**REVIEW PAPER** 



# Valorization of digestates from urban or centralized biogas plants: a critical review

F. Guilayn D · M. Rouez · M. Crest · D. Patureau · J. Jimenez

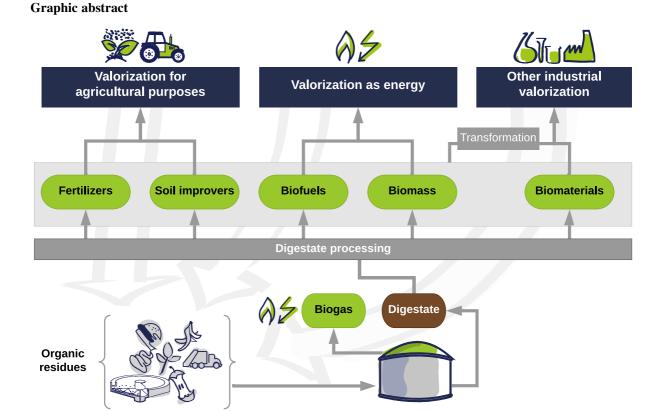
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Abstract Direct landspreading of anaerobic digestates is the most common digestate management strategy. Nevertheless, digestate post-treatment can be unavoidable, especially for environmental services providers operating large-scale anaerobic digestion (AD) facilities. This review aims to assess the technical feasibility of achieving value-added products from digestates from urban and/or centralized AD plants (UC-AD). An exhaustive effort was dedicated to identifying and clarifying the available processing technologies and specific issues that can be related to UC-AD digestates. The valorization options were classified according to the final product destination. The result is a useful information source for assessing digestate valorization pathway given a local market and context. Agriculture was the first destination to be considered, as it allows a more direct closing of nutrient and carbon cycles. Several processes exist either for concentrating desirable characteristics of

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F. Guilayn · D. Patureau · J. Jimenez LBE, INRAE, Univ Montpellier, 102 avenue des étangs, 11100 Narbonne, France digestates, enhancing organic matter stability or producing pure and reformulated fertilizers. Thermal conversion processes are either under development or full-scale demonstration. They allow to valorize the solids through the production of biofuels and/or biochar and in the coming future, to start a whole biorefinery system. Similarly, biomass harvesting processes such as microalgae are under upscaling, enabling to valorize the nutrients of the digestate liquid phase while producing renewable biomass from sunlight. Several value-added products were already obtained in laboratory to pilot conditions from UC-AD digestates, for example, biopesticides, biosurfactants and composite materials. Adding to technical challenges, the quality variation of digestates, regulation barriers, public acceptance and the difficult access to new markets are among the main obstacles to UC-AD digestates valorization into value-added products.



**Keywords** Anaerobic digestion · Biogas effluent · Digestate valorization · Biogas slurry · Nutrient recovery · Resource recovery · Circular economy · Upcycling · Biowaste · Food waste · Sewage sludge · Wastewater treatment plant

# Abbreviations

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ABP	Animal by-products
AC	Activated carbon
AD	AnAerobic digestion
DM	Dry matter
FAO	Food and Agriculture Organization of the
	United Nations
GCV	Gross calorific value
HLS	Humic-like substances
HM	Heavy metals
MBT	Mechanical biological treatment
MSW	Municipal solid waste
NA	Not available
NUE	Nutrient uptake efficiency
OFMSW	Organic fraction of municipal solid waste
OLR	Organic loading rate

OM	Organic matter
OMP	Organic micropollutants
RT	Retention time
SS	Sewage sludge
TAN	Total ammoniacal nitrogen
TSS	Total suspended solids
UC-AD	Urban or centralized AD plants
TKN	Total Kjhedal nitrogen
VFA	Volatile fatty acids
VFA VS	Volatile fatty acids Volatile solids
	•

# 1 Introduction and objectives of the review

Anaerobic digestion (AD) of organic residues is a well-developed and growing solution for both waste upcycling and green energy production. The European Biogas Association defines digestate as the solid or liquid material from controlled anaerobic fermentation processes of biodegradable material (European

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Biogas Association 2015). For the treatment of organic residues, AD present several advantages, such as:

- Waste stabilization with mass and volume reduction (Möller et al. 2010; Kothari et al. 2014);
- Increased recycling of organic matter and nutrients, thus promoting the conservation of natural resources (Drosg et al. 2015);
- A cost-effective (Kothari et al. 2014) and relatively simple technology capable of processing a wide range of substrates (Ağdağ and Sponza 2005; Appels et al. 2008; Capson-Tojo et al. 2016);
- Resulting valuable products: biogas, a renewable energy source and *digestate*, recognized by its application as a fertilizer and/or soil improver.

This review is oriented to digestates from urban and/or centralized AD plants (UC-AD). These plants are here defined as those receiving mostly urban or industrial inputs (such as sewage/industrial sludge, food/household/commercial waste, municipal solid waste, among other). They are usually not inserted in the context of farms though in many cases the centralized plants<sup>1</sup> do receive agricultural inputs. In opposition, agricultural digestates receive, essentially, agricultural residues, livestock manure/slurry and/or energy crops. Compared to agricultural digestates, UC-AD digestates present particular issues and more challenging management, which will be addressed in this review.

As in the context of sewage sludge management in wastewater treatment plants (WWTP), the choice of the digestate treatment pathway on biogas plants should be evaluated in an early stage of AD projects conception. For the definition of a treatment line/ process, the designers should consider the economic and environmental aspects of the local legislation, as well as the quality requirements from the possible consumers and disposal sites nearby the plant. However, compared to digestates, sewage sludges have characteristics that can be more easily generalized. Actually, the word *digestate* refers to a set of very heterogeneous matter (Guilayn et al. 2019a). Digestate processing equipment are being adapted from previous technologies developed for other substrates such as

raw manure, wastewater and sewage sludge (Al Seadi et al. 2013). As AD is being intensively encouraged, policymakers, public and private environmental sectors can deal with a difficult decision-making scenario.

A significant number of digestate-dedicated scientific reviews have been already published. Some of them are digestate post-treatment inventories, including commercial solutions and promising techniques (Fuchs and Drosg 2013; Sheets et al. 2015). Other reviews have more special focus; such as the effects of AD on the digestate characteristics (Möller and Müller 2012), valorization of digestates from the organic fraction of municipal solid waste<sup>2</sup> (OFMSW) (Logan and Visvanathan 2019), valorization of agricultural digestates (Monlau et al. 2015), the risks associated to the agricultural spreading of digestates (Nkoa 2014), characterization methods (Teglia et al. 2010), nutrient recovery techniques (Makádi et al. 2012; Lebuf et al. 2013; Lin et al. 2015; Tao et al. 2016; Vaneeckhaute et al. 2017), algae production (Uggetti et al. 2014; Monlau et al. 2015), energy valorization and anaerobic biorefinery (Uggetti et al. 2014; Sawatdeenarunat et al. 2016). However, to the best of the authors' knowledge, no review has a special focus on urban and centralized AD facilities, the broad possibility of value-added end-products and their technical feasibility. This review aims to fill up this gap, while:

- Applying a reverse approach, giving insights and assessing the technical feasibility of achieving value-added products with existing and developing technologies.
- Classifying the valorization options according to the final product (instead of process), for providing a piece of practical information for urban and centralized AD plants needing to better valorize digestates.

Before addressing the valorization techniques, more detailed contextualization is provided: first a global UC-AD digestate production prospection (Sect. 2), then a review of digestate properties (Sect. 3). The latter includes the effects of AD on digestate fertilizing value and innocuity. Following (Sect. 4), the specific challenges of UC-AD digestate

 $<sup>\</sup>overline{1}$  Internally defined as large scale AD plants receiving a wide diversity of waste streams and with an installed capacity superior to 30–50 kt/y.

 $<sup>^2</sup>$  OFMSW: not source-separated. Defined operationally by the authors as the organic material obtained after mixed collection of municipal solid waste and separation by mechanical biological treatments (MBT).

valorization are discussed. Finally (Sect. 5), the two objectives above are addressed throughout a critical review on UC-AD digestate valorization backed up by both scientific and technical expertise from co-authors counting with academic and industrial backgrounds. Attached to this review: (i) a detailed summary of digestate processing technologies (Appendix A in ESM), which includes its by-products, advantages, bottlenecks and readiness levels; and, (ii) the methodology used for gathering an initial scientific publication library along with a short bibliometric study on digestate valorization technologies (Appendix B in ESM).

#### 2 Digestate production: situation and prospection

Currently, the world's biggest economies present either important biogas sectors or specific policies that will boost centralized and urban AD (Table 1). Those might include direct economic incentives or indirect legislation such as the international trend for minimizing organic waste landfilling and incineration. Besides, AD is more than a trending solution to agricultural issues in both developed and developing countries. It is also between the most suitable solutions to treat and recover the increasing volumes of urban residues such as food waste (Capson-Tojo et al. 2016). There is certainly a trend for increasingly global production of UC-AD digestates,thus raising the concerns around its destination.

#### **3** Digestate properties

#### 3.1 Fertilizing and amendment value

The same AD process mechanisms make both agricultural and UC-AD digestates recognized as fertilizers, despite the possible different associated risks. The effects of AD on the fertilizing and amendment value of digestates are summarized in Table 2.

Due to a conversion of the more easily biodegradable organic matter into biogas (mostly  $CH_4$  and  $CO_2$ ), AD increases the biological stability of the digested material and the concentration of recalcitrant OM such as lignin and humic-like substances. Humification degree is therefore increased during AD, which was demonstrated by several advanced techniques (Massaccesi et al. 2013). Some authors suggest the occurrence of humification processes during AD (Polak et al. 2005; Brunetti et al. 2012). In any case, AD engenders an improvement in the organic soil amendment value of the input residues.

OM abatement also implies the reduction of dry matter (DM) and viscosity (Möller et al. 2008; Möller and Müller 2012). DM reduction is about 25% (Smith et al. 2010). Final DM values greatly depend on the applied process moisture. Moreover, as small-sized particles are first consumed, AD tends to lead to better dewatering properties of the digestate if compared to the raw material (Hjorth et al. 2010).

In AD, organic matter conversion rates may greatly vary (13–65% (Monlau et al. 2015), 20–95% (Möller and Müller 2012)), depending mostly on the type of substrate fed to the digester, AD parameters (organic loading rate, retention time and temperature) and the resulting performance (Bauer et al. 2009). Easily biodegradable inputs such as food waste, some types of OFMSW, animal slurries and cereal grains, will tend to induce greater OM reduction (Fuchs and Drosg 2013). On the contrary, since lignocellulosic material is more difficulty digested (Labatut et al. 2011), fibrous lignocellulosic material such as litter bed, silages and cattle manure will lead to lower DM and OM reduction and digestates with a greater VS/DM ratio if not pre-treated (Möller and Müller 2012).

For application as a soil amendment, the C/N ratio of organic material must be compatible with the soil microorganism's requirements. This is necessary to avoid excessive N release (from OM decomposition, if the C/N is too low) or, in the opposite scenario, soil-N immobilization (microbiological uptake, if the C/N is too high). Following OM mineralization, a significant fraction of the input C is converted to CO<sub>2</sub> and CH<sub>4</sub> (biogas), the C/N ratio is thus reduced during AD (Möller et al. 2008). For the same configuration, higher retention times will lead to greater C consumption thus lower C/N ratios on digestate. Desired digestate C/N stability thresholds are reported between 10 and 20 (Teglia et al. 2010). This value can be highly variable depending on the digestate type, ranging from about 3 to 20 (Guilayn et al. 2019a).

Digestates inherit the nutrients content from the AD feedstock. However, due to the mass reduction (OM biodegradation), digestate nutrients concentrations tend to increase. Nonetheless, the fertilizer value of a given product not only depends on the total nutrient

Table 1 AD situation and prospect in some of the world's biggest economies

Country/ region	Remarkable information	References
European Union	World's biggest producer of electricity from biogas (half of the world's production)	Council Directive (1975), Deremince and Königsberger (2017), Liebetrau et al. (2017),
	AD boosted through extensive national support schemes such as feed-in-tariffs	Scarlat et al. (2018)
	Over 16.000 AD plants (2017), Germany is the leading country with 8000 agricultural plants and over 1200 sludge digestion plants (2015)	
	European policies favor organic waste prevention while encouraging recovery techniques such as AD over landfilling	
United	1480 organic waste AD plants producing biogas	USDA, U.S. EPA and U.S. DOE (2014)
States of America	With proper support, over 11,000 existing sites could host an AD process to generate biogas in farms or WWTP	
	The US is currently the second-fastest-growing market for renewable energies after China	
China	One of the first countries to have implemented massive AD policies	Scarlat et al. (2018)
	AD is now being applied for almost a century	
	Over 40 million of familiar AD units (agricultural)	
	Large-scale engineered biogas installations are also numerous, estimated above 100,000 in 2014	
	AD tends to be boosted with the Chinese Medium-and-Long Term Development Plan for Renewable Energy. This plan aims 8000 new large-scale biogas plants and almost doubling the number of domestic digesters	
Japan	At least 63 AD plants treating animal waste to produce electricity and/or heat. Food waste AD is also reported	Ministry of Land Infrastructure Transport and Tourism (2013), Yokoyama and Matsumura
	300 Japanese WWTP had AD tanks in 2010	(2015)
	Among national incentives, Japan government established a generous feed-in tariff system back in 2012 aiming to boost renewable electricity injection in the grid	
India	With the implementation of a National Biogas and Manure Management Program back in the 1980s': the country counted with 4.75 million household digesters in 2014 and had an estimated potential for additional 12 million	Ministry of New and Renewable Energy (2014)
	Biogas production from centralized AD is reported to be growing fast	
Brazil	Biorefinery pioneer with the bioethanol production from sugarcane after the National Alcohol Program in the 1970s'	Mariani (2015), ANP (2017)
	Biogas technologies are underdeveloped. A recent survey in Brazil has listed only about 130 AD plants producing more than 1250 m <sup>3</sup> of biogas/day	
	Important regulatory landmarks should boost AD in the coming years, such as the recent resolution authorizing biomethane injection in the natural gas grid	

content. It also depends on the nutrient availability to the plants and their uptake efficiency, which also tends to be enhanced with AD. During the digestion process, the organic fraction of nutrients is mineralized and released from complex organic compounds, thus increasing N, P, K, Ca and Mg accessibility. This effect is highly relevant for organic N and organic P, and notably the first (Möller and Müller 2012; Al Seadi

Characs.	Effect	Resulting change	Main mechanisms	References
DM	Decrease	- 1.5% to - 5.5% (absolute)	Destruction of biodegradable matter	Möller et al. (2008), Pognani et al. (2009), Vaneeckhaute et al. (2013a)
VS or OM	Decrease	- 5 to -15% DM (absolute)	OM conversion to biogas	Möller and Müller (2012)
Viscosity	Decrease	NA	Destruction of small particles	Carballa et al. (2009), Hjorth et al. (2010), Möller and Müller (2012)
Dewater- ability	Possible increase	NA	Prior destruction of small particles that are commonly slow-settling/non-filterable	Carballa et al. (2009), Hjorth et al. (2010)
Biodegrad.	Decrease	Highly depends on RT	OM conversion to biogas	Teglia et al. (2011), Menardo et al. (2011b)
рН	Increase	+ 0.5 to + 2 units	VFA degradation, ammoniacal nitrogen release	Kiely et al. (1997), Möller and Müller (2012), Batstone et al. (2015)
Fibers content	Increase	Final lignin content about 10 to 20% DM basis	Lignocellulose is poorly biodegradable	Pognani et al. (2009), Menardo et al. (2011b), Dabert (2015), Sambusiti et al. (2015, 2016), Cavalli et al. (2016)
Amend. value	Increase	Highly variable. No consensus on indicators.	Increase in concentration of recalcitrant OM such as humus precursors	Pognani et al. (2009), Tambone et al. (2009)
Nutrient availability	May vary, overall positive effect for N, P and K	Highly variable	Increase if organic N and P mineralization Decrease if formation of Ca/Mg/ Fe–P compounds	Möller and Müller (2012), Bachmann et al. (2016)
TAN	Increase	+ 25 to 55%	Mineralization of proteins and amino acids. Released TAN is partially used for biomass growth	Möller and Müller (2012)
Total N	From concentration increase to possible significant losses	Usually little/no change	Increase: mass reduction due to OM conversion Loss: struvite incrustation/ settling and ammonia volatilization	Möller et al. (2008, 2010), Banks et al (2011), Möller and Müller (2012), Zirkler et al. (2014)
Total P, K, Ca, Mg	From concentration increase to possible significant losses	Usually little/no change	Increase: mass reduction due to OM conversion Loss: precipitation and incrustation inside the digester	Marcato et al. (2008), Schievano et al. (2011), Banks et al. (2011), Zirkler et al. (2014)
S	No change/possible loss	Usually little/no change	Degradation of OM containing S, reduction to $H_2S$	Schievano et al. (2011), Zirkler et al. (2014)
C/N	Decrease	-3 to $-5$ units	C mineralization	Möller and Müller (2012)
Heavy metals	Increase of concentration	Highly variable	Concentration: digestate mass and volume reduction	Appels et al. (2008), Marcato et al. (2009), Stefaniuk et al. (2015)
	Reduced bioaccessibility		Accessibility: biological uptake and pH increase reducing metals solubility	
Heavy metals	Possible loss of Cd and Zn	Highly variable	Precipitation of Cd and Zn sulfides, which has low solubility and a high density	Zirkler et al. (2014)

Table 2 Summary of AD effects on inputs and resulting digestate quality

Table 2 continued

Characs.	Effect	Resulting change	Main mechanisms	References
Bad odor	Decrease	up to—98%	VFA degradation	Powers et al. (1999), Hjorth et al. (2009)
Pathogens	Partial to complete kill	30% up to > 99% abatement	Temperature and time; competition for substrate with the adapted AD microorganisms	Carballa et al. (2009), Bonetta et al. (2014)
OMP	Highly variable	Highly variable	Highly compound-dependent	Brändli et al. (2007), Govasmark et al. (2011), Stasinakis (2012), De Moor et al. (2013), Mailler et al. (2014, 2017)

AD anaerobic digestion, *Bio-degrad* biodegradability, *Characs* characteristics, *DM* dry matter, *NA* not available, *OM* organic matter, *OMP* organic micropollutants, *RT* retention time, *TAN* total ammoniacal nitrogen, *VFA* volatile fatty acids, *VS* volatile solids

et al. 2012; Mehta and Batstone 2013; Bachmann et al. 2016). However, the pH rise may also lead to the formation of low plant-available compounds such as calcium phosphates (Wahal et al. 2010; Möller and Müller 2012; Mehta and Batstone 2013).

Along with the OM digestion, highly biodegradable material tends to produce digestates with a higher Total Ammoniacal Nitrogen to Total Nitrogen (TAN/TN) ratio and the opposite for fibrous materials. N-rich digestates are greatly associated with animal waste such as pig slurry and poultry manure, slaughterhouse waste, sewage sludge, cereal residues and some agriindustrial residues. Some specific biowaste and many fibrous materials (e.g. maize silage, cattle manure) are related to lower N-content digestates (Benoît et al. 2014). In fact, the protein content on the feedstock is the greatest source of the N and S contents of digestates (Straka et al. 2007; Möller and Müller 2012; Park and Kim 2016).

Throughout the AD process, several mechanisms promote a pH increase from about 0.5 to 2 units (Möller and Müller 2012). Final pH in digestates ranges from 7 to 9 (internal databases). In usual conditions, the pH increase is driven by the consumption of volatile fatty acids (VFA) and the production of ammonium (Kiely et al. 1997; Möller and Müller 2012; Batstone et al. 2015). Moreover, increasing the pH buffering capacity by adding an alkali is usually performed in AD operation, in order to avoid the inhibition of methanogenic archaea by acidification (Nguyen et al. 2015).

For storage and spreading purposes, the conversion of VFA and the destruction of odor components (e.g. benzaldehyde and phenols) are responsible for a significant decrement of odor nuisances in digestates if compared to raw inputs (Hjorth et al. 2009).

On the negative side, several mechanisms during AD may lead to serious nutrient losses in full-scale operational systems, even if many authors relate negligible unbalances. First of all, the mineralization of N during AD produces ammoniacal nitrogen, which can represent a + 10 to + 33% TAN share on TN (Möller and Müller 2012). Higher TAN concentrations coupled with a pH increase favors the conversion of NH<sub>4</sub><sup>+</sup> to free ammonia (NH<sub>3</sub>). This effect results in an important increase of ammonia volatilization risk during the AD process or digestate storage, transport, spreading and processing (Nkoa 2014), but not systematically (Risberg et al. 2017). Also due to the pH rise, carbonates, phosphates and cations such as  $NH_4^+$ ,  $K^+$ ,  $Ca^{2+}$  and  $Mg^{2+}$  may precipitate and incrust (e.g. as struvite) inside the digester and equipment downstream (Marcato et al. 2008), which might represent non-negligible nutrient loss.

Globally, AD tends to significantly upgrade the fertilizing value of the input waste in both organic and mineral aspects. Resulting digestates may present nutrient uptake performances equivalent or better than those of industrial mineral fertilizers (Vaneeckhaute et al. 2013a, b), as well as amendment characteristics that can be similar to those of composts (Tambone et al. 2010). However, if no specific quality criteria are available for raw digestates, they can be outside the scope of standards and regulations conceived for conventional fertilizers and soil amendments. For example, for France and EU, Guilayn et al. (2019a) observed that raw digestates were too wet (low DM) for meeting "soil improvers" regulations and with

poor nutrient content (wet weight basis) for meeting several "(organic-)fertilizer" regulations.

# 3.2 Digestate innocuity

The conversion of OM to biogas represents a mass loss that might concentrate not only the valuable nutrients but undesirable components such as heavy metals (HM) and organic micropollutants (OMP) (Lehmann et al. 2015). Nevertheless, a prevailing absence of exceeding legislation limits can be observed in the literature, in studies from several countries and for various types of digestates (Benoît et al. 2014; Zirkler et al. 2014; Dabert 2015; Koszel and Lorencowicz 2015). For HM, it was confirmed with a previously published meta-analytic study by the same authors of the present review (Guilayn et al. 2019a). Nevertheless, an important number of studies present concerns regarding the ecotoxicological effects and the environmental safety of digestates for land application. Gas emissions, heavy metals, ammonium toxicity, high salinity and pathogens are among the most frequent concerns. (Teglia et al. 2011; Alburquerque et al. 2012; Bonetta et al. 2014; Owamah et al. 2014; Tigini et al. 2016). The content of non-biodegradable compounds such as plastics, metals, glasses and stones, also called "physical/inert impurities", is often related to digestates from urban and commercial waste, including OFMSW, source-separated biowaste (such as household food waste) and depackaging unities (Dabert 2015, SUEZ expertise).

Regarding HM, several authors have reported hazardous characteristics of animal manures, sewage sludge and OFMSW (Zirkler et al. 2014; Dabert 2015). Indeed, animal manure is one of the main sources of HM because Cu, Zn and other heavy metals are widely applied in animal feed due to antimicrobial and growth-stimulating effects (Poulsen 1998). For example, copper sulfate is widely used on dairy disease-preventing footbaths. Crop residues may also be related to chemical contamination due to the application of pesticides, impurities of mineral fertilizers and the accumulation of heavy metals on the soil. Cadmium, for example, is a major contaminant issue in agriculture as it is easily assimilated by plants and it is between the most toxic heavy metals to humans. Cadmium soil contamination in fields is related to the application of P fertilizers from phosphate rocks, spreading of sewage sludge and atmospheric transport of mining dust (Van Bruwaene et al. 1984; Robson et al. 2014). In the case of OFMSW, a study has associated higher HM of Cr, Hg and Pb (Dabert 2015). Zirkler et al. (2014) reported higher levels of Zn, Pb, Cd and Ni for digestate with sewage sludge inputs compared to agricultural digestates. Interestingly, they report Cd and Zn loss which was associated with precipitation as sulfides.

Despite the concerns on total HM concentrations, several authors indicate that the chemical accessibility of HM is decreased with AD. Apart from biological accumulation (e.g. biofilm complexation, chelation or translocation to the cells' interior), the decreasing of heavy metals' accessibility is suggested to be driven by the pH increase that reduces the solubilities of metals and promotes precipitation processes with carbonates and sulfides (Bloomfield and McGrath 1982; Zandvoort et al. 2006; Marcato et al. 2009). Nevertheless, the HM bioavailability to plants will greatly depend on soil characteristics (Almeida et al. 2019). Little attention seems to be dedicated to plant growth experiments for assessing the actual plant HM uptake after spreading (Marcato et al. 2009).

As for HM, OMP concentrations on digestates will depend on the effective control on the use of such substances at the source of the AD feedstock and its respective production chain. Apart from sewage sludge (SS) and manure AD, little attention seems to be dedicated to the fate of OMP during AD. Papers dedicated on sewage sludge AD (Stasinakis 2012; Mailler et al. 2014, 2017) assessing pharmaceuticals, estrogens, flame retardants, phthalates, detergents, hydrocarbons, among others, concluded that the fate of these contaminants in AD is generally influenced by solids retention time (SRT), temperature, sludge composition, bioaccessibility of the compounds and the adaptation of the microbial biomass. Results are variable and highly compound-dependent.

Regarding biological contamination, weed seeds, crop disease spores, resistance genes, bacteria, viruses, fungi, and other pathogens present in the feedstock are partially eliminated during AD, depending on SRT and temperature for a possible full abatement (Al Seadi et al. 2012; Kjerstadius et al. 2013; Youngquist et al. 2016; Tian et al. 2016; Seruga et al. 2020). For safety reasons, even if some AD configurations are being demonstrated to insure sanitation, legislations as the European (The Commission of the European Communities 2011), for Animal By-Products (ABPs),

may require the pasteurization of specific AD feedstocks (before AD) or the whole digestate, regardless the following digestate processing.

# 3.3 Time variability of digestate quality

AD feedstock may greatly vary on quality and quantity during an annual basis, which may impact the composition of digestate within the same biogas plant, especially if those variations provoke process instabilities. Zirkler et al. (2014) monitored 4 full-scale biogas plants in Germany during one year concluded that digestates from the same plant present important heterogeneity over time, discouraging studies bases on single samples.

It must be stated, however, that AD can be regarded as a tool for reducing the time variability of waste streams for better organic waste management within a territory. For example, a French long-term research project (Project DIVA, Dabert 2015) monitored 5 fullscale plants with diversified inputs and processes and noted a variability usually inferior to 10% and always inferior to 20% for all the monitored characteristics (including agri-value and OMP). In parallel, much greater variability in the feedstock was observed during the same period. Banks et al. (2011) collected AD input (mostly food waste) and output data for a mass balance study under a period of 426 days. They observed less variability of N, P and K output (digestate) than the input (food waste). These observations can be explained by an overall buffering effect of the AD plant.

Quality warranty is necessary for a trusted and legal marketing of bio-based products. For marketing purposes, it is usually attained by labeling the products based on existing quality criteria. However, UC-AD digestates are, most of the time, out of the current regulations conceived for composts or classical fertilizers (Guilayn et al. 2019a). Additionally, UC-AD digestates can present non-source separated inputs that are stricter regulated in the EU (Guilayn et al. 2019a). Both are reasons why digestate post-treatment might be necessary to ensure digestate valorization.

#### 3.4 Types of UC-AD digestates

Supported by the works of Guilayn et al. (2019a), two great types of UC-AD digestates can be defined: those coming from wet and dry AD processes. This

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classification appeared as a first factor explaining the clustering analysis.

Except for sewage sludge mono-digestion, UC-AD digestates from wet AD processes are originated mainly from co-digestion plants and regional facilities, many times integrated to WWTP. In many cases, process water is used to dilute the inputs, which implies an increased capacity of digestate post-treatment downstream. A rising configuration is the dilution of inputs during some depackaging techniques of packed biowaste. Common inputs are sewage sludge, food waste, source-separated organic waste, OFMSW, food-agri industrial residues and even agricultural residues. The reactors are usually Continuously Stirred Tank Reactor (CSTR) working in mesophilic conditions.

According to Guilayn et al. (2019a) these digestates present a low C/N ratio (< 10 and usually < 8), moderate to low VS content (55–70%, DM basis) and high TN (50 up to > 150 g/kg DM) (Guilayn et al. 2019a). When presenting higher inputs of sewage sludge and other P-rich material such as pig slurry, they present a high P-content (20  $\pm$  10 g/kg DM). When presenting great amounts of protein-rich feedstock they present a particularly high TAN/TN (64  $\pm$  11%), which is regularly the case of food waste and livestock slurry.

UC-AD digestates from dry-AD plants are mostly originated from OFMSW and source-separated organic waste (biowaste). The digestion is performed in high-solids conditions (DM > 15% but up to 30-40%) (Li et al. 2011). Most of the centralized Dry-AD plants in Europe seem to be performed in continuous plug flow reactors but several commercial batch systems exist (Li et al. 2011; André et al. 2018). In dry conditions, the reduced content of water allows a better energy efficiency to heat the digesters. Dry-AD is thus commonly performed in thermophilic conditions (Li et al. 2011). The higher process temperature induces faster hydrolysis kinetics that compensates for the lower mass transfer rates and also results in greater pathogen inactivation. On a dry matter basis, they present lower VS (40-50% DM basis) and poorer nutrient content (< 50 g N/kg DM, < 10 g P/kg DM and < 25 g K/kg DM), which is probably linked to inputs. However, if comparing nutrient contents of dry and wet-AD UC-AD digestate on a wet weight basis, no clear difference can be observed (Guilayn et al. 2019a). Adding to that, digestates from Dry-AD were observed to present C/N ratio greater than 8–10, which can be necessary for several soil amendment regulation and quality criteria.

In both cases, a common inconvenience of UC-AD digestates is the presence of inert impurities that will affect negatively the processes downstream and the quality/acceptability of final products. The content of inert impurities greatly depends on the effectiveness of source-separation and/or on an effective removal/ depackaging step before AD. Effective pre-treatment of OFMSW can be achieved to remove this kind of material, producing a digestate that can be processed to achieve quality standards for landspreading (SUEZ 2017a).

As a rule of thumb, UC-AD digestates present inferior content of fibers compared to agricultural digestates, as it can be observed in Table 3. Agricultural digestates may present a lignocellulosic content up to 875% OM due to large quantities of silage, manure and crop residues. In its turn, are mostly between 25 and 30%. This is strikingly important for the choice of post-treatment equipment and the possible valorization pathways, as discussed later.

It must be noticed, though, that UC-AD may also receive important volumes of (ligno-)cellulosic material such as green waste, paper waste, cardboards and even agricultural residues. All these inputs can be recalcitrant. As it can be observed in Table 3, the UC-AD digestates presenting the highest content of residual fibers are (i) a digestate from a biowaste greatly composed of green waste and (ii) an OFMSW digestate. The OFMSW from different locations are highly variable but can be composed of a significant amount of paper waste and cardboards (Demirbas 2006 (Turkey); Zhang and Banks 2013 (UK); Li et al. 2016 (USA)).

# 4 UC-AD digestates management: challenge, particularities and opportunities

Land spreading of digestates is the most common valorization route. However, for the providers of environmental service operating urban and regional facilities (non-agricultural), several legal, logistical and technical bottlenecks along with environmental concerns can be listed:

- (i) Digestate spreading can be strictly controlled by environmental authorities. In some cases, it can be associated with important environmental risks including ammonia volatilization, pathogens, organic micropollutants and heavy metal content, over-fertilization, nutrient runoff, among other (Nkoa 2014);
- (ii) Digestate spreading regulation is usually stricter for digestates originated from non-source-separated AD inputs. For example, in the new EU regulation on fertilizers (2019/1009), digestates are an authorized component material but excluding those from sewage sludge, OFMSW and other mixed-stream substrates as AD feedstock;
- (iii) Production of UC-AD digestates is permanent and relatively constant on a yearly basis (in quantity), while the agricultural needs are highly seasonal. Digestates must thus be stored for a long time until the growing season, or even transported to distant regions (King et al. 2013; Gong et al. 2013);
- (iv) Adding to the time-quantity issue, digestate characteristics might be highly time-variable (Zirkler et al. 2014), depending on feedstock variations and process performance stability. In this matter, UC-AD digestates may be more problematic than agricultural ones. In order to operate in full capacity and to maximize methane production, the operators may receive a very wide variety of organic waste streams within the territory (internal expertise). Quality variation may be problematic in terms of quality control for machinery operation, landspreading and marketing purposes (Dahlin et al. 2015);
- (v) The production of digestate may exceed the capacity of the local available arable lands for receiving nutrients (Vaneeckhaute et al. 2013a; Nkoa 2014). Increasing land competition implies in high and growing digestate transportation costs. According to Dahlin et al. (2015) the digestate transport distance is reported to have doubled in the last years to distances reaching 150 km. Moreover, the number of large and centralized facilities exceeding local nutrient spreading capacity tends to grow since they present better economic feasibility.

Main inputs as designed by the source	Type	Actual digestate destination	Innovative research: final product	Fraction	DM % ww	VS % DM	HC % VS	C % VS	HC + C % VS	LG % VS	Source
Three digestates mainly composed of cattle manure, cattle/swine slurry and silage	Agri	NA	Methane (i)	SF	12.4–29	83.6–89.7	13.9–26.3	36.1–43.1	56.9-63.2	21–27.9	Menardo et al. (2011a)
95% cattle manure, 5% millfeed (% ww)	Agri	Landspreading	NA	SF	26.5	72.8	23.8	34.3	58.1	14.3	Dabert (2015)
51% pig slurry, 49% FAI <sup>a</sup> (% ww)	Agri	Landspreading	NA	RW	6.4	70.9	20.3	24.2	44.5	16.9	Dabert (2015)
84% OFMSW, 16% source- separated urban biowaste	UC-AD	(Co- )composting	NA	RD	25.1	43.7	19.7	22.6	42.3	34.6	Dabert (2015)
9% groats, 29% olive oil cake, 57% triticale silage and 5% chicken manure <sup>b</sup>	Agri	Not available	Ethanol (ii)	SF	94.4°	89.9	19.4	22.7	42.0	35.3	Sambusiti et al. (2016)
95% cattle manure, 5% millfeed	Agri	Spreading	NA	RD	17.4	65.5	17.5	23.5	41.0	18.3	Dabert (2015)
86% biowaste <sup>d</sup> , 14% FAI <sup>e</sup> (% ww) UC-AD	UC-AD	(Co- )composting	NA	RD	19.3	53.1	8.6	32.3	40.9	30.5	Dabert (2015)
42% cattle slurry, 18% cattle manure, 17% pig slurry, 14% liquid FAI <sup>f</sup> , 9% other <sup>g</sup>	Agri	Landspreading	NA	RW	5.7	68.9	13.2	23.6	36.8	17.1	Dabert (2015)
Source selected biowaste	UC-AD	(Co- )composting	Enzymes, biosurfactants, biopesticides (iii)	SF	24.4	63.0	16.0	16.5	32.5	27.8	Cerda et al. (2019)
59% OFMSW, 22% cow manure slurry, 18% agro-industrial waste and 2% energy crops (%ww)	UC- AD/ Agri	NA	NA	RW	5.8	75.1	9.1	13.0	22.1	26.8	Pognani et al. (2009)
Source-separated food waste	UC-AD	NA	NA	RD	19.9	61.8	7.6	3.3	10.8	3.1	Tampio (2016)
Source-separated domestic food waste	UC-AD (lab- scale)	NA	NA	RW	6.7	67.7	5.0-6.0	3.6-5.3	8.6–11.3	2.6–2.8	Tampio (2016)
VW and waste activated sludge	UC-AD (pilot)	NA	NA	RD	34.2	6.69	6.7	2.5	9.2	3.2	Tampio

Main inputs as designed by the source	Type	Actual digestate destination	Innovative research: final product	Fraction	DM % ww	VS % DM	HC % VS	C % VS	HC + C % VS	LG % VS	Source
Source-separated OFMSW <sup>h</sup>	UC-AD	NA	NA	RD	32.2	58.7	4.8	3.0	7.8	2.1	Tampio (2016)
C cellulose, DM dry matter, FAI food/agri-industrial waste, HC hemicellulose, LG lignin, NA not available or not applicable, OFMSW organic fraction of municipal solid waste, RD wholraw digestate from dry-AD, RW whole raw digestate form wet-AD, SF solid fraction, SOL soluble matter, UC-AD urban and/or centralized AD plant, VS volatile solids,	d/agri-ind , RW whol	lustrial waste, HC	7 hemicellulose, LG orm wet-AD, SF so	f lignin, NA 1 lid fraction,	not availat SOL solut	ole or not ap	plicable, <i>O</i> <sup>1</sup> <i>\C-AD</i> urba	<i>FMSW</i> orgar	ic fraction of ralized AD p	f municipal dant, VS vc	solid waste, datile solids,
ww wet weight											
<sup>a</sup> FAI: sludge from slaughterhouse, grease sludg	grease sluc	lge, swine digest	e, swine digestive contents, Millfeed, among other	sed, among	other						
<sup>b</sup> NOt clear if %ww											
<sup>c</sup> Dried milled											
<sup>d</sup> Biowaste: OFMSW, paper, cardboard, textile,	ard, textilé	e, green waste									
<sup>e</sup> FAI: yogurt, dough, grease, among other	other										
<sup>f</sup> FAI: no more information											
<sup>g</sup> Other: cereals residues, silage, among other	ong other										
<sup>h</sup> not clear since the original author also defined	also define		OFMSW as being originated by MSW mixed collection followed by MBT	MSW mixed	1 collectio	n followed	by MBT				
Innovative research topics											
(i) Thermal post-treat. to produce CH4 (AD): successful	(H4 (AD):	successful									
(ii) Post-treatment for bioethanol fermentation:	rmentation		successful (about 9% of whole digestate's COD)	gestate's C(	(QC						
(iii) Fermentation for producing biosurfactants	vsurfactant	s and other value	and other value-added compounds: low soluble carbohydrate content thus poor yields	s: low solub	le carbohy	drate contei	nt thus poor	· yields			

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A holistic view of a biogas plant considering digestate treatment and destination is necessary to avoid inappropriate management that could lead to negative results in terms of carbon print, energy balance, economic viability and public acceptance of AD as a waste valorization option. As discussed, land spreading is not individually enough to overcome the digestate challenge, especially for UC-AD digestates. The application of digestate upgrading technologies is essential and such processes might have different and multiple objectives, including:

- Treating or removing undesired digestate characteristics as a waste/wastewater treatment strategy (e.g. chemical precipitation, nitrification/denitrification), prior to digestate valorization or simply for reaching disposal requirements,
- Reducing transportation costs by concentrating the valuable components and properties of the digestate (e.g. solid/liquid separation, thermal drying, membrane filtration, evapo-concentration);
- Producing relatively pure and value-added products (e.g. N-stripping and struvite crystallization);
- Ensuring controlled and homogenous quality of products over time;
- Increasing market acceptance and
- Creating/reaching new markets.

To achieve these objectives, compared to agricultural AD plants, UC-AD designers and operators may benefit from higher capital investment, economies of scale, proximity to industrial clusters and better synergy with industrial actors.

# 5 Achieving value-added products from nonagricultural digestates

The literature review on digestate valorization processes started by a systematic literature research and a bibliometric study presented on Appendix B in ESM. In brief, a research query was developed for identifying publications containing, in the title, multiple variations of the words "digestate" (e.g. biogas effluent/slurry, anaerobically digested) and "treatment" or "valorization" (e.g. processing, recovery, removal). This query resulted in 1362 papers that were manually verified, resulting in a first library of 520 publications. For complementing the review, this original library has been extensively expanded by looking into citations and by performing specific queries on the identified processes and products.

The valorization of digestates into value-added products was separated into three great categories: agriculture, energy and other industrial valorization. The products cited in the coming sections are summarized in Tables 4, 5, 6 and 7, with a focus on feasibility. No product prices were added either because of their incertitude for fertilizers (highly volatile N/P/K world commodity market) or due to the lack of established market and market prices for other components (notably the carbon content) and byproducts.

This review classifies digestate products according to their destinations, not according to processes that generate them. Products and processes are thus repeated when necessary, with different focus depending on their destinations. A same processe can generate different products for different destinations. Processes and their general advantages and bottlenecks are thus introduced only once during the review, usually, the first timethey are cited but clearly indicated in the few exceptions. Digestate processing techniques are summarized in Appendix A in ESM, where inputs, outputs, advantages, digestate limiting (necessary) characteristics, bottlenecks, costs and readiness levels are included.

### 5.1 Agricultural valorization

The following sections summarize a wide range of options for generating value-added products from UC-AD digestates, aiming to reach the agricultural market. Agriculture is considered a priority since closing the food production loop is essential for meeting our civilization challenge of sustainable agriculture and food supply. Adding to that, returning organic matter to soil is one of the most promising climate change mitigation strategies (e.g. in the frame of the "4 per mille initiative") (Minasny et al. 2017).

As numerous reviews have addressed nutrient recovery from digestates (Lebuf et al. 2013; Romero Güiza et al. 2016; Vaneeckhaute et al. 2017; Monfet et al. 2018), our focus is to address the technical issues related to UC-AD digestates.

Agricultural valorization of digestates will be classified, in the following sections, into three product categories: N/P/K fertilizers (Sect. 5.1.1) and soil improvers & organo-mineral fertilizers (Sect. 5.1.2).

Table 4 Summary of	Table 4 Summary of agricultural valorization: value-added pro-	ducts from UC-AI	value-added products from UC-AD digestates and qualitative considerations	
Final product designations	Config.	TRL	Major product concerns, not including legislation <sup>a</sup>	References <sup>b</sup>
Liquid fraction, liquid phase, supernatant	Raw digestate mechanical separation	Commercial	<ul> <li>N-concentration → gaseous emissions, especially NH<sub>3</sub> and N<sub>2</sub>O</li> <li>Relative low nutrient concentrations- &gt; short transportation -distances</li> <li>Salinity</li> <li>Chemical and organic contaminants</li> <li>Pathogens</li> <li>Market value can be &lt; 0</li> </ul>	Al Seadi et al. (2012), Nkoa (2014)
Concentrates, retentates (from membrane or evaporation processes)	Pressure-driven membrane filtration of (pre-treated) LF (Vacuum)-evapoconcentration of LF or membrane retentates	Commercial	Acidification needed to avoid N-loss thus N-depleted product	Vaneeckhaute et al. (2017)
Adsorption products	Adsorption columns or batch reactions starting from LF	Not clear	Nutrient accessibility can be poor	Vaneeckhaute et al. (2017)
N/S solutions: Ammonium sulfate or ammonium nitrate	NH <sub>3</sub> from direct or indirect stripping processes (stripping columns, evaporators, dryers or composting)	Commercial	The resulting solution is often too acid and corrosive	Vaneeckhaute et al. (2017)
Struvite (magnesium ammonium phosphate, MAP)	Crystallization in raw digestate LF mainly	Commercial	<ul> <li>- Nutrient accessibility can be low especially in alkaline soils</li> <li>- Heavy metal contents</li> </ul>	Desmidt et al. (2015), Yetilmezsoy et al. (2017), Vaneeckhaute et al. (2017)
K-struvite	c.f. struvite	Not clear	c.f. struvite	Desmidt et al. (2015), Vaneeckhaute et al. (2017)
Ca-P, mainly calcium c.f. struvite phosphate or hydroxyapatite	c.f. struvite	Commercial	c.f. struvite	Desmidt et al. (2015), Vaneeckhaute et al. (2017)
Incineration ashes	Combustion of dried digestate pellets	NA	No nitrogen P-accessibility tends to be poor	Ehmann and Lewandowski (2013), Christel et al. (2014)

oduct

16)

Mulbry et al. (2005), Uggetti

Simple use as fertilizer does not allow the economic

ot

References<sup>b</sup>

Major product concerns, not including legislation<sup>a</sup>

The different options are summarized in Fig. 1 (N/P/K fertilizers) and Fig. 2 (soil improvers and organomineral fertilizers). Other agricultural products such as biopesticides are discussed in the "Industrial valorization" section.

According to studies from different countries, those are characteristics that the farmers are concerned when dealing with organic fertilizers and/or soil improvers (Tur Cardona et al. 2015; Dahlin et al. 2015; Case et al. 2017): nutrient accessibility (equivalency to traditional mineral fertilizers), consistency in the nutrient content, odor nuisances, capacity to enhance soil structure, salinity, pathogens, possible application with current available machinery and price (often free to farmers). Based on literature material, an effort has been made to address these points for the different products under the agriculture section.

# 5.1.1 N/P/K fertilizers

5.1.1.1 Liquid fraction from phase separation (dewatering) To begin with, a relatively simple dewatering process can be a tool to enhance digestate value. It produces a liquid fraction (LF) and a dewatered fraction also called "solid fraction" (SF). The most common phase separation equipment are screw presses and decanter centrifuges, either isolated or combined in this sequence (Guilayn et al. 2019b). Phases separation of digestates tends to concentrate nutrients into the LF and the organic matter into the SF, but the global mass balances are highly dependent on the separation technique whose choice depends on digestate characteristics thus AD input type (Guilayn et al. 2019b). Centrifuges present a much greater separation performance but are costly to operate and not adapted to digestates with big particles and long fibers (Guilayn et al. 2019b and internal industrial expertise). Globally, digestate LF present fertilizing properties equivalent/close to mineral fertilizers (Sigurnjak et al. 2017) and the SF is closer to organic amendments such as composts, but with a greater amount of nutrients (Tambone et al. 2015). The SF, as a product, will be discussed within the organic soil improvers Sect. 5.1.2.

In the case of certain separators, the separation of N from P can be a tool for a better nutrient management, following crop needs. N tends to concentrate into de liquid fraction, as TAN follows water. P tends to concentrate into the SF, as P is mostly present as or

Table 4 continued		
Final product designations	Config.	TRL
Microalgae and macrophyte hiomass	Phototropic raceway pound with pre- treated LF	Mostly on pilc and demonstratio

Table 5 Summary of se	Summary of soil improvers and organic fertilizers for agricultural valorization: value-added products from digestates and qualitative considerations	tural valorization: va	llue-added	l products from digestates and	qualitative considerations
Final product	Config.	Application	TRL	Major product concerns, not including legislation <sup>a</sup>	References <sup>b</sup>
Solid fraction fibers, cake <sup>c</sup>			Comm.	<ul> <li>- C/N can be low</li> <li>- Inerts (stones, plastics, glass and metal parts)</li> <li>- Relative low nutrient concentrations - short transportation distances</li> <li>- Not as stable as compost</li> <li>- Market value can be &lt; 0</li> </ul>	Teglia et al. (2010), Al Seadi et al. (2012)
(Enriched-) dried digestate	Dying of raw digestate or SF	<ul> <li>Organic fertilizer</li> <li>Litter bed</li> <li>Culture support</li> </ul>	Сотт.	<ul> <li>DM &gt; 75–85% for to allowing stable long term storage</li> <li>Bulk density &gt; 300 kg/ m<sup>3</sup> required for long- distance transportation (might require pelletizing to be achieved)</li> <li>Acidification needed to avoid N-loss</li> </ul>	Delfosse et al. (2011), Dahlin et al. (2015)
Compost	Composting digestate or SF with addition of bulking material	<ul> <li>Soil improver</li> <li>Culture support</li> </ul>	Comm.	<ul> <li>Market value can be close to 0</li> <li>Heavy metal contents</li> <li>Inerts (stones, plastics, glass and metal parts)</li> </ul>	Teglia et al. (2010), Dahlin et al. (2015)
Vermi-compost, vermicast	c.f. compost	<ul> <li>Organic fertilizer</li> <li>Soil improver</li> <li>Litter bed</li> <li>Culture support</li> </ul>	Comm.	<ul><li>Heavy metals contents</li><li>Pathogens</li></ul>	Quintern and Morley (2017)
Biochar	Pyrolysis of dried digestate pellets	<ul> <li>Organic fertilizer</li> <li>Soil improver</li> </ul>	Comm.	<ul> <li>- Nutrient accessibility can be low, decreasing with increasing temperatures</li> <li>- Currently too expensive for simple land spreading in agriculture</li> </ul>	Christel et al. (2014), Al-Wabel et al. (2018)

Table 2 Colletinger					
Final product	Config.	Application	TRL	Major product concerns, not including legislation <sup>a</sup>	References <sup>b</sup>
Humic-like substances	Alkaline extraction from raw digestate, or - Soil improver composts, possibly followed by - Biostimulant acidification and drying to isolate humic acids	- Soil improver - Biostimulant	Lab.	Lab. Research needed	Montoneri (2017)
Comm. commercial, Conf TRL technology readiness	<i>Comm.</i> commercial, <i>Config.</i> most common or most feasible configuration, <i>Lab.</i> laboratory conditions, <i>LF</i> liquid fraction, <i>NA</i> not available, <i>UC-AD</i> urban or centralized plants, <i>TRL</i> technology readiness level of product application	tion, <i>Lab</i> . laboratory c	conditions	, <i>LF</i> liquid fraction, <i>NA</i> not av	ailable, UC-AD urban or centralized plants,
<sup>a</sup> See Appendix A in ESM for process limitations	[ for process limitations related to UC-AD digestates	ligestates			
<sup>b</sup> Prioritizing specific liters	<sup>b</sup> Prioritizing specific literature reviews on processes or products when available	ı available			
<sup>c</sup> In literature, "fibers" are usually referred to as	b usually referred to as the solid fractions fro	om agricultural digest	ates, whil	e "cake" is mainly used to de	the solid fractions from agricultural digestates, while "cake" is mainly used to designate solid fractions from WWTP sludge

ligestate

adsorbed into particles, thus following DM distribution (Guilayn et al. 2019b, Hjorth et al. 2010). However, this N/P fractionation effect cannot be taken as a rule. It is highly depending on the matrix composition and separation performance (Guilayn et al. 2019b). Adding to that, the resulting LF present enhanced physical characteristics: from a slurry viscous digestate, one can produce a pumpable and injectable liquid fraction (Fuchs and Drosg 2013). More recently, a growing number of articles propose short-circuit urban farming to recycle urban waste digestates. For example, the LF can be diluted and used and applied to hydroponic systems for providing nutrients and even boosting plant yields (Krishnasamy et al. 2012; Stoknes et al. 2016; Antón et al. 2017; Fuldauer et al. 2018; Takemura et al. 2019).

Finally, phases separation is a common process before advanced post-treatments. These processes are oriented and more developed to the LF for two main reasons. Firstly, LF represents up to 90% of the absolute input mass (Guilayn et al. 2019b) and, as previously stated, it is where most of inorganic nutrients are distributed.

5.1.1.2 Membrane filtration From the separated LF, membrane filtration are fully commercial to further fractionate and concentrate the nutrient content of digestates until reuse (c.f. section 5.3) or disposal water is produced. They include microfiltration  $(0.1-2 \ \mu m),$ ultrafiltration  $(0.01-0.1 \ \mu m),$ nanofiltration (0.001-0.01 µm) and reverse osmosis  $(< 0.001 \ \mu m)$ . However, they require low total suspended solids (TSS) concentrations to avoid fouling and damage (< 1% DM) (Frischmann 2012). Capital expenditures are high (up to 1.5 M€ for a 40 kt/y facility) as well as operational costs due to energy requirement, cleaning maintenances and membrane replacement (4 to 7 €/t). The investment on membrane filtration are normally limited to large facilities (> 10 to 15,000 t/y) with great local nutrient surpluses (Levasseur et al. 2017). Few full-scale/industrial pilot units treating digestates are reported in literature (Chiumenti et al. 2013b; Bolzonella et al. 2018; Adam et al. 2018).

5.1.1.3 Adsorption and ion exchange products A lot of attention has been dedicated to the selective separation of soluble nutrients through adsorption, mostly ammoniacal nitrogen and orthophosphates

Table 6 Su	Table 6 Summary of energy valorization: value-added products from UC-AD digestates and qualitative considerations	-added products from UC-AD dig	sestates and qualit	ative considerations	
Final product	Config.	Application	TRL	Major product concerns, not including legislation <sup>a</sup>	References <sup>b</sup>
Dried pellets	Drying and pelletization of raw digestate or SF (mainly)	<ul> <li>Bio-fuel</li> <li>Thermal conversion</li> </ul>	Industrial pilot	High ashes content $\rightarrow$ low calorific value	Kratzeisen et al. (2010), Pedrazzi et al. (2015)
Bio-oil or bio-crude	<ul> <li>Pyrolysis of dried digestate pellets</li> <li>Hydrothermal liquefaction of digestate</li> </ul>	<ul><li>Bio-fuel</li><li>Investigated to replace crude oil in cracking refineries</li></ul>	Industrial pilot	<ul><li> Removal of tars</li><li> Poor quality for refining compared to crude oil</li></ul>	Balat et al. (2009), Yuste (2016)
Syngas	Pyrolysis or gasification of dried pellets	<ul> <li>Bio-fuel</li> <li>Conversion to biomethanol, biodiesel</li> <li>Fermentation to bioethanol or several other metabolites</li> </ul>	Gasification: commercial	<ul> <li>Necessary gas conditioning, especially the removal of tars</li> <li>Fermentation: low gas solubility</li> </ul>	Balat et al. (2009)
Bio- methane Bio- ethanol	Post-treatment for digestate post- digestion or recirculation - Post-treatment for digestate fermentation	Bio-fuel Bio-fuel	Commercial Lab.	Minimum 35% methane in biogas needed for combustion in Stirling engines Research needed	Monlau et al. (2015), Sun et al. (2015) Monlau et al. (2015)
Bio- hydrogen	<ul> <li>Syngas fermentation</li> <li>Post-treatment for digestate fermentation</li> <li>Syngas murification</li> </ul>	Bio-fuel	Lab.	Research needed	Uggetti et al. (2014)
Bio-diesel	<ul> <li>Microalgae harvesting and extraction</li> <li>Syngas conversion</li> </ul>	Bio-fuel	Industrial pilot	Research needed	Uggetti et al. (2014)
<i>Config.</i> mos process and	<i>Config.</i> most common or most feasible configurat process and product application	tion, Lab. laboratory conditions, SI	F solid fraction, U	<i>Config.</i> most common or most feasible configuration, <i>Lab.</i> laboratory conditions, <i>SF</i> solid fraction, <i>UC-AD</i> urban or centralized AD plants, <i>TRL</i> technological readiness level of process and product application	nnological readiness level of

<sup>b</sup>Prioritizing specific literature reviews on processes or products when available

<sup>a</sup>See Appendix A in ESM for process limitations related to UC-AD digestates

Table 7 Sum	Summary of industrial valorization: value-added products from UC-AD digestates and qualitative considerations	ducts from UC-AD digestate	es and qualitativ	e considerations	
Final product	Config.	Application	TRL	Major product concerns, not including legislation <sup>a</sup>	References <sup>b</sup>
Reuse water	Membrane filtration or distillation after evaporation processes	Several (irrigation, livestock watering, industry)	Not clear	Salinity and suspended solids	Ayers et al. (1985), Chiumenti et al. (2013b), Adam et al. (2018)
Biopesticides	Raw digestate or SF as growing media after inoculation or intrinsic properties of digestates or digested compost	Plant protection	Lab.	Research needed	Raymond and Federici (2017), Cerda et al. (2019)
Duckweed or microalgae biomass	Phototropic raceway pound with pre-treated LF	Livestock feed	Not clear	Protein content	Uggetti et al. (2014), Vaneeckhaute et al. (2017)
Mushrooms	Composting digestate or SF with the addition of bulking material	Livestock feed	Not clear	R&D needed	Stoknes et al. (2013, 2016)
Earthworms	Vermi-composting	Livestock feed	Commercial	R&D needed	Edwards (1985), Quintern and Morley (2017)
Biochar (raw or activated)	Dried pellets pyrolysis or gasification	<ul> <li>Livestock feed additive</li> <li>TAN adsorbant for litter beds</li> <li>Replacing activated carbon for several purposes</li> <li>Enhancement of biological processes</li> </ul>	Commercial (mostly wood biochar)	Biochar commercial production is still mainly derived from wood biomass	Schmidt (2012), Fagbohungbe et al. (2016), Wu et al. (2017), Hagemann et al. (2018)
Ashes	Incineration of dried pellets	(AD and composting) Portland cement manufacturing (input in kilns) or comment additive	Not clear	R&D needed	Donatello and Cheeseman (2013)
Flame retardants	Struvite or	Wood composites and textile	NA	R&D needed	Yetilmezsoy et al. (2018), Guo et al. (2019)
Flame retardants	Ammonium sulfate	Wood and mulch protection	Theoretical possibility	R&D needed	George and Susott (1971), Hickman and Perry (1996)
Explosives	Ammonium nitrate from N-stripping processes	Mining and civil construction	Theoretical possibility	R&D needed	Meyer et al. (2007)
Humic-like substances	Raw digestate	<ul> <li>Biosurfactants</li> <li>Bioplastic composite</li> </ul>	Lab.	R&D needed	Montoneri (2017)
<i>Config.</i> most c <sup>a</sup> See Appendiy <sup>b</sup> Prioritizing s <sub>1</sub>	<i>Config.</i> most common or most feasible origin configuration, <i>Lab.</i> laboratory conditions, <i>LF</i> liquid fraction, <i>TRL</i> readiness level of process and product application <sup>a</sup> See Appendix A in ESM for process limitations related to UC-AD digestates <sup>b</sup> Prioritizing specific literature reviews on processes or products when available	<i>ab</i> . laboratory conditions, <i>LI</i> C-AD digestates ts when available	F liquid fraction	, TRL readiness level of process	and product application

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adsorption. The concept consists on driving the effluent through adsorption columns, then recover the adsorbed nutrients. The most common adsorbents are natural zeolites, biochar, clays and resins (He et al. 2016). As some relatively low-cost adsorbents as zeolites and biochar are investigated, some studies propose the enriched adsorbent as a final fertilizing product after simple adsorption through batch reactions (Kizito et al. 2015). In a deep review of nutrient recovery options from digestates, Vaneeckhaute et al. (2017) pointed out that the recovery of ammoniacal nitrogen as N-rich zeolites is potentially the lowest-cost technology for N-recovery. This is not developed at full-scale and it is limited by the availability of the adsorbent at a low price and current limited performances. One of the main concerns around this topic is the plant availability (desorption) of the sorbed nutrients, which was not found within digestate studies. With <sup>15</sup>N labeled ammonia, Taghizadeh-Toosi et al. (2012) proved that ammonia adsorbed to wood biochar was accessible to plants.

Another low-cost adsorption strategy that has been investigated is an adsorption contact reaction followed

by phase separation (Olga et al. 2014). However, it seems more appropriate to classify this strategy as a phase separation enhancement.

### 5.1.1.4 Evapo-concentration

processes Concentrating LF, SF and membrane retentates can be a useful technique to reach fertilizer product categories (Guilayn et al. 2019a), while possibly complying to recognized ABP hygienization when thermal treatments are applied with temperatures above 70 °C and over > 1 h. LF and membrane retentates can be concentrated with evaporation to achieve up to 50–90% volume and mass reduction (Chiumenti et al. 2013a; Guercini et al. 2014). Vacuum-evaporation needs an input DM below 1 to 3% (Frischmann 2012; Levasseur et al. 2017). However, evaporators need the removal of large particles otherwise the fouling of heat exchangers will be a current operational problem (Vondra et al. 2018, internal industrial expertise).

In evaporation, the effect of ammoniacal nitrogen volatilization can be an option to recover N (next section). Countering this effect, another option is to acidify the input to pH 5 to 6, which may allow to

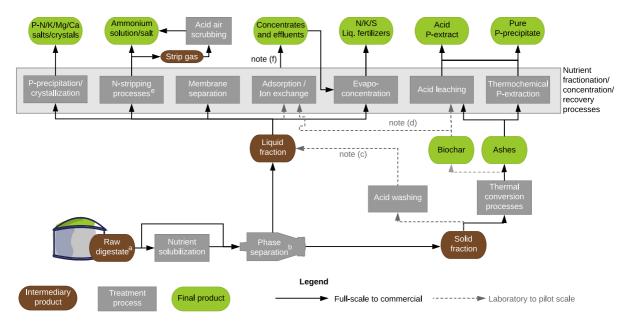


Fig. 1 Processes to produce N/P/K fertilizers from digestate. Major process inputs and outputs may be omitted. **a** Usually comprises a storage step. **b** Some plants or research might directly treat whole digestates, which depends mostly on suspended solids and rheology to match downstream equipment requirements. **c** The washing solution can be treated by most

processes applicable to digestate LF (or mixed to it).  $\mathbf{d}$  As the adsorbent.  $\mathbf{e}$  N-stripping processes are here considered as those specifically designed for N-stripping (e.g. stripping towers), but also indirect processes such as thermal drying, evaporation and even composting.  $\mathbf{f}$  concentrates and effluents from most of the previous processes

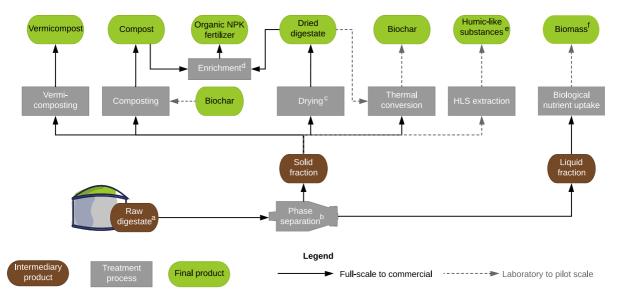


Fig. 2 Processes to produce soil improvers and organo-mineral fertilizers from digestates. Major process inputs and outputs may be omitted. **a** Usually comprises a storage step. **b** Some plants or research might directly treat whole digestates, which depends mostly on suspended solids and rheology to match downstream equipment requirements. **c** Thermal drying might include a pelletizing/granulation step downstream.

maintain more than 95% of the total N in the concentrated or dried product (Chiumenti et al. 2013a; Pantelopoulos et al. 2016).

For operators of UC-AD, simple digestate concentration processes such as phase separation and evaporation can be relatively low cost and robust solutions. However, they present a much smaller opportunity to create value and overcome the difficulties of reaching the fertilizer market from raw digestate. Advanced solutions such as membrane filtration can be used to further fractionate nutrients but will need economies in scale.

5.1.1.5 Nitrogen/sulfur solutions from ammoniacal nitrogen removal process Digestates present up to 80% of the nitrogen in digestates as ammoniacal nitrogen (Guilayn et al. 2019a) and an alkaline pH, which makes them particularly attractive for promoting N-stripping. Ammoniacal nitrogen is present in water as the equilibrium of two species: NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> (free ammonia and ammonium). N-stripping is achieved by favoring free ammonia in the equilibrium through pH and/or temperature increase while enhancing liquid/gas transfer. The temperature and pH increase tend both to promote hydrolysis of proteins, increasing ammoniacal

**d** "Enrichment" refers to adding mineral nutrients (conventional or recovered from digestates) usually for matching specific formulations. **e** Humic-like substances are also applied as biostimulants, which must be differentiated from nutrient supply and soil amendment. **f** Most of publications are related to microalgae. As an agricultural product microalgae biomass is usually cited as a slow-release fertilizer

nitrogen content, which might extend N-recovery efficiency beyond the original TAN content (Serna-Maza et al. 2015). In practice, alkalization is often performed with the addition of NaOH or KOH and/or through  $CO_2$  stripping, which also enhances gas transfer.

Battista and Bolzonella (2019) conceived an innovative low-cost process based on solar energy. A greenhouse equipped with solar-powered fans promote digestate concentration (> 60% DM) and N-stripping process. The preliminary tests on OFMSW digestate resulted in over 95% stripping efficiency (TAN concentration reduction corrected for DM content) and about 40% NH<sub>3</sub> recovery (limited by the acid trap efficiency).

Many commercial solutions already exist. The main operational problem is scaling of stripping columns and corrosion (Hidalgo et al. 2015). Indeed, TSS must be low to avoid stripping column clogging. For this reason, stripping is often performed in the centrifuge liquid fraction or with screw press followed by an additional step as sieving or settling.

After N-stripping, N-recovery is performed through scrubbing in acidic solutions or water to produce ammonium sulfate, ammonium nitrate (Jamaludin et al. 2018) or ammonium water (Gasum 2016). Ammonium nitrate is a common fertilizer but usually submitted to stricter regulation due to its explosive ignition reaction (Lallanilla 2013). In vapor stripping, condensation can be used to form directly ammonia in water. Recent research has demonstrated the interest of using alternative and safer acids such as citric acid to produce ammonium citrate (Jamaludin et al. 2018).

Alternatively, hydrophobic membrane contactors can be placed directly into solution (raw digestate or liquid fraction) either in the operating digester (Lauterböck et al. 2012) or in a dedicated removal tank (Vanotti et al. 2017).

In any case, final ammonium sulfate solution, the most common product, is acidic and can present at up to 6-10% of TN which is diluted compared to commercial fertilizers (Vaneeckhaute et al. 2016; Bolzonella et al. 2018). It can be difficult to commercialize.

5.1.1.6 Precipitation/crystallization P-products The studies on precipitating nutrients from digestates are normally oriented to P-precipitation. This is due to unique features of P among the major macro-nutrients: (1) there is no volatile gaseous form of P in the global P cycle; (2) global production of P-fertilizers are concentrated into a very few counties and (3) it is estimated that we can reach a peak in P production still in this century. Additionally, as the quality of remaining phosphate rocks is rapidly degrading, the concentration of impurities such as heavy metals is increasing (Desmidt et al. 2015).

The most cited precipitation product that can be produced from digestates is magnesium ammonium phosphate hexahydrate (MgNH<sub>4</sub>PO<sub>4</sub>·6H<sub>2</sub>O), known simply as struvite. Several commercial processes worldwide allow an efficient recovery of high-quality struvite crystals from phosphate rich effluents, including: PHOSPAQ<sup>TM</sup> (Paques), AirPrex<sup>®</sup> (CentriSys), PEARL<sup>®</sup> (Ostara), NuReSys<sup>®</sup> (NuReSys), Phosphogreen<sup>TM</sup> (SUEZ), among other. Other include K-struvite (KMgPO<sub>4</sub>·6H<sub>2</sub>O), calcium phosphate and hydroxyapatite (Monballiu et al. 2019). Ca-P products seems to be considered less valuable due to an overall lower P-accessibility to plants (Cabeza et al. 2011).

High consumption of chemicals is one of the main disadvantages of (K-)struvite crystallization. Indeed, pH must be increased from about 7–8 to 9–10. As in N-stripping; alkali requirements can be reduced

through  $CO_2$  stripping (Fattah et al. 2010). Since the formation of struvite is a proton releasing reaction, constant addition of an alkali is necessary to keep a constant high pH. Additionally, from both economic and environmental perspectives, a great limitation of (K-)struvite recovery is the necessary source of Mg, which is also listed as critical raw material by the EU (European Comission 2011).

Any choice of a particular process and optimization criteria for struvite recovery should be assessed caseby-case. They have been extensively studied and reviewed (Kataki et al. 2016; Tao et al. 2016; Kataki and Baruah 2018). In brief, pH, temperature and the molar ratios of  $Mg^{2+}$ ,  $NH_4^+$ ,  $PO_4^{3-}$  and competitive ions such as  $Ca^{2+}$  must be carefully controlled to promote struvite supersaturation levels. Adding to that, to produce a pure product with proper crystal growth, TSS must be low (< 1 g/L) and the possible disturbing effect of dissolved OM on ionic dissociation must be considered (Capdevielle et al. 2015; Aleta et al. 2018). According to several technology providers, struvite feasibility need orthophosphate minimal concentration threshold of 50 to 70 mg  $P-PO_4^{3-}/L$  (P-REX 2017; SUEZ 2017b; PAQUES 2019), which is largely overpassed by most of UC-AD digestate liquid fractions (Akhiar et al. 2017). It must be noticed that either in R&D a full-scale operation, many large/pilotscale producers of "struvite" do not actually have a precise control of struvite supersaturation and quality control (internal expertise). This fact can jeopardize struvite image, for example, if the product contains large amounts of Ca-P and it is tested for agricultural use while referred as "struvite".

Full-scale P-recovery as struvite is still mostly limited to WWTP, including the previously cited commercial processes. Struvite from sewage sludge digestate has been demonstrated to have very low concentrations of heavy metals and organic micropollutants (Uysal et al. 2010). The WWTP sector is promising because its economic feasibility does not rely on struvite sales. In WWTP, struvite crystallization reduces spontaneous incrustation problems and P-removal operational costs. In traditional WWTP configurations, about 20% of the P input load can be due to the return of digestate's centrate to head of the plant (Evans 2007).

Full-scale recovery of soluble phosphates through P-crystallization can achieve over 90% efficiency (Desmidt et al. 2015). Thus, nowadays, the limiting step for achieving great total recovery efficiencies relies in a solubilization step prior to crystallization. For this reason, several pre-treatments are proposed and the most addressed seems to be chemical/biological acidification (Braak et al. 2015; Szogi and Vanotti 2015; Vaneeckhaute et al. 2016; Piveteau et al. 2017; Guilayn et al. 2017). UC-AD digestates from codigestion plants accepting high charges of effluents with high alkalinity and calcium content tend to produce low-quality struvite. Two examples of such co-substrates are pig slurry (Piveteau et al. 2017) and dairy manure (Tao et al. 2016). In such cases, the necessary steps of P dissolution and the following removal/chelation of competitive ions can thus hinder struvite recovery feasibility. The same applies for sewage sludge from WWTP with coagulation steps based on aluminum. They might contain Al-P salts that are highly difficult to solubilize (Braak et al. 2015).

Oliveira et al. (2018) are possibly the first to evaluate struvite recovery from OFMSW digestate. The procedure included dilution with distilled water or 1.1 M nitric acid (2.5 S/L ratio), followed by electrodialysis for recovering the negatively charged phosphates in the anode side and, finally, struvite precipitation. Under the best conditions, they have achieved 43% total P extraction and up to 100% precipitation.

Struvite is a slow-release mineral fertilizer. Research seems inconsistent about whether struvite needs an acidic soil or not to be equivalent or even better than conventional mineral fertilizers (Ackerman et al. 2013; Talboys et al. 2016). This controversy might reflect a product quality heterogeneity among studies.

Struvite selling prices are related from 40 up to > 1400  $\notin$ /t (Lebuf et al. 2012; Desmidt et al. 2015). In most of the cases, struvite price is considered below its nutrient market value calculated to be around 690  $\notin$ /t in 2012 (Desmidt et al. 2015). The study of Yetilmezsoy et al. (2017), based on laboratory conditions, estimated a minimum sale price of 560  $\notin$ /ton for a 6-year payback.

Vivianite (Fe(II)<sub>3</sub>(PO4)2•8H<sub>2</sub>O) recovery is a new promising solution whose R&D is being led by researchers of the Delft University of Technology. As iron coagulants are widely applied in WWTP, the reducing environment of AD favors the formation of Fe(II) and, more particularly, it can be bound with phosphates as vivianite. Prot et al. (2019) have demonstrated the feasibility of a magnetic separation system for recovering vivianite from sewage sludge digestate.

5.1.1.7 Ashes and biochar as source of mineral nutrients Thermal conversion processes such as incineration (combustion) or pyrolysis can be effective options for digestate valorization. Some definitions and operational aspects regarding thermal conversion of digestates will be discussed later in the section "Energy valorization".

Ashes are the product resulting from incineration and char (referred as biochar when produced from renewable biomass) are the solid final products from carbonization processes such as pyrolysis. Biochars though can present itself a high content of measured ashes (up to 40–60%). The ash content is increased with increasing process temperatures (Neumann et al. 2016; Opatokun et al. 2017).

Thermal conversion processes concentrate the elements that are non-volatile at the applied temperature, that may reach more than 1000 °C for combustion. For this reason, P and K in digestate ashes can be as high as 25% and 15%, respectively (Kratzeisen et al. 2010). However, several studies indicate that the P in ashes present poor accessibility to plants. Cabeza et al. (2011) studied heavy metal depleted sewage sludge ashes and concluded that they are not effective P-fertilizers due to low P accessibility.

Biochar tends to present a better nutrient bioaccessibility than ashes. Christel et al. (2014) observed that after pyrolysis at > 700 °C the biochar P content was not extractable with water. However, at lower temperatures biochar had more water-extractable P than ashes produced at the same combustion temperatures.

Commercial processes are already in place to recover high-grade phosphates from ashes through chemical or thermochemical extraction but are still not economically interesting (Vaneeckhaute et al. 2017).

#### 5.1.2 Soil improvers and organo-mineral fertilizers

5.1.2.1 Biomass harvested from digestates as slowrelease organo-mineral fertilizers The nutrients from digestates can be recovered through several biomass harvesting processes. As the nutrients are trapped within the biomass, these products are commonly referred as slow-release fertilizers (Mulbry et al. 2005). The most common biomasses that can be produced from digestate nutrients are duckweed and microalgae. They are the only category of products under the "Soil improvers and organomineral fertilizers" section that can be originated from the liquid fraction of digestates. They greatly differ as the organic matter content is produced from photosynthesis rather than originated from the digestate.

*Microalgae* have been used in engineered ponds to produce biomass either in photoautotrophic, heterotrophic or mixotrophic systems (Xia and Murphy 2016). Given the species and environmental conditions, they can be used to selectively produce proteins (up to 60% in biomass), lipids (up to 60%), carbohydrates (up to 30%) and a large range of possible valuable and unique compounds (Uggetti et al. 2014). For UC-AD digestates, microalgae biomass concentration can reach up to 3 g/L and productivity up to 0.6 g/L .d (Xia and Murphy 2016).

Microalgae have been successfully harvested from digestate liquid fractions, but digestates present a series of negative effects that may jeopardize microalgae growth. The most recurrent are high turbidity and ammonia toxicity (Xia and Murphy 2016). Because of it, most of the studies use diluted digestates at ratios ranging from around 1:5 to 1:50 (Erkelens et al. 2014; Marcilhac et al. 2014, 2015; Koutra et al. 2017). No consensus on TAN inhibition concentrations could be found since it is highly depending on microalgae species and ionic strength. Other than dilution, recurrent considered treatments are N-stripping (Marazzi et al. 2017), adsorption (Marazzi et al. 2017) and coagulation (Chen et al. 2012). As summarized by Uggetti et al. (2014), other potential problems of digestates are the presence of heavy metals and OMP (potential biological toxicity), competitive microorganisms and pathogens, VFA (stimulation of competing heterotrophic bacteria) and long-chain fatty acids (potential toxicity).

Full-scale economic feasibility of microalgae still depends on strong price reduction in each step of the production chain, which also include the source of nutrients and water. Both can be replaced by digestates. The application of microalgae biomass as slowrelease fertilizer can be efficient (Mulbry et al. 2005; Coppens et al. 2016), but the economic feasibility of microalgae harvesting will depend on the establishment of true biorefinery systems allowing to profit from the whole biochemistry of the biomass. Some insights on microalgae biorefinery are provided in "Energy valorization" and "Other industrial valorization possibilities".

*Duckweed* are small macrophytes from different species classified as the *Araceae* subfamily *Lemnoideae*. They have indeed been used for decades for wastewater treatment (Alaerts et al. 1996). In Europe, the LEMNA project claimed to build the first full-scale nutrient recovery plant based on duckweed (Pascual 2016). The plant will operate on pig slurry digestates and the output biomass is intended to be used as fertilizer and for feed (high protein content).

There is little research on duckweed applications for UC-AD digestates. This is probably because duckweed have extremely high land requirement. For example, given a maximum potential to remove 600 kg N/ha (Leng 1999), a small/medium-sized urban AD facility producing only 1 kt/y of digestate with a 10 gN/kg concentration would require at least 15 ha of duckweed ponds.

5.1.2.2 Solid fraction from phase separation (dewatering) The digestate dewatered fraction, or solid fraction (SF) of digestates is obtained from the same phase separation processes described previously for the LF (Sect. 5.1.1.1). As for the LF, the separation process enhances the biochemical and physical characteristics of the SF compared to the whole digestate. Indeed, after separation, an initially liquid to viscous digestates can be turned into a well-stackable product, presenting better handling properties, lower transportation costs and with a better bulking capacity for returning to soil (Fuchs and Drosg 2013). While the LF present properties closer to mineral fertilizers (Sigurnjak et al. 2017), the SF is more similar to organic amendments such as composts, but with a greater amount of nutrients (Tambone et al. 2015).

The literature results seem controversial about SF effectiveness as an amendment product compared to compost. For example, Tambone et al. (2015) concluded that digestate SF (from pig slurry, energy crops and agro-industrial residues) were already stable and further composting did not enhance remarkably its characteristics. Teglia et al. (2011) evaluated SF from WWTP sludge and food processing waste digestates. The authors concluded that the products were not stable and recommended composting as a post-treatment.

Again, phase separation is a key-enabling technology prior to OM valorization. In the case of SF, composting, thermal drying and thermal conversion are usually applied or investigated as following processes. They will be discussed and described in the following sections.

5.1.2.3 Dried digestates Dried digestates are produced from thermal drying process which usually require a DM concentration input higherthan 15%. To achieve this level, UC-AD digestates from wet AD processes might need a previous phase separation step. Some plants apply a recirculation of the dried product, which implies a loss of equipment capacity (Frischmann 2012).

As in evaporation process, thermal drying promotes ammoniacal nitrogen volatilization, which can be seen as an opportunity to recover N. In the opposite, in order to maintain up to 95% of the total N in the dried product, digestates can be previously acidified (Pantelopoulos et al. 2016).

Dried UC-AD digestates can be nutrient-rich, but it seems that there is no consensus for their applicability as organic soil amendments (stable OM). For example, Tambone et al. (2010) demonstrated that a dried WWTP sludge digestate presented much inferior amendment properties than composts. However, in the same study, the authors indicated that other different raw digestates (including OFMSW, animal slurry and FAI among inputs) were similar to composts as amendments, which should be expected to be maintained after thermal drying. Indeed, OM stability seems to be the more controversial digestate quality parameter around digestates (Alburquerque et al. 2012). It reflects the lack of consensus on the indicator to be used thus a lack of homogenized data for performing conclusive meta-analysis.

Many types of thermal dryers are available in the market. The most common for digestate treatment are rotary, disk and belt dryers. The latest have been the more indicated to digestates presenting large particles, which can be the case of UC-AD digestates (Arlabosse et al. 2010). All of them require high thermal energy, relevant capital investments (600 k€ for a 10 kt/y facility) and intensive air treatment for safe air disposal and to avoid atmosphere ignition. Indeed, an explosive atmosphere due to organic dust can be a constant threat for operators, especially in the case of mixing-drying equipment such as rotary driers

(internal expertise). In some cases, the fire risk can be aggravated by intensive self-heating of dried digestate (internal expertise).

The quantity of digestate to be dried and the final moisture depend on the available heat from cogeneration. Dried products should achieve DM > 75-85% for allowing long term stability (VALDI-PRO 2015; Dahlin et al. 2015). Energy consumption for thermal drying ranges from 1.2 to 1.3 kWh/kg of removed water. In biogas plants, the heat from cogenerators is normally not enough to dry the whole digestate flow. It usually allows drying less than half of the whole digestate flow (Bolzonella et al. 2018). In addition, raw dried digestate may present a low bulk weight (around 100 kg/m<sup>3</sup>) making long-distance transportation costly (Dahlin et al. 2015). Longdistance transportation is indicated to require more than 300 kg/m<sup>3</sup>. Pelletization/granulation can be used to increase dried digestate bulk weight up to 600 kg/ m<sup>3</sup> (Dahlin et al. 2015). Pelletization/granulation also enhances handling ease of final product, making it more easily applicable with equipment designed for conventional mineral fertilizers. Nagy et al. (2018) performed an economic evaluation on the production of dried pellets and concluded that the production costs would difficultly justify a simple use in agriculture. In such cases, the economics will drive digestate dried pellets to be seen as an energy product more than a fertilizer.

5.1.2.4 *Composts* Composting is an aerobic process that decomposes the biodegradable fraction of the OM and microbial biomass. Digestates into  $CO_2$ composting is а developed technology in agricultural, urban and industrial AD plants. It is commonly performed either in situ or ex situ. Due to the more favorable thermodynamics, the OM decomposition in the composting aerobic conditions can achieve greater extents than AD. The composted material is then cured (maturation phase) and grinded. The main operational parameters of industrial composting are the duration (from 3 weeks up to 1 or 2 months), C/N ratio (20 to 40), temperature, moisture (ideally around 50%) and aeration method and intensity (Epstein 2011). Composting is an intensive self-heating process. Temperatures can reach more than 70 °C within the piles. For this under certain criteria, composting is reason. authorized in the EU as a hygienization process for digestates containing Category 2 and 3 ABP (Amlinger and Blytt 2013).

For wet digestates, composting is conventionally performed on the SF after dewatering. Additionally, raw digestates or SF need, usually, co-substrates for one or more of these reasons: (i) lack of residual biodegradable OM allowing the bioprocess requirements for an effective temperature increase, (ii) digestates are normally too wet for composting and/ or not sufficiently physically structured when dewatered, and (iii) C/N of digestates is too low for composting (Tremier et al. 2014; Zeng et al. 2014, 2015). Common co-substrates and bulking agents are green waste, wood chips, sawdust and recirculated compost grinding refuse (Epstein 2011, internal industrial expertise). An emerging composting technique useful for liquid residues such as UC-AD digestates from wet-AD consists of spreading and constantly turning the digestate into a saturated support/bulking material (Chiumenti 2015; Levasseur et al. 2017).

Due to heating, great water loss during composting represent a volume and mass reduction thus an advantage in terms of transportation costs. Compare to the compost feed, a massic reduction from about 30 to 50% can be achieved (Levasseur et al. 2017). However, for UC-AD, the need for co-substrates represent a substantial increase of final volume to be managed in comparison to initial digestate volume.

Moreover, eventual physical impurities and trace metals present in UC-AD digestates are not removed during the composting process. These contaminants can either be concentrated or "diluted" depending on water loss and co-substrate quantity and quality. It is consensual, though, that the bioaccessibility and solubility of heavy metals are highly limited with composting due to strong bonding to the compost organic matter matrix (Smith 2009).

UC-AD operators must consider that digestate composting is not always a low-cost process and that compost price is commonly low outside the retail marketing. According to the survey of Dahlin et al. (2015), composts are sold to agriculture from 0 to  $7 \notin /t$ , which is far below other products that can be recovered from digestate. Prices up to 80  $\notin /t$  could be achieved through the application of fine processing for meeting the horticultural market.

Due to longer RT and the co-substrate volume, the composting installations need as much as 4 times the

surface of AD installations (internal industrial expertise). In the case of UC-AD land price is normally higher in urban/industrial areas than agricultural ones. Moreover, in urban areas, the composting facility are usually required to be enclosed. It is also the case in some countries as UK and Ireland for ABP processing regardless the zone where the installations are localized (Amlinger and Blytt 2013). In such cases, ventilation for air collection in a high surface and subsequent air treatment can be extremely costly. Adding to that, several composting regulators require industrial composters to apply forced aeration and/or mechanical turning such as the U.S. Environmental Protection Agency (Epstein 2011), which adds significant operational costs compared to the low-cost convective aeration composting.

5.1.2.5 Vermicomposts/Vermicast Although less developed than simple composting, digestate *vermicomposting* is already performed at industrial scale. Only in New Zealand, 200,000 tons of dewatered sludge from municipalities and industries was valorized through vermicomposting (Quintern and Morley 2017). Compared to composting, the main operational advantages are the promotion of aeration, turning and process acceleration by the earthworms. Additionally, it is reported to achieve greater volume reduction (65 to 85%) (Quintern and Morley 2017).

Digestate vermicomposting was demonstrated to achieve great pathogen reduction and compliance with spreading standards (Rajpal et al. 2014). However, it is often preceded by a short thermophilic composting phase. The composting phase allows high temperature securing hygienization and desactivation of weed seeds. Moreover, pre-composting allows the removal of ammoniacal nitrogen, a highly toxic compound to earthworms (Krishnasamy et al. 2014). As for composting, structuration is also important so digestates are usually vermicomposted with a bulking agent. With pre-composting, Krishnasamy et al. (2014) experimented food waste digestate vermicomposting and get the best results with 7:3 digestate:sawdust in 75 days. Tesfamichael and Stoknes (2017) reported a sample of a commercial vermicompost produced from SF of food waste digestate only (no co-substrate). The product is related to be high-quality: stable, structured and nitrified.

Vermicomposting is an emerging alternative for digestate composting allowing to add more value to digestates. However, little information is available in literature. The high ammoniacal nitrogen content of digestates is fatal to earthworms which can be an important cause of process disturbances. If pre-treated by composting, most of the exposed composting drawbacks need to be considered.

5.1.2.6 Biochar Biochars contain a significant amount of transformed and stabilized organic matter. It is seen much more as a soil improver than a mineral fertilizer. A food waste digestate biochar, for example, can present up to 61% volatile solids, 45% C and 6% N (Opatokun et al. 2016). Digestate biochars present a relatively high specific surface, pore volume and polar functional groups that confer it adsorbent properties (Inyang et al. 2012; Stefaniuk and Oleszczuk 2015; Opatokun et al. 2016; Wongrod et al. 2018; Jiang et al. 2018). Biochars are considered an emerging organic amendment and slow-release fertilizer with a large list of advantages to the soil and plants. These advantage include soil structuration, the reduction of greenhouse gases emissions, adsorption of contaminants and many of the benefits attributed to soil humus (Tan et al. 2017). The scientific literature is richer in biochars from wood and other carbon-rich residues such as rice straw (Tan et al. 2017). Nevertheless, the agronomic interest of biochars originated from different types of digestate, including industrial food waste had already been demonstrated (Opatokun et al. 2017).

Integrate thermal conversion with AD is a promising alternative. It allows enhancing the digestion and the AD plant energy efficiency while increasing the digestate value-chain. The produced digestate biochar can be used to increase soil fertility while promoting carbon sequestration for fighting climate change.

It is indicated though, that current biochar prices are over-expensive for field application. Nowadays, the main large crops present specific costs of over  $1000 \notin$ / ha (Desbois and Legris 2007). The recommended biochar doses from 5 to more than 100 t/ha (Major 2010; Someus 2015) are extremely high considering biochar prices reported from 500 to over 1000  $\notin$ /t (Jirka and Tomlinson 2014). The application frequency is not annual, but the farmers must calculate indirect long-term fertilization benefits thus long term payback periods. From an economic point-of-view, many other applications of biochars can be prioritized (Schmidt 2012), some of them being addressed in this review in next Sect. 5.3. 5.1.2.7 Humic-like substances for soil amendment or biostimulation Humic substances are complex organic compounds abundant in nature originated from the decomposition and reorganization of organic matter. This domain is still of a great interest pushed by the recognized importance of humic substances for understanding the fertility of natural soils, but also due to its remarkable applications on agriculture or horticulture as soil conditioners and biostimulants (Muscolo et al. 2013). Indeed, depletion of soil OM is a worldwide problem resulting from intensive agricultural systems with low reintroduction of stable organic matter. The benefits of adding humiclike substances (HLS) to soil are widely recognized, especially in the case of OM-poor soils (Lyons and Genc 2016).

For commercial purposes, HLS are commonly extracted from natural fossil sources such as leonardite and peat. They can be found, similarly, in anthropogenic organic material such as (vermi-)composts, digestates and landfill leachates (Atiyeh et al. 2002; Eyheraguibel et al. 2008; Morard et al. 2011; Calvo et al. 2014; Fascella et al. 2015, 2018; Silva and Brás 2016; Palumbo et al. 2018). Digestates from UC-AD have been successfully applied for the extraction of a pool of soluble organic compounds referred to as HLS, "soluble biopolymers", "biobased organic substances", "biowaste derived soluble substances", among other variants. To maximize the extraction of HLS from digestates, the digestate must undergo a strong alkaline treatment allowing to solubilize the humic-like acids (Salati et al. 2011; Fascella et al. 2015; Prevot et al. 2015; Montoneri 2017).

In one hand, the intrinsic lack of standards for the quality of HLS is one of the major problems of this sector for a wider agriculture adoption. In the other hand, it can be an opportunity for upcycling complex organic matrices such as digestates from UC-AD. For agriculture, another problem is the lack of consensus on application doses. The current doses recommended by product furnishers can be ineffective (Lyons and Genc 2016).

If doses are uncertain for conventional fossil HS, the scenario is even blurrier considering the small literature around digestate HLS as biostimulants. To the best of our knowledge, only Guilayn et al. (2020) compared HLS from different digestates (sludge and manure) at different doses for biostimulation. Extraction costs of HLS from UC-AD digestates and other substrates were estimated from 100 to 500 USD/t in the frame of the BioChemEnergy project (Montoneri et al. 2011). As a reference, global wholesale internet prices of soluble dried humic acids extracted from leonardite range from 100 up to 1300 USD/t.

#### 5.1.3 Conclusions on agricultural valorization

In UC-AD, effective fertilizers and soil improvers can be produced from digestates. Phase separation is both a valorization process itself and a key technology enabling more advanced treatments. From the LF, membrane filtration, evaporation, N-stripping and P-precipitation are full-scale feasible options allowing producing value-added products. From the SF, the most used processes are composting and thermal drying. Adding to that, thermal conversion for biochar production and HLS extraction seems promising solutions that need to be further investigated for product comprehension, process optimization and cost reduction.

Adding to the technical challenges, some important bottlenecks hamper the development of the agricultural valorization of UC-AD digestates: (i) in the case of non-source-separated AD feedstock, marketing of fertilizing products are usually more limited by regulatory frameworks; (ii) in the conventional agriculture, nutrient value depends on highly volatile global prices of N, P and K; (iii) there is still no clear market value to be associated for the OM content and quality; (iv) the presence of inert impurities separation (e.g. glass and plastics) in the case of poor source/postseparation can be a particular problem of UC-AD digestates.

#### 5.2 Energy valorization

The following sections address the valorization of UC-AD digestates as energy products. Figure 3 present the most recurrent pathways in scientific literature.

## 5.2.1 Biofuels from thermal conversion processes

The European Biogas Association positioned itself against the promotion of digestate combustion mostly due to the interruption of the carbon cycle and the loss of nitrogen during drying (European Biogas Association 2013). However, as previously discussed, the nitrogen can be recovered after acid scrubbing and the waste treatment service itself (removing pollution) must not be neglected. The innocuity of digestates depend above all on the innocuity of the AD feedstock. Some organic waste innocuity will be a concern until the civilians and the production sectors do not provide safe residual streams and/or an effective waste sourceseparation. As this can take several decades, combustion and thermal conversion processes in such cases can be effective to destroy pollutants while recovering energy and biofuels.

Thermal conversion processes are usually classified as combustion, pyrolysis and gasification. Other variants include torrefaction, hydrothermal carbonization, vapothermal carbonization and hydrothermal liquefaction. All these processes share the principle of promoting an irreversible thermal decomposition of the organic matter, followed by different reactions of reorganization. They differ in terms of temperature range, oxygen level, pressure and water/vapor presence to drive the quality and yield of the different valuable products: heat and power, syngas, bio-oil and biochar. Beyond energy valorization, syngas and biooil (or biocrude) are investigated as inputs for complex biorefineries, based either on fermentation or thermochemical fractioning (Balat et al. 2009).

It must be highlighted that digestate biochar contain great amount of ashes (up to 40-60%), which is increased by higher process temperatures (Neumann et al. 2016; Opatokun et al. 2017). Indeed, digestate biochar from digestates is difficultly an energy product replacing charcoal. To illustrate it, solid fossil fuels present a gross calorific value (GCV) of 22-37 MJ/kg, while dry hardwood has a GCV of 18-19 MJ/kg (Osborn 1985). Peng et al. (2020) summarized biochar from different digestates and thermal conversion processes. They present GCV usually below 10 MJ/ kg and only two values exceeding 25 MJ/kg (both from agricultural inputs). Digestate biochar applications in agriculture and industry as soil improver, adsorbent and bioprocess enhancer should be more suitable than energy valorizaiton.

Conventional combustion, pyrolysis and gasification processes are performed in dried digestate, usually pelletized (Opatokun et al. 2014; Wiśniewski et al. 2015; Gusiatin et al. 2016; Morero et al. 2017). The more recent hydrothermal conversion processes are similar to pyrolysis as they occur in the absence of oxygen. For wet biomasses as digestates, they present the important advantage of not requiring the intensive energy consuming drying pre-treatment (Mumme et al. 2011; Funke et al. 2013; Reza et al. 2016). Among all these processes, only gasification was found to be fully commercial on UC-AD digestates as several gasification installations are operating in the US for treating sewage sludge digestates (U.S. EPA 2012).

From a climate change and carbon cycle point-ofview, thermal conversion can be compared to AD but in a greater extent: a greater fraction of the carbon is converted to fuels that will eventually be combusted, but the remaining carbon is much more stable than previously. In soil, biochar is indicated to be stable for thousands of years, greatly contributing to the soil carbon sequestration strategy for fighting climate change (Vaccari et al. 2011).

#### 5.2.2 Biofuels from fermentation processes

Lignocellulosic biomass is composed of carbohydrate polymers (cellulose and hemicellulose) structured in lignin (a phenolic polymer). AD alone is not able to break lignocellulose structure in order to access and monomerize its sugars. For this reason, the literature is abundant in pre-treatments and post-treatment to enhance methane yields. Adding to that, a lot of attention has been dedicated for valorizing the digestate residual OM as biofuels through fermentation.

In scientific literature, after several types of digestate post-treatment, two distinct approaches are commonly found to further produce fermentation biofuels from digestate: recirculation/post-digestion for biomethane production or alcoholic fermentation. These two options have been demonstrated to be interesting for agricultural digestates. These treatments are often alkaline, enzymatic or thermochemical (Monlau et al. 2015). Recently, Brémond et al. (2020) explored fungal treatment and had promising results. However, UC-AD digestates tend to present much lesser fibrous contents. As observed in Table 3, most of them are below 30% (hemicellulose plus cellulose, % of OM) while agricultural digestates can reach more than 60%. Unless the UC-AD receives important amounts of recalcitrant fibrous material, this valorization pathway tends to be ineffective. To integrate non-agricultural AD with the production of fermentation metabolites non-fibrous from residues, the best-known

configuration is to perform a two-step process where intermediary fermentation products such as hydrogen and VFAs are recovered prior to the biogas-producing methanogenic reactor (Capson-Tojo et al. 2016).

Another approach linking digestate to fermentation biofuels is the use of digestate LF as a source of moisture and nutrients for fermentation, which will consist more of a nutrient recovery strategy rather than digestate OM valorization (Zhang et al. 2010; Bashiri et al. 2016).

#### 5.2.3 Biofuels from harvested biomass

Biodiesel is produced from the transesterification of vegetable or animal lipids, oil and fats with an alcohol under the presence of a catalyzer. From UC-AD digestates, the best approach for producing biodiesel seems to be the production of lipid-accumulating biomass such as microalgae (Uggetti et al. 2014). Operational aspects and bottlenecks on microalgae harvesting have been previously discussed (Sect. 5.1.2.1). According to Shalaby (2014), algae can produce 30 time more lipids and oils than oilseed crops in terms of footprint. Uggetti et al. (2014) reported a lipid content between up to 60% of the biomass. Other than lipids to biodiesel, carbohydraterich microalgae have been studied for producing bioethanol through fermentation.Additionally, microalgae biomass can be used as inputs for the thermal conversion processes discussed above (Uggetti et al. 2014).

#### 5.2.4 Conclusion of energy valorization

UC-AD digestates energy valorization via direct fermentative processes seems unlikely. Differently than agricultural digestates, UC-AD digestates tend to present a low content of lignocellulosic fibers that could be post-treated to release carbohydrates. Using the LF as a nutrient source for fermentation or to harvest biomass is possible but it does not consist of valorizing the energy contained in the digestate's residual OM.

Thermal conversion processes represent a huge potential to produce biofuels (and far beyond) from UC-AD digestates, regardless the presence of impurities and contaminants. Thermal conversion can enhance AD thermal efficiency but, from an energetic point-of-view, no discussion is provided about the

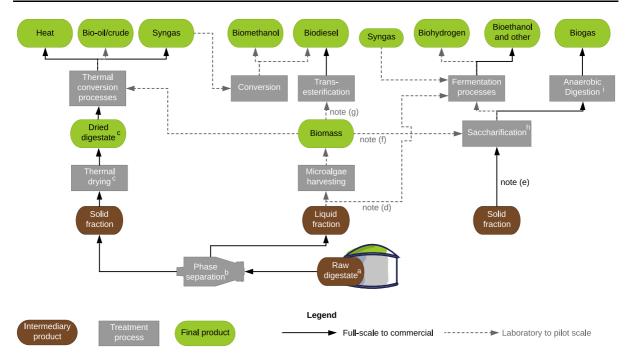


Fig. 3 Biofuels from digestate. Major process inputs and outputs may be omitted. a Usually comprises a storage step. b Some plants or research might directly treat whole digestates, which depends mostly on suspended solids and rheology to match downstream equipment requirements. c Thermal drying might include a pelletizing/granulation step downstream. d As

interest of including the AD step if the thermal conversion system is conceived from the beginning (Funke et al. 2013; Reza et al. 2016). Additionally, there is a lack of experience feedback when it comes to thermal conversion of digestates. Low development can be linked to the fact that thermal conversion processes require very high capital expenditures and an specific engineering expertise for operating machinery under high temperature and/or pressure.

# 5.3 Other industrial valorization possibilities

Little attention has been dedicated to industrial valorization of digestates beyond agriculture and energy. The following sections will describe few opportunities related in literature, along with some conceptual ideas.

# 5.3.1 Reuse water

Water scarcity for agriculture is a reality. In many regions, water canal systems are overexploited as well

the culture medium (source of nutrients and moisture). **e** If rich in residual fibers. **f** If rich in carbohydrates. **g** If rich in lipids. **h** Saccharification is referring to post-treatments aiming to release the carbohydrates from the structured organic matter. **i** Through a post-digester or simply recirculation

as aquifers. Simultaneously, there is an increasing water demand for industry and growing cities (Fischer et al. 2014).

Most of digestate liquid streams could be a source of water for irrigation purposes, but clean water is needed for irrigation due to limits on nutrients and pollutants. Moreover, traditional spraying irrigation techniques are not indicated for digestates due to the high ammoniacal nitrogen content. From digestates, irrigation water can be produced with reverse osmosis or condensation from evaporation processes, which were both previously discussed (Sect. 5.1.1).

In many cases, UC-AD are localized far from agriculture. Depending on transporting distances, in situ or industrial digestate water reuse can be a better option. Among the few large scale experiments in literature, Chiumenti et al. (2013b) observed 1.7 t/h of UF + RO permeate for 3.6 ton/h of digestates, which represented 48% of the digestate mass. The final permeate water had 1300 mg/L of DM and less than 100 mg/L of COD (single RO step). Adam et al. (2018) obtained 10–12% of final permeate after NF

followed by two consecutive RO. The permeates had about 2 mg/L of suspended solids and 27–32 mgO<sub>2</sub>/L of COD. As a quality reference, the Food and Agriculture Organization of the United Nations (FAO) recommends less than 50 mg/L of suspended solids to avoid clogging of drip irrigation systems (Ayers et al. 1985). For unrestricted use of irrigation water, the FAO recommends water salinity lower than 0.7 dS/m (Ayers et al. 1985), which is respected by the permeate water from Chiumenti et al. but not from Adam et al. For "class A" reuse water (all food crops), the new European Legislation proposal on water reuse specifies limits of 10 mg/L of BOD<sub>5</sub> and total suspended solids, among a few other parameters (European Comission 2018).

Fortunately, water is not sufficiently scarce to have an important added value. If it was the case, dilution of AD inputs would also tend to be avoided thus less water would be recovered from digestates. Much likely, the feasibility of the reuse water processes relies on the sales of concentration fractions and/or cost reduction compared to other options.

#### 5.3.2 Animal feed products

In most of the countries, regulation framework would not allow the commercialization of waste-derived products for animal feed, especially outside the agricultural sphere such as digestates from mixedsource feedstock. The following possibilities are strongly hypothetical, depending on legislation evolution along with research programs to confirm their innocuity.

5.3.2.1 Harvested biomass for animal feeding Protein-rich microalgae and macrophyte (discussed in Sect. 5.1.2.1) are often indicated as potential source for primary animal feed. They are often reported as high-grade nutritional food for livestock and aquaculture (Uggetti et al. 2014; Pascual 2016). No information on marketing prices could be found as no industrial production of such biomasses from digestate is available. As a reference, high protein animal feed (50–70% protein) can be found in global international wholesale websites ranging from 400 up to 1300 USD/t.

Little attention seems to be dedicated to the treatment of digestate with mushrooms composting. According to Stoknes et al. (2013), common edible

*Agaricus bisporus* is conventionally produced in a mixtures of compost, straw, horse manure, chicken manure and gypsum. They were able to substitute manure from the mixture with a dewatered digestate from municipal source-separated food waste, without affecting mushroom yield. To achieve thermogenesis in early process stages, they had to add only 30 g/kg (dry basis) of chicken manure with as much as 470 g/ kg of digestate.

5.3.2.2 Earthworms from vermicomposting for animal feeding Al Seadi et al. (2013) indicated that earthworms from digestate vermicomposting (Sect. 5.1.2.5) can be used to feed chickens but no further information was provided. Indeed, earthworms are known from decades for its protein content (60–70%) and its application for organic waste treatment and animal feed (Edwards 1985).

5.3.2.3 Feed additives for livestock Biochar is already used as a feed supplement for livestock. Positive effects include rapid decrease on diarrhea incidence, improvement of feed intake, reduction of allergies, among other benefits. However, no specific information is available on the biomass origin of these biochar (Gerlach and Schmidt 2012; Schmidt 2012).

Humic substances from leonardite are widely commercialized as feed additive (HUMINTECH 2015a). In the academy, Montoneri et al. (2013) tested as feed additives different "soluble biobased products" extracted from several streams of an UC-AD treating biowaste (including digestate) (as discussed in Sect. 5.1.2.7). The products performed similarly to fossil humic substances for the reduction of ammonia production. The tests were performed under simulated cecal fermentation and in vitro intestinal fermentation.

# 5.3.3 Biopesticides

The evolution of disease-causing organisms towards plant resistance genes and defense agents is one of the greatest hazard to the sustainability of modern agriculture and food safety (Fischer et al. 2014). Along with that, concerns to improve environmental safety, food quality and human health tends to boost the demand for eco-friendly alternatives to the conventional chemical pesticides.

*Bacillus thuringiensis,* referred usually as "Bt", are the most usual bacteria do produce biopesticides for

insect control. Bt-based products represent about 75% of the global biopesticide use (Olson 2015). They are currently commercialized by various biotech companies under different names and configurations (Rosas-Garcia 2009). By 2013, the biopesticide market represented a 3 billion USD industry but mere 5% of the pesticides market. Biopesticide are expected to grow to above 4.5 billion USD by 2023. Around 2050, the use of biopesticides is expected to overpass the use of synthetic pesticides (Olson 2015).

Support media is an important limitation for the cost-effectiveness to produce Bt-biopesticides. They were estimated to contribute up to 25% of the total production cost (Brar et al. 2007). Digestates can be used as nutrient source and growth support for producing Bt-products. Certainly one of the pioneers of this possibility were Cerda et al. (2019), as part of the DECISIVE project (H2020-EU.3.5.4, ID 689229). They have proofed the concept by successfully producing biopesticides through solid-state fermentation with a biowaste digestate from a UC-AD plant. The conditions, though, remain distant from full-scale implementation.

It is important to mentation that the regulatory framework is favorable towards biopesticides in the USA and in the EU (Villaverde et al. 2014; U.S. EPA 2019).

#### 5.3.4 Adsorbents: biochar

For almost every activated carbon (AC) application, digestate biochars can be and has been assessed as an alternative. It implies in a long list of potential biochar industrial applicability (Schmidt 2012). As previously stated, digestate biochars present a relatively high specific surface (up to 470 m<sup>2</sup>/g BET surface), high pore volume (0.06 to 0.55 cm<sup>3</sup>/g) (Inyang et al. 2012; Stefaniuk and Oleszczuk 2015; Opatokun et al. 2016; Wongrod et al. 2018; Jiang et al. 2018; Liu et al. 2020) and polar functional groups conferring to it adsorbent properties (Jiang et al. 2018).

As AC surface is usually above  $1000 \text{ m}^2/\text{g}$  (Tadda et al. 2016), biochar performance could be expected to be poorer than AC, if the biochar is not activated before or after pyrolysis through physical and chemical treatments (Sizmur et al. 2017). However, in comparison to carbonaceous materials such as wood, digestate biochar tend to present much higher N and O content and much more polar functional groups on its

surface. For this reason, depending on the adsorbate, biochar can present better adsorption results than AC despite its smaller surface. Moreover, some studies indicate the advantage of using digestate instead of raw substrate for pyrolysis. Yao et al. (2011) compared the adsorption of phosphates with biochar from sugar beet railing and its digestate to a commercial AC. Among the three, the digestate biochar had the best phosphate removal efficiency (73% compared to almost 0 for the rest). Moreover, production of biochar from the digestate was significantly higher (45.5% compared to 36.3%), with similar bio-oil production (12.5 and 10.9%) and a greater specific surface.

According to Schmidt (2012), the high prices of biochar previously discussed are due to its valuable application in animal farming. The author states that around 90% of the biochar produced in Europe (mostly from wood biomass) is used either as silage agent (no official reference), in animal litter (adsorption of ammonia), animal feed additive or effluent treatment (composting).

# 5.3.5 Engineered materials

5.3.5.1 Bioplastics After hydrolysis and extraction of complex soluble OM from a UC-AD digestate (as discussed in Sect. 5.1.2.7), Franzoso et al. (2016) investigated the production of poly(vinyl alcohol-co-ethylene). With up to 10% digestate soluble OM, the resulting blend presented lower melt viscosity and similar or better mechanical properties. To produce bioplastics from UC-AD digestates, this option seems more feasible than the chain-elongation pathway after fermentation. This is due to the same reason of the unlike production of bioethanol: lack of residual biodegradable OM (intrinsic to digestates) and low residual fiber content (if low fibrous inputs).

5.3.5.2 Civil construction Sewage sludge (dried or as ashes) has been investigated for integration with civil construction for several decades (Tay 1987). Sludge incineration ashes can be integrated in mainly two manners to civil construction materials: (i) as a constituent of sintered materials such as bricks, tiles and pavers; (ii) as part of the Portland cement manufacturing either as an additive to cement composition or included as clinker material in furnaces to optimize cement mineral composition. The main limitation is the content of P that can be include undesirable cement characteristics (Donatello and Cheeseman 2013). No specific studies could be found on UC-AD digestates.

# 5.3.6 Biosurfactants

Biosurfactants are traditionally defined as amphiphilic organic compounds that can be naturally produced by plants and microorganisms (Vijayakuma and Saravanan 2015). Due to higher biodegradability and less toxicity, they are an emerging alternative to chemical surfactants. Indeed, most of the surfactants in the market are produced by the petrochemical industry. In 2011, global biosurfactants market worthen around 1.7 billion USD (Reis et al. 2013). Surfactants have a wide range of industrial applications including food industry, pharmaceutical, cosmetics, textile and pollution remediation. Biosurfactants are usually classified as glycolipids and lipopeptides and produced through various aerobic microorganisms consuming mainly carbohydrates and lipids. Their production cost is about 3 to 10 times higher than that of chemical surfactants (Reis et al. 2013).

Cerda et al. (2019) explored the production of sophorolipids (a glycolipid) from a biowaste digestate from a UC-AD by using *Starmella bombicola* (a yeast) as inoculum under aerated conditions. However, two major bottlenecks were identified: (i) optimal fermentation needed a pH around 3.5 but digestate had an important buffering capacity for acidification and (ii) the yeast used in the study requires initial sugar concentration above 100 g/L, which is far from digestate composition as they are consumed during AD.

Apparently neglected by biosurfactant reviews, a more practical approach has been extensively investigated: the application of humic-like substances (HLS) extracted from organic residues as biosurfactants (as discussed in Sect. 5.1.2.7). Extracted from UC-AD digestate, composts, sewage sludge or OFMSW, different uses for HLS-biosurfactants were demonstrated, including: heavy oil removal (Baxter et al. 2014), soil remediation from aromatic hydrocarbons (Conte et al. 2005; Montoneri et al. 2009, 2014), textile dyeing (Montoneri et al. 2009; Savarino et al. 2009), emulsions (Vargas et al. 2014), among other (Montoneri et al. 2011). Many of these publications seems to trace back to researchers from the University of Turin (Italy) and more precisely to the BiochemEnergy project (Montoneri et al. 2011). Indeed, these research teams have been investigating this subject from over a decade (Quagliotto et al. 2006). The BiochemEnergy project estimated a product value ranging from 1 to 100 USD/kg which is high compared to an operational cost estimated from 0.10 to 0.50 USD/kg.

Fossil-based humic substances are industrially used due to its surface-active properties, for example, as drilling fluid (HUMINTECH 2015b). However, no cases of commercial application of HLS extracted from digestates were found. Industrial biosurfactants are expected to change water surface tension from 72 down to 35 mN/m (Akbari et al. 2018). The best performing HLS-biosurfactants from organic residues reduced surface tension to as low as 30–36 mN/m (Quagliotto et al. 2006; Savarino et al. 2009).

With proven applicability, favorable economics and increasing environmental awareness, HLS from UC-AD digestates as biosurfactants are a promising value-added product.

#### 5.3.7 Flame retardants: struvite or ammonium sulfate

The currently conventional wood treatment for fire prevention relies on chemicals that contain halogenated compounds, which are harmful to the human health and the environment (Guo et al. 2019).

Wastewater recovered struvite (discussed in Sect. 5.1.1.6) was demonstrated to be an alternative eco-friendly flame retardant to textile and wood (Yetilmezsoy et al. 2018; Guo et al. 2019). Guo et al. (2019) indicated that the main mechanism of struvite wood fire protection was the enhancement of char formation due to two mechanisms: (i) heat absorption with the releasing of a non-flammable gas (preventing temperature increase) and (ii) the amorphous MgHPO<sub>4</sub> resulting from struvite thermal decomposition promotes a gas barrier and increase wood structure preventing wood devolatilization (Guo et al. 2019). In both studies, however, the performance was not compared to traditional commercial products.

Ammonium sulfate (discussed in Sect. 5.1.1.5) is another common digestate derived product. In the 1970s it gained interest for its use as flame retardant for wood (George and Susott 1971) and later for organic mulch protection in fire-risk regions such as California (Hickman and Perry 1996). No recent publication is available on this subject.

# 5.3.8 Ammonium nitrate for the manufacturing of explosives

Ammonium nitrate can be obtained from digestates after  $NH_3$  stripping and scrubbing with nitric acid instead of the usual sulfuric acid (producing ammonium sulfate) (Sect. 5.1.1.5). Ammonium nitrate is currently the most important constituent for the manufacturing of modern explosives. However, no study could be identified to assess the compatibility and feasibility of producing prilled, porous and pure ammonium nitrate from digestates while aiming the manufacturing of industrial explosives (Meyer et al. 2007).

# 5.3.9 Functional landfill cover layer

If there is no better option for the destination of a digestate or a partial stream, they can be used as engineered cover layers to promote nutrient removal from leachates in landfills. For example, Peng et al. (2018) demonstrated this concept at laboratory scale with an OFMSW digestate. They succeeded on removing leachate nitrates, which was associated mainly to denitrification, but also adsorption. This can be a useful destination to environmental services providers operating both landfills and UC-AD plants.

# 5.3.10 Conclusion of industrial valorization possibilities

Several options other than agriculture and energy could be identified to valorize UC-AD digestates. From these options, effectively, it seems that only biochar can be considered as a full-scale developed product with industrial applications. However, no case of UC-AD digestates biochar marketing could be identified. After that, another promising strategy is the use of humic-like substances as biosurfactants, which is still under development or demonstration. There are indeed several opportunities for industrial synergy if the AD plant is localized nearby an industrial area, but most of them still require extensive research and development.

# 6 Conclusion

Technically, there are several possibilities to reach value-added products from UC-AD digestates. Agriculture is the first destination to be considered due to (i) a more advanced technological state-of-the-art, (ii) the positive effects of AD on digestate fertilizing value and (iii) for closing nutrient cycles and promoting carbon sequestration in soil. Aiming agriculture, several processes are already available either to concentrate desirable characteristics of digestates, enhance OM stability or to produce pure and reformulated fertilizers. However, most of the processes still have major drawbacks and great marge for optimization. Globally, reflecting the AD industrial sector, most of the research literature is oriented to WWTP (sludge) digestates or agricultural digestates. Many specific issues of UC-AD digestates from other organic residues are poorly explored. Adding to that, OM valorization seems much less developed and explored than nutrient recovery. The only full-scale OM transformation techniques seem to be composting (fully commercial) or thermal conversion (few installations, mostly demonstrators). Despite the full development of the agricultural destination for digestate byproducts, there is a wide range of urgent subjects for R&D, including process optimization, environmental safety/performance of final products (e.g., pathogens, emerging pollutants, nutrient leaching, atmospheric emissions and life cycle assessments), assessment and enhancement of product performance regarding crop needs (e.g., nutrient balance, nutrient accessibility, OM stability) and market research (e.g. market size and final consumer needs).

Especially for UC-AD designers and operators, when the agricultural destination is not possible, thermal conversion processes are a technically advanced option to valorize the solids through the production of biofuels and/or biochar. In the near future, they could be used to start a whole biorefinery system. Similarly, biomass harvesting processes such as microalgae are rapidly upscaling, enabling to valorize the nutrients on the digestate liquid phase while capturing atmospheric  $CO_2$  and producing renewable biomass for biorefinery. In both cases, the main underlying mechanisms seem to be clearly understood, but R&D programs are still necessary to overcome major bottlenecks and provide more industrial pilot demonstration and validation.

Several other products were successfully obtained from UC-AD digestates such as biopesticides, biosurfactants and composite materials. Many of these approaches seem promising, but they seem limited to a few research groups working under bench to pilot scale. They need to be further investigated and upscaled from process to product application.

If UC-AD digestates could be effectively valorized into value-added products, would it be possible to drive AD inputs and/or parameters for enhancing digestate value (reverse engineering)? What balance to be found with revenues from gate fees and/or optimization for biogas production? Few researchers seem to address these questions.

As regulations and public acceptation are constantly evolving, perhaps one of the greatest recalcitrant barriers can be the price competition with traditional fossil/ore-based products. Many of them such as crude oil, coal, N and P fertilizers are internationally traded as commodities under extremely volatile prices. Adding to that, traditional industrial products have been optimized for application performance and cost reduction. With increasing environmental awareness, a consumer-driven market change towards upcycled and eco-friendly products is fundamental and might enhance digestate intrinsic value, thus UC-AD economic feasibility and environmental performance.

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#### Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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