

1 **Validated innovative approaches for energy-efficient resource recovery and re-use from**  
2 **municipal wastewater: from anaerobic treatment systems to a biorefinery concept**

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1 **Abstract**

2 The development of innovative technologies in wastewater treatment create the concept of biorefinery  
3 in wastewater treatment plants (WWTPs), placing anaerobic processes in the highlight. Starting from  
4 the conventional anaerobic treatment processes to “closing the loop” scheme, next generation  
5 WWTPs are ready to serve for water, energy and materials mining. While bioenergy is still  
6 dominating the resource recovery, recovery of value-added materials (i.e. struvite, biopolymers,  
7 cellulose) are receiving significant attention in recent years. So, what are the state-of-the-art  
8 approaches for energy-efficient resource recovery and re-use from municipal wastewater? This paper  
9 follows a critical review on the validated technologies in operational environment available and  
10 further suggests possible market routes for the recovered materials in WWTPs. Considering the  
11 development and verification of a novel technology together with the valorisation of the obtained  
12 products, biorefinery and resource recovery approaches were gathered in this review paper from a  
13 circular economy point of view. General currently-faced barriers were briefly addressed to pave the  
14 way to a create to-the-point establishments of resource recovery facilities in the future.

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25 **Keywords:** anaerobic treatment, biorefinery, energy recovery, material recovery, municipal  
26 wastewater, valorisation

## 1 **1. Introduction**

2 During the last years, wastewater treatment plants (WWTP) have moved from the concept of “waste  
3 treatment”, aimed at discharging treated wastewater into surface waters, to the concept of “water  
4 resource recovery facility” (WRRF). This transformation from pollutants removal to valuable  
5 resources frames wastewater management in the broader context of the circular economy. The  
6 question that arises is which are the possible recovered and safely reusable resources to help closing  
7 the loop in WRRF?

8 First of all, the reclaimed water: Water reuse is particularly important because it is considered as an  
9 effective approach to address water shortage problems and water quality deterioration issues (Sun et  
10 al. 2016). Water reuse can be one of the methods of recycling treated wastewater for beneficial  
11 purposes, such as agricultural and landscape irrigation, industrial processes, non-potable domestic  
12 use (e.g. toilet flushing), and groundwater replenishing. At EU level the minimum quality standards  
13 for water reuse have been proposed in May 2018; this proposal for regulation lays down minimum  
14 requirements for water quality and monitoring and the obligation to carry out specified key risk  
15 management tasks. Classes of reclaimed water quality, minimum treatment requirements, allowed  
16 uses, irrigation methods and minimum requirements for water quality are set  
17 ([http://www.europarl.europa.eu/RegData/etudes/BRIE/2018/625171/EPRS\\_BRI\(2018\)625171\\_EN.pdf](http://www.europarl.europa.eu/RegData/etudes/BRIE/2018/625171/EPRS_BRI(2018)625171_EN.pdf)).  
18 pdf).

19 On site energy recovery, particularly as biogas production, in WWTP is widely diffused as an  
20 alternative source of energy, for the recovery of thermal, electrical and mechanical energy, to be  
21 consumed either inside (also achieving energy self-sufficiency) or outside the plant. Nowadays  
22 energy recovery takes place mostly in the sludge line and actions in water line are much rarer but  
23 more and more of interest (Papa et al. 2017). Biogas is the main resource of anaerobic treatment  
24 systems. In the last years; however, two-step bioconversion comes into prominence as more value is  
25 derived to volatile fatty acids (VFAs) production before ending up to other end-products. Moreover,  
26 anaerobic processes offer much more than conventional wastewater treatment, provide recovering

1 sustainable energy and valuable biochemicals. This scenario helps to recognize conventional and  
2 innovative (i.e. anaerobic membrane bioreactor - AnMBR) anaerobic processes as core of biorefinery  
3 general concept (Puyol et al., 2017, Krzeminski et al., 2017).

4 Nutrient recovery and recycling take an important role in circular economy. Recovered nutrients  
5 from the wastewater can be utilized as soil amendments or fertilizers for beneficial uses in agriculture.  
6 In particular,  $\text{NH}_4^+$  form is advantageous because it predominates in anaerobic reactor effluents and  
7 can be useful for fertigation purposes. Phosphorus recovery (i.e. in the form struvite or phosphorous  
8 salts) becomes essential for preventing eutrophication in the aquatic environment and for alleviating  
9 economic dependence on phosphate rocks. Addressing raw materials conservation, arising from  
10 phosphorus scarcity is described as one of the greatest global challenges of the 21<sup>st</sup> Century (Peng et  
11 al. 2018).

12 The resources mentioned above are those most commonly recovered in WWTPs; in addition to them  
13 there are more innovative ones that can be originated from cellulosic primary sludge (CPS) and  
14 polyhydroxyalkanoate (PHA) rich sludge. The cellulosic sludge can be separated by upstream  
15 dynamic sieving. The CPS can then be anaerobically digested to produce biogas, or, under optimal  
16 acidogenic fermenting conditions it produces VFAs. Here the propionate content can be more than  
17 30% and can optimize the enhanced biological phosphorus removal (BPR) processes (Crutchik et al.  
18 2018). Long-term operation indicated that anaerobic alkaline fermentation for VFA production from  
19 sewage sludge is both technically and economically feasible (Liu et al., 2018). Regarding PHA  
20 recovery, primary and secondary sewage sludges are potential feedstock for its recovery (Kumar et  
21 al. 2018). PHAs have comparable properties to petrochemical plastics and can also serve as a biofuel  
22 or building blocks for the synthesis of various chemicals (Kleerebezem et al. 2015).

23 While some of the above-mentioned reuse and recovery approaches towards wastewater are already  
24 efficiently implemented, some of them still lack the convenient technology together with social-  
25 technological planning and design methodology to identify their potential end-use and market  
26 requirements (Van Der Hoek et al. 2016).

1 At European level there are several EU projects founded by Horizon 2020 that aim at recovering  
2 materials from centralized and decentralized WWTPs. For example, SMART-Plant is an Innovation  
3 Action that aims at reducing the energy and environmental footprint and, contemporary, at recovering  
4 valuable materials (SMART-Products are water, cellulose, biopolymers, nutrients) that are valued in  
5 construction, chemical and agriculture supply chain (smart-plant.eu). POWERSTEP is another  
6 Innovation Action aims at energy positive WWTP, biogas production and carbon extraction  
7 (powerstep.eu). P-REX, similarly, aimed at sustainable sewage sludge management promoting  
8 phosphorus recycling and energy efficiency (p-rer.eu).

9 This review critically analyses innovative anaerobic processes to recover materials and energy from  
10 municipal wastewater, state of the art WWTPs and future aspects. The energy-efficient resource  
11 recovery is examined by the critical analysis only of the anaerobic processes. Moreover, the discussed  
12 technologies are selected based on the validation criteria of the demonstrative or full scales  
13 applications to ensure the robustness of the technologies supporting the characteristics, as quantity  
14 and quality, of the products to be marketed. Hence, this review paper aims to provide a comprehensive  
15 overview to the biorefinery concept, recent leading technologies and further sustainable scenarios to  
16 fulfil circular economy goals. Although great previous efforts have been done towards anaerobic  
17 processes and the concept of resource recovery, environmental technology verification (ETV) has  
18 further reviewed innovative technologies together with the possible valorisation market alternatives,  
19 bottlenecks or barriers of recovered products.

## 20 **2. Brief evolution of anaerobic schemes as the core of the biorefinery approach**

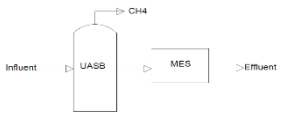
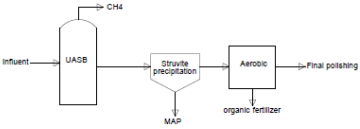
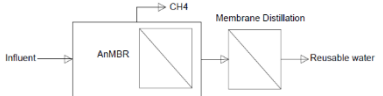
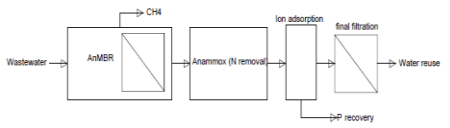
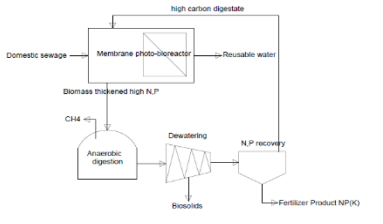
21 Anaerobic treatment is one of the most promising treatment technologies for developing more  
22 sustainable sanitation; it is considered to be the core technology for resource and energy recovery  
23 (Stazi and Tomei 2018). Upflow anaerobic sludge blanket (UASB) was successfully implemented  
24 and established within a wide acceptance in municipal WWTPs, especially in tropical and subtropical  
25 regions where the temperature of the wastewater is usually above 20 °C (Lohani et al. 2016).  
26 Expanded granular sludge bed (EGSB) was further developed to enhance substrate-biomass

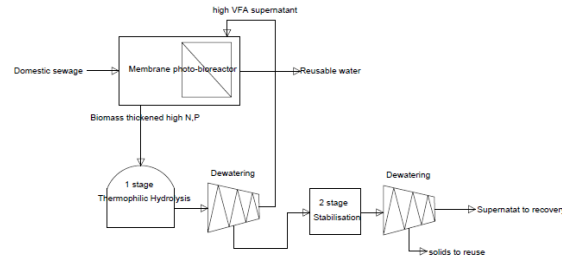
1 interaction within the treatment system by expanding the sludge bed and increasing hydraulic mixing  
2 compared to UASB (Niwa et al. 2016; Cuff et al. 2018). Although well-established UASB and/or  
3 EGSB configurations mostly meet the requirements necessary for anaerobic treatment, unfavourable  
4 conditions regarding the disintegration of granules led to the development of AnMBR by coupling  
5 membrane technology with anaerobic treatment. Meanwhile, combined heat and power (CHP)  
6 systems using the anaerobic digestion (AD) of sludge has become the most adopted technology in the  
7 existing energy self-sufficient WWTPs. Based on life cycle comparison, AnMBR technology was  
8 found to produce more net energy as biogas compared to conventional activated sludge coupled with  
9 AD (Gu et al. 2017). The main advantage of AnMBR as a mainline wastewater treatment process is  
10 the capacity to recover most of the energy potential in the wastewater rather than the fraction currently  
11 recovered by the aerobic-anaerobic treatment. The recent results obtained from pilot-scale AnMBRs  
12 treating domestic wastewaters were reviewed and discussed in detail (Shin and Bae 2018).

13 Table 1 provides a schematic representation of different flow schemes to enhance the role of  
14 anaerobic processes as core of biorefinery approach. Table 2 refers to resource recovery associated  
15 with the schemes in Table 1; all of them recover methane, which is the main produced-resource in  
16 WWTPs where anaerobic processes are implemented. Moreover, some of them recover N, P and  
17 VFAs due to coupling with the membrane technology. The optimization of anaerobic processes brings  
18 the key towards energy self-sufficient WWTPs.

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**Table 1.** Flow schemes with resource recovery - role of anaerobic processes

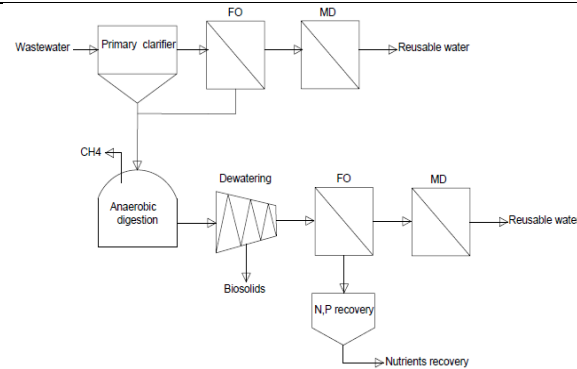
| Main units                         | Scheme and resource recovery   | Anaerobic process  | Reference                  | Number scheme |
|------------------------------------|--|--------------------|----------------------------|---------------|
| Anaerobic Process + Post-treatment |   | UASB in main line  | (Li and Yu 2016)           | 1a*           |
|                                    |   | UASB in main line  | (Verstraete et al. 2009)   | 1b*           |
| AnMBR + Post-treatment             |   | AnMBR in main line | (Song et al. 2018)         | 2a*           |
|                                    |  | AnMBR in main line | (Batstone and Virdis 2014) | 2b*           |
| Aerobic Membrane + AD + dewatering |  | AD in sludge line  | (Batstone et al. 2015)     | 3a*           |



Double stage AD in sludge line

(Batstone et al. 2015)

3b\*

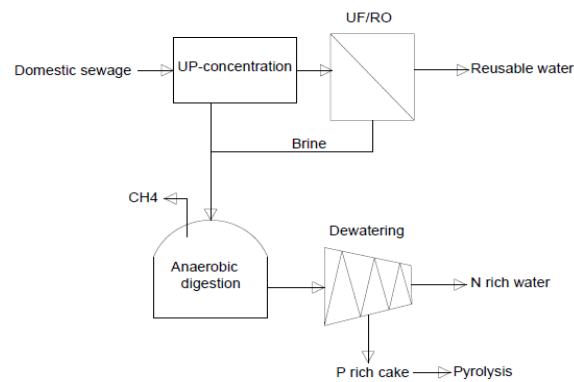


AD in sludge line

(Ansari et al. 2017)

4a\*\*

Membrane (double stage) + AD + dewatering



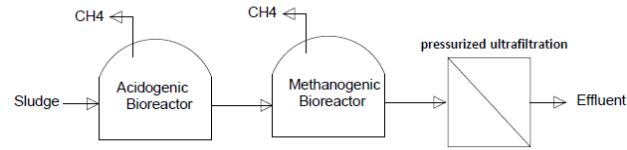
AD in sludge line

(Verstraete et al. 2009)

4b\*\*



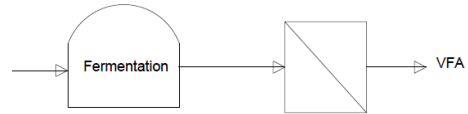
AD/Fermentation + Membrane



AD in sludge line

(Joo et al. 2016)

5a\*\*



Fermentation in sludge line

(Longo et al. 2015)

5b\*\*

- 1 Legend: MES=microbial electrochemical systems; MAP= magnesium ammonium phosphate; MD=membrane distillation, UF=ultrafiltration; RO=reverse osmosis; FO= forward  
 2 osmosis; VFA=volatile fatty acids. \*Lab/Pilot scale; \*\*Pilot/demonstrative scale

**Table 2.** Resource recovery for different plant schemes.

| Resource recovery | Item            | Scheme numbers |
|-------------------|-----------------|----------------|
| Methane           | CH <sub>4</sub> | 1,2,3,4,5      |
| Water Reuse       | WR              | 2,3,4          |
| VFA               | VFA             | 5b             |
| N rich sludge     | NS              | 3,4            |
| P rich sludge     | PS              | 3,4            |
| N rich water      | NW              | 4              |
| P rich water      | PW              | None           |
| Struvite          | MAP             | 1b             |

### 3. Resource recovery in anaerobic processes

#### 3.1. Wastewater treatment line

The biogas produced during anaerobic treatment of wastewater can be utilized as an energy source (Table 3). However, a significant amount of methane cannot be recovered since a major proportion is dissolved in the effluent, even if the biogas exhibits high CH<sub>4</sub> content (Liu et al., 2018; Souza et al., 2011). Anaerobic wastewater treatment processes such as UASB is therefore limited because of low liquid upflow velocity and inadequate mixing (Yeo and Lee 2013). CH<sub>4</sub> losses recorded in different anaerobic reactors at a pilot-scale were listed and discussed and average CH<sub>4</sub> loss in the effluents were stated between 19-85% (Crone et al. 2017). In municipal wastewater treatment up to 30-40% of the produced methane was reported to be loss in the application of AnMBR (Krzeminski et al. 2017). Although many efforts were directed towards recovering dissolved CH<sub>4</sub> from anaerobic effluents, such as hollow fibre membrane contactors (Cookney et al. 2016) or down-flow hanging sponge reactors (Hatamoto et al. 2011), scaling up is still missing and validation is required.

In the concept of valorisation of municipal wastewater, two-step bioconversion stands as an attractive alternative route (Li and Yu 2011) in the fermentation reactors (Table 3). Complex organic matter in wastewater is simply converted to VFAs before ending up as other valuable end-products (Zhou et al. 2018). This allows a separated optimization of bioconversion mechanisms into a more straight-

1 forward bioproduction process (Pan et al. 2018). The concentration and speciation of VFAs during  
2 this process often determines the desired quality of the end-products (Reyhanitash et al. 2017).

3 Although VFAs are so-called intermediate products, they have various potential applications within  
4 the WWTP. Acetate is the most-preferred VFA product for denitrification within WWTP followed  
5 by butyrate and propionate (Elefsiniotis and Wareham 2007). Propionate can enhance BPR processes  
6 in biological nutrient removal systems (Chen et al. 2004). However, further application, either within  
7 WWTP or the product value, will determine the desired VFA concentration and speciation (Peces et  
8 al. 2016) as discussed in the following section. Microbial fuel cells (MFCs) is also another option to  
9 produce electricity from VFAs by fermentative hydrogen production (Teng et al. 2010); however,  
10 this technology has not yet been validated. Low power density and high operating cost of MFCs limits  
11 their implementations on a large-scale (He et al. 2017).

12 AnMBRs have been successfully implemented to treat municipal wastewaters with high COD  
13 removal rates for the main water reuse purpose. However, the discharge of the treated effluents into  
14 the aquatic environment or water reuse is usually not possible without further nutrient removal (Ruiz-  
15 Martinez et al. 2012; Batstone et al. 2015). In this regard, nutrient removal from AnMBR effluent  
16 using microalgae was proposed (Viruela et al. 2016). In addition, within the context of EU LIFE  
17 Project MEMORY (life-memory.eu), submerged AnMBR was demonstrated to combine AD and  
18 membrane technology. Such innovative pilot-scale implementations suggest promising technologies  
19 for municipal wastewater treatment and resource recovery ([https://ec.europa.eu/info/research-and-  
20 innovation/law-and-regulations/identifying-barriers-innovation\\_en](https://ec.europa.eu/info/research-and-innovation/law-and-regulations/identifying-barriers-innovation_en)).

1 **Table 3.** Recovered products in anaerobic wastewater line.

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| Wastewater line Process | Influent  | Scale | Volume m <sup>3</sup> xline | HRT h       | OLR kg COD/m <sup>3</sup> /d | T °C | Recovered products | Amounts/Composition                               | Location | Reference                      |
|-------------------------|-----------|-------|-----------------------------|-------------|------------------------------|------|--------------------|---|----------|--------------------------------|
| Fermentation reactor    | sewage WW | Pilot | 0.5                         | 4.6–5.9 (d) |                              | 35   | VFA                | 7453 mg COD/L                                     | Italy    | (Longo et al. 2015)            |
| Fermentation reactor    | sewage WW | Pilot | 1                           |             |                              | 42   | VFA                | 120 mg VFA/g VS/d                                 | Belgium  | (Morgan-Sagastume et al. 2015) |
| Fermentation reactor    | sewage WW | Pilot | 0.05                        | 8 (d)       |                              | 25   | VFA                | 2825 mg COD/L                                     | China    | (Li et al. 2011)               |
| UASB                    | sewage WW | Full  | 605x8                       | 8           |                              |      | CH <sub>4</sub>    | 390.1 Nm <sup>3</sup> /d, 78.2% CH <sub>4</sub>   | Brazil   | (Rosa et al. 2018)             |
| UASB                    | sewage WW | Pilot | 0.12                        | 4           |                              | 24   | CH <sub>4</sub>    | 0.072 L CH <sub>4</sub> /g COD <sub>removed</sub> | Brazil   | (Barbosa and Sant'Anna 1989)   |
| UASB                    | sewage WW | Pilot | 20.36                       | 10          | 0.5                          | 20   | CH <sub>4</sub>    | 0.3-0.9 Nm <sup>3</sup> /d, 78% CH <sub>4</sub>   | Spain    | (Álvarez et al. 2008)          |
| UASB                    | sewage WW | Pilot | 0.021                       | 4.7         |                              |      | CH <sub>4</sub>    | 0.22 L CH <sub>4</sub> /g COD <sub>removed</sub>  |          | (Uemura and Harada 2000)       |
| UASB                    | sewage WW | Pilot | 0.11                        | 9           | 0.73                         | 25   | CH <sub>4</sub>    | 212 L CH <sub>4</sub> /g COD <sub>removed</sub>   |          | (Agrawal et al. 1997)          |
| UASB                    | sewage WW | Full  | 14                          | 12          | 0.88                         |      | CH <sub>4</sub>    | 0.24 L CH <sub>4</sub> /g COD <sub>removed</sub>  | Brazil   | (Souza et al. 2011)            |
| UASB                    | sewage WW | Pilot | 0.36                        | 5           | 2.12                         | 25   | CH <sub>4</sub>    | 0.22 L CH <sub>4</sub> /g COD <sub>removed</sub>  | Brazil   | (Souza et al. 2011)            |
| UASB                    | sewage WW | Full  | 1200                        | 6           | 2.35                         | 25   | CH <sub>4</sub>    | 0.075 L CH <sub>4</sub> /g COD <sub>removed</sub> | India    | (Draaijer et al. 1992)         |

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**Table 4.** Recovered products in anaerobic sludge line

| Sludge line | Influent      | Scale | Flow | Volume              | HRT   | OLR                     | T    | Residual concentrations/Recovered products | Amount   | Location                    | Reference               |
|-------------|---------------|-------|------|---------------------|-------|-------------------------|------|--|--|-----------------------------|-------------------------|
| Process     |               |       | t/d  | m <sup>3</sup> line | d     | kg VS/m <sup>3</sup> /d | °C   | °C   |  |                             |                         |
| FERM        | WAS           | Full  |      | 30                  |       | 3                       | 35   | VFA  | 1261 kg VFA/d  | Wuxi city of China          | (Liu et al., 2018)      |
| FERM+AD     | Cellulosic PS | Pilot | n.a  | n.a.                | n.a   |                         | n.a. | VFA  | 100-120 mg COD/g VS-d  | Verona, Italy               | (Crutchik et al. 2018)  |
|             |               |       |      |                     |       |                         |      | Struvite                                   | 0.15 kg struvite per capita/year   |                             |                         |
|             |               |       |      |                     |       |                         |      | CH <sub>4</sub>                            | n.a.   |                             |                         |
| AD          | PS+WAS        | Full  | n.a  | 4400x2              | 20    | 1-2                     | M    | Electricity                                | 25-55% COD <sub>converted</sub>  | Tilburg-Noord, Netherlands  | (De Vrieze et al. 2016) |
|             |               |       |      |                     |       |                         |      | Ammonia                                    | 143 kg N/d   |                             |                         |
|             |               |       |      |                     |       |                         |      | Struvite                                   | 36 kg P/d  |                             |                         |
| AD          | WAS           | Full  | n.a  | 2900                | 25    | 1-2                     | M    | Electricity                                | 38% COD <sub>converted</sub>   | Land van Cuijk, Netherlands | (De Vrieze et al. 2016) |
|             |               |       |      |                     |       |                         |      | Ammonia                                    | 175 kg N/d   |                             |                         |
|             |               |       |      |                     |       |                         |      | Struvite                                   | 137 kg P/d   |                             |                         |
| AD          | PS+WAS        | Full  | n.a  | 5430x2              | 20    | 1-2                     | M    | Electricity                                | 0.82 m <sup>3</sup> CH <sub>4</sub> /m <sup>3</sup> /d ,<br>25-55%COD <sub>converted</sub> | Bath, Netherlands           | (De Vrieze et al. 2016) |
|             |               |       |      |                     |       |                         |      | Ammonia                                    | 440 kg N/d   |                             |                         |
|             |               |       |      |                     |       |                         |      | Struvite                                   | 112 kg P/d   |                             |                         |
| AD          | WAS           | Full  | n.a  | 12000x6             | 20    |                         | M    | Biogas                                     | 0.4 m <sup>3</sup> biogas/VSS  | Castiglione Torinese, Italy | (Traversi et al. 2015)  |
| AD          | PS+WAS        | Full  | n.a  | 12000x6             | 25    |                         | M    | Biogas                                     | 0.49 m <sup>3</sup> biogas/VSS   | Castiglione Torinese, Italy | (Traversi et al. 2015)  |
| TH+AD       | Sewage sludge | Full  | n.a  | 100                 | 15-18 | 3.5                     | 55   | CH <sub>4</sub>                            | 0.35 m <sup>3</sup> CH <sub>4</sub> /kg VS   | Namyangju city, Korea       | (Han et al. 2017)       |
| Co-DA       | PS+WAS+OFM SW | Full  | n.a  | n.a.                | 45    | 1.3                     | 35   | Electricity                                | 400 kW   | Rovereto, Italy             | (Nghiem et al. 2017)    |
| Co-DA       | PS+WAS+OFM SW | Full  | 120  | n.a.                |       | 0.78                    | 70   | Electricity                                | 125 kW   | Treviso, Italy              | (Nghiem et al. 2017)    |
| Co-DA       | PS+WAS+OFM SW | Full  | 95   | n.a.                |       | n.a.                    | 55   | Electricity                                | 95 kW  | Kurobe, Japan               | (Nghiem et al. 2017)    |
| Co-DA       | PS+WAS+OFM SW | Full  | 2700 | n.a.                | 18    | n.a.                    | 35   | Electricity                                | 11000 kW   | East Bay MUD, USA           | (Nghiem et al. 2017)    |

## 1 **3.2. Sludge treatment line**

2 Sewage sludge management constitutes a major part of the operating expenses of municipal WWTPs.  
3 In full-scale WWTPs sewage sludge usually undergoes anaerobic digestion to recover energy (CH<sub>4</sub>-  
4 rich biogas), and thus produce heat and electricity within the concept of combined heat and power  
5 plants. There is also a growing trend to use sludge as a feedstock in other value-added processes  
6 together with bioenergy (Zacharof and Lovitt, 2013) as shown in Table 4.

### 7 **3.2.1 Recovered materials: VFA**

8 More attention has been paid in the recent years to recover VFAs through the acidogenic fermentation  
9 of sewage sludge on down-stream processes (Liu et al. 2012; Longo et al. 2015). In a wider  
10 biorefinery concept, carbon upgrading to VFAs seems an energy-efficient and cost-effective strategy.  
11 However, utmost importance lies here when considering the WWTP as an integrated process, since  
12 extracting VFA will reduce the amount of organic matter fed to AD, which will eventually decrease  
13 the energy recovery (Peces et al. 2016). In this regard, the benefits of VFA production and extraction  
14 from sewage sludge should be well-designed and optimized in order not to outshadow methane  
15 recovery. The optimization should focus on two main criteria: (i) the cost (capital investment and  
16 operating expenses) of the fermentation and extraction process and further earnings from VFA use or  
17 sale; and (ii) the impact on CH<sub>4</sub> generation (Peces et al. 2016).

18 Depending on the selective production of VFAs from the acidogenic fermentation of sewage sludge,  
19 VFAs also have high economic values such as materials used in the production of bioplastics and  
20 biotextiles (Zacharof and Lovitt 2013; Lin et al. 2018). For instance, acetate and butyrate are preferred  
21 for polyhydroxybutyrate (PHB) production, while propionate is required when producing  
22 polyhydroxyvalerate (PHV) (Shen et al. 2014; Peces et al. 2016). In addition, some important  
23 characteristics such as higher flexibility, low stiffness and brittleness, and higher tensile strength and  
24 toughness are highlighted to promote the production of co-polymers using VFAs with higher  
25 propionate/acetate ratio (Frison et al. 2015; Crutchik et al. 2018). However, establishing consistence  
26 VFA concentrations and proportion remains a significant challenge.

### 1 **3.2.2 Recovered materials: Nutrients**

2 As mentioned earlier, there is large interest in decreasing costs and elevating sustainability by energy-  
3 efficient resource recovery in the concept of biorefinery (Raheem et al. 2018). So far, validated  
4 biorefinery products from WWTPs include nutrients (i.e. N, P), biopolymers (i.e. PHA) and cellulose  
5 (Zijp et al. 2017; Raheem et al. 2018).

6 The recovered nutrients from WWTPs can be utilized for struvite and/or Ca-P precipitation (Cieřlik  
7 and Konieczka 2017; Melia et al. 2017) or biochar adsorption (Huggins et al. 2016). Struvite  
8 ( $\text{MgNH}_4\text{PO}_4$ ) crystallization has been successfully used for simultaneous recovery of nutrients from  
9 wastewater (Hermassi et al. 2018) together with calcium phosphate ( $\text{Ca}_3(\text{PO}_4)_2$ ) (Le Corre et al.  
10 2009). Struvite is more preferred in agricultural use due to the fact that magnesium (Mg), N and P are  
11 released simultaneously (1:1:1 M ratio), and that the rate of nutrient release is slow compared to other  
12 fertilizers (Puchongkawarin et al. 2015). Deficient concentrations of phosphorus, on the other hand,  
13 limit the struvite precipitation. Even the presence of toxic compounds and/or micropollutants in  
14 wastewater restrain its purity and further agricultural application. Hence, alternative nutrient recovery  
15 technologies (i.e. membrane, electrodialysis) should be considered to improve the quality of the  
16 recovered nutrients (Xie et al. 2016). Several other benefits are also associated with P recovery by  
17 crystallization. For instance, the volume of the sludge produced together with other undesired  
18 precipitates diminishes, which eventually decreases the cost of sludge disposal (Hermassi et al. 2018).  
19 Integration of the Short Cut Enhanced Nutrient Abatement (SCENA) system into the Carbonera  
20 WWTP, Italy, was previously evaluated (Longo et al. 2017) and the results motivated the Horizon  
21 2020 ‘SMART-Plant’ action which is currently running and investigates the optimization of the best  
22 scenario for SCENA within SMARTech4a and SMARTech4b. Briefly, the SCENA system integrates  
23 the following processes: optional upstream concentration of cellulosic sludge, fermentation of  
24 dynamic thickened sewage sludge to produce VFAs as carbon source, and nitrogen and phosphorus  
25 removal (by P-bioaccumulation) via nitrite from sludge reject water using an SBR. In this  
26 configuration, nitrogen is removed through the bioprocesses of nitrification/denitrification, and Enhanced

1 BPR (EBPR) via nitrite using the VFAs from sludge fermentation liquid as carbon source.  
2 SMARTech4b is another validated SCENA pilot-scale system at the WWTP of Psytalia, Greece. It  
3 enables the integration of the enhanced biogas recovery (by thermal pressure hydrolysis) of sewage  
4 sludge with side stream energy-efficient and compact nitrogen removal and phosphorus recovery.  
5 The CAMBI™ thermal hydrolysis process has been installed to treat 50% of the produced sludge  
6 before this is sent for AD. The integration of CAMBI™ with anaerobic digestion produces, after  
7 dewatering, a reject water stream that has a very high ammonium nitrogen concentration (>1.2 gN/L)  
8 to be removed in the SCENA unit.

9 Furthermore, many technologies are applied in full size for specific objective of phosphorous salts  
10 recovery. In fact, CalPrex™ reactor (Pre-digestion P-recovery) placed between the acid phase and  
11 gas phase digesters enables dissolved phosphorus in dewatered centrate precipitates and is recovered  
12 as a brushite crystal. Similarly, the AirPrex™ reactor (Sludge optimization and P-recovery) placed  
13 between the anaerobic digester and the dewatering equipment converts the orthophosphate into  
14 struvite crystals, which are harvested from the bottom of the reactor. Both the WASSTRIP™ and  
15 Ostara Pearl™ processes are already in operation in a number of municipal installations and achieve  
16 efficient P recovery (Point et al. 2017). Examples of commercial processes for P recovery and the  
17 different final P products derived are thoroughly listed and discussed (Melia et al. 2017). Potassium  
18 recovery from wastewater has not been substantially considered and is an emerging issue (Batstone  
19 et al. 2015). There are also quite number of other promising nutrient recovery technologies that are  
20 yet invalidated, such as microbial recovery cell- anaerobic osmotic membrane bioreactor ((MRC)-  
21 AnOMBR system (Hou et al. 2017), reactive sorbents (Hermassi et al. 2018) and microalgae (Viruela  
22 et al. 2016).

### 23 **3.2.2 Recovered materials: PHA**

24 Biopolymers are a group of polymers with similar properties to petroleum-based plastics, produced  
25 from renewable sources also by different types of bacteria using carbon as a substrate (Raheem et al.  
26 2018). The main advantages of PHAs are the possibility of being completely biodegradable and non-



1 toxic. PHA-storing bacteria are well-known to grow in activated sludge processes of WWTPs that  
2 store these polymers as carbon source and energy reserve (Frison et al. 2015). The series of operations  
3 needed for microbial production of PHAs are substantially described in the literature (Tamis et al.  
4 2014; Anjum et al. 2016). Ongoing pilot-scale demonstrations in recent years offer fundamental  
5 experience to produce PHA from waste materials in enough quantities to inspire value chains and  
6 investment within first bio-based value chains. To launch the private and public relationships that  
7 will drive the economic and regulatory framework, it is a crucial to verify and explore technology  
8 process basis, to validate recovered material flows to marketable renewable resources (Valentino et  
9 al. 2017). SMARTech2b stands as the key to enable secondary mainstream energy-efficient resource  
10 recovery in Manresa WRRF, Spain. It applies the mainstream SCEPPHAR (Short-cut Enhanced  
11 Phosphorus and PHA Recovery) and consists of two sequencing batch reactors (SBRs); one for  
12 heterotrophic bacterial growth operated under anaerobic/anoxic/aerobic sequence (HET-SBR), and  
13 another SBR for autotrophic nitrifiers growth (AUT-SBR), an interchange vessel and a chemical  
14 system for P-recovery as struvite. The integrated system accomplishes enhanced N-removal and P-  
15 recovery in municipal WWTP. PHA is recovered from the anaerobic purge of the SBR. SMARTech5  
16 also applies the SCEPPHAR concept in Carbonera WWTP, Italy, which was conceived as a modified  
17 version of SCENA where PHA recovery is an economically sustainable option. It accounts of the  
18 following subprocesses: (i) cellulosic primary sludge fermentation to enhance the production of VFAs  
19 and release nitrogen and phosphorus in soluble forms (i.e. ammonia and phosphate); (ii) solid and  
20 liquid separation of the fermentation products and recovery of struvite from the sewage sludge  
21 fermentation liquid by the addition of  $Mg(OH)_2$  to favour the precipitation; (iii) ammonium  
22 conversion to nitrite accomplished in a SBR; (iv) selection of PHA storing biomass in a SBR by the  
23 alternation of aerobic feast conditions and followed by anoxic famine conditions for denitritation  
24 driven by internally stored PHA as carbon source; (v) PHA accumulation using a fed-batch reactor to  
25 maximize the cellular PHA content of the biomass harvested from the selection stage. Within the  
26 context of the INCOVER Project, pilot-scale mainstream phototrophic PHA recovery is conducted in

1 Viladecans, Spain. PHA is produced through photo-bioreactors in which microalgae and  
2 cyanobacteria communities grow in a symbiotic relationship, removing pollutants from urban and  
3 agricultural wastewaters and accumulating PHA. Produced biomass is then fed into the AD with  
4 sewage sludge or other biomass sources as co-substrate for biogas production (incover-project.eu).  
5 An innovative biogas upgrading technology is also implemented, based on the symbiosis between  
6 microalgae and bacteria and the photosynthetic fixation of CO<sub>2</sub>, which removes CO<sub>2</sub> and H<sub>2</sub>S to produce  
7 biomethane of 92%. In El Torno WWTP, Spain, PHA production is produced through two-stage  
8 anaerobic-photosynthetic high rate algae pond systems that are consisting of pulse feeding of  
9 municipal wastewater pre-treated in an UASB reactor with molasses as COD source. Similarly, after  
10 PHA production, the remaining biomass is converted into biogas using thermal pre-treatment and an  
11 anaerobic co-digestion process followed by biogas upgrade.

### 12 **3.2.2 Recovered materials: Cellulose**

13 Municipal wastewater contains high amounts of cellulose fibre (30–50% of the total suspended solids)  
14 that is mainly originated from toilet papers (Behera et al. 2018). These cellulose fibres easily enter  
15 biological treatment systems of WWTPs if they are not separated during the primary treatment;  
16 biodegradation of cellulose is comparatively difficult and depends on many factors (Ruiken et al.  
17 2013; Crutchik et al. 2018). On the other hand, cellulose fibres hold a great potential as a resource  
18 which can be recovered from wastewater by sieving (Ruiken et al. 2013). The benefits of cellulose  
19 dewatering sludge are: minimization of chemical consumption, lower electricity consumption for  
20 aeration, less chance of phosphate release and much lower sludge volume to discharge that reduces  
21 sludge handling and management cost. Cellulose harvesting is expected to have added benefits to the  
22 WWTP's downstream biological process and provided outside the WWTP for the downstream  
23 blending with PHA and processing for final biocomposite production. SMARTech1 comprises an  
24 innovative integration of dynamic fine-sieving together with in-situ post-processing that is currently  
25 validated in the municipal WWTP of Geestmerambacht, Netherlands. CirTec has developed flow  
26 scheme with filter for primary treatment (Salsnes Filter<sup>TM</sup>) and separating cellulosic fibres to produce

1 a highly-concentrated sludge. The produced cake layer or fine sieved fraction (FSF) harvested from  
2 Salsnes Filter<sup>TM</sup> has a very heterogeneous composition containing mainly cellulosic fibres originating  
3 from toilet paper. The result is a market-ready cellulose that has been cleaned, dried and disinfected.  
4 Examples of the recovered materials from WWTPs in SMART-Plant are shown in Fig. 1.

#### 5 **4. Technologies to the market: focus on the environmental technology verification and other** 6 **performance certifications**

7 At EU level, innovative environmental technologies are validated by Environmental Technology  
8 Verification (ETV) Program to prove the reliability of the developed claims and help technology  
9 purchasers identify innovations that suit their needs. Hence, the best long-term technical,  
10 environmental and economic performances are validated by ETV protocol. ETV ensures that the  
11 performance claims are as structured and complete in order to present a clear assessment of the  
12 technology's potential and value. However, it does not cover the evaluation of the technology's  
13 performance against standard or pre-defined criteria. More information can be found at ETV's  
14 official website ([https://ec.europa.eu/environment/ecoap/etv\\_en](https://ec.europa.eu/environment/ecoap/etv_en)).

15 In addition to validation of a technology, the functional properties of recovered materials should be  
16 determined using specific functional tests to compare recovered products with industrial products.

17 The use of phosphate salts, biochar and pyrolysis materials, is more controlled and regulated  
18 compared to other recovered materials. The European Sustainable Phosphorus Platform (ESPP) are  
19 implementing many activities for the sustainable management of phosphorus and other nutrients.

20 STRUBIAS - EU Fertilisers Regulation - sets criteria for nutrient recovery rules within EU  
21 Fertilising Products Regulation. At national level, authorisations of struvite/recovered phosphates as  
22 fertilisers together with phosphorus recycling legislations are in force in some EU countries. The  
23 main challenge of these organic-based fertilizers is to ensure that their application is not resulting in  
24 an accumulation of different organic non-biogenic and inorganic compounds (e.g., toxic metals and  
25 non-metals) (Hermassi et al. 2018). Strong debates continue regarding the social awareness of  
26 consumers and framework regularities about food security (3rd European Nutrient Event, 2018).

1 Inconsistency and high-variability of available sources for recovered PHA quality in routine  
2 production is still a hidden gem (Valentino et al. 2017). For instance, the extracted PHA from  
3 municipal secondary wastewater was examined using  $^{13}\text{C}$  NMR spectroscopy (Kumar et al. 2018).  
4 Size exclusion chromatography (SEC) and differential scanning calorimetry (SDC) were also used  
5 for the characterization of the recovered PHA (molecular number, molecular weight, glass transition  
6 temperature etc.) (Frison et al. 2015). However, consistent quality of recovered PHA derived from  
7 wastewater as feedstock has not been proven or refuted presenting a big challenge in scaled-up  
8 implementations (Valentino et al. 2017).

## 9 **5. Valorisation of recovered materials to consumer/industrial products**

10 Sustainability assessment of the recovered materials from wastewater was conducted (Zijp et al.  
11 2017) with respect to 6 different categories as follows: economic welfare, resource depletion,  
12 environmental and biological quality, technical welfare, human health and social welfare. It was  
13 concluded that PHA and struvite seemed economically feasible in terms of production costs and  
14 market values. However, PHA needs urgent and further investigations as it exhibits some critical  
15 barriers, which have to do with the possible emissions of toxic compounds during the production  
16 stage and concerns regarding the perception of the market on the food security. Similarly, struvite  
17 utilization also depends on location-specific aspects and legislations and needs further assessment.  
18 Cellulose recovery and application, on the other hand, seems less feasible due to the costs of extra  
19 hygiene step when used in the paper and carton industry. This step is not required in construction  
20 applications which makes cellulose recovery more beneficial for all resource themes. In a recent  
21 study, better value was derived from valorising CPS to VFAs and struvite from the fermentation  
22 liquid, then  $\text{CH}_4$  was further recovered after AD of remaining fermentation solids (Crutchik et al.  
23 2018). The authors made a simple comparison by assuming  $\text{CH}_4$  market price of  $0.11 \text{ €/m}^3$ , the best  
24 valorisation of  $\text{CH}_4$  from CPS could be up to  $0.46 \text{ €/capita}\cdot\text{year}$ . Acetate and propionate price could  
25 be as high as  $0.45$  and  $1.01 \text{ €/kg}$ , respectively, meanwhile struvite could be sold up to  $0.76 \text{ €/kg}$ .  
26 Therefore, the VFAs and struvite route before biomethanization have the potential to increase the

1 market value potential of CPS up to 1.55-1.95 €/capita-year (Crutchik et al. 2018). Overall, potential  
2 end-use of the recovered materials with respect to market requirements highly influence its role within  
3 circular economy. Some of the potential end-uses of the recovered materials from WWTPs are  
4 discussed with existing market values and possible valorisation alternatives.

5 The different market possibilities and the discussion of the advantages and disadvantages for the  
6 recovered materials commercialization opportunities are summarized in Table 5.

### 7 **5.1. Market possibilities for recovered materials: Nutrients**

8 Adding nutrient-loaded sorbents enhances the soil quality in terms of agricultural yield and nutritional  
9 quality. However, socioeconomic conditions highly influence whether such materials can be  
10 applicable in commercial agriculture. The key factors that influence the application of post-sorbent  
11 fertilizers are the availability of feedstock, the technology to manufacture fertilizers, and the  
12 investment costs and capacity. (Hermassi et al. 2018). In addition, soil measurements together with  
13 plant bioavailability indices can help to determine fertilizer performance (Peng et al. 2018). Effects  
14 of struvite as fertilizer on various plants can be found (Kataki et al. 2016).

15 The market value of struvite varies from 188 to 763 €/t struvite in the recent years (Molinos-Senante  
16 et al. 2011; Desmidt et al. 2015). Although economic feasibility of struvite recovery is limited by  
17 high operational costs, it was also determined that when the struvite sale price is assumed as 560  
18 €/ton, the net profit of 445.62 €/day was obtained for a full-scale fertilizer production industry with a  
19 500 m<sup>3</sup>/day capacity. (Yetilmezsoy et al. 2017). The European Commission's draft "market study"  
20 assesses the possible sources of raw materials for nutrient recycling, STRUBIAS technologies and  
21 economic aspects. High quality of these struvite-based products enables them to be used as effective  
22 slow-release fertilizers for agriculture practices. Furthermore, P recovery also aids to cease  
23 eutrophication in aquatic environments. In this regard, if economic aspects for P recovery are not  
24 satisfactory, environmental benefits and government regulations could be the driving force (Peng et  
25 al. 2018). During the market development strategy for struvite, focus should be based on a holistic  
26 approach considering pricing, demand, purity, size, storage, transportation and distribution with

1 respect to the existing regulatory framework of contaminants and eco-toxicity (Desmidt et al. 2015;  
2 Katakai et al. 2016).

## 3 **5.2. Market possibilities for recovered materials: Biopolymers**

4 Biopolymers must compete with petroleum-based polymers, which are available in high amounts at  
5 relatively low prices. Biogas could be also considered the main competitor for biopolymer production  
6 in WWTPs since organic carbon from waste material will not be diverted for the production of  
7 biopolymers when the production of biogas is more convenient (Kleerebezem et al. 2015). Thus, the  
8 market potential of bioplastics seems limited so far (Van Der Hoek et al. 2016). However, (EEA  
9 report No 8/2018) reported that the global production of plastics is estimated to account for about 7  
10 % of the world's fossil fuel consumption. The proportion of bioplastics is still low, currently below 1  
11 %. However, the worldwide biopolymer production capacity is forecast to increase from 6.6 million  
12 tonnes in 2016 to 8.5 million tonnes in 2021.

13 Production of biopolymers from waste feedstock seems advantageous and economical depending on  
14 the market requirements. It has multiple applications especially in material and packaging industries  
15 and utilisation of waste feedstock as substrate makes a great contribution to waste management and  
16 reduces environmental pollution. For instance, these waste materials are proved to be efficient  
17 substrates producing significant amounts of PHA or extracellular polymeric substances (EPS) that  
18 can help to reduce the production cost by eliminating the usage of pure carbon sources. Research is  
19 still on-going for the lower-cost production of PHAs by utilization of such low cost wastes and using  
20 wild and mutant strains of microorganisms (Anjum et al. 2016). Optimization of the processing  
21 techniques can pave the way to take PHA formation from waste materials to industrial level and then  
22 into the market (Pakalapati et al. 2018). At the moment, the bottleneck of the process seems to be the  
23 extraction of PHA from the biomass which requires thermal and/or chemical processes which are  
24 usually expensive.

### 25 **5.2.1 Market possibilities for recovered materials: PHA**

1 PHA has gained greater attraction in the recent years due to their many advantages such as  
2 biodegradability, biocompatibility, controllable thermal and mechanical properties as well as  
3 molecular weight diversity, which allow them to be used as bioimplant materials for medical and  
4 therapeutic applications (Zhang et al. 2018). Although the utilization of waste materials for the  
5 synthesis of high-class materials such as PHAs has led to cost reduction as previously mentioned, the  
6 final products cannot be used in medical applications where high purity products with non-toxic  
7 nature are of utmost considerations (Raza et al. 2018). PHAs recovered from waste materials can  
8 contain viral, plasmid, bacterial or genetic contaminations that hinder their potential usage for medical  
9 applications. Impurities in PHA regarding proteins, lipids, endotoxins, antifoam agents, DNA and  
10 hypochlorite have been previously reported (Koller et al. 2013b, a). Such impurities require specific  
11 post recovery washing procedures that eventually cause a major increase in product cost (Raza et al.  
12 2018).

13 In this regard, the majority of recovered PHA applications take place in a wide range of products  
14 including paper coatings, bags, containers, food packaging materials, bottles, cup etc. (Muhammadi  
15 et al. 2015). For instance, water-resistant layer for paper, film or cardboard can be produced out of  
16 the latex of PHAs (Anderson and Dawes 1990; Bourbonnais and Marchessault 2010). PHAs can also  
17 be used to replace petrochemical polymers in toner and developer compositions as well as ion-  
18 conducting polymers (Muhammadi et al. 2015).

19 Industrial PHAs and their applications were discussed by Anjum and colleagues (Anjum et al. 2016).  
20 Among these materials and their applications, recovered PHA can find its own place in such practices:  
21 Biopol (co-polymer of poly (3-hydroxybutyrate-co-3-hydroxyvalerate)), currently produced by  
22 Metabolix (Cambridge, MA, USA), can be used in packaging materials, shampoo bottles, disposable  
23 razors, disposable cups, disposable knives and forks. Nodax (PHA copolymer family consisting of 3-  
24 hydroxybutyrate) (P&G Chemicals, USA/Japan) is available as foams, fibres or nonwovens, films  
25 and latex among others. Biogreen (Mitsubishi Gas Chemicals) developed the production of P (3HB)  
26 from methanol, and markets it under the trade name Biogreen. Furthermore, PHAs can be used to

1 make foils and diaphragms, pressure sensors for keyboards, stretch and acceleration measuring  
2 instruments, material testing, shockwave sensors, lighters, gas lighters; acoustics, and for ultrasonic  
3 therapy and atomization of liquids (Anjum et al. 2016). A high performance of PHA biopolymer,  
4 Minerv-PHA (Minerv, Italy), takes the place of highly pollutant materials such as PET, PP, PE, HDPE  
5 and LDPE. The most known commercially-available PHA products can be found elsewhere  
6 (Bugnicourt et al. 2014).

7 Other than being used mainly as environmentally friendly plastics for packaging purposes, PHAs are  
8 considered as a source for the synthesis of chiral compounds which highlights them as raw materials  
9 for the production of paints (Reddy et al. 2003; Mohammadi et al. 2015). Furthermore, PHA can be  
10 hydrolysed chemically, and the monomers can then be converted into molecules such as 2-alkenoic  
11 acids,  $\beta$ -hydroxy acids,  $\beta$ -hydroxyalkanols,  $\beta$ -hydroxyacid esters,  $\beta$ -acyllactones,  $\beta$ -amino acids,  
12 which hold great potential as biodegradable solvents (Madison and Huisman 1999; Mohammadi et  
13 al. 2015). Other important, industrial applications of PHAs are printing and photography, art-smart  
14 gels, heat-sensitive-adhesives and also fishing equipment (Pakalapati et al. 2018).

15 Blending PHAs, in particular polyhydroxybutyrate (PHB), with other polymers, or with plasticizers,  
16 creates opportunities to enhance their properties by decreasing the processing temperature and  
17 lowering the brittleness of PHAs based plastics. So far many blends containing PHB/PHAs have been  
18 investigated and several types of plasticizers have been proposed (Bugnicourt et al. 2014).

19 In addition, nanocomposites of PHA are also reported (Anjum et al. 2016). For example, the  
20 preparation of biodegradable nanocomposites using NFC and poly(3-hydroxybutyrate-co-3-  
21 hydroxyvalerate, PHBV) as the polymer matrix has been investigated (Srithep et al. 2013). This can  
22 be a good alternative to valorise two types of recovered materials from WWTPs in one application.

23 PHA yield on carbon source, its productivity and downstream costs determine their introduction into  
24 the global market (Możejko-Ciesielska and Kiewisz 2016). The selected end-use of PHA often  
25 determines the market specifications and requirements.



1 The current cost of PHAs production and recovery with aqueous two-phase extraction method is  
2 stated as 5.77 USD/kg. However, utilizing a cheaper carbon source such as sludge has the potential  
3 to reduce the final PHA production price significantly (Leong et al. 2017). Hence, the theoretical  
4 price of PHAs produced in fed-batch mode using waste materials could reach up to 3.51 €/kg. Yet,  
5 they are still not cost-efficient when compared to their synthetic alternatives such as polypropylene  
6 and polyethylene, which cost 1.47 and 1.15 €/kg, respectively (Możejko-Ciesielska and Kiewisz  
7 2016). The current PHA price ranges between 2.2-5.0 €/kg, which depends on monomer composition  
8 and is usually higher for the copolymers. In spite of having several environmental advantages, the  
9 PHA prices are still not commercially-competitive with conventional petroleum-based polymers,  
10 which typically cost less than 1.0 €/kg (Valentino et al. 2017).

### 11 **5.2.2 Market possibilities for recovered materials: EPS**

12 EPS are biopolymers that are considered eco-friendly, cost effective and sustainable alternatives to  
13 substitute the existing chemical flocculants. Potential environmental applications of EPS can be listed  
14 as follows as summarized from More et al., 2014): Water treatment (Wang et al. 2012), wastewater  
15 treatment (Li et al. 2013), colour removal from wastewater (Liu et al. 2009), sludge dewatering (Yang  
16 et al. 2012), metal removal or recovery (Mikutta et al. 2012), removal of toxic organic compounds  
17 (Zhang et al. 2011), landfill leachate treatment (Zouboulis et al. 2004) and soil remediation and  
18 reclamation (Chandran and Das 2011). Existing literature is limited to lab-scale applications and  
19 further research is still needed to be scaled up to field applications since EPS can be used as a cost-  
20 effective treatment alternative. The cost of the EPS extraction and purification can be the limiting  
21 factor in the field application considered of various sectors with chemistry, structure and properties  
22 of interest (More et al. 2014).

### 23 **5.3. Market possibilities for recovered materials: Cellulose**

24 Classification of cellulosic materials as well as current and emerging markets for cellulose-based  
25 products have been thoroughly discussed (Keijsers et al. 2013). Accordingly, cellulose markets were  
26 classified into 9 categories: Textile, non-woven, wood and timber, pulp/paper and board, cellulose

1 dissolving pulp, cellulosic films, building materials, cellulosic fibre composites and green chemicals.  
2 The selected end-use of a certain lignocellulosic raw material often specifies the market requirements.  
3 Hence, the end-use of cellulose determines the market prices and volumes, which are directly linked  
4 to the cellulose quality and defined over physical properties, chemical composition, unwanted  
5 components, prior treatments of raw material, and physical, chemical, biological stability of the  
6 cellulose. For instance, end-uses for cellulose such as pulp, paper and board are pointed on brightness,  
7 tensile and tear, freeness, write-ability; the price range is between 450-650 raw material price €/ton.  
8 Polymeric cellulose will eventually have higher chemical purity requirements. Meanwhile, market  
9 requirement of building materials for extracted cellulose is based on its strength, moisture absorbency  
10 and fire retardancy. Details of market volume and market price of purified celluloses can be found in  
11 the literature (Keijsers et al. 2013).

12 Considering its nature and high energy content, fine sieved fraction (FSF) has started to gain attraction  
13 in countries such as the Netherlands (Ghasimi et al. 2016). The recovered cellulose can be used either  
14 as raw material to make paper products, adhesion binders for asphalts (Crutchik et al. 2018) or as the  
15 fibrous reinforcement material in bricks (Kim et al. 2017) when properly separated and refined. For  
16 instance, cellulosic material recovered from screenings were used as an ingredient in the production  
17 of asphalt to create a bike path near Beemster WWTP, the Netherlands (Selster A.S., 2018). Similarly,  
18 Makron (Finland) uses recycled cellulose fibre additives for asphalt.

19 The use of natural fibres as the adsorbent is another emerging trend in environmental engineering  
20 applications including environmental remediation and water filtration membranes as the fibres are  
21 abundant, readily available and are more environmentally friendly compared to carbon based  
22 materials (Carpenter et al. 2015). They are used as adsorbents in wastewater treatment (i.e. in the  
23 form of membrane) and for the removal of adsorbates such as oil, dyes, heavy metals and ionic  
24 compounds (Rahman et al. 2018). As the production of effective adsorbents at low cost and low  
25 energy consumption is placed at the centre of many researches, the properties and possible  
26 modifications of recovered cellulose need to be thoroughly investigated and understood.

1 Furthermore, market projections of cellulose nanomaterial-enabled products are estimated (Cowie et  
2 al. 2014) and recent developments in production of cellulose nanofibrils (CNF) were discussed  
3 (Nechyporchuk et al. 2016). Fluorescent cellulose bio-based plastics were successfully fabricated  
4 based on the strong hydrogen bonding interaction between cellulose chains and conjugated dye  
5 molecules, and further suggested as a good candidate for making anti-counterfeiting banknotes (Wang  
6 et al. 2016). In another study, the transparent and flexible cellulose-based nanocomposite papers were  
7 fabricated to be used as solar cell substrates (Cheng et al. 2018). However, smooth surface was  
8 proposed to be maintained on the cellulosic material to avoid problems during the coating process.  
9 The use of cellulose nanocrystals for thermal insulation can be another option to value recovered  
10 cellulose from WWTPs (Septevani et al. 2017). CNF can be combined with clay for the preparation  
11 of nanopaper to obtain unique brick-and mortar structure (Carosio et al. 2016).

12 Cellulose can be efficiently used to produce valuable chemicals or biofuels, such as VFAs (Guo et  
13 al. 2015), poly lactic acid (Graupner et al. 2009; Jamshidian et al. 2010) and bioethanol (Zabed et  
14 al. 2017). However, it should be noted that the use of cellulose for biogas and bioenergy production  
15 are positioned at the bottom of the biomass value pyramid (Gavrilescu 2014).

16 The possibility to use the separated materials to safely produce toilet paper was also suggested as a  
17 real cradle to-cradle application, but difficulties in relation to social acceptance were also highlighted  
18 (Ruiken et al. 2013). In all conditions and possible applications, extraction and purification methods  
19 of cellulose must be thoroughly studied to assess its feasibility to meet the criteria of end-use markets.

1

2 **Table 5.** Market possibilities, quality indicators and market price range for recovered struvite, PHA and cellulose.

3

| Product   | Market possibilities  | Quality indicators   | Price range (€/t)  | Reference                                       |
|-----------|---|--|--|---|
| Struvite  | <ul style="list-style-type: none"> <li>• Fertilizer</li> </ul>  | <ul style="list-style-type: none"> <li>• Solubility, purity, plant bioavailability, metal content, crystal size</li> </ul>   | <ul style="list-style-type: none"> <li>• 188-763</li> </ul>  | (Molinos-Senante et al. 2011; Peng et al. 2018) |
| PHA       | <ul style="list-style-type: none"> <li>• Paper (coating)</li> <li>• Packaging (foils)</li> <li>• Electronics (headphones, keyboards)</li> <li>• Printing and photography</li> <li>• Others</li> </ul>   | <ul style="list-style-type: none"> <li>• Purity, toxicity, chemical constituents, blending</li> </ul>  | <ul style="list-style-type: none"> <li>• 2.2-5.0 €/kg</li> </ul>   | (Anjum et al. 2016; Valentino et al. 2017)      |
| Cellulose | <ul style="list-style-type: none"> <li>• Textiles</li> <li>• Non-woven</li> <li>• Wood, timber</li> <li>• Pulp, paper and board</li> <li>• Cellulose dissolving pulp</li> <li>• Cellulosic films</li> <li>• Building materials</li> <li>• Cellulosic fibre composites</li> <li>• Green chemicals</li> </ul> | <ul style="list-style-type: none"> <li>• Purity/colour/fibre length/distribution/lustre/softness/hygienic</li> <li>• Purity/fibre length/ distribution/ absorbency</li> <li>• Density/strength and modulus/durability/hardness/colour</li> <li>• Brightness, tensile and tear, freeness, writability</li> <li>• <math>\alpha</math> Cellulose %, polymerisation degree</li> <li>• <math>\alpha</math> Cellulose %, polymerisation degree</li> <li>• Strength, moisture absorbency, fire retardancy</li> <li>• Compatibility</li> <li>• Glucose yield/extractability</li> </ul> | <ul style="list-style-type: none"> <li>• 1200-1900</li> <li>• 200-400</li> <li>• 450-600 €/m<sup>3</sup></li> <li>• 450-650</li> <li>• 1600-2000</li> <li>• 3000-3500</li> <li>• -</li> <li>• 200-400</li> <li>• 50-100</li> </ul> | (Keijsers et al. 2013)                          |

## 1 **5 Barriers to resource recovery and reuse and solutions to overcome**

2 Actually, notwithstanding the important research activities and the developed technologies for  
3 resources recovery from the real WWTPs, many bottlenecks for the market uptake and for their  
4 application could be identified.

5 In fact, the law for water reuse, actually, regulates only the irrigation purpose. None specific  
6 indications and legislations were clearly promoted for fertigation objective as highlighted even by  
7 EU Innovation Deal on sustainable wastewater treatment combining anaerobic membrane technology  
8 and water reuse. Many lacks have been identified both in the legal definition of the term discharge  
9 and for quality standards provisions adopted for wastewater effluents to be used for agriculture.  
10 Moreover, recognition of the economic and environmental benefits of water reuse within reclaimed  
11 water pricing have to be implemented.

12 For phosphorous and ammonia salts, more detailed studies and programs have been developed at  
13 European level to overcome regulatory barriers. Not similar evidences have been identified for PHA  
14 and cellulose potential recovery.

15 Moreover, the quality, the purity and the characteristics of the recovered resources change on the  
16 basis of the implemented process in the WWTPs. From the other hand, the different market sectors  
17 request inlet materials with diverse standards on the basis of the final productive application. For this  
18 reason the certification of the technologies, which also has to include the main properties of the  
19 recovered products, seems necessary to couple the recovery processes to the industrial sectors.

20 In this direction, the European criteria of “End-of-waste” could be identified as possible legislative  
21 solution to support the resources recovery application in the WWTPs. In fact, this approach (Waste  
22 Framework Directive 2008/98/EC) specifies when certain waste ceases to be waste and obtains a  
23 status of a product or a secondary raw material. The obtainment of the end of waste status has to be  
24 supported by several conditions: 1- the substance or object is commonly used for specific purposes;  
25 2- there is an existing market or demand for the substance or object; 3- the use is lawful (substance

1 or object fulfils the technical requirements for the specific purposes and meets the existing legislation  
2 and standards applicable to products); 4- the use will not lead to overall adverse environmental or  
3 human health impacts. Starting from this point, specific regulations, centred on the end of waste  
4 concept, could be implemented to support the regulatory framework of the resources recovery. This  
5 approach can justify and encourage the technological investments in the WWTPs to economically  
6 address and support the resources recovery and to promote the circular economy in the water sector  
7 (Guest, et al., 2009).

8 Finally, public perception and social acceptance are insufficiently developed for all the described  
9 materials. Therefore, specific formative and public dissemination activities have to be strongly  
10 supported.

## 11 **6 Conclusions**

12 This paper has provided a commentary on recent advances in energy-efficient resource recovery  
13 approaches in WWTPs. Anaerobic processes stand as the “gold mine” in wastewater mining strategy  
14 while AnMBRs are the “gold diggers”. While the biorefinery concept has been widely recognized,  
15 lab-scale studies are gradually evolving into validated pilot/full scale implementations. Onsite energy  
16 recovery is still getting most attention. However, recently-developed and validated processes, such  
17 as SCENA and SCEPPHAR, derive more value to VFAs, while achieving satisfactory nutrients and  
18 PHA recovery, respectively. Together with nutrients and PHA, cellulose is another value-added  
19 material to be recovered in WWTPs. Among all the energy and material recovery methods, some are  
20 consistently considered to be beneficial to improve sustainability, and some of them still need further  
21 research to achieve desired feasibility. Struvite has a comparatively large market, also brings strong  
22 debates on food security that needs to be addressed in the near future. Valorisation of PHA and  
23 cellulose, by the way, should not be overlooked since there are huge market alternatives. Therefore,  
24 there is a need to develop the regulatory framework for resource recovery and carry out  
25 socioeconomic assessments considering the market potential and specific requirements.

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1 **Figure captions**

2 **Fig. 1.** Examples of the recovered products in the context of SMART-Plant Project **(a)** Recovered  
3 PHA from SMARTech2b in Manresa WWTP, Spain **(b)** Recovered struvite from SMARTech5 in  
4 Carbonera WWTP, Italy **(c)** Recovered cellulosic sludge from Geestmerambacht WWTP,  
5 Netherlands



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