sectors (Tyagi and Lo, 2013).

When using sludge for PHA production benefits such as the recovery and use of waste materials as biodegradable plastics and the reduction of production costs due to use of easily available sludge may be mentioned (Tyagi and Lo, 2013). However, more studies are required to clarify the technical and economical issuses (Brar et al., 2009).

4.6. Proteins

Sewage sludge contains 61% proteins, 11% carbohydrates, 1% lipids and 27% other components and may be considered a protein source (Chen et al., 2007). Considering that aproximatively 50% of dry weight of bacterial cells are proteins, and also, that proteins are constituents in animal feed and providing energy and nitrogen, the recovery of this macromolecules is important. Therefore, a number of mechanical, chemical and thermal processes have been applied. Hwang et al. (2008) applied the ultrasonic-alkaline pretreatment followed by precipitation and drying and observed a supernatant protein concentration of 3177.5 mg/L, with a protein recovery of 80%. The authors reported that the proteins obtained can be comparable with the commercially proteins in terms of nutrient composition.

According to the study of Xiao et al. (2017), the main stages for protein recovery from sludge include screening, treatments, filtration, protein precipitation from the protein solution, drying of protein precipitate and the recovery of final protein product. Some authors, affirm that prior to proteins recovery is the solubilisation of waste activated sludge by ultrasonication and alkaline treatment, the second one giving better results for protein recovery. Garcia et al. (2017), tried to recover proteins from solubilized sludge by two hydrothermal treatments. The results obtained showed that ammonium sulphate addition is the best separation method, achieving a protein recovery of 87% in the case of thermal hydrolysis and 86% in the case of wet-oxidation, respectively. An inconvenience of recovery processes are the heavy metals, which are recovered together with proteins. This becomes a problem especially if the purpose is to use proteins as nutritional supplements for animals (Tyagi and Lo, 2013). However, the use of sludge in the production of protein remains a subject that can show promising outcomes.

4.7. Enzymes

Different types of enzymes (i.e., *protease, glycosidase, dehydrogenase, catalase, peroxidase, α-amylase, α-glucosidase*) are presented in sludge, being considered valuable products, that need to be recovered. The use of enzymes in different sectors such as food, detergents, pharmaceuticals and chemical industries makes their recovery a priority. For this purpose, different types of wastewater (such as municipal or industrial: resulted from paper production or from printing and dyeing) were used for isolation, characterization and distribution of extracellular enzyme-producing yeasts (Balasubramanian and Tyagi, 2017).

Researchers have focused on wastewater sludge as a source for enzyme production, various methods being used to extract enzymes from activated sludge in order to measure their activity, including stirring with additives (detergents and cation exchange resins), ultrasonication and combined processes (Guanghui et al., 2009; Nabarlatz et al., 2010). Nabarlatz et al. (2010) using the ultrasonication assisted extraction method to recover enzymes from activate sludge, reported that a power intensity of 3.9 W/cm^2 and a sonication time of 10-20 min were enough to achieve the highest rate of enzymes recovery. Plattes et al. (2017) extracted enzymes from activated sludge in an ultrasonic cleaning bath, and affirmed that the enzyme activity of the extracts increased with increasing sonication time and can it can be reduced because of the storage conditions (freezing drastically). Significant extraction results have been obtained by Sethupathy and Sivashanmugam (2017) for a consortium of hydrolytic enzymes from waste activated sludge using ultrasonication and stirring with surfactants. Nevertheless, the enzymes recovered from sludge are not used yet at large-scale. Further studies must be carried out to explore the extraction of enzymes from sludge, considering also the techno-economical issues and eco-friendly approaches.

5. Energy recovery from wastewater sludge

5.1. Energy from biogas

The main source of energy in WWTPs is the biogas produced by AD, with a content of methane (50-70%) and carbon dioxide (30-50%), and some traces of nitrogen, hydrogen, hydrogen sulfide and water vapor (Tyagi and Lo, 2013; Shen et al., 2015). AD is one of the most applied technologies for biogas generation in WWTPs; Silvestre et al. (2015) proving that 52% of energy from sludge was transformed into biogas. Nevertheless, to increase the biogas generation, many pre-treatment methods such as microwave irradiation, ozonation, ultrasonification, enzymatic treatment, treatment with alkali or acids, wet oxidation, usage of liquid jets were investigated (Tyagi and Lo, 2011; Cano et al., 2015). Sludge thermal pretreatment and AD, is a good combination for biogas generation, being used for the co-generation of heat and power (CHP) (Carlsson et al., 2016). Countries like Germany, Austria, The Netherlands and USA, use this CHP-AD combination in the existing energy self-sufficient WWTPs (Gu et al., 2017).

The study of Ruffino et al. (2015) showed that the production of methane increased with 21-31% when thermal pre-treatment was used. Another study (Farno et al., 2017) demonstrated that from the total energy saved (585 kW), 159 kW energy was produced due to the increase in biogas generation by using thermal pre-treatment, and 82 kW and 344 kW were saved from the mixing and pumping system, respectively. Based on life cycle comparisons, Smith et al. (2014) consider that anaerobic membrane bioreactor (AnMBR) technology could produce more net energy as biogas than conventional activated sludge with AD. Furthermore, Wei et al. (2014), using AnMBR technology combined with a heat pump and osmosis, obtained a high methane production, energy equivalent being 1.57 kWh/m^3 for wastewater with COD of 500 mg/L.

Analyzing the improvement of biogas production (by AD) through the concept of circular economy, co-digestion of food waste and wastewater sludge is a feasible solution (Nghiem et al., 2017). This solution not only raises the available carbon concentration and increases digester gas production (improving energy balance), but also provides savings in the overall energy costs of plant operation (Di Maria et al., 2016; Maragkaki et al., 2017). According to Schafer et al. (2013), in Europe the biogas production increased from 2.5 to 4.0 m^3 in the WWTPs which have implemented co-digestion. Some exemples of WWTPs that have implamented the AD of sewage sludge with co-digestion of organic waste are presented by Shen et al. (2015). Considering the fact that biogas can be used for electricity generation, production of heat and steam, fuel gas vehicles and others, its recovery and conversion is really essential.

5.2. Energy from biofuels

Since biofuels have the potential to replace the non-renewable petroleums fuels in future, the use of waste sludge as a substrate for their production gained attention in recent years. Hydrogen represents one of the gaseous biofuels that can be recovered from sludge, being a sustainable alternative due to its high energy yield and clean combustion result (water). To recover it and moreover, to improve the production of hydrogen-rich fuel gas from sewage sludge, different thermochemical treatments, like drying, pyrolysis and gasification were investigated. Manara and Zabaniotou (2012) affirmed that a gaseous product with higher H_2 percentage is produced by pyrolysis rather than by drying of wet sludge. Other two methods used to produce hydrogen from activated sludge are photosynthesis and fermentation, which are more environmental friendly than the chemical processes. Massanet-Nicolau et al. (2010) reported a value of 18.14 L H_2/kg dry solids, produced by fermentation of primary sewage sludge. In the same study, the hydrogen production via mesophilic anaerobic fermentation in a continuously fed bioreactor was 27 L H_2/kg volatile solids. Wang et al. (2010) used UV iradiation as pre-treatment for waste activated sludge and observed a hydrogen production of 138.8 mL/gTS during the batch anaerobic fermentation. Guo et al. (2008) used microwave as pretreatment for waste activated sludge and obtained a hydrogen production during AD of 14.65 mL (11.44 mL/g total COD). The effects of ultrasonification (Elbeshbishy et al., 2010) and combined co-digestion of rice straw and sewage sludge (Kim et al., 2012) on hydrogen production were studied, both of them increasing the production process.

Furthermore, hydrogen in combination with carbon monoxide forms *syngas*, which can be a clean alternative for fossil fuels in electricity generation or in production of liquid fuels (Lv et al., 2007). According to Tyagi and Lo (2013), syngas production takes place in two steps: pyrolysis of sewage sludge and gasification of char in the presence of oxygen or air. While investigating the pyrolysis process of sewage sludge, Lv et al. (2007) reported that at a temperature of 1040°C, the production of syngas reached the maximum value (66%). Zuo et al. (2011) stated that the use of activated carbon enhanced the concentration of syngas in the pyrolysis gas. Also, through sludge pyrolysis process, at intermediate temperatures bio-oils were produced (Cao and Pawlowski, 2012). A limitation of the process is the presence of PAHs in the oil, which has carcinogenic or mutagenic characteristics. A possible solution for this problem can be the microwave-induction pyrolysis;

Tian et al. (2011), reporting a maximum oil yield of 49.8 wt% (time: 6 min), and negligible quantities of PAHs. According to literature data, significant oil yields can be obtained as high as 13% by using anaerobically digested sludge or 46% when mixed raw sludge is used (Tyagi and Lo, 2013). Another biofuel that can be recovered from sludge is *biodiesel*. Municipal sludge is a lipid feedstock for biodiesel production, due to its higher content of lipids (phospholipids, monoglycerides, diglycerides, triglycerides and free fatty acids) (Kargbo, 2010). In order to enhance the biodiesel production, it is important to use the microorganisms that are selected for their oil-producing capabilities and to use the pre-treatment methods (ultrasonification, thermal treatment or alkaline/acid hydrolisis) (Tyagi and Lo, 2013). Differently, Pastore et al. (2013) proposed a two-step process for the production of fatty acid methyl esters (FAMEs). The preliminary dewatered sludge extraction using hexane in acidic ambient followed by methanolysis allows the yield of FAMEs to be maximized, while minimizing the associated total energy consumption and costs. The final purification of biodiesel by vacuum distillation allows biodiesel to be recovered together with sterols, waxes, aliphatic alcohols, carotene and lycopene, increasing the economic gain of the overall process (Pastore et al., 2013).

The advantages of biofuel obtained from sludge is its availability with low costs and the abundance of sludge supply (Massanet-Nicolau et al., 2010).

5.3. Electricity production from sludge by microbial fuel cells

The use of microbial fuel cells (MFC) for electricity production is considered a sustainable solution for different problems such as excess sludge and water-energy crisis (Nikhil et al., 2018). According to Lefebvre et al. (2011), when the fraction of electron charge that contribuies to electricity generation is 40% and the hydraulic retention times is 20 h, the potential of energy that can be recoverd from wastewater by MFC, can reach 0.65 kWh/m^3. Furthermore, Plappally and Lienhard (2012) stated that the use of MFC increases the potential to achieve energy efficiency in a WWTP, the energy consumption being between 0.3 and 0.6 kWh/m^3. To improve the energy performances, MFCs technologies were combined with membrane treatment processes (Gu et al., 2017). Tian et al. (2014), using AnMBR system developed with microfiltration membranes which serve as cathodic chamber for MFCs, reported that this combination produced stable electricity for over 600 h operation time. Electricity production can also be increased from 3 W/m^3 to 11.5 W/m^3 by combining MFCs and osmotic membrane bioreactor (Hou et al., 2016). Combination of MFCs technologies with fluidized bed membrane bioreactor (MBR) (Li et al., 2014) and aerated biological filter system were also investigated (Dong et al., 2015). The literature analysis shows that researchers studied the MFCs technologies with application at pilot and real scale (Feng et al., 2014; Dong et al., 2015; Oon et al., 2017). This applicability at full-scale is due to the fact that

MFCs can remove pollutants and generate electricity under ambient temperature, neutral pH and normal pressure (Raheem et al., 2018). Also, MFCs can carry out several microbial processes (such as organic matter removal, nitrification and denitrification) for wastewater treatment inside the same bioreactor (Gonzalez-Martínez et al., 2018).

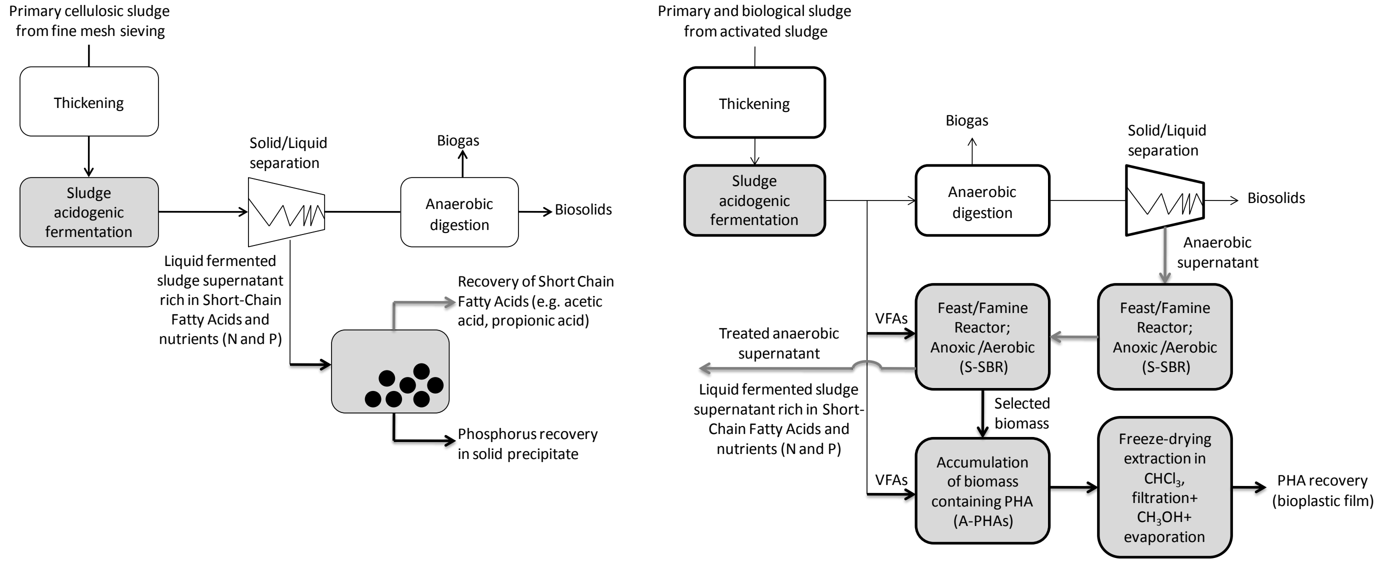
Although the use of MFCs technologies in WWTPs can improve the treatment performances, their application is limited due to the electrode materials that are expensive (Lefebvre et al., 2011). This means that further substantial research in terms of cost and yields increase is needed.

However, there are other energy recovery technologies that can be used such as the *Anaerobic ammonium oxidation (Anammox)*, because a significant amount of energy can be recovered during nitrogen removal from WWTPs. Anammox, besides the fact that decreases the aeration rates and reclaims the maximum organics from water, can improve the energy efficiency in WWTPs (Gao and Tao, 2011). The partial nitrification/anammox (PN/A) can save energy by reducing the oxygen demand for the nitrification process and minimize the quantity of excess sludge (Bauer et al., 2016). According to Gao et al. (2014), the energy in the anammox reactor can be recovered through a combination of AD and autotrophic nitrogen removal, converting WWTPs in energy-producing systems (Kartal et al., 2010). According to Tyagi and Lo (2013), the heat energy produced from the treatment processes is higher than the required heating energy in the plant. The heat energy in a WWTP can be used as an energy source of heat pumps for heat supply and electricity saving. With all these technologies used for energy recovery or energy saving, many challenges still exist and more studies are necessary, in terms of technology, cost and environmental issues.

6. Materials and energy recovery in urban biorefineries

The technologies presented until now are aimed at the recovery from sludge of clearly defined resources or energy; however, this approach does not consider the totality of a WWTP. In recent years, for WWTPs, it was observed a radical change of vision of the plant itself, this change being considered as a “paradigm shift” (Puchongkawarin et al., 2015). The WWTP is no longer considered only for its environmental protection and sanitation functions, but also as the starting point for the exploitation of potential resources (including sludge) that are now considered wastes. In this context, the current WWTPs should be understood as self-sufficient systems (energetically and economically) and secondly as a “factory” (a biorefinery) of new compounds for the market.

An “urban” biorefinery involves the recovery of primary cellulosic sludge (PCS) as a starting point. By introducing a fine sieving phase (< 0.35 mm) downstream of the coarse sieving, it is possible to recover the cellulose from PCS. Present in urban wastewaters due to the discharge of toilet paper, cellulose is an important polysaccharide consisting of a large number of glucose molecules joined together by a β (1$\rightarrow$4) glycoside bond. Ruiken et al. (2013) showed that the introduction of such a sieve allows to obtain removals efficiencies of: TSS = 50%; COD = 35%; N_{TOT} = 1%; P_{TOT}< 1%, higher than those achievable with primary sedimentation. In addition, the cellulose content in the removed suspended solids is equal to 79%, which is very high. Furthermore, since cellulose is not completely biodegradable in a conventional WWTP, its removal by fine sieving would allow an improvement of biological processes (i.e., activated sludge and AD), as highlighted by Ruiken et al. (2013). The cellulosic sludge thus removed can be enhanced with different solutions, such as those presented in Fig. 4.



a) **b)**

Fig. 4. Process diagram for the valorisation (**a**) of the "primary cellulosic sludge" for phosphorus recovery via struvite production (amended from Crutchik et al., 2018) and (**b**) of the mixed sludge (primary + secondary) – for bioplastics recovery (amended from Frison et al., 2015).

In the first solution (Fig. 4a), the solid part of the previously thickened primary sludge is sent to the acidogenic fermenter. This is a SBR reactor operating under temperature and pH conditions that maximize the production of short-chain fatty acids (SCFA), including acetic and propionic acid. Under optimal conditions of temperature (37°C, mesophilic regime) and pH (pH=8 in the inlet sludge), Crutchik et al. (2018) estimated a per capita SCFA production of 2.92 kg COD/ year. In addition, the fermentation process allows the release of nutrients (nitrogen and phosphorus) present in the inlet sludge. Subsequently, the sludge coming out of the fermenter is subjected to solid/liquid separation, where the solid part is sent to AD. In terms of biogas production, Crutchik et al. (2018) estimated a per capita value of 3 m^3/year. Instead, with reference to the liquid part, the high concentrations of SCFA and nutrients suggest a material recovery; through propionic acid (>30%), the post-fermentation liquid can be used both for phosphorus recovery in EBPR (Enhanced Biological Phosphorus Removal) or chemical-physical processes (through struvite production) and both for high added value compounds production such as PHAs used in the production of bioplastics (Frison et al., 2015). In the specific case of Fig. 4a, the fermentation liquid was used for the recovery of phosphorus through the struvite production; Crutchik et al. (2018) estimated a per capita struvite production of 0.15 kg/ year having adopted a molar ratio (PO_4^{3-}: Mg^{2+}) of (1.0:1.5), a dosage of 5 g/L of magnesium hydroxide, $Mg(OH)_2$ as well as by adjusting the initial pH to 8.5 with NaOH (0.1 M).

In the second solution (Fig. 4b), the mixed “primary and secondary” sludge, previously thickened, is sent to the acidogenic fermenter. Also in this case, fermentation allows obtaining fermented sludge rich in SCFA and nutrients. Differently from Fig. 4a, this scheme allows PHAs recovery as well as nutrients removal from the anaerobic supernatant. For the scope, Frison et al. (2015) tested a new process in which the alternation of aerobic-feast (of abundance) and anoxic-famine (of famine) conditions allows selecting the biomass containing PHA and the removal of nitrogen by nitritation/denitritation. Another way of removing nitrogen, different from the conventional denitrification/nitrification, nitritation/denitritation involves the removal of nitrogen by oxidation of ammonia (NH_4^+) into nitrite (NO_2^-) and its subsequent reduction into gaseous nitrogen (N_2); in these processes, the amount of carbon that must be supplied to the microorganisms is lower than that needed in the conventional treatment (Malamis et al., 2014). Since the anaerobic supernatant is usually hot (mesophilic, 30-40°C) and the high temperatures favour the growth of ammonium oxidising bacteria over nitrite-oxidising bacteria (Hellinga et al., 1998), nitritation/denitritation is the ideal treatment to remove nitrogen from the anaerobic supernatant (Malpei et al., 2008). Consequently, nitrogen removal is facilitated and a reduced route (a shortcut), as compared to conventional denitrification/nitrification, can be used.

In terms of equipment, the layout of Fig. 4b includes two SBR reactors. The first (N-SBR) is dedicated almost exclusively to the nitritation process (aerobic). The second SBR (indicated as S-SBR) is intended for the denitritation process, as well as for the selection of biomass containing PHA. In particular, the S-SBR operating cycle consists of 50 minutes of aerobic conditions and 250 minutes of anoxic conditions, for a total of 300 minutes, excluding the feeding, settling and discharge phases typical of SBR. In addition, the liquid effluent from the N-SBR reactor (with high nitrite concentrations as compared to ammonium) is sent to the S-SBR reactor during the first 10-12 minutes of the anoxic phase (famine). Frison et al. (2015) showed that during aerobic conditions ammonium is oxidized to nitrite and volatile fatty acids (VFA), from fermented sludge, are converted to PHA; during conditions of anoxic famine, nitrite is reduced to N_2. Microorganisms leading to this reduction use PHA stored in the reactor's internal biomass (representing the carbon source); the external source of carbon (VFA) is therefore added only at the beginning of the aerobic phase. Following sedimentation in the S-SBR reactor, the biomass containing PHA is separated from the treated supernatant; the latter, which is nitrogen-free, is recirculated in the WWTP water line. Instead, the biomass thus selected is sent to the third reactor (indicated in Figure 4b as A-PHAs), which makes it possible the PHAs accumulation. By maintaining a concentration of dissolved oxygen constantly equal to 2 mg/L, the reactor requires the addition of a carbonaceous substrate. Considering, for example, the addition of sludge fermentation liquid with wollastonite (a very common mineral in metamorphic contact rocks from dolomite and impure limestone), Frison et al. (2015) show that A-PHAs reactor biomass can accumulate up to 21±5% PHA (gPHA/gVSS x100), with a COD/N/P ratio of (100:7.8:0.06) and after 8 hours of operation, the observed yields in PHA production were about 0.40 g COD PHA/g COD VFA.

The biopolymers produced in this way are characterized by a prevalent presence of 3HB (3-hydroxybutyrate) and 3HV (3-hydroxyvalerate); 3HB represents the majority of the PHA produced (57%) while the percentage of 3HV is equal to 41%. The composition of the PHAs suggests their recovery during thermoplastic processing. The characterization of the material revealed that the biopolymer is composed of long molecular chains with a mean molecular weight (M_W) of 7.4×10^5 g/mol and a similar distribution of the chain length (polydispersion index of 1.25 M_W/M_n where M_n is the number of moles). In general, low crystallinity in combination with a low Tg (glass transition temperature, equal to -1.6°C) is an index of amorphous biopolymers (Frison et al., 2015).

According to Frison et al. (2015), there will be a real revolution in the coming years, the WWTPs used today will be real biorefineries.

7. Conclusions and future research directions

The excess sewage sludge production is a serious concern for WWTPs due to environmental and socio-economic factors. The use of conventional sludge disposal methods such as landfilling, disposal in oceans, land application or incineration is limited due to stingent legislation and pressure from environmental authorities and public domain. Solutions that can replace with success the sludge landfill and incineration from the ecological and economical points of view are reuse and recycling of sewage sludge and sludge residues ash into new marketable materials. This application can bring major economic benefits and are consistent with the concept of circular economy.

Many treatment technologies are applied at pilot or full scale to recover resources and energy from wastewater sludge systems. Considering the circular economy principles, the development and regular update of information databases with new advanced treatment and recovery technologies, or with the evolution of economic, environmental and social-cultural contexts should be realized.

Currently, most of the WWTPs use biogas from the sludge anaerobic digestion for digester heating and electricity generation. To improve energy production, co-digestion was adopted by adding external organic waste into the digester. Although energy self-sufficient WWTPs are definitely feasible, many challenges still exist, particularly in developing countries and future efforts are needed in terms of addressing technology, costs and environmental protection issues (Gu et al., 2017). The selection of the best sludge management scenario for a particular area needs the usage of decision-making tools, like LCA (Life Cycle Assessment) that allow for the assessment of a probable environmental impact of proposed strategies.

Even if the products obtained from sludge increase the profitability, the high-volume fuels support to fulfill national energy demands, and the power generation decrease the costs and sidesteps greenhouse-gas releases, the major issue with the resources recovered from sludge being related to the manufacturing cost of value added products versus the market price. In other words, the succes of the resources derivated from sludge and utilization of recovery technologies, will depend mainly upon the technical and economical feasibility (catching investors' interest in developing such technologies), environmental sustainability, market aspects and public acceptance.

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